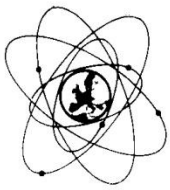
CRITICALITY CONTROL
IN CHEMICAL AND METALLURGICAL PLANT

CONTROLE DE LA CRITICALITE
DANS LES INSTALLATIONS CHIMIQUES ET MÉTALLURGIQUES



KARLSRUHE SYMPOSIUM

1961

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT
EUROPEAN NUCLEAR ENERGY AGENCY

ORGANISATION DE COOPÉRATION ET DE DÉVELOPPEMENT ÉCONOMIQUES
AGENCE EUROPÉENNE POUR L'ÉNERGIE NUCLÉAIRE

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L'Organisation de Coopération et de Développement Économiques a été instituée par une Convention signée le 14 décembre 1960, à Paris, par les Membres de l'Organisation Européenne de Coopération Économique, ainsi que par le Canada et les États-Unis. Aux termes de cette Convention, l'O.C.D.E. a pour objectif de promouvoir des politiques visant :

- *à réaliser la plus forte expansion possible de l'économie et de l'emploi et une progression du niveau de vie dans les pays Membres, tout en maintenant la stabilité financière, et à contribuer ainsi au développement de l'économie mondiale ;*
- *à contribuer à une saine expansion économique dans les pays Membres, ainsi que non membres, en voie de développement économique ;*
- *à contribuer à l'expansion du commerce mondial sur une base multilatérale et non discriminatoire, conformément aux obligations internationales.*

La personnalité juridique que possédait l'Organisation Européenne de Coopération Économique se continue dans l'O.C.D.E., dont la création a pris effet le 30 septembre 1961.

Les Membres de l'O.C.D.E. sont : la République fédérale d'Allemagne, l'Autriche, la Belgique, le Canada, le Danemark, l'Espagne, les États-Unis, la France, la Grèce, l'Irlande, l'Islande, l'Italie, le Luxembourg, la Norvège, les Pays-Bas, le Portugal, le Royaume-Uni, la Suède, la Suisse, la Turquie.

L'Agence Européenne pour l'Énergie Nucléaire (ENEA) a été créée en décembre 1957 dans le cadre de l'O.E.C.E. afin de développer la collaboration atomique entre l'ensemble des pays de l'Europe occidentale. Un Comité de Direction de l'Énergie Nucléaire comprenant des représentants de tous les pays Membres et Associés est l'organe directeur de l'ENEA.

L'Agence a pour fonctions de (a) *créer des entreprises communes ; trois entreprises ont été constituées : la Société Eurochemic pour le retraitement des combustibles irradiés (à Mol, Belgique), le Projet de réacteur à eau lourde bouillante de Halden (Norvège), le Projet Dragon de réacteur à haute température refroidi par gaz (à Winfrith, Royaume-Uni) ;* (b) *harmoniser les programmes de recherches en facilitant la coopération entre les pays Membres dans les domaines scientifiques et techniques, les échanges de personnel et d'informations ;* (c) *élaborer des règles uniformes en matière nucléaire pour l'ensemble de l'Europe, notamment dans les domaines de la santé et de la sécurité, du transport des matières radioactives, de la responsabilité civile et des assurances ;* (d) *étudier les aspects économiques de l'énergie nucléaire en examinant périodiquement les programmes nationaux, la place de l'énergie nucléaire dans la balance énergétique de l'Europe et le marché des combustibles, matériaux et équipements nucléaires.*

L'ENEA travaille en liaison avec les autres organisations internationales intéressées, particulièrement avec l'Euratom et l'Agence Internationale de l'Énergie Atomique.

The Organisation for Economic Co-operation and Development was set up under a Convention signed in Paris on 14th December 1960 by the Member countries of the Organisation for European Economic Co-operation and by Canada and the United States. This Convention provides that the O.E.C.D. shall promote policies designed:

- *to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;*
- *to contribute to sound economic expansion in Member as well as non-Member countries in the process of economic development;*
- *to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.*

The legal personality possessed by the Organisation for European Economic Co-operation continues in the O.E.C.D., which came into being on 30th September 1961.

The Members of O.E.C.D. are: Austria, Belgium, Canada, Denmark, France, the Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States.

The European Nuclear Energy Agency (ENEA) was set up in December 1957 as part of the O.E.E.C. to develop nuclear collaboration between the countries of Western Europe. A Steering Committee for Nuclear Energy, composed of representatives from all Member and Associated countries, is the controlling body of ENEA.

The work of the Agency comprises (a) *creation of joint undertakings, three of which — the Eurochemic Company for reprocessing irradiated fuels at Mol in Belgium, the Halden boiling heavy water reactor project in Norway, and the Dragon high-temperature gas-cooled reactor project at Winfrith in the United Kingdom — are already in operation;* (b) *the harmonisation of research programmes, encouraging scientific and technical co-operation between Member countries and the exchange of information and personnel;* (c) *the establishment of uniform atomic regulations for Europe, especially in the fields of health and safety, liability and insurance in case of accident, and the transport of radioactive materials;* (d) *the study of the economic aspects of nuclear energy, by a regular examination of national programmes, of the place of nuclear energy in Europe's overall energy balance sheet, and of the markets for nuclear fuels, materials and equipment.*

ENEA works in liaison with the other international organisations concerned, especially Euratom and the International Atomic Energy Agency.

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CONTENTS

TABLE DES MATIÈRES

OPENING ADDRESS 11

Allocution

Prof. Dr. Werner HEISENBERG, Director of the Max-Planck Institute for Physics and Astrophysics

Session I

FUNDAMENTALS

PRINCIPES FONDAMENTAUX

Chairman: / *Président :* Mr. J. L. SCHWENNESEN (United States)

FUNDAMENTALS OF CRITICALITY CONTROL, E. R. Woodcock (U.K.A.E.A.)..... 15

Fondement de l'étude de la criticalité

SURVEY OF THEORETICAL CALCULATION METHODS, R. T. Ackroyd and E. D. Pendlebury (U.K.A.E.A.)..... 47

Examen des méthodes de calcul théoriques

MÉTHODES THÉORIQUES UTILISÉES DANS LES CALCULS DE CRITICALITÉ, P. Benoist, R. Caizergues, C. Clouet d'Orval et B. Duchemin (C.E.A., Saclay - France) 119

Theoretical Methods used in Criticality Calculations

Session II

FUNDAMENTALS (continued)

PRINCIPES FONDAMENTAUX (suite)

Chairman : / *Président :* M. H. DOPCHIE (Belgique)

A REVIEW OF CRITICALITY DATA OBTAINED BY EXPERIMENTAL METHODS, D. Callihan (Oak Ridge - U.S.)..... 139

Examen des données relatives à la criticalité obtenues au moyen de méthodes expérimentales

CORRELATIONS OF EXPERIMENTAL AND THEORETICAL CRITICAL DATA, Hugh C. Paxton (Los Alamos - U.S.) 173

Corrélation entre les données expérimentales et théoriques en matière de criticalité

Session III

APPLICATION OF CRITICALITY CONTROL PRINCIPLES TO PLANT DESIGN

APPLICATION DES PRINCIPES DE CONTROLE DE LA CRITICALITÉ DANS LA CONCEPTION DES INSTALLATIONS

Chairman :
Président : Prof. M. ZIFFERERO (Italie)

SAFE-BY-SHAPE CHEMICAL PLANTS, K. M. Hill and H. E. Parker (U.K.A.E.A.)... 209
Contrôle de la géométrie dans une usine de chimie

CONTROLE DE LA CONCENTRATION ET APPLICATIONS CORRESPONDANTES, J. Gunther (C.E.A. - France)..... 233
Concentration Control and its Applications

DISCUSSION ON PROGRAMMES FOR CRITICALITY RESEARCH AND ECONOMIC ASPECTS. 251
Discussion sur les programmes de recherches sur la criticalité et les aspects économiques

Session IV

APPLICATION OF CRITICALITY CONTROL PRINCIPLES TO PLANT DESIGN (continued)

APPLICATION DES PRINCIPES DE CONTROLE DE LA CRITICALITÉ DANS LA CONCEPTION DES INSTALLATIONS (suite)

Chairman :
Président : Prof. A. F. ORLICEK (Austria)

MASS (BATCH) CONTROL AND ITS APPLICATIONS, A. J. Mallett (Oak Ridge - U.S.).. 263
Contrôle de la masse (du chargement) et applications correspondantes

HOMOGENEOUS AND HETEROGENEOUS POISONING AS POTENTIAL METHODS FOR CONTROLLING CRITICALITY, J. W. Wachter (Oak Ridge - U.S.)..... 289
Possibilités de contrôle de la criticalité par empoisonnement homogène et hétérogène

ÉTUDE D'UNE MÉTHODE D'EMPOISONNEMENT HÉTÉROGÈNE ET DE SON APPLICATION PRATIQUE, R. Caizergues (C.E.A., Saclay - France) 315
Study of a Heterogeneous Poisoning Method and its Application

Session V

CRITICALITY CONTROL IN THE FABRICATION OF FUEL ELEMENTS

CONTROLE DE LA CRITICALITÉ DANS LA FABRICATION DES ÉLÉMENTS COMBUSTIBLES

Chairman :
Président : Mr. H. HOWELLS (United Kingdom)

A TYPICAL MANUFACTURING PLANT FOR ENRICHED UO_2 FUEL ELEMENTS, J. W. Wachter and W. T. Mee (Oak Ridge - U.S.)..... 345
Usine type de fabrication d'éléments combustibles en UO_2 enrichi

NUCLEAR SAFETY IN CHEMICAL AND METALLURGICAL PROCESSING OF PLUTONIUM, E. D. Clayton (Hanford - U.S.)..... 367
Mesures de sécurité applicables au traitement chimique et métallurgique du plutonium

Session VI

STORAGE AND TRANSPORT PROBLEMS

PROBLÈMES DE STOCKAGE ET DE TRANSPORT

Chairman : *Président :* Mr. A. LARSSON (Sweden)

METHODS OF CALCULATION FOR INTERACTING ARRAYS, WITH SPECIAL REFERENCE TO STORAGE PROBLEMS, R. T. Ackroyd, F. R. Charlesworth and A. F. Thomas (U.K.A.E.A.) 395

Méthodes de calcul des arrangements avec interaction et applications aux problèmes de stockage

CRITICALITY ASPECTS OF THE TRANSPORT OF FISSILE MATERIALS - THE TECHNICAL BASIS OF THE PROPOSED I.A.E.A. REGULATIONS, R. F. Baker (International Atomic Energy Agency) 433

Problèmes de criticalité dans le transport des matières fissiles - les bases techniques de la réglementation proposée par l'Agence Internationale de l'Énergie Atomique

PRESENTATION OF " CRITICALITY ASPECTS OF THE TRANSPORT OF FISSILE MATERIALS ", M. C. Erginsoy (International Atomic Energy Agency) 441

Présentation des problèmes de criticalité dans le transport des matières fissiles

Session VII

OPERATIONAL PROBLEMS

PROBLÈMES D'EXPLOITATION

Chairman : *Président :* Dr. H. K. SAUER (Germany)

ASPECTS ADMINISTRATIFS DU CONTROLE DE LA CRITICALITÉ, P. Lecorché et R. Caizergues (C.E.A., Saclay - France) 459

Administrative Aspects of Criticality Control

ORGANISATION FOR OPERATIONAL CONTROL OF CRITICALITY, H. Howells and J. T. Daniels (U.K.A.E.A.) 465

Organisation du contrôle de la criticalité dans les conditions d'exploitation

ADMINISTRATIVE AND OPERATIONAL NUCLEAR SAFETY CONTROLS IN A CHEMICAL PROCESSING PLANT, C. L. Schuske (Rocky Flats Plant - U.S.) 477

Contrôle de sécurité dans une usine de traitement chimique - règlement administratif et conditions d'exploitation

A REVIEW OF CRITICALITY INCIDENTS, W. R. Stratton (Los Alamos - U.S.) 491

Examen des accidents dus à la criticalité

Session VIII

ORGANISATION OF EXPERIMENTAL CRITICALITY RESEARCH

ORGANISATION DE LA RECHERCHE EXPÉRIMENTALE EN MATIÈRE DE CRITICALITÉ

Chairman : *Président :* M. R. GALLEY (France)

ORGANISATION DE LA RECHERCHE EXPÉRIMENTALE EN MATIÈRE DE CRITICALITÉ, J. Bertrand, D. Breton, R. Caizergues, C. Clouet d'Orval, E. Deilgat, M. Molbert et P. Verriere (C.E.A., Saclay - France) 537

Organisation of Experimental Criticality Research

THE EQUIPMENT AND METHODS USED IN BRITISH CRITICALITY LABORATORIES, J. G. Walford and A. F. Thomas (U.K.A.E.A.) 558
Équipement et méthodes utilisés pour l'étude de la criticalité dans les laboratoires britanniques

EXPERIMENTS FOR CRITICALITY CONTROL, D. Callihan (Oak Ridge - U.S.) 589
Expériences sur le contrôle de la criticalité

ANNEX - *ANNEXE*

LIST OF PARTICIPANTS BY COUNTRY 617
Liste des participants par pays

The Opening Session of the Conference (left to right):

Dr. SCHNURR, Mr. SCHWENNESEN, Prof. Dr. HEISENBERG, Mr. HUET, Prof. NICOLAIDIS, His Exc. Dr. BALKE, His Exc. Dr. WERKMEISTER, Mr. KRIELE.

OPENING SESSION

under the Chairmanship of

Professor Dr. Werner HEISENBERG
Director of the Max-Planck Institute for Physics and Astrophysics

in the presence of

Professor Dr. Ing. Siegfried BALKE
Federal Minister of Atomic Energy and Water Power

Dr. Karl WERKMEISTER
Ambassador, Head of the Delegation of the German Federal Republic to the Organisation for European Economic Co-operation

Professor Léandre NICOLAIDIS
Chairman of the Steering Committee of the O.E.E.C. European Nuclear Energy Agency

M. Pierre HUET
Director of the O.E.E.C. European Nuclear Energy Agency

Dr. Walter SCHNURR
General Manager of the Kernreaktor Bau- und Betriebsgesellschaft m.b.H

Dr. Rudolf GREIFELD
General Manager of the Kernreaktor Bau- und Betriebsgesellschaft m.b.H

OPENING ADDRESS

by

Prof. Dr. Werner HEISENBERG

Director of the Max-Planck Institute for Physics and Astrophysics

May I welcome you on behalf of the physicists of this country and I would like to say first of all how much I appreciate the honour of acting as Honorary Chairman of the European Nuclear Energy Agency's Symposium on Criticality Control in Karlsruhe.

Twenty years ago, when the concept of criticality was discovered and discussed by the theoreticians and experimental physicists, the diffusion of neutrons in solid and liquid matter still belonged in the domain of fundamental physics. It was a somewhat new field and it was eagerly discussed. I remember how satisfactory it was to find that even rather unexpected theoretical calculations were supported by the ensuing experiments and how the experiments in turn clarified the theoretical aspects. In those days physicists, by comparing successive experimental and theoretical results, succeeded surprisingly quickly in getting a quite clear insight into the essential features of the criticality phenomenon. Now, as you all know, in the twenty years that have followed this first development, the problems have swiftly changed from questions of fundamental physics to problems of engineering, of design and construction, and all these varied detailed questions in connection with the control of criticality in reactors have been very thoroughly investigated and in the majority of cases solved.

But this problem of criticality does not exist only in the reactor field. It comes up wherever fissionable material is accumulated in large amounts, for example in reprocessing plants, in conversion plants for enriched uranium, or in factories where fuel elements are produced. We are here at a point where questions of great practical importance need further clarification. And it is at this point that the discussions of the present Symposium will come in, and will, as we hope, lead to further clarification and to some progress.

Recent problems are similar to the old ones in principle, but they are very different in practical detail. For instance, this interesting dependence of criticality on geometrical conditions is still the same subject which puzzled us so much in the early years; but now, of course, the geometry cannot be arbitrarily chosen, and one has to take into account other problems which have to be solved along with it. The absorption and reflection of neutrons has also to be studied under new conditions.

However, I should certainly not launch into a discussion of problems of which I understand so little, and in conclusion I should, perhaps, just

add a few words to express my great satisfaction that these problems are now discussed quite freely and openly in an international conference of experts, in an atmosphere of complete mutual confidence and common interest. I have absolutely no doubt that this is by far the best and quickest way to success and progress.

Session I

FUNDAMENTALS
PRINCIPES FONDAMENTAUX

Chairman - Président:

Mr. J. L. SCHWENNESEN
(United States)

FUNDAMENTALS OF CRITICALITY CONTROL

E. R. WOODCOCK
United Kingdom Atomic Energy Authority

INTRODUCTION

It would perhaps be appropriate in this first paper to be presented at a symposium on criticality control to attempt to lay the firm foundations of the subject on which subsequent contributors can build. To start *ab initio* from the basic nuclear physics would, however, only be repeating what is fairly well known and readily available. The core of the edifice on this foundation is largely mathematical in character, though leavened and restrained by the work of the experimental physicist. But it is the facade, consisting of the resulting principles, rules and recipes, which is probably of most interest in the present context, as this is the information needed by the practical man who is designing and operating plants for the processing of nuclear fuels.

The structure we are studying is by no means complete. Nevertheless, it is valuable at the present time to stand back and survey the stage that has been reached. It will not be possible to explore the whole building, but the facade will be covered in detail together with the public rooms that lie immediately behind. This introductory paper can then best help by summarizing the most important fundamental results that have appeared from the mathematical and experimental analyses and these can perhaps form the pegs on which the subsequent discussions can be hung.

Consequently we will discuss, with illustrations as far as possible, three main aspects of the criticality problem. In the first place it will be considered what physical or mathematical parameters are best to measure or calculate in order to determine how near a given assembly is to the critical state; secondly, the main factors affecting the criticality of a system will be outlined and the magnitude of their effects indicated; and thirdly, the basic principles which can be applied to ensure criticality safety will be briefly described.

CRITICALITY

When a reactor is loaded with sufficient nuclear fuel, so that a self-sustaining fission chain reaction takes place, it is said to have become critical. The chain reaction is accompanied by the evolution of heat and of penetrating radiation. To prevent the attainment of temperatures harmful to the materials of the reactor, the heat developed is removed by a gaseous or liquid coolant and may be made to do useful work in a power plant. The radiations are reflected back into the reactor core or largely absorbed by massive shields designed to protect the operating personnel from their dangerous biological effects.

In a reactor the approach to critical is carefully controlled and its state kept under careful surveillance by elaborate instrumentation so that criticality is never exceeded by more than a very small amount. If, however, a critical state were to be achieved or exceeded accidentally in a plant processing nuclear fuel, it would almost certainly occur in equipment and under conditions where no provision had been made for control or for the removal of the heat generated, and where there might not be shielding for the protection of the operators from the effects of radiation. Even were heat production so small that the plant suffered minor or no damage, the amount of radiation received by nearby personnel could easily reach lethal proportions, and since the safety of the men operating such plants is of primary importance, every precaution must be taken to prevent the occurrence of even minor criticality incidents. With larger amounts of heat produced there are not only greater radiation hazards, but there is the possibility of the release of radioactive and toxic materials over a wide area.

Measures of Criticality

Before discussing how to measure criticality it is first necessary to give a precise definition of the term. If a neutron is inserted into an assembly containing fissionable material it will in general initiate a chain reaction. This chain will terminate because eventually more neutrons are lost by capture within the assembly or escape from it than descendants are born in fission. It may not, however, terminate until the composition of the materials in the assembly has changed by neutron-induced transmutation of its atoms or physical changes have been effected by the energy released in the fissions. The phenomenon of criticality depends on these neutron chain reactions and since it is desirable to define it in relation to a specific assembly, we must state the definition in terms of a fictitious assembly obtained by adding the property that it does not undergo any changes as the result of a neutron chain reaction. In such a fictitious assembly it is either possible or it is impossible for a neutron chain reaction to continue indefinitely. If a continuing neutron chain reaction is impossible the assembly is subcritical and if it is possible the assembly is supercritical. A system which lies on the dividing line between subcritical and supercritical is said to be critical.

The equivalent definition expressed mathematically is that criticality is the condition that a certain equation — the neutron transport equation — has a non-trivial, time-independent solution.

Neither of these forms of definition is of much practical value as it stands. The fact that a continuing neutron chain reaction is possible does not mean that it will necessarily be always initiated within any finite interval of time, nor can a condition for the existence of a non-trivial solution of the neutron transport equation be expressed in a form convenient for evaluation.

It is desirable, therefore, to devise some parameter to measure the criticality of a system, i.e. how near it is to critical, which can be measured experimentally or calculated to test a given assembly. Such a criticality parameter should change smoothly as the system passes through critical and should be such that the critical state can be determined either by interpolation between values for subcritical and for supercritical systems, or by extrapolation from subcritical systems.

The Time Constant λ

It can be shown mathematically that there is an eigendistribution of neutrons possible in any finite system such that the number of neutrons in any given state changes only with time according to the law

$$N = N_0 e^{\lambda t}$$

and that this distribution will be steadily approached so long as N is large enough for the random variations in neutron behaviour to be negligible. This eigendistribution is approached closely in a very short time for a supercritical system but for a subcritical system can only be approached if N_0 is large enough.

The parameter λ is called the " time constant " and is positive, zero or negative according to whether the system is supercritical, critical or subcritical.

The time constant is a possible criticality parameter. Its behaviour as a system passes through critical is illustrated in Figure 1 which has been calculated for spheres containing mixtures of water and fissile element X, which approximates to U 235, surrounded by a reflector of water. From the calculation depicted by curve I, λ appears to have the properties desirable in a criticality parameter. However, curve II is a case where λ is only a good parameter for finding a critical size by extrapolation from supercritical values, or by interpolation if its values are known for near-critical systems and, although curve III allows extrapolation to critical from subcritical values, it is only good for interpolation over short ranges. Furthermore this curve does not give a good measure of the degree of criticality for well subcritical systems.

This rather odd behaviour of λ near critical arises from the changing relative importance of neutrons of high and low velocity. For a system such as I, where the slow neutrons are always of predominant importance, or for one which only contains fast neutrons, λ will behave smoothly and is quite a suitable parameter to use, but for other systems its properties are not ideal.

Figure 1 is a theoretical result. The use of experimental values of λ as a criticality parameter is made somewhat different by the phenomenon of delayed neutrons from fission. For calculation of critical size it is quite legitimate to ignore the time delay in the emission of these neutrons and this has been done for Figure 2. When the delayed neutrons are correctly allowed for, the result is as shown in Figure 2 which is for the same assembly as Curve I in Figure 1. The so-called prompt critical size is shown, which is the size which would be critical if the delayed neutrons were non-existent. The portion of the curve near critical on the scale of this Figure is almost indistinguishable from a horizontal straight line. In fact it varies smoothly enough to enable the critical size to be determined from measurements of λ close to critical. Also the small value of time constant for slightly supercritical systems makes it feasible to obtain the critical size experimentally by interpolation from measurements on slightly subcritical and slightly supercritical systems.

The Generation Constant μ

Although λ is a possible parameter to adopt as a measure of criticality, its shortcomings encourage the search for a more suitable quantity. These shortcomings arise to a large extent both from the time delay in the emission of some of the neutrons from a fission and from the wide range of the velo-

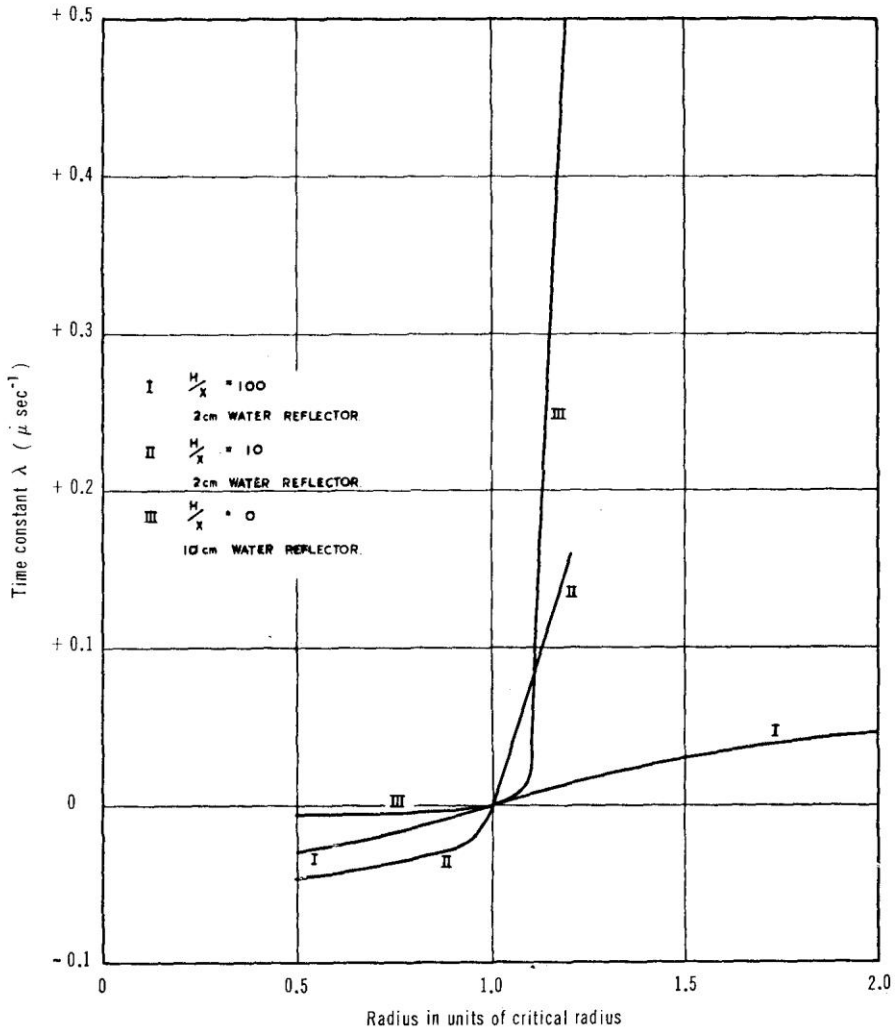


Figure 1. *Variation of time constant through critical, no delayed neutrons.*

cities of the neutrons which can be present in nearly all the systems of interest. That is to say, they are mainly associated with the use of time as an independent variable. The only way to avoid time being an independent variable in experimental work is to set up steady-state systems, but in a theoretical treatment there is no such restriction. An alternative approach is to use generation, sometimes called " collision generation ", as the variable. This is defined in such a way that if a neutron of generation g interacts with a nucleus, the emergent neutrons are of generation $(g + 1)$.

Exactly as for the time constant it can be shown that there is an eigendistribution of the neutrons which are in the same generation, such that the number of neutrons of generation g, in any given state, is given by

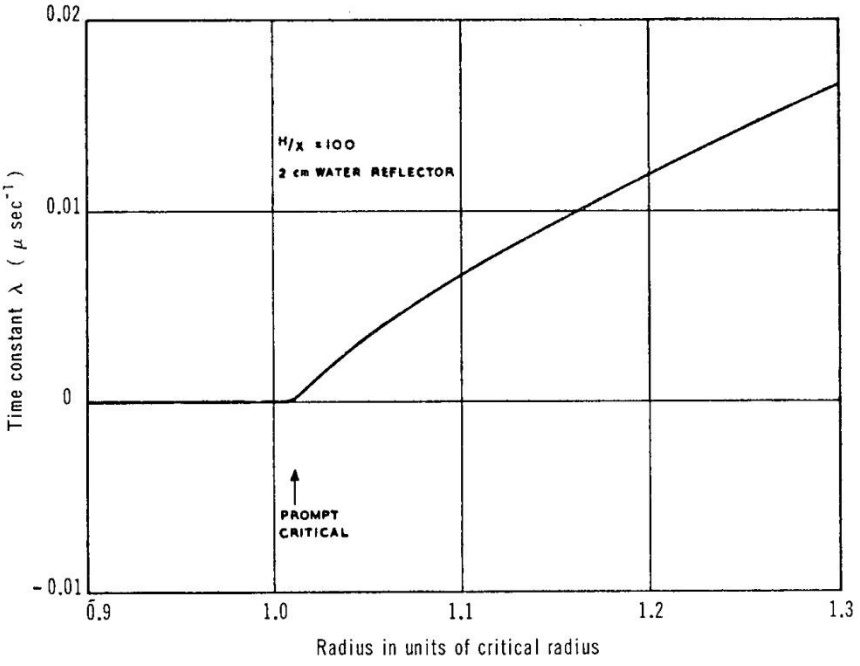


Figure 2. *Variation of time constant through critical allowing for delayed neutrons.*

$$N_g \propto e^{\mu g}$$

where μ can be called the " generation constant ". This is calculated, in practice, by calculating the ratio A of neutrons in generation g to neutrons in generation $(g + 1)$, i.e.

$$A = \frac{N_g}{N_{g+1}}$$

When an eigendistribution has been achieved, A is a constant independent of g. In fact

$$A = e^{-\mu}$$

Unless the system is a long way from critical, A is not greatly different from unity, so

$$\mu \approx 1 - A$$

and there is little difference between taking μ or $(1 - A)$ as the criticality parameter.

Figure 3 shows how μ varies through critical for the same three assemblies as were used to illustrate the behaviour of the time constant. It will be seen that this parameter varies more smoothly than the time constant and is therefore a preferable quantity to use.

The Multiplication Factor k

For the determination of the values of λ or μ it is necessary first to set up an eigendistribution of neutrons, and indeed this is true for most methods

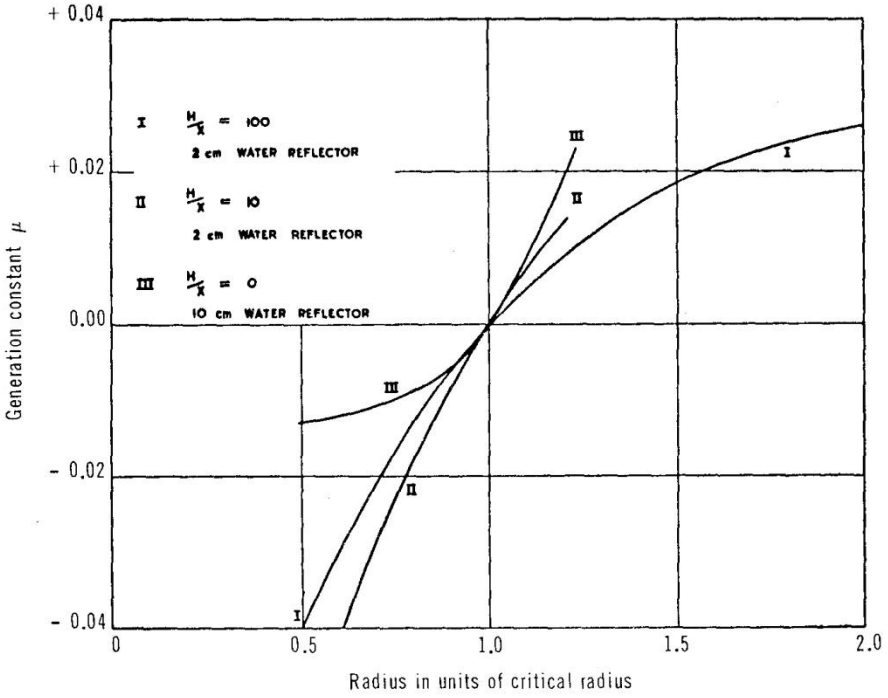


Figure 3. *Variation of generation constant through critical.*

of assessing criticality. The eigendistributions are not the same in each case, though for critical systems they are closely related. The logarithmic rate of change of the total number of neutrons in the system with time or with generation is sensitive to divergences from the true eigendistribution. This sensitivity can be reduced by giving more weight to neutrons in certain regions, but it is often not easily possible to reduce it as much as is desirable. The difficulty can be alleviated to some extent by focussing attention more strongly on the neutrons that are important in maintaining the chain reaction.

Since fission must be a link in the chain, one way of doing this is to introduce the concept of " fission generation ". The descendants of a neutron in fission generation f remain in the same generation until they cause a fission. Neutrons emerging from that fission are then in generation $(f+1)$. With this definition the same analysis as for collision generation can be followed. In this case, however, by virtue of long usage, the exponential form is not usually quoted but a multiplication factor k is defined by

$$k = \frac{N'_{f+1}}{N'_f}$$

where N'_f is the total number of neutrons of generation f emitted from fissions.

The variation of k as a system passes through critical is shown in Figure 4. Only one curve is drawn, as this is typical. As can be seen, k is an acceptable criticality parameter. It also has a useful interpretation, for a system with

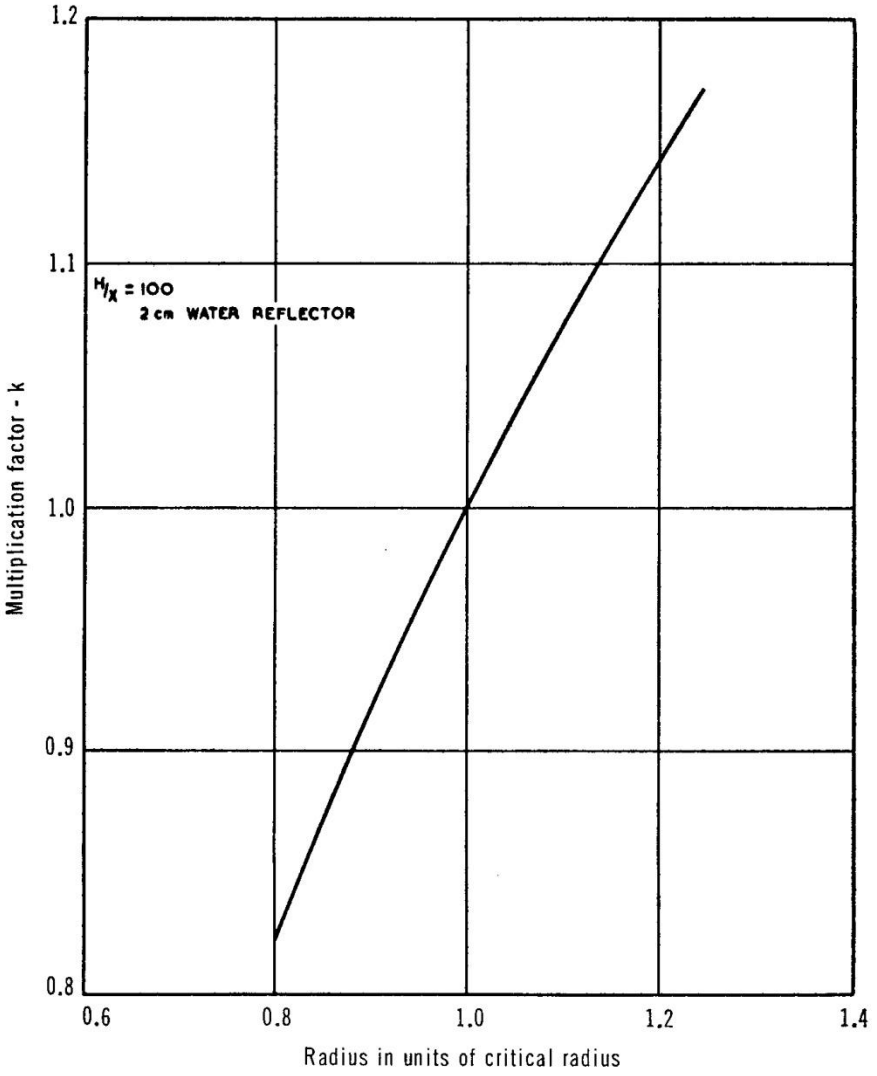


Figure 4. *Variation of multiplication factor through critical.*

criticality k would become critical if the number of neutrons from a fission were divided by k. Thus, when the fraction of delayed neutrons from a fission is known, it is immediately possible to find prompt critical which, as may be deduced from Figure 2, is the dividing line between supercritical systems that can readily be controlled and those for which control is difficult.

Reflection R *and Surface Multiplication* M

By emphasizing the fissions and using N'_f, a count of generation f neutrons as they emerge from fissions initiated by generation $(f-1)$ neutrons, the

sensitivity to the degree of settling into an eigendistribution has been much reduced. An alternative approach, which appears to be equally effective and also has certain other advantages, is to consider the balance of neutrons crossing a boundary Γ which divides the system into two regions.

If, when an eigendistribution has been achieved, a neutron crossing Γ in one direction gives rise, on the average, to X descendants that return to the boundary, and if a neutron crossing Γ in the opposite direction results, on the average, in Y descendants returning, then the quantity XY is a measure of criticality. The system is supercritical, critical or subcritical according to whether this quantity is greater than, equal to or less than unity.

It is convenient, in systems which consist of a core containing fissile material surrounded by a reflector, to choose Γ as the boundary between core and reflector. Then X becomes the reflection R, or albedo, of the reflector, and Y becomes the surface multiplication M of the core. This approach has been used for a wide variety of systems from which two related results have emerged. In the first place it is only necessary in order to evaluate M and R that the eigendistribution of neutrons be known at the boundary Γ and both these quantities have been found to be insensitive to small changes in this distribution. Secondly, the value of M is found to be predominantly a function of the core and to depend to a much less extent on the reflector, whereas the value of R is predominantly a function of the reflector.

As criticality parameters it is best to use R and 1/M, as these vary smoothly and nearly linearly as the system passes through critical. The critical state is then that for which $R = 1/M$. A typical example is given in Figure 5 which is for one of the systems used for the earlier illustrations.

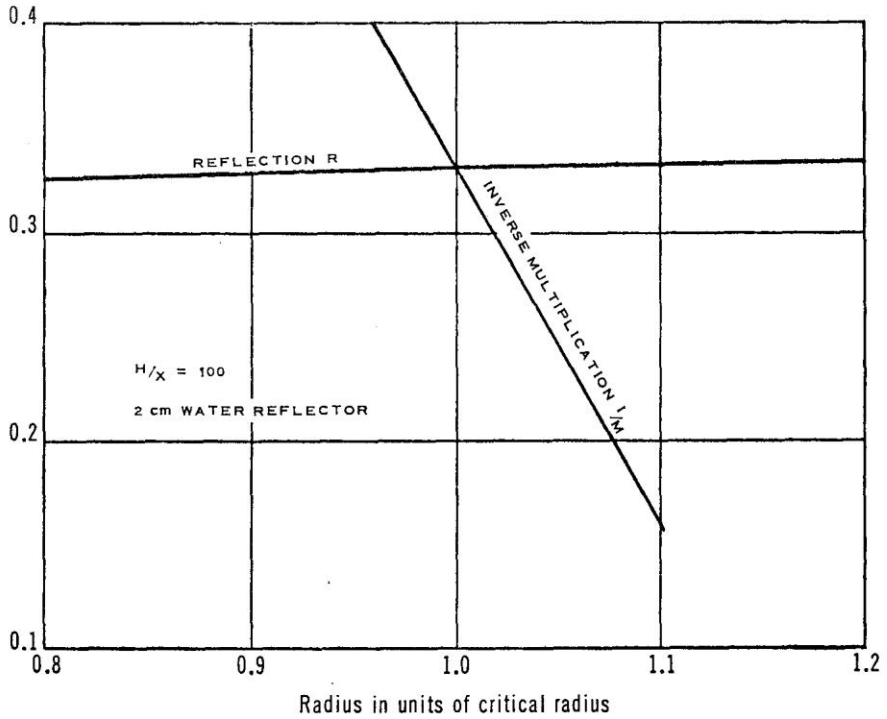


Figure 5. *Variation of surface multiplication M and reflection R through critical.*

Experimental Multiplications

The criticality parameters described in the last three sections are useful for the theoretical calculation of a critical size but are not easily adaptable to experimental measurement. However, the surface multiplication introduced above is an example of a more general concept of multiplication which is the basis of most experimental measurements of critical size.

Let us suppose that a source S of neutrons and a counter C to detect neutrons are placed inside or in the vicinity of an assembly containing fissile material, and that the counter reading when corrected for background is C_1. Suppose also that when the fissile material is removed, the corrected counter reading is C_0. Then the quantity

$$M_e = \frac{C_1}{C_0}$$

is a multiplication which measures the effect of the presence of the fissile material. Furthermore, when the system becomes critical M_e becomes infinite, so a graph of $1/M_e$ plotted against the variable dimension can be used to extrapolate to critical size. This is a crude outline of principle and there are numerous refinements necessary to devise an acceptable procedure. In particular it is desirable to continue measurements near to the critical size and to so position the source and counter that a reliable extrapolation can be made.

A particular case, by way of illustration, is to find the critical size of a system consisting of a spherical core surrounded by a reflector. For this experiment it is best to place the source at the centre of the core. The multiplication M_e is then given by the ratio

$$M_e = \frac{\text{rate at which neutrons cross core boundary outwards}}{\text{rate of emission of neutrons from source}}$$

If we define a central source multiplication M_c as the value of this M_e for an unreflected core, we have the relation

$$M_e = \frac{M_c}{1 - RM}$$

where R and M are the reflection and surface multiplication described in the preceding section.

The behaviour of M_e in a typical case for an unmoderated spherical core is illustrated in Figure 6 [1] which demonstrates that this experimental approach is a reliable method of measuring critical size for these spherical systems.

Properties of R and M

The use of reflection and surface multiplication as criticality parameters is not yet well documented in the literature, so it may be useful to include here some further discussion on their properties. The surface multiplication M of a core depends upon the core size, shape and material and also on the angular distribution and energy spectrum of the neutrons entering. The angular distribution is not an important factor in practice, since neutrons

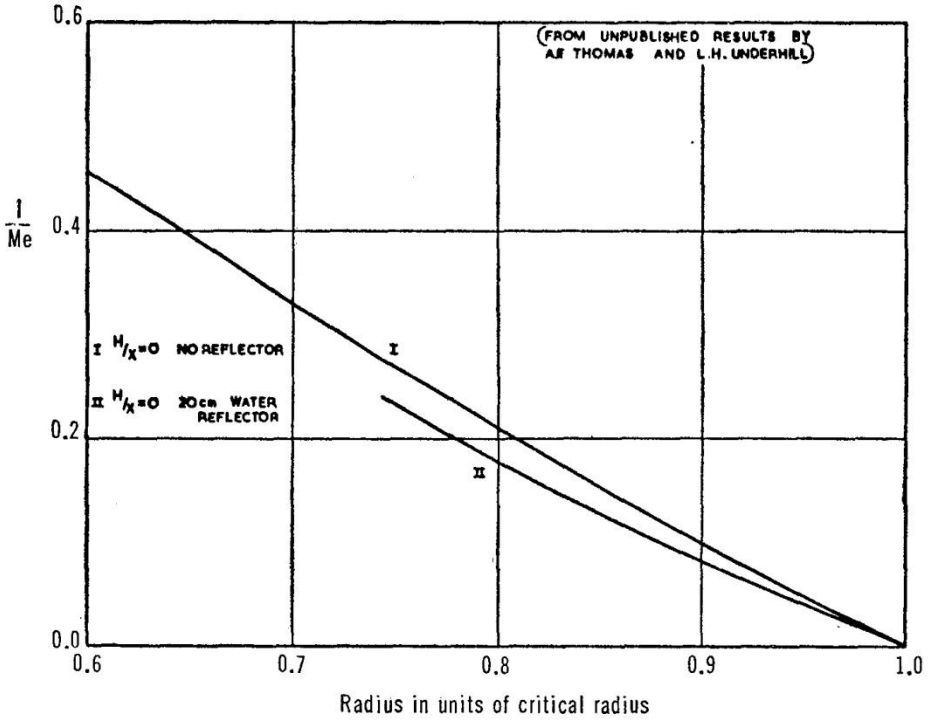


Figure 6. *Variation with size of the multiplication (Me) from a central source in a spherical system.*

returning from a reflector of any material are distributed in angle in substantially the same way. The incident neutron energy can be anywhere between that of thermal neutrons and that of newly-born fission neutrons. An extreme effect will occur when the core is of fissile metal, e.g. U 235 or Pu 239. Then a thermal neutron returning will be absorbed very close to the core boundary giving, on the average, about two neutrons of fission energy. One of these, on the average, will escape immediately and the other enter the core and be multiplied. So the multiplication of thermal neutrons is given by

$$\text{M(thermal)} = \text{M(fission)} + 1.$$

However, in such a case the neutrons reflected back to the core will always contain a high proportion of neutrons above thermal energies, so in practice this surface multiplication always lies in the range

$$\text{M(fission)} \leqslant \text{M} \leqslant \text{M(fission)} + 0.5.$$

Another extreme will occur if the core absorbs strongly neutrons which have been slowed down from their original fission energies. This will result in

$$\text{M} \leqslant \text{M(fission)}.$$

So in all cases an upper bound to M is M(fission) + 0.5 and very often M(fission) will be a lower bound. Hence the effect of the reflector on the value of M is very limited.

The size, shape and material of the core are the most important factors influencing M. The extreme values of 1/M are 1 for a core of infinitesimal size and 0 for a core that would be critical if unreflected. It appears from the results of a number of calculations that, for cores of the same material of geometrically similar shape, 1/M varies almost linearly with linear dimension between these limits. Some of this evidence is given in Figure 7 which is for spheres and infinite cylinders of a variety of materials, cadmium-plated and reflected by wood, so that the incoming neutrons included none of thermal energies. The individual points were calculated by a Monte Carlo method which inevitably leaves some statistical uncertainty, the amount of which is indicated by marking the standard deviation in each case.

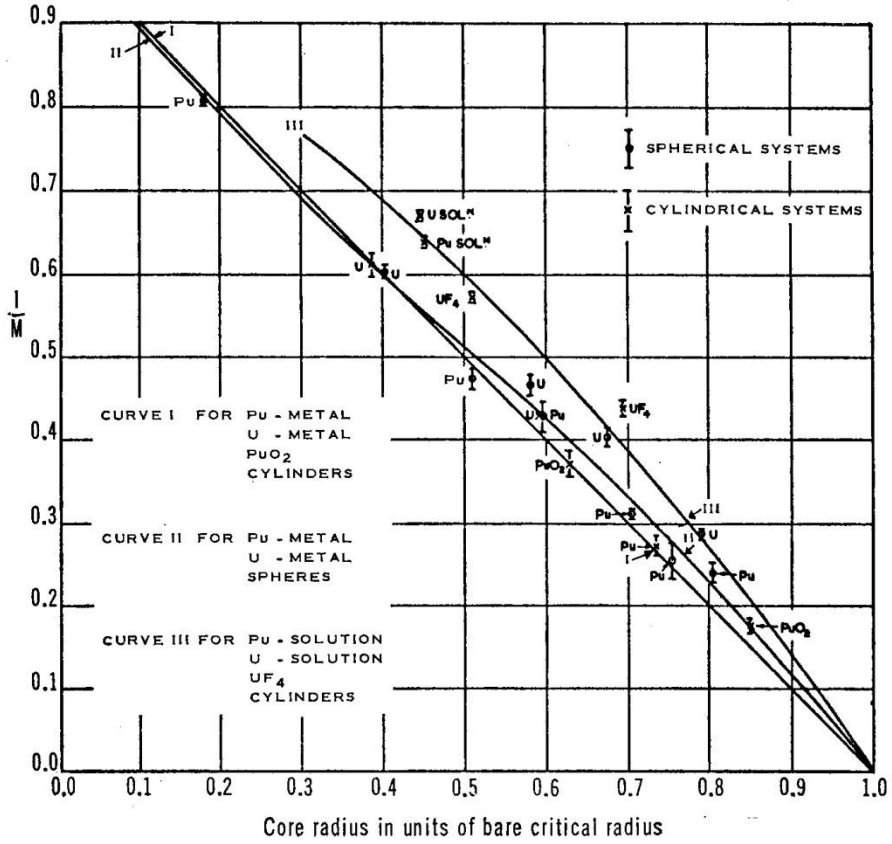


Figure 7. *Surface multiplication M.*

It follows, therefore, that not only is M a good criticality parameter but also that a reasonably good estimate of its value can be made from no more knowledge than the unreflected critical size of a core of the same material and geometrically similar shape.

The reflection R depends on the same factors that have been discussed in their influence on M, i.e. the angular distribution and energy spectrum of the incident neutrons and the material, size and shape of the reflector.

Figure 8 shows some typical values which arose from the same calculations that give Figure 7. The full lines are for very thick reflectors. The dashed line indicates the result of limiting the wood thickness so that (outer wood radius) $\times$ (wood density) $= 8.3$ g/cm^2, and if continued the dashed line would reach $R = 0$ at this point. As for multiplication, the angular distribution of the incident neutrons can be ignored. The effect of energy spectrum in one particular case is indicated in the Figure.

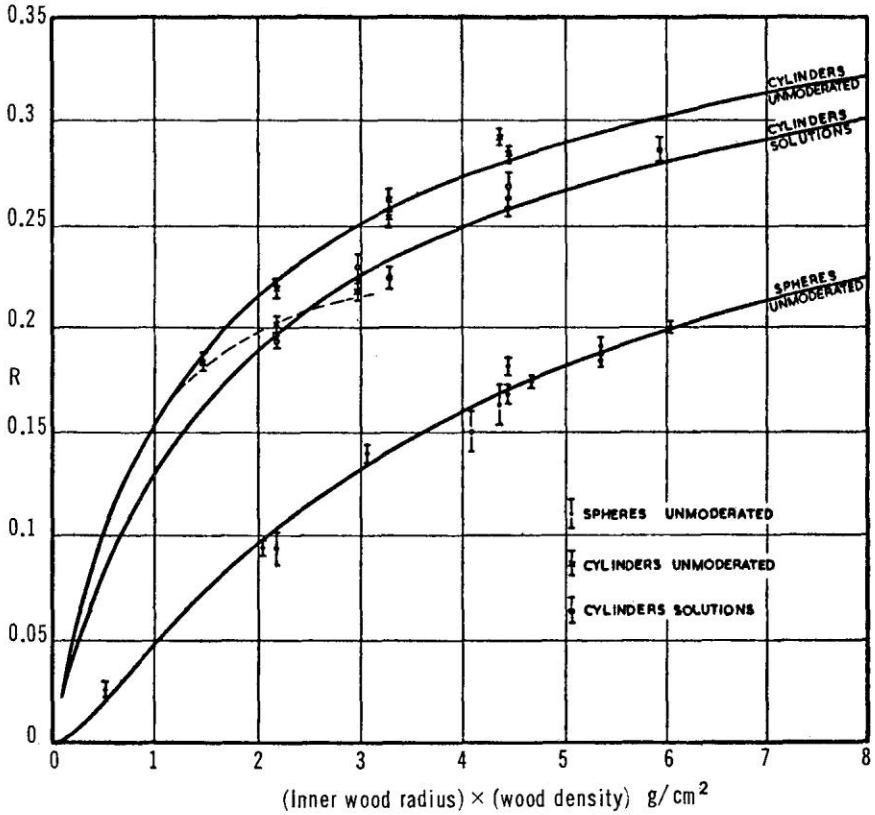


Figure 8. *Reflection R.*

For a reflector of fixed thickness, which may be infinite, the value of R increases monotonically as the radius of the inner surface increases (i.e. as the core size increases) reaching an asymptotic value which is the reflection from an infinite slab R_∞. This value depends on the reflector material and thickness, increasing from zero at zero thickness and reaching an asymptotic value for large thicknesses. Table I shows some typical values of R_∞ where the incident spectrum is that from a U 235 metal core.

It will be noted that R_∞ reaches its maximum value for water at a thickness of 16 cm and has nearly reached this value at 8 cm, whereas graphite needs 60 cm at least to become effectively infinite. This difference will be referred to in a later section when the effect of reflectors on critical size is discussed.

TABLE I. SOME VALUES OF R_∞ FOR INCIDENT NEUTRONS FROM U 235 METAL

Material	Thickness of Reflector *cm*	R_∞
Wood (Cd-plated) (0.6 g/cm³)	20	0.46
Graphite (1.7 g/cm³) [2] ...	8	0.59
	16	0.73
	32	0.84
	64	0.92
Water (1.0 g/cm³) [3] ...	4	0.45
	8	0.55
	16 & over	0.565

FACTORS AFFECTING CRITICALITY

The critical size of an assembly of fissile material depends on all the materials present, whether fissile or non-fissile, their shape, disposition and density. In order to sort out and catalogue the effects of these variables to some extent, we will focus the attention mainly on systems which can be divided into a core, containing all the fissile material, surrounded by a reflector. We will then look in turn at the effect of the shape of the core, the materials which comprise the core and their density, and finally the reflector. In order to keep the discussion within bounds, only homogeneous cores will be considered.

Effect of Shape - Basic Parameters

It can be shown that a spherical core or, exceptionally, a core that is very nearly a sphere, will give the smallest critical volume and mass for these single core-reflector systems. The effect of variation from this simple shape is illustrated by the curves of Figure 9, which were obtained experimentally [4]. These curves are for cylinders of varying elongation or squatness and give critical masses in units of the spherical critical mass. They show an appreciable increase in critical mass, or volume, as the core departs appreciably from a near isometric cylinder. Another way of interpreting these results is to say that it is possible to increase the critical mass of the core by restricting its diameter, on the one hand, or, on the other, by restricting its height.

It is pertinent, therefore, to ask whether there is a diameter of core which can never become critical, whatever its height, or whether there is a height of core which can never become critical, whatever its diameter. This proves indeed to be possible and the relevant values are important to know as a basis for ensuring safety. There are thus four basic parameters which can be listed for each core of given composition and density surrounded by a given reflector. These are the critical mass and volume of the spherical core, the critical diameter of the infinite cylindrical core and the critical thickness of the infinite slab core. By way of illustration, the results of some recent calculations of these basic parameters for idealized plutonium solutions are

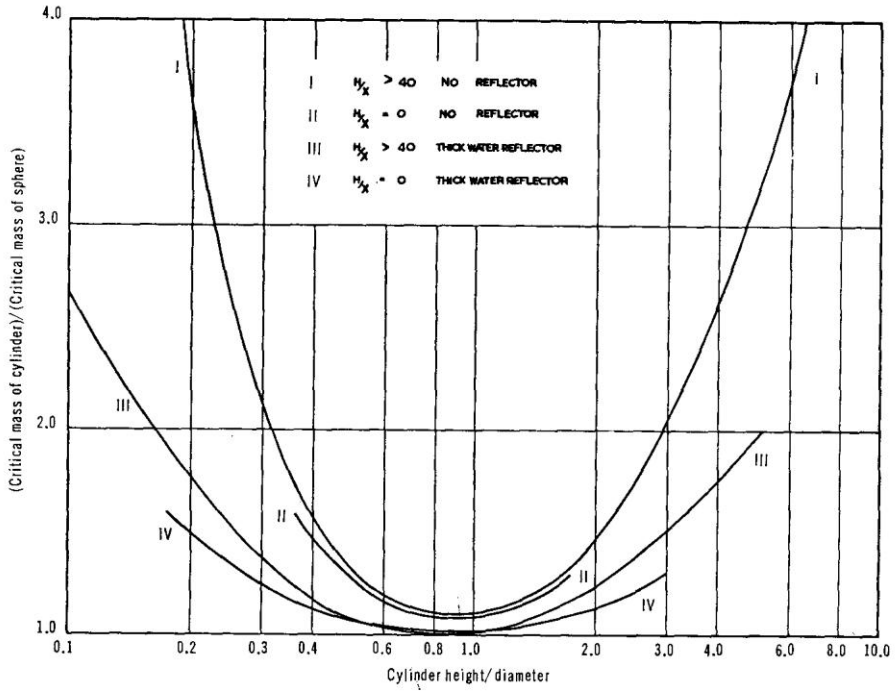


Figure 9. *Effect of shape on critical mass. (From Paxton, LAMS 2415).*

shown in Figures 10 and 11. The cores are in each case reflected by effectively infinite water and show the variation of the basic parameters with x when the core material is $Pu(NO_3)_4.xH_2O$ and $x \geqslant 5$; together with, for spherical cores only, values for $PuO_2.xH_2O$ and $x \leqslant 5$. To prevent confusion it is perhaps worth noting that log (1 + H/Pu ratio) has been used as abcissa in Figure 10 in order to enable the points at H/Pu = 0 to be included.

Density Scaling

In order to separate the various effects resulting from changing the material composition of the core, it is useful to consider the density of the fissile isotope as fixed and examine the effect of adding other non-fissile materials to such a core. To relate these results, therefore, to real systems, it will be necessary to know the effect of changing the core density, and to introduce the density scaling laws.

Probably the only law in the whole subject of criticality which is simple, useful and exact is that which, when formulated in terms of critical size, states:

> In a critical system, if the densities are increased everywhere to x times their initial value and all the linear dimensions are reduced to $1/x$ times their initial value, the system will remain critical.

If r is a typical linear dimension of the core of such a critical system, ϱ_c the core density and ϱ_t the reflector density, this law states that

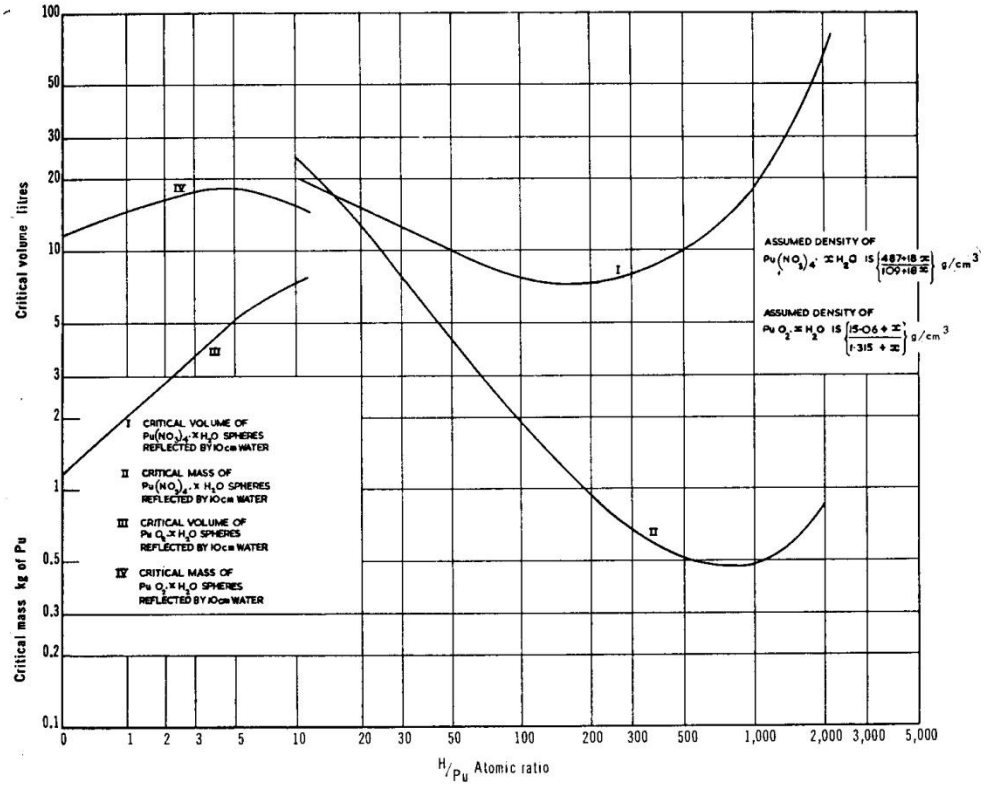


Figure 10. *Critical mass and volume of water-reflected spheres of plutonium " solutions ".*

$$r \propto \rho_c^{-1} \tag{1}$$

provided the ratio ρ_c/ρ_t remains constant.

This law is sometimes stated in terms of critical mass as

$$\text{critical mass} \propto \rho_c^{-2}.$$

The scaling law of which a special case is quoted above, can be applied to the core and reflector separately. For the core it states that the surface multiplication M does not change if the density and dimensions of the core are changed in inverse proportion. For the reflector it states that the reflection R does not change if the density and dimensions of the reflector are changed in inverse proportion.

Using this formulation and the fact that $MR = 1$ for a critical system, a more general density scaling law can be deduced, that for a critical system

$$r \propto \rho_c^{-(1-\Phi)}\, \rho_t^{-\Phi} \tag{2}$$

where

$$1/\Phi = 1 - \frac{\partial(1/M)}{\partial r} \Big/ \frac{\partial R}{\partial r}$$

provided Φ is independent of r.

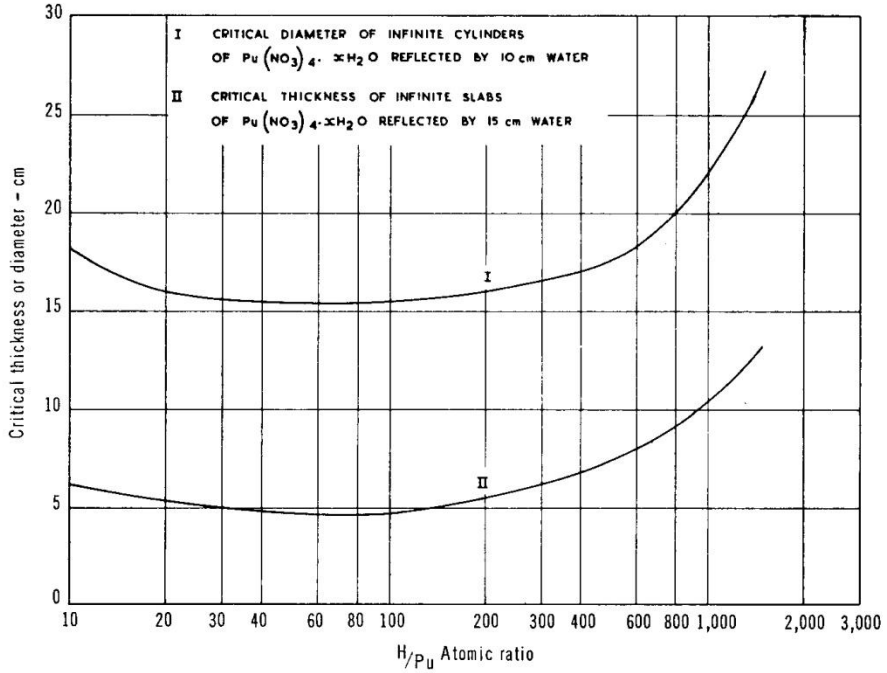


Figure 11. *Critical thickness of infinite slabs and diameter of infinite cylinders of water-reflected plutonium " solutions ".*

The partial differentials are taken at the critical value of r, the densities remaining constant. That is, they represent the slopes of the curves of Figure 5 at their point of intersection.

It is not difficult to see that Φ cannot be entirely independent* of r, since for very large values R becomes nearly constant and so $\Phi = 0$, but Φ cannot vanish for smaller r except perhaps at $r = 0$. Hence for very large cores, equation (1) gives the effect of density changes in the core and, in this case, density changes in a thick reflector are unimportant. However, it has been shown by experiment and calculation that in all cases tested, Φ retains a substantially constant value over a wide range of density variation, and therefore equation (2) represents a valuable density scaling law. Some values of Φ are given in Table II.

The last four results are deduced from experiment and quoted by H. C. Paxton [4]. The other results are from calculation.

Effect of Added Scatterers

Having discussed the density scaling laws, we will now consider the effect of adding different diluents to the fissile core without changing the density of the fissile isotope. The addition of materials which scatter neutrons without

* Except for an infinite slab for which $\frac{\partial R}{\partial r} = 0$ and $\Phi = 0$.

TABLE II. VALUES OF Φ FOR USE IN THE DENSITY SCALING LAW EQUATION (2)

Core	Geometry	Reflector	Φ
U 235, Pu 239	sphere or	Cd plate surrounded	
U 235 F_4	infinite cylinder	by wood	0.11
PuO_2	infinite		
U $235O_2F_2.25H_2O$	cylinder	» »	0.11
$Pu(NO_3)_4.40H_2O$.			
$Pu(NO_3)_4.400H_2O$	sphere	Water	0.058
$Pu(NO_3)_4.5H_2O$ ..	sphere	Water	0.10
U 235	sphere	Natural uranium	0.27
U $235H_3C$	sphere	Natural uranium	0.14
U $235O_2F_2,115H_2O$	isometric cylinder	Water	~ 0.04
Pu 239	sphere	Natural uranium	~ 0.3

appreciably changing their energy will, in most cases, make it more difficult for the neutrons in the core to escape. They will thus increase the chance that a neutron in the core can give birth to further neutrons by causing fission. In this way the multiplication M of the core will increase and consequently the critical mass will decrease.

If the core is in the form of a thin disc or slab, however, the reverse effect will occur. The additional scattering centres will now tend to scatter neutrons out of the core and the critical size will increase.

As an illustration, for spherical cores, the results shown in Figure 10 at an H/Pu atomic ratio of 10 can be used. The nitrate core contains, for each Pu atom,

4 atoms of nitrogen
17 atoms of oxygen
10 atoms of hydrogen.

The Pu density is 1.20 g/cm³, and its critical mass is 24 kg of Pu. The oxide core contains, for each Pu atom,

7 atoms of oxygen
10 atoms of hydrogen.

The Pu density is 2.10 g/cm³ and its critical mass is 15.5 kg of Pu.

Applying the density scaling law to the oxide core, we deduce that at a Pu density of 1.20 g/cm³ its critical mass would be 40 kg of Pu.

The difference between the two cores is that the nitrate core contains for each Pu atom,

4 additional atoms of nitrogen
and 10 additional atoms of oxygen

and the addition of these atoms to the oxide core therefore reduces the critical mass from 40 kg to 24 kg, i.e. by 40 %. The oxygen atoms are almost pure scatterers. The nitrogen atoms give a little neutron absorption as well as scattering, which would tend to increase the critical mass. The reduction from 40 kg to 24 kg therefore represents a slight underestimate of the effect of adding additional scatterers. This and some other pairs of comparative results are given in Table III.

TABLE III. EFFECT OF ADDED SCATTERING ATOMS ON CRITICAL SIZE

Critical Parameter	Density of Pu or U g/cm^3	Reflector	Core I		Core II		Percentage change due to added scatterers
Slab thickness ..	10.1	Water	Pu	1.6 cm	PuO_2	1.9 cm	18% increase
Cylinder diameter	10.1	None	Pu	13 cm	PuO_2	12 cm	10% decrease
»	10.1	Cd & Wood	Pu	9.6 cm	PuO_2	8.6 cm	10% decrease
»	5.1	None	*U	47 cm	*UF_4	29 cm	38% decrease
»	5.1	Cd & Wood	*U	34 cm	*UF_4	21 cm	38% decrease
Sphere, mass of Pu	10.1	Water	Pu	16.5 kg	PuO_2	12 kg	28% decrease
Sphere, mass of U	5.1	None	*U	710 kg	*UF_4	160 kg	77% decrease
Sphere, mass of Pu	1.2	Water	$PuO_2.5H_2O$	40 kg	$Pu(NO_3)_4.5H_2O$	24 kg	40% decrease

* 92.5% U 235, 7.5% U 238.

Effect of Added Moderators

Of the possible effects of adding diluent to a fissile core, that of neutron moderation is the most spectacular and important. The fast neutrons born in fission are moderated, or slowed down, by scattering in any material. For materials of high atomic weight, the possible slowing down is limited in extent. For materials of low atomic weight, and in particular for hydrogen, the moderation will be sufficient to reduce neutrons to energies where they will have a bigger chance of causing a fission and will thus decrease the critical size. Hydrogen is the most effective moderator and is commonly present in the form of water. For other elements the moderating power is much less, and often balanced to some extent by neutron absorption. So, in practice, carbon, beryllium and deuterium are the only important moderators apart from hydrogen.

The effect of adding water to a core of plutonium nitrate solution for which the plutonium density is maintained constant, is illustrated in Figure 12. This is derived from Figure 10 by removing the plutonium density variation, and shows that the critical mass can vary by a factor of nearly 10^5 as the H/Pu ratio increases from 0 to 2,000.

Effect of Added Absorbers

The third possible effect of adding a diluent to the core is to absorb neutrons. Such a process can only increase the critical size. The addition of nitrogen to the $Pu(NO_3)_4.4OOH_2O$ spherical core so as to make the N/Pu ratio 20 increases* the critical mass by 5%. This demonstrates that even a weak poison like nitrogen (thermal absorption cross section 1.9 barns)

* Deduced from reference [5].

can have a detectable effect on critical mass. The addition of a strong poison like boron (thermal absorption cross section 755 barns) will make a very appreciable difference.

The small neutron absorption of hydrogen itself (thermal absorption cross section 0.33 barns) is important where dilute solutions, i.e. with a high H/X ratio, are concerned. The minimum shown in the curve of Figure 12

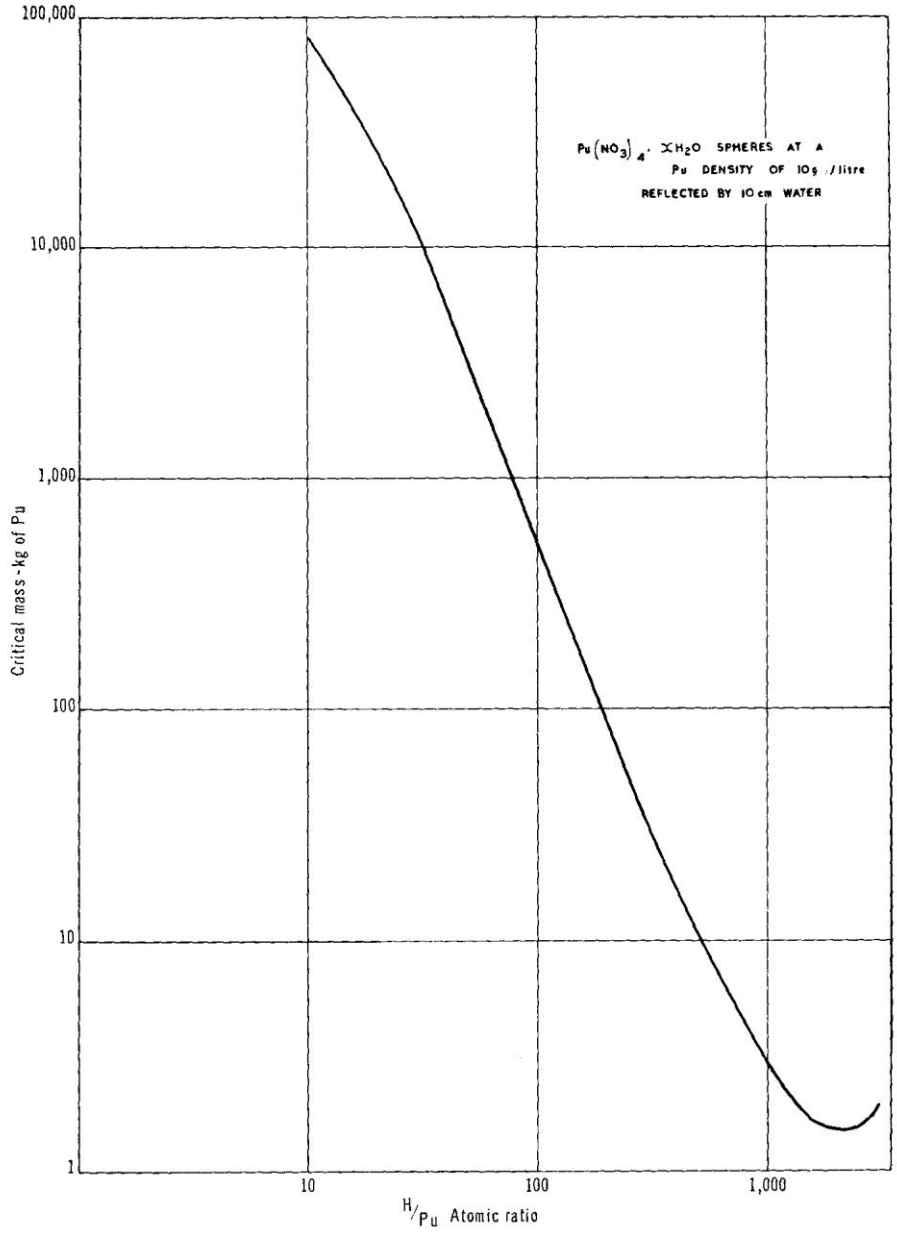


Figure 12. *Variation of critical mass with moderation at constant plutonium density.*

is due to this effect. In fact a plutonium solution cannot be made critical at any size if the H/Pu ratio is greater than about 3,600.

Overall Effect of Core Dilution

The preceding sections have explained separately the three effects that occur as the result of core dilution. In any real physical problem not only will the effects of additional scattering, moderation and absorption be present to varying degrees but also the density of the fissile material will be reduced. The overall picture can be illustrated qualitatively by considering the particular cores shown in Figure 10.

Suppose water is added steadily to a spherical core of PuO_2 in such a way as to keep the fissile material always homogeneously disposed. First, when a very little water has been added, both the critical mass and the critical volume increase. At this stage the effect of the change of density of the Pu predominates, the additional scattering has only a small effect and there is not enough hydrogen present to give any appreciable moderation. Soon, however, when the H/Pu ratio exceeds about 4 the effect of neutron moderation begins to be felt and the critical mass begins to decrease, although the critical volume continues to increase. At an H/Pu between 10 and 20 the moderation causes the critical volume to decrease also. The plutonium oxide curves will be of similar shape to the nitrate curves for the higher moderations. Both the critical mass and critical volume decrease to a minimum and then start to rise again, as the absorption of neutrons by the hydrogen begins to take effect, and both become infinite at H/Pu of about 3,600.

Since it is encountered in many of the common uranium fuels, the isotope U 238 is a diluent of particular importance. U 238 dilution results in a reduction in the density of the U 235 fuel, additional centres for elastic scattering, absorption and neutron moderation by inelastic scattering, the absorption being mainly in the region of neutron energies from 5 ev to a few Kev, the socalled " resonance region ". For uranium metal cores the overall result is that the mass of U 235 in a critical sphere follows the law, derived empirically by Paxton [6], that

$$\text{U 235 critical mass} \propto (\text{U 235 concentration})^{-0.7}$$

for U 235 isotopic concentrations greater than 25 %, either unreflected or with a thick uranium reflector. For lower U 235 metal isotopic concentrations the critical mass rises more quickly, becoming infinite at a concentration between 5 and 6 %.

If there is also sufficient hydrogen in the core as moderator, the mass of U 235 in a critical sphere is reduced and the limiting concentration below which such a core cannot become critical occurs when the uranium consists of about 1 % U 235 and 99 % U 238. This limiting concentration can be further reduced to about that in natural uranium by separating the uranium from the hydrogenous moderator to form a latticed arrangement. In such a core the neutrons are, to some extent, separated from the U 238 while they are being slowed down and thus have a greater chance of reaching thermal energies without being caught in the U 238 resonance region on the way.

Effect of Reflectors

The effect of reflection of neutrons back to the fissile core was mentioned in the discussion of criticality parameters (See pages 21 and 23) and from

such graphs as Figures 7 and 8 the effect of a given reflection R on critical mass can be deduced.

The reflection depends on the thickness of the reflector and Figure 13 illustrates how this affects the critical mass of spheres of two of the plutonium "solutions" calculated in the case of a water reflector. It will be seen that water is effectively infinite at a thickness of 8 cm if H/Pu = 800 and at a thickness of 9 cm for H/Pu = 10.

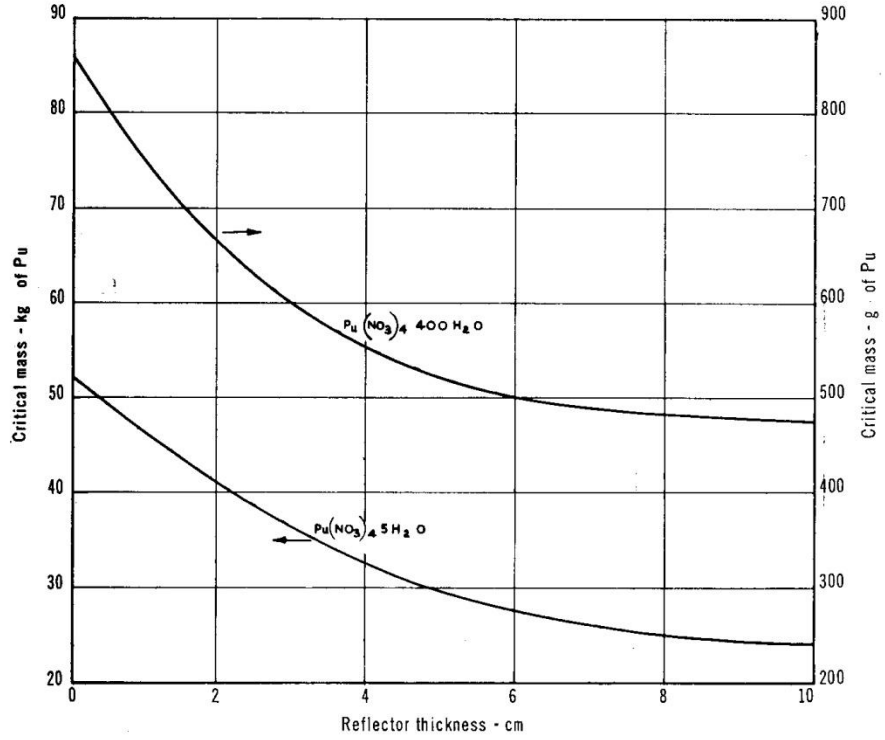


Figure 13. *Effect of reflector thickness on critical mass of plutonium solutions.*

Some more comprehensive results are given in Figure 14 [4] which shows the effect on the critical mass of a U 235 metal sphere of reflectors of various thicknesses and material. Beryllium, together with BeO, is the most efficient non-fissile reflector known. The curve for aluminium is typical of common structural materials. Uranium is a reflector whose efficiency is enhanced somewhat by fission. Water is peculiar in that up to a thickness of 2 in. it is a better reflector than most other materials of the same thickness, but any increase in thickness beyond 2 in. results in very little increased reflection. This peculiarity is shared by many hydrogeneous compounds such as oils or waxes. On the other hand, graphite is not an effective reflector in small thicknesses but is one of the best in large thicknesses. A similar difference was noticed for the slab reflections listed in Table 1, though for this case it needed more than a 2-in. thickness of water to be effectively infinite.

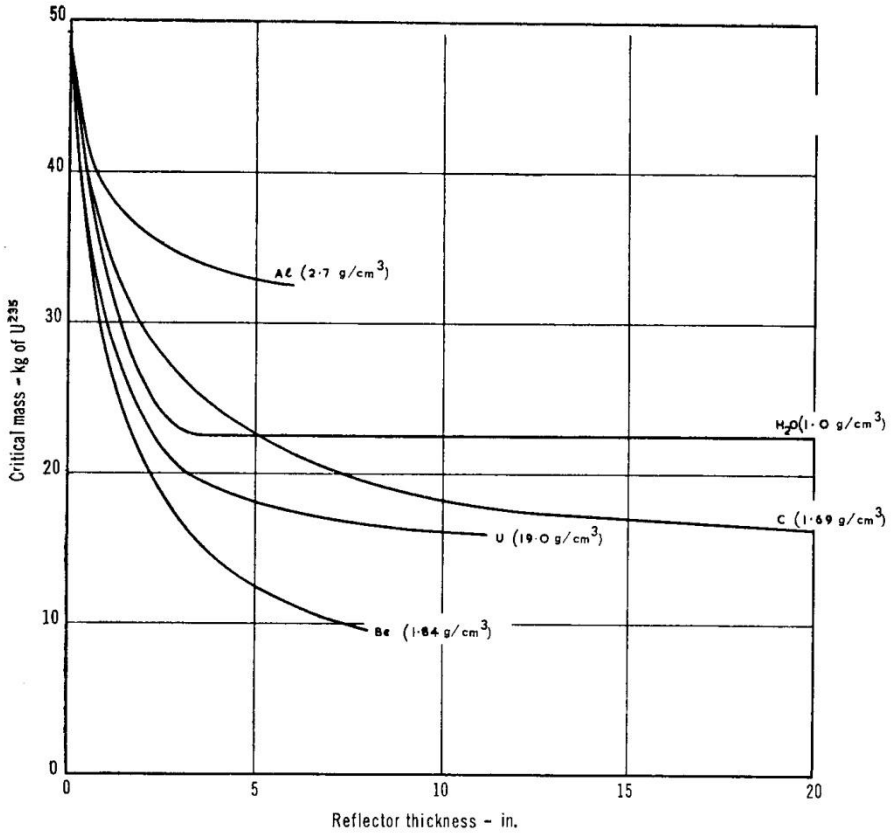


Figure 14. *Critical mass of U 235 (93.5 %) metal (18.8 g/cm³) spheres in various reflectors.*

The term " better reflector " is used above in the sense that it gives the smaller critical size. This does not necessarily mean that it gives the larger value of R since there may also be some effect on M. Thus an infinite slab of U 235 (92.5 %) metal reflected on each side by 8 cm thick graphite is critical at a thickness of 2.7 cm, R being 0.59 and M being 1.7. However, a U 235 slab of this thickness if reflected by 8 cm thick water would have a surface multiplication M of 2.1 and the critical thickness is only 1.8 cm. The enhanced multiplication arises because neutrons reflected from the water have an appreciably softer spectrum than those reflected from this thickness of graphite. Thus, although R for 8 cm thick water is 0.55, which is less than that for a similar thickness of graphite, the water is in this case the " better reflector ".

Reflectors will often occur which consist of two or more layers of different materials. Such mixed reflectors often give rise to phenomena which are not apparent from a knowledge of the effects of the materials separately. For example, there is nothing peculiar about pure cadmium as a reflector of a core of plutonium or enriched uranium metal, yet a mixed reflector consisting of a thin layer of cadmium, next to the core, surrounded by thick

water, results in a critical size appreciably larger than that for either water or cadmium alone. This phenomenon can be readily explained by the fact that many of the reflected neutrons returning from a thick water reflector are of thermal energies and are absorbed in the cadmium, whereas for a pure cadmium reflector practically all of the reflected neutrons are of high energy and are not absorbed. More difficult to explain are the two phenomena depicted in Figure 15. Curve I [7] shows the effect of a layer of steel between the core and a thick water reflector, and here again a larger critical size is obtained with a mixed reflector than with a pure one of either steel or water. The opposite effect is illustrated in curve II [8] of Figure 15 which shows a

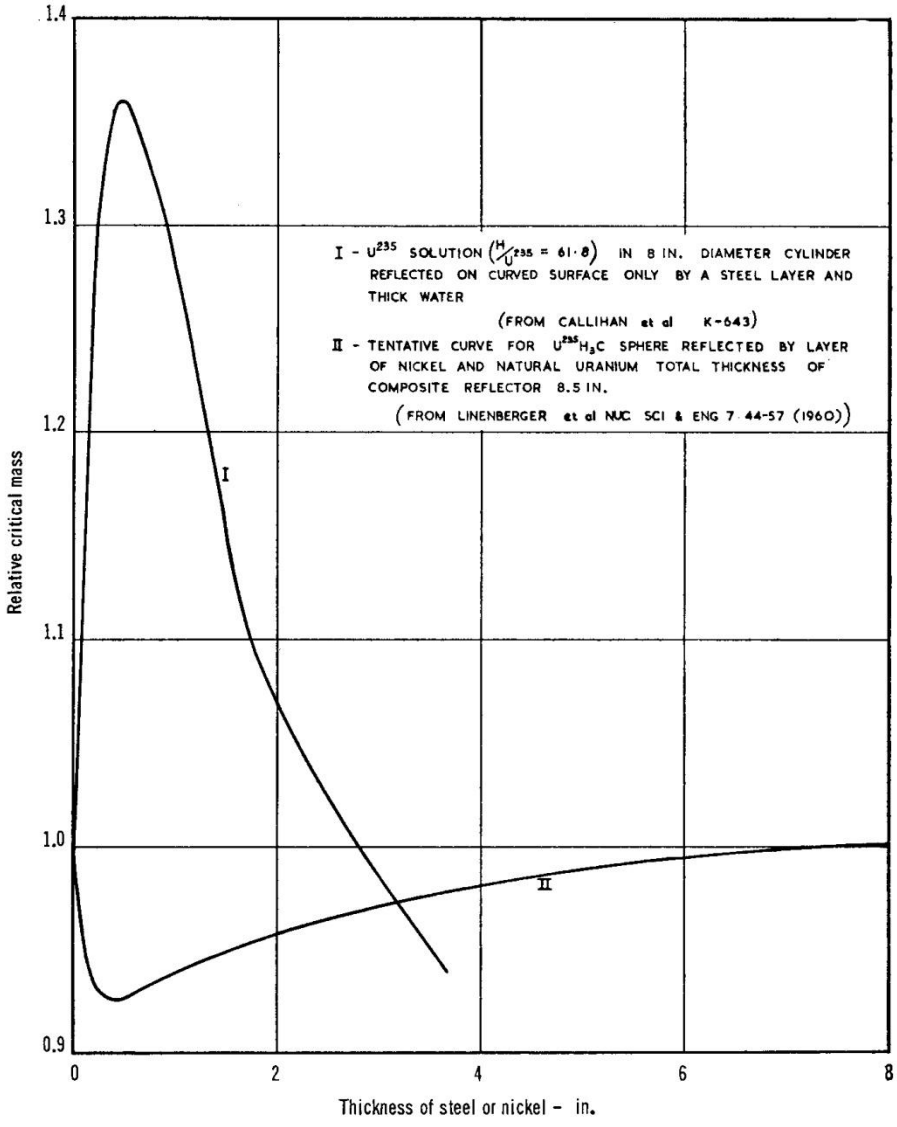


Figure 15. *Effect of composite reflectors on critical mass.*

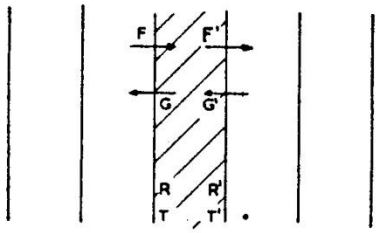
case where a mixed reflector consisting of nickel, next to the core, surrounded by uranium, gives a smaller critical mass than a reflector of either material alone.

A reflector is most effective in reducing critical size when it is in contact with the core. A small gap between reflector and core increases the critical size by a significant amount. Very roughly, in spherical or cylindrical geometry, a gap of width 10 % of the core radius increases the critical mass by 20-30 %.

Interaction

So far we have discussed systems consisting of a single core surrounded by a reflector. It will often happen in practice that the fissionable material is divided into a number of separate cores. This will occur, for example, in a chemical processing plant where the fissile material is confined to several separate vessels, in a storage vault, or in many other circumstances. The new phenomenon that arises is that neutrons can now pass backwards and forwards between one core and another with the possibility of multiplying.

It is of interest, by way of illustration, to study the one-dimensional model of a series of parallel slabs each of which is of finite thickness but of infinite face area:



Suppose that for a typical slab, when the system is critical, a flux of neutrons F is entering its left-hand face; a flux G is leaving its left-hand face; a flux F′ is leaving its right-hand face and a flux G′ is entering its right-hand face.

Then
$$G = RF + T'G'$$
$$F' = R'G' + TF$$

where R is the fraction reflected of neutrons entering from the left and T the fraction transmitted and R′ and T′ are reflections and transmissions of neutrons entering from the right. Since fissions will occur in some of the slabs the reflections and transmissions are not necessarily less than unity.

These equations can be written as

$$G' = (G - RF)/T' \qquad (3)$$
$$F' = R'G' + TF$$

So, knowing F, G and the quantities R, R′, T and T′, the values of F′ and G′ can be calculated. For the left-hand boundary slab of the array $F_e = 0$ and G_e can be given an arbitrary value, say $G_e = 1$. The above equations (3) then enable the fluxes to be calculated step by step through the system. If such a calculation yields, for the right-hand boundary slab, $G'_r = 0$, the

system is critical. So, if R, R′, T and T′ for each slab are known, critical configurations can be calculated.

If the array consists of an equispaced series of similar slabs of fissile material separated by layers of shielding material, the equations can be simplified. For each slab we will have $R' = R$ and $T' = T$. Let us now introduce the surface multiplication for the fissile slabs

$$M = R + T$$

and let R_0 and T_0 be the reflection and transmission of the shielding slabs. We define an interaction parameter

$$q = \frac{\frac{1}{2} T_0 M}{1 - R_0 M} \tag{4}$$

and, by making the simplifying assumption that

$$F' = G \tag{5}$$

for each fissile slab, equations (3) can be reduced to

$$F_j = q \{F_{j+1} + F_{j-1}\}$$

where F_j is the flux leaving one side of the j^{-th} fissile slab.

The critical number of slabs then only depends on q, and some values are given in Table IV.

TABLE IV

Interaction Parameter q	Critical Number of Slabs
∞	1
1.000	2
0.707	3
0.618	4
0.577	5
0.555	6
0.521	10
0.500	∞

The isotropic assumption, equation (5), can lead to underestimates or overestimates of critical number, depending on the size and material of the individual slabs. It is therefore not particularly valuable for interacting slab problems for which equations (3) can be solved directly. However, as will appear in a later paper, it is a useful parameter for the assessment of interacting arrays of more general geometry, and conservatism can be assured by overestimating the value of q. This can be done for slabs, for example, by using, instead of M, the quantity

$$2x \max (R, T).$$

Some results have been calculated [3] by equations (3) for two similar parallel slabs of uranium metal, density 18.7 g/cm³ containing 92.5 % U 235 separated by water and reflected by thick water on their outer surfaces. These are given in Table V where the uranium thickness quoted is the sum of the thicknesses of the two uranium slabs.

TABLE V

Separation of Slabs *(cm)*	Critical Uranium Thickness *(cm)*
0	1.71
2	0.76
4	0.40
6	0.51
8	1.00
10	1.50

It will be seen that the critical uranium thickness reaches a minimum at a separation of about 4 cm and thereafter increases as the separation increases.

It is of interest then to ask how the criticality of the system changes if the uranium goes slowly into solution in the water so that we have uranium metal slabs separated by 4 cm of a uranium/water mixture and reflected on their outer faces by a uranium/water mixture. The results in Table VI are for 4 cm thickness of outer reflector, the H/U ratios refer to the uranium/-water mixtures and the total uranium thickness is the thickness of the slab that would be obtained if all the uranium present were gathered together into a single slab of metal.

TABLE VI

'SOLUTION' OF TWO URANIUM SLABS 4 cm APART

H/U ratio	Total Amount of Uranium when Critical *(cm)*
∞	1.84
2,000	1.50
1,000	1.46
500	1.44
200	1.30
50	0.78
20	1.01

It will be seen that as 'solution' proceeds the critical uranium thickness goes through a further minimum.

These examples illustrate that a more dangerous system from a criticality point of view can be achieved when the fissile material is in separate lumps than when it is all gathered together, and also suggest that, during solution of uranium in a hydrogeneous liquid, a system may appear at an intermediate stage which is more dangerous than at either end point.

The whole subject of interacting arrays has not yet been explored thoroughly enough to formalize the factors affecting criticality as fully as for systems with a single core. There is scope for further experimental work in this field. The theoretical side is the subject of a lecture at a later session and so it will not be further discussed here.

Precautions to Avoid Criticality

In the preceding sections we have discussed at some length the factors that affect the criticality of a system containing fissile material. With this knowledge, the next question that arises is how to devise a system of control, in a practical case such as a plant processing fissile material, to ensure that a critical state is not reached accidentally. There are basically two possible ways of effecting such a control. One is to divide the plant into units of such a restricted size that a critical assembly cannot be formed whatever material is present in these units. The other is to impose a restriction on the amount of fissile material that can enter the various parts of the plant. These can be called respectively geometrical and operational control. It is clearly ideal to have a plant for which the safety is ensured entirely by geometrical control, but this ideal is rarely entirely achieved in practice. It will often happen that it is impossible or grossly uneconomical to carry out the desired processes in vessels which are geometrically safe. So, as a rule, some degree of operational control has to be imposed. In particular, certain materials can be readily excluded from the plant or may need to be excluded to make its operation possible. For example, the presence of plutonium can often be excluded from a uranium processing plant, or that of uranium metal from a plant handling uranium compounds and solutions.

Geometrical Control

As an example, suppose that in a particular chemical plant it is possible to ensure that the only materials which can be present are plutonium nitrate solutions, water and precipitates of plutonium oxide (PuO_2). Reference to Figures 10 and 11 shows that for plutonium nitrate solutions the minimum critical volume is 7.2 litres, the minimum critical cylinder diameter is 15.4 cm and the minimum critical slab thickness is 4.6 cm. The addition of further nitric acid to this solution will increase the minimum critical parameters since the poisoning effect of nitrogen by neutron absorption is the predominant result of this change. Plutonium oxide precipitates, however, give lower values of these critical parameters at zero moderation. At high moderations they give minimum values which are very little different from those for the nitrate solutions. Consequently, for the plant to be geometrically safe, it must be designed to be subcritical even if filled with dry PuO_2, the minimum critical parameters for which are volume 1.18 litres, diameter 8.3 cm* and thickness 1.9 cm. Since the plant envisaged will contain a number of vessels, interaction between them will affect the criticality. To allow for this the individual vessels will have to be separated by distances which will be greater the nearer each vessel is to critical. In order to keep the plant within bounds the individual vessels must therefore not be allowed to approach too near to the critical state. Let us say that a factor of 0.8 should be applied to the volume and 0.92 to the diameter and thickness.

The result of this analysis is that if the plant is built so that no vessel size exceeds either a volume of 0.94 litre or a diameter of 7.6 cm* or a thickness of 1.75 cm and if the vessels are separated by at least a specified distance (which will give about the usual separation for this type of plant), and are

* Approximate figure for illustration only.

connected by pipework of no more than, say, 4 cm diameter, then it will be geometrically safe.

Mass Limitation

It may well happen, however, that a plant built to these specifications is not satisfactory and it proves necessary to introduce some form of operational control to enable the geometrical limitations to be relaxed. In some cases it may be desirable to go to the extreme and remove all geometrical limitation. Control may then be achieved by restricting the mass of fissile material in each vessel of the plant. However well it is applied, this type of control cannot be as rigid as a geometrical limitation. So it is desirable, and has become customary in many places, to apply in addition a safety factor of 0.5, giving 200 g of plutonium as the maximum allowed in any vessel.

It might be possible to relax this limitation on mass if some geometrical control were also applied. Figure 10 indicates that the critical conditions giving minimum mass need more than the minimum volume, so some restriction on volume may allow the mass limitation to be increased. Figure 16 has been prepared for the plutonium nitrate solutions to explore this possibility and from this it is clear that in this case no worthwhile relaxation of the mass limit can result from a volume restriction.

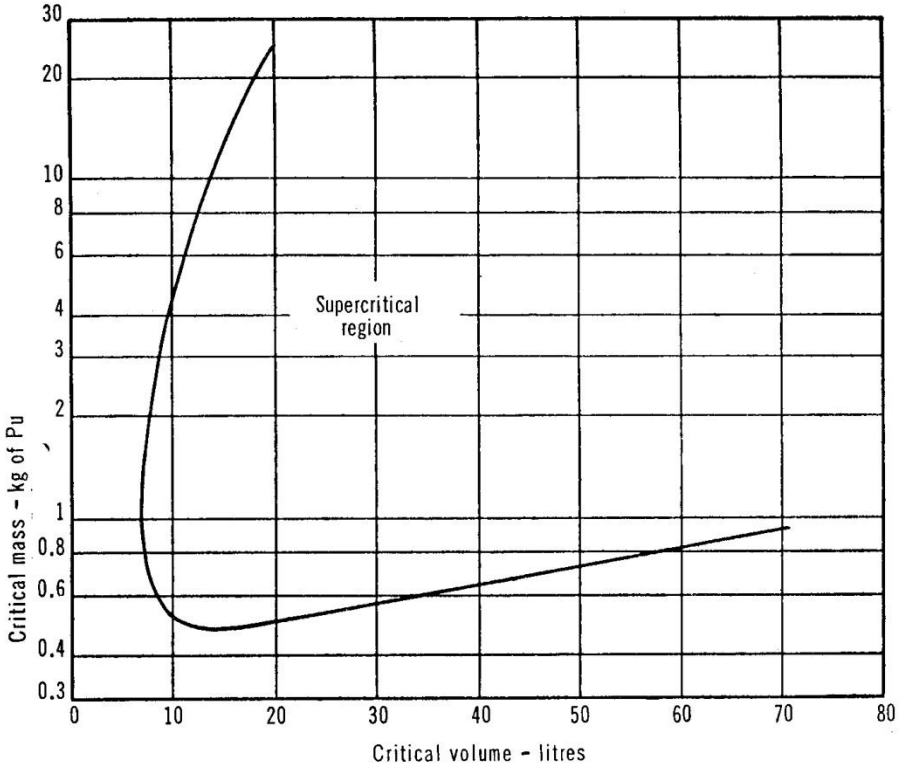


Figure 16. *Critical conditions for* $Pu(NO_3)_4$ $\times$ H_2O *spheres, water-reflected.*

It is more worthwhile, however, to impose a restriction on the diameter of a vessel and Figure 17 illustrates this [9]. It is a curve showing diameter versus minimum critical mass deduced from experiments with aqueous solutions of uranyl fluoride, highly enriched in U 235. The minimum critical mass of this solution is 0.88 kg*. If the vessel is restricted to a diameter of 7 in. this rises to 1.6 kg, whereas it is necessary to restrict the diameter to 5.5 in. to ensure that such a solution cannot become critical in any amount.

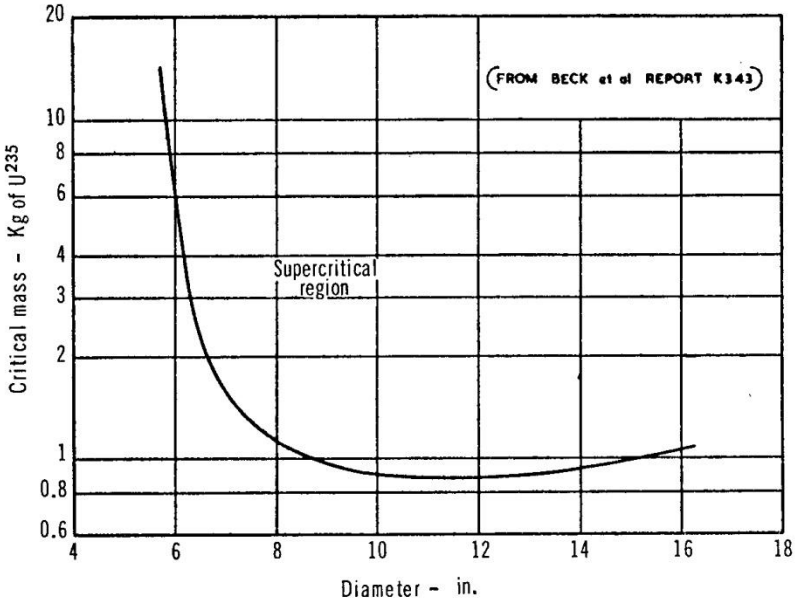


Figure 17. *Critical conditions for UO_2F_2 aqueous solutions (93.4 % U 235) cylinders water-reflected.*

Poison Control

Another method of criticality control is to design the processing plant in such a way that neutron absorbers are present, intermingled with the fissile material. It is necessary to guarantee the presence of these poisons in sufficient amount to ensure that criticality cannot be reached in any vessel. A particular example of this is to use hydrogen as the poison. If a solution of any fissile material in a hydrogeneous solvent is made dilute enough, a critical assembly cannot be formed in any circumstances. These conditions are determined by a H/Pu ratio exceeding about 3,600 for plutonium solutions or by a H/U 235 ratio exceeding about 2,400 for uranium solutions. If the latter contain U 233 the amount of this isotope should be included with the U 235 in forming the ratio. This method is in effect a ' concentration ' control and is usually known by that name.

* These figures and the graph Figure 17 are taken directly from a paper [9] written in 1949. Views on the precise values of the minimum critical parameters have been modified since then and slightly smaller figures are customarily quoted.

Poison control is valuable provided that the poison is known to be always present. In particular where concentration control is applied, precautions must be taken to ensure that the limiting concentration is never exceeded, even momentarily, over any significant volume.

Other Factors

The example used to illustrate the arguments on geometrical control and mass limitation (see page 44) used critical sizes for reflection by thick water. It may happen exceptionally that a better reflector than thick water is present — concrete is an example — and, if so, suitable allowance must be made by imposing tighter restrictions. On the other hand, in many cases it can be demonstrated that the only reflectors that can be present are less effective than thick water. For instance, the only possible reflector may be the thin walls of a tank. In such circumstances the restrictions can be relaxed.

In general, in order not to impose restrictions which are more onerous than is essential, it is worthwhile examining the plant so as to exclude impossible circumstances. The aim is to find the worst credible conditions and to base the necessary criticality restrictions on these, though in doing this only those situations should be rejected which can be proved impossible or so unlikely that a tolerable risk is run in ignoring them.

Concluding Remarks

This paper has endeavoured to present the fundamentals of criticality in such a way as to form an introduction to a deeper and fuller discussion. The author is conscious that in no part is the treatment anywhere near complete. Those sections which are to be taken up in later papers have been discussed only briefly and superficially. Although such topics as measures of criticality have been covered more fully, even here a large number of loose ends have been left. This is in part because of the gaps in our knowledge of a developing subject, but also because it was felt necessary to enforce some volume limitation on this contribution which, unless firmly restricted, shows a tendency to behave like neutrons in a supercritical system and grow with no apparent limit.

REFERENCES

1. A.F. Thomas and L.H. Underhill, Unpublished.
2. E.R. Woodcock and J.B. Parker, " Infinite Slab Criticality Calculations ", *AWRE-014/60* (1960).
3. E.R. Woodcock, " Critical Assemblies of Infinite Slabs of Highly Enriched Uranium and Water ", *AHSB Report 17* (1959).
4. H.C. Paxton, " Critical Data for Nuclear Safety Guidance ", *LAMS-2415* (1960).
5. F.E. Kruesi, J.O. Erkman and D.D. Lanning, " Critical Mass Studies of Plutonium Solutions ", *HW-24514 (Del)* (1952).
6. H.C. Paxton, " Critical Masses of Fissionable Metal as Basic Nuclear Safety Data ", *LA-1958* (1955).
7. D. Callihan, D.F. Cronin, J.K. Fox and J.W. Morfitt. " Critical Mass Studies, Part V ", *K-643* (1950).
8. G.A. Linenberger, J.D. Orndoff and H.C. Paxton, " Enriched-Uranium Hydride Critical Assemblies ", *Nucl. Sci. Eng. 7.* 44 (1960).
9. C.K. Beck, D. Callihan, J.W. Morfitt and R.L. Murray, " Critical Mass Studies, Part III ", *K-343* (1949).

DISCUSSION

H. DOPCHIE, Belgium:

I was quite astonished at the result, that adding a scatterer which is non-absorbent and non-moderator would increase the critical mass of a thin slab. Would you not say that the statement could only be true for an under-moderated core with a reflector which slows down the neutrons?

E.R. WOODCOCK:

It is certainly not true that the effect of a scatterer added to a slab depends on the moderating reflector. This effect exists whatever the reflector is. There may be a case where there is a different effect, that is where the core consists of a slab of a large thickness. If one has a critical system with a very thick slab, one may possibly get the opposite effect, but this has not been investigated as yet.

G. BLÄSSER, Euratom:

I wonder how one obtains the formulae giving the density of plutonium solutions as a function of the water content as found in Figure 10, "Assumed density of $Pu(NO_3)_4 . xH_2O$ is $\left(\frac{487 + 18x}{109 + 18x}\right)$ g/cm^3"?

E.R. WOODCOCK:

These formulae, which give the overall densities of the plutonium solutions, are found by knowing the density of the dry material on the case of the plutonium oxide, and of $Pu(NO_3)_4 . 5H_2O$ in the plutonium nitrate case, which have been estimated at maximum crystal density, and just adding water as a pure mechanical mixture. It was not necessary to try to tie them up with any real experimental values because we have the density scaling factor which enables any small correction in density to be made quite accurately.

W. SCHÜLLER, Eurochemic:

I should like to raise a question concerning Figure 15: " Effect of Composite Reflectors on Critical Mass ", and particularly the curve No. II where the critical mass for composite reflectors is shown to be smaller than the critical mass for the individual reflectors. Has any attempt been made to explain this effect and are there other examples where this effect has been observed?

E.R. WOODCOCK:

I would like to pass this question over to Dr. Paxton. This is a result which he has given in a recent paper. I know of no explanation: presumably there is one, but I have not come across any other case in which this effect has been observed.

H.C. PAXTON, United States:

We have no verified explanation for this effect but we suspect that it may be associated with a strong scattering resonance that nickel has at about

16 keV. The effect was first demonstrated simply because a small sample of nickel seemed to be surprisingly efficient just outside the core interface in a uranium-reflected hydride system. Indeed, in adding a layer of nickel within the uranium-reflected system, the critical mass decreased; if it is not a scattering effect, I do not know what it could be.

J.L. SCHWENNESEN, United States:

Gentlemen, we have of course a very wide audience here, representing not only specialists in the nuclear safety field, but also design engineers, persons who are concerned with operational aspects, etc. Recognising that the subject of fundamentals in this field is exceedingly difficult to discuss anyway, I am wondering if there are any speakers who would perhaps characterize the fundamentals a little differently or perhaps stress, from their particular point of view, certain of Mr. Woodcock's observations or emphasise them differently.

H.C. PAXTON:

I might simply remark that surface multiplication is a beautiful concept. One of the difficulties has been with the measurement problem : it is simply that other types of multiplication are much easier to measure, and I think that what Mr. Woodcock says should hopefully encourage more attempts at measuring and computing surface multiplication. It is extremely useful.

SURVEY OF THEORETICAL CALCULATION METHODS

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INTRODUCTION

In this survey of methods of calculation for critical sizes and masses of isolated fissile assemblies, stress is laid on those simple methods which have been found useful over a period of many years and continue to give good service, and those advanced methods which are increasing in practical importance with the growing power of digital machines. The choice of a simple or an advanced method for a particular problem demands some experience if it is to be a wise one. One can get probably the best results for money spent by a balanced application of simple and advanced methods according to the following scheme:

a) Establish accurate critical sizes of the system for a few values of the significant parameters;
b) Interpolate between the established points by a simple calculation which may be based for some problems on the appropriate mathematical model, and for other problems on perturbation theory.

The best way of establishing critical sizes for a few values of the significant parameters is in principle the experimental one, but for a variety of reasons experiment is seldom feasible for establishing all the points required. Recourse to an advanced method should be made if the significant physics of the problem is not well understood. Advanced methods require careful preparation and can be misleading if good nuclear data are not available. If the physics of the problem is well understood a simple method can sometimes be used with advantage, but there are pitfalls for the unwary.

In balancing the claims of simple and advanced methods it is well worth remembering that in plants processing fissile material the physical state of a potentially critical system may not be known with anything like the accuracy usually met in the reactor field. Consequently, in assessing the criticality of a vessel, a variety of potentially critical systems may have to be examined. Extreme accuracy in the calculations is not called for. As long as the calculated critical sizes are slightly conservative, simple methods are good enough. However, in the absence of suitable check experiments it would be most unwise to rely on simple methods only, and advanced calculations are often called for. Simple methods are considered to be those based on few-group diffusion theory and age theory. Advanced methods, in principle, stem either directly from transport theory or the Monte Carlo method, and their appli-

cation is possible where there are good facilities for computing. Examples from transport theory are the spherical harmonics and Carlson S_n methods. Multi-group diffusion is regarded as an advanced method for it requires, in practice, the services of a digital computer.

Criticality problems for isolated vessels can be regarded as falling into two classes: (*i*) spectral problems; and (*ii*) problems of shape.

A typical problem of class (*i*) would be to determine the critical size of a water-reflected sphere of 60 % uranium (U 235) oxide and paraffin wax with an H/U 235 ratio of 3, since the emphasis here is on spectrum and not on shape. The critical dimensions of a water-reflected tee-junction for a dilute aqueous solution of highly enriched uranyl fluoride is an example of the second class of problem. Some methods could be suitable for both types of problem, e.g. the Monte Carlo method; others, such as the variational methods and buckling conversions of diffusion theory, are very convenient for dealing with changes of shape which involve only insignificant changes in neutron spectra. The Carlson S_n version of transport theory is an example of an advanced method which can deal with problems of class (*i*), but is limited for problems of class (*ii*) to spherical, finite and infinite circular cylindrical, and slab systems.

Criticality problems on isolated systems can be classified also on the basis of: (*i*) homogeneous systems; and (*ii*) heterogeneous systems.

Class (*i*) includes systems with cores of solid metal or solutions. Class (*ii*) includes conventional lattices of fissile rods or sheets in a moderator, solutions containing sludges, encrustations or severe gradients in the concentration of fissile material. Most of the work on heterogeneous systems employs diffusion theory. Originally, diffusion theory was chosen because of its simplicity, and subsequently many experiments were fitted into the diffusion treatment of lattices by judicious choice of constants. Consequently, there are many practical reasons for adhering to this simple model for lattices. In recent years the Monte Carlo method has been applied to lattice theory, but this work is perhaps to be regarded as supplementing rather than supplanting the earlier work on lattices.

In this review we are principally concerned with outlining the basic methods used for criticality calculations, and not with the details of a method when applied to a specific field. For example, we indicate how diffusion theory is applied in lattice theory, but we refer the reader elsewhere for details when there is a suitable review.

The two fundamental methods for criticality calculations are the Monte Carlo method and the method of transport theory. The former is in principle the more accurate of the two, since the intention in the calculation is to mimic the actual situation in every detail. Since the method is a statistical method, the answers obtained are only probable ones. The uncertainty in the answer can be reduced to negligible proportions if one can put in the numerical effort required. The numerical effort required can be reduced by using certain statistical ' dodges ', but it may not be clear always that when labour has been saved, unwarranted assumptions have not crept in. We also consider the simplification of Monte Carlo calculations by use of matrix algebra and the neutron traffic model. Provided good nuclear data are used, and there are good computing facilities available, the Monte Carlo method is an excellent means of establishing basic data. The method has one serious disadvantage which it shares with experimental methods (since the methods

can be roughly regarded as experiments with numbers and neutrons respectively), that the results obtained do not always shed much light on the physics of the problems considered. This disadvantage is not peculiar to the Monte Carlo method; it is shared to some extent by most advanced methods. An advanced method which does give considerable insight is the spherical harmonics method of transport theory, but unfortunately this is not well suited to machine methods of computation. Simple methods can be helpful in understanding the physics of criticality problems, if sometimes it is only in a negative way.

In the transport method the statistical fluctuations in the neutron population of a system are ignored. This is a reasonable assumption for critical systems. Transport theory can be formulated either as an integro-differential equation, the Boltzmann equation, or in terms of integral equations. In the sequel, we show how one of the approximate methods of diffusion theory is obtained from transport theory, and particular stress is laid on the so-called $1\frac{1}{2}$ group or albedo method, as this attractive version of diffusion theory can be very useful for some criticality calculations. Diffusion theory is usually derived assuming that the linear dimensions of the system are large compared with the neutron mean free path. We give an outline of a diffusion theory for finite systems, and show by numerical comparison with Carlson S_n and Monte Carlo results that it can give in some problems significantly better results than classical diffusion theory. In keeping with the policy of adding a little spice to our review of established methods, we consider the problem of checking the accuracy of advanced methods from an entirely new standpoint, which employs the geometry of a function space appropriate to the Boltzmann equation.

In the sequel we shall refer to a number of standard works and papers, and, for convenience, we usually adopt the notation of the reference. Thus, the notation in this paper is not consistent throughout.

The Boltzmann Equation of Transport Theory

If $N(\underset{\sim}{r}, v, \underset{\sim}{\Omega})dv\, d\Omega$ denotes the number of neutrons per unit volume at the point with position vector $\underset{\sim}{r}$, with speeds in the range v to $v + dv$, and directions lying in cones of solid angle $\delta\Omega$ with axes parallel to the direction $\underset{\sim}{\Omega}$, then the Boltzmann equation for a region V can be written (Davison [1]):

$$v\,\underset{\sim}{\Omega}.\text{grad}\, N(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) + v\alpha(v)\, N(\underset{\sim}{r}, v, \underset{\sim}{\Omega})$$
$$= \int_{\Omega'}\int_{v'} v'\,\beta(v')\, N(\underset{\sim}{r}, v', \underset{\sim}{\Omega}')\, f(\underset{\sim}{r}; v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})dv'\, d\Omega' \qquad (1)$$
$$+ \text{source terms}$$

In this equation α is the inverse mean free path and β the number of secondary neutrons per unit path for neutrons of speed v. The integration on the right-hand side of (1) is over all directions $\underset{\sim}{\Omega}'$ at position r, and all neutron speeds v'. The physical meaning of the kernel $f(\underset{\sim}{r}; v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})$ is as follows: $\frac{\beta}{\alpha} f(\underset{\sim}{r}; v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})dv\, d\Omega$ is the number of neutrons at $\underset{\sim}{r}$ produced in the speed range dv with directions in the cone of solid angle $d\Omega$ and axis $\underset{\sim}{\Omega}$, when a neutron of speed v' and direction $\underset{\sim}{\Omega}'$ undergoes a collision. The products of such a collision (secondary neutrons) are in general fission neutrons, elasti-

cally and inelastically scattered neutrons. The kernel f is in general represented as a sum of three kernels, one for each of three basic types of secondary neutron produced as the result of fission, elastic and inelastic scattering. The parameters α and β can be expressed in terms of the microscopic cross sections as follows.

Suppose there are n nuclei per unit volume, then:

$$\left.\begin{aligned}\alpha &= n(\sigma_e + \sigma_{in} + \sigma_f + \sigma_c)\\ \beta &= c\alpha = n(\sigma_e + \sigma_{in} + \nu\,\sigma_f)\end{aligned}\right\} \tag{2}$$

where σ_e, σ_{in}, σ_f and σ_c denote the cross sections for the elastic scattering, inelastic scattering, fission and capture processes. The number of secondaries per collision is c and ν is the number of neutrons emitted per fission.

For the three processes producing secondary neutrons the function f takes the following forms for a homogeneous medium. In fission, the neutrons are emitted isotropically in the laboratory system, and f reduces to:

$$f_f(v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega}) = \mathrm{F}(v)/4\pi \tag{3}$$

where $\mathrm{F}(v)$ denotes the fission spectrum. Neutrons are also assumed to have an isotropic distribution in the laboratory system after inelastic collisions, i.e.:

$$f_{in}(v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega}) = \frac{1}{4\pi}\, g(v' \to v) \tag{4}$$

where $g(v' \to v)dv$ is the probability that a neutron of speed v is scattered inelastically in the speed range v to $v + dv$.

Since neutrons can be scattered anisotropically in an elastic collision, and many of the simpler theories for calculating critical sizes are strictly valid for isotropic scattering in the laboratory system, a correction factor is applied to the cross section for elastic scattering. Subsequently, we shall see how this correction factor is obtained.

If $f_e(\mu)$ is the angular distribution of the scattered neutrons, where μ is the cosine of the angle between $\underset{\sim}{\Omega}$ and $\underset{\sim}{\Omega}'$, then the correction factor for the scattering cross section is:

$$1 - \int_{-1}^{1} \mu\, f_e(\mu) d\mu,$$

i.e. the effective elastic scattering cross section σ'_e is given in terms of σ_e the cross section for elastic scattering by:

$$\sigma'_e = \sigma_e \left(1 - \int_{-1}^{1} \mu\, f_e(\mu) d\mu\right) \tag{5}$$

The effective cross section for both scattering processes, i.e. the transport cross section, is then defined to be:

$$\sigma_{tr} = \sigma'_e + \sigma_{in} \tag{6}$$

The expression for $f(\underset{\sim}{r}; v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})$, in the case of a homogeneous medium, can be written:

$$f = \frac{\alpha}{\beta}\left\{\nu\, p_f f_f + p_{in} f_{in} + p_e f_e\right\} \tag{7}$$

where p_f, p_{in} and p_e are the probabilities that a collision results in fission, inelastic scattering, and elastic scattering.

A Simple Form of the Integral Equation of Transport Theory

Instead of formulating transport theory in terms of the Boltzmann integro-differential equation (1), it can be expressed in terms of integral equations. The details can be found in Davison [1]. There is, however, a particularly simple form of the integral equation approach to transport theory which it is instructive to consider. If the neutrons are all of one speed, or the cross sections are independent of energy, the neutron flux $\Phi(\underset{\sim}{r})$ at a point $\underset{\sim}{r}$ satisfies the integral equation:

$$\Phi(\underset{\sim}{r}) = \frac{c\alpha}{4\pi}\int_{\text{reactor}} \Phi(\underset{\sim}{r}') \frac{e^{-\alpha|\underset{\sim}{r}-\underset{\sim}{r}'|}}{|\underset{\sim}{r}-\underset{\sim}{r}'|^2}\, dV'$$

where α is the inverse mean free path and c is the number of secondaries per collision. It is assumed in (8) that the medium is homogeneous and scattering is isotropic in the laboratory system. The extension of (8) to the case of a core surrounded by a reflector is given by Davison [2], Fuchs [3] and Frankel and Goldberg [4]. If the scattering is anisotropic, a first approximation is to retain the functional form of (8) but to use a modified α. Instead of using the scattering cross section to calculate α, one uses the transport cross section.

DIFFUSION THEORY

To determine the critical size of a reactor with reasonable accuracy, it is often essential to know the distribution of the neutrons in space and energy. In diffusion theory the maxim ' divide and rule ' is adopted. First of all the migration of neutrons in space is considered, assuming that all the neutrons have either the same speed or the cross sections are independent of speed (Davison [1]; Case, de Hoffman and Placzek [5]). This idealization leads to one-group diffusion theory. Secondly, a determination is made of the energy distribution of neutrons from a fission source which is uniformly distributed over an infinite moderator and it is discussed in some detail by Weinberg and Wigner [6], Glasstone and Edlund [7]. Thirdly, the spatial distribution of the neutrons, as a result of slowing down, is determined. One approach leads to the Fermi age theory [6, 7]. The Fermi age treatment is strictly applicable to slowing down in media other than those containing very light nuclei. There is also a computational difficulty which has tended to restrict the use of age theory to bare reactors. The difficulty arises because the multiplying and slowing down properties of a reflector are, in general, different from those of the reactor core. The neutron energy spectrum, which is fairly uniform throughout a bare reactor, can change markedly in the neighbourhood of a core-reflector interface. The computational difficulty can be overcome if direct numerical methods are employed. These methods (Mandl and Howlett [8], Hurwitz and Ehrlich [9]) can be useful where there are good computing facilities. The final stage in the development of diffusion theory brings in the concept of many energy groups. In an m group theory the slowing down of neutrons is accounted for by an approximate method for which the energy range is divided into m discrete intervals or ' groups '. The neutrons in each group are regarded as diffusing without energy loss until they have undergone a number of collisions which, on the average, would decrease their energy so that they enter the next lower energy group*. In discussing the slowing down of neutrons it is often convenient

* In a further generalization a neutron can enter any of the lower energy groups.

to express the energy in a logarithmic, dimensionless form by using the quantity lethargy or logarithmic energy decrement. Thus for example, if u_g is the width of the gth lethargy interval, then the average number of collisions a neutron requires to traverse the pth interval is u_g/ξ where ξ is the average logarithmic energy decrement per collision. Since the number of collisions per second in the gth group can be written as $\Sigma_g \Phi_g(\underset{\sim}{r})$ at the point with position vector $\underset{\sim}{r}$, if the neutron flux for the gth group is $\Phi_g(\underset{\sim}{r})$ and Σ_g is the average scattering cross section for the gth group, the number of gth group neutrons which become $g+1^{th}$ group neutrons per second is:

$$\frac{\Sigma_g \xi}{u_g} \Phi_g(\underset{\sim}{r}).$$

Thus, the quantity $\Sigma_g \xi/u_g$ can be shown to play the rôle of an absorption cross section, as it gives the rate at which p^{th} group neutrons are removed to become neutrons of group $p=1$. In this way the multi-group model of diffusion theory, in principle, converts the general transport problem for neutrons, with energy and spatial co-ordinates as independent variables, into a system of coupled partial differential equations with the spatial co-ordinates as the only independent variables.

One-Group Diffusion Theory

There are two well-known ways of developing a one-group diffusion theory granted that the scattering is isotropic in the laboratory system, viz. a physical approach and a mathematical treatment based on the integral equation of transport theory. The first form is often called elementary diffusion theory and the second is usually referred to as modified or infinite-medium diffusion theory. If, as is sometimes true, the scattering is isotropic in the centre of mass system, the assumption of isotropic scattering in the laboratory system is only a good approximation for heavy nuclei*. A correction is sometimes made to elementary diffusion theory to allow for the scattering being isotropic in the centre of mass system. The diffusion coefficient of the elementary theory is divided by $1-\bar{\mu}_0$, where $\bar{\mu}_0$ is the average cosine of the scattering angle in the laboratory system. The correction factor is got [6, 7] by comparing the elementary theory with a diffusion theory which is obtained as a first approximation to the spherical harmonic form of the Boltzmann equation, which is sometimes called the ' second form of Boltzmann's equation ' [6] to distinguish it from the form (1). The correction factor is obtained assuming that the medium is not a strong absorber.

One-group modified diffusion theory can be obtained by considering solutions of the integral equation (8) when the region of integration extends over the whole of space. The neutron flux $\Phi(\underset{\sim}{r}')$ is expanded as a Taylor series about the point $\underset{\sim}{r}$. Substituting the Taylor series in (8) and integrating gives:

$$\Phi = \frac{\beta}{\alpha}(\Phi + \nabla^2\Phi/3\alpha^2 + \nabla^4\Phi/5\alpha^4 + \ldots\ldots) \qquad (9)$$

This equation is satisfied by:

$$\left.\begin{array}{l} \nabla^2\Phi + k^2\Phi = 0 \text{ for a multiplying medium} \\ \text{and} \\ \nabla^2\Phi - \mu^2\Phi = 0 \text{ for an absorbing medium} \end{array}\right\} \qquad (10)$$

* For incident neutrons above 1 MeV, scattering by most nuclei is usually anisotropic in the centre of mass system.

provided

$$\left.\begin{aligned} k/\alpha &= \tan(k/\beta) \\ \mu/\alpha &= \tanh(\mu/\beta) \end{aligned}\right\} \quad (11)$$

For slightly multiplying media (β/α a little greater than unity) and for slightly absorbing media (β/α a little less than unity) the formulae (11) reduce to:

$$\left.\begin{aligned} k^2 &\simeq 3\alpha(\beta - \alpha) \\ \mu^2 &\simeq 3\alpha(\alpha - \beta) \end{aligned}\right\} \quad (12)$$

The approximate formulae (12) for k and σ can also be obtained by solving the Boltzmann equation in the P_1 spherical harmonic approximation (see page 80).

Davison [2] has shown that the neutron current vector $j(\underset{\sim}{r})$ at any point $\underset{\sim}{r}$ in the infinite medium is given by:

$$j(\underset{\sim}{r}) = -\,\mathrm{D}\ \mathrm{grad}\ \Phi(\underset{\sim}{r}) \quad (13)$$

where

$$\left.\begin{aligned} \mathrm{D} &= (\beta - \alpha)/k^2 \text{ for a multiplying medium} \\ \mathrm{D} &= (\alpha - \beta)/\mu^2 \text{ for an absorbing medium} \end{aligned}\right\} \quad (14)$$

For weakly absorbing and multiplying systems, (14) become approximately:

$$\mathrm{D} = 1/3\alpha \quad (15)$$

It is reasonable to expect that the infinite medium equations can be used to give fairly good results on the critical size and the neutron flux distribution for reactors which are large compared with the mean free path. This intuitive conclusion is confirmed by a rigorous analysis [1] based on the one-group Boltzmann equation (1), which shows that the exact solution for the neutron flux in a uniform isotropically-scattering medium can always be represented in the form:

$$\Phi(\underset{\sim}{r}) = \Phi_{as}(\underset{\sim}{r}) + \Phi_{tr}(\underset{\sim}{r}) \quad (16)$$

The ' asymptotic solution ' $\Phi_{as}(\underset{\sim}{r})$ satisfies one of the infinite medium equations (10), and the transient term $\Phi_{tr}(\underset{\sim}{r})$ is given by:

$$\Phi_{tr}(\underset{\sim}{r}) = 0(e^{-\alpha d}) \quad (17)$$

where d is the distance of the point from the nearest source or boundary. Thus, if the dimensions of the region considered are large compared with the mean free path, then $\Phi_{tr}(\underset{\sim}{r})$ can be neglected in comparison with $\Phi_{as}(\underset{\sim}{r})$, except in the neighbourhood of sources and boundaries. The modified diffusion equations are solved for each region of the reactor. The solutions for the separate regions are then matched by imposing boundary conditions (to be discussed) at the interfaces of regions and at a bare surface. The resulting equations give rise to an eigenvalue problem for the critical size. The requisite eigenvalue is distinguished from the others since it is the smallest, and its associated eigenfunction is non-negative.

Boundary Conditions

Interfaces. To illustrate the boundary conditions that arise, consider a simple reactor consisting of a weakly multiplying core surrounded by a weakly absorbing reflector. In this case the approximate boundary conditions are (using suffixes c and r to indicate core and reflector quantities respectively):

$$\left.\begin{aligned} \Phi_c &= \Phi_r \\ \frac{1}{\alpha_c}\,\mathrm{grad}\,\Phi_c &= \frac{1}{\alpha_r}\,\mathrm{grad}\,\Phi_r \end{aligned}\right\} \tag{18}$$

These boundary conditions are a special from of Davison's [2] boundary conditions for modified diffusion theory. The same result as (18) is obtained if the Boltzmann equation is solved to the P_1 approximation (see page 80) under the assumption that scattering is isotropic in the laboratory system.

If the scattering is not isotropic in the laboratory system the last member of (18) can be replaced by:

$$D_c \frac{\partial \Phi_c}{\partial_n} = D_r \frac{\partial \Phi_r}{\partial_n} \tag{19}$$

where

$$\left.\begin{aligned} D_c &= 1/3\alpha_c(1 - \bar{\mu}_c) \\ D_r &= 1/3\alpha_r(1 - \bar{\mu}_r) \end{aligned}\right\} \tag{20}$$

and n is in a direction normal to the interface.

Bare surface. There are two well-known approximate formulations of the boundary condition for a bare surface (convex), viz. that no neutrons cross the surface inwards. They are the ' extrapolated end point ' and the ' linear extrapolation distance ' conditions. The first states that the function representing the flux in the reactor should vanish at a distance d_0 in free space from the bare surface. Over a wide range of β/α, i.e. c, the quantity $\beta\, d_0$ in Table I is almost constant.

TABLE I

c	βd_0
0	1.000
0.5	0.721
1.0	0.710
3.0	0.720
∞	0.750

The second form of the boundary condition states that at the boundary the ratio of the flux to the normal derivative of the flux is a constant, i.e.:

$$\left\{ \Phi \Big/ \frac{\partial \Phi}{\partial_n} \right\}_{\textit{bare surface}} = -d \tag{21}$$

The extrapolation distance d can be shown from transport theory [1, 5, 6, 7] to be given by the semi-empirical result $0.710\ \lambda_t$, where λ_t is the transport mean free path.

Multi-Group Diffusion Theory

The P_1 approximation of the multi-group version of the spherical harmonics form of the Boltzmann equation (1) has been used by Mandl [10, 11] to obtain a simple multi-group diffusion theory. Assuming that the scattering is isotropic in the laboratory system, the multi-group equations for spherical systems are of the form:

$$-\frac{1}{3\alpha_g}\frac{d^2}{dr^2}(r\Phi_g) + r\alpha_g\Phi_g = r\sum_{g'}\gamma_{g'g}\Phi_g + rF_g\sum_{g'}\alpha_{f,g'}\Phi_{g'} \tag{22}$$

for the flux Φ_g in the gth group, for which the inverse mean free path is α_g. In (22) $\gamma_{g'g}$ is the cross section per unit volume for transfer from group g' to group g by scattering, and γ_{gg} is the cross section per unit volume for the neutrons scattered in this group to remain in this group. The number of fission neutrons liberated in a particular energy group g is

$$F_g \sum_{g'} \alpha_{,} \Phi_{g'}$$

where $\alpha_{f,g}$ is the cross section per unit volume for neutrons in group g and F_g is the number of neutrons in the fission spectrum which are emitted in that group.

Alternatively the multi-group equations are derived directly from physical considerations. The same assumptions are made as regards the leakage and absorption terms in the continuity equation for each energy group as are used in deriving one-group diffusion theory. The contribution of fission neutrons to the sources for each energy group can be expressed in a similar way to that used in (22). In each continuity equation excepting that for the lowest energy group there is also a sink due to the removal of neutrons by slowing down.

There is also for each group, excepting the highest, a source term representing the influx of those neutrons to the group which have been slowed down from high-energy groups. The strength of the slowing down sources are calculated on the lines indicated in the introductory section on diffusion theory. In calculating the leakage term the diffusion coefficient has to be corrected for anisotropic scattering if this is significant.

The simplest form of multi-group diffusion theory is the well-known two-group theory, which can be very useful, despite its simplicity, if carefully applied. For a reactor with a reflector, the two-group diffusion equations often take the form [7]:

Core

$$\left.\begin{aligned} D_{1c}\nabla^2\Phi_{1c} - \Sigma_{1c}\Phi_{1c} + k\Sigma_{2c}\Phi_{2c} &= 0 \text{ for fast neutrons} \\ D_{2c}\nabla^2\Phi_{2c} - \Sigma_{2c}\Phi_{2c} + \Sigma_{1c}\Phi_{1c} &= 0 \text{ for thermal neutrons} \end{aligned}\right\} \quad (23)$$

Reflector

$$\left.\begin{aligned} D_{1r}\nabla^2\Phi_{1r} - \Sigma_{1r}\Phi_{1r} &= 0 \text{ for fast neutrons} \\ D_{2r}\nabla^2\Phi_{2r} - \Sigma_{2r}\Phi_{2r} + \Sigma_{1r}\Phi_{1r} &= 0 \text{ for thermal neutrons} \end{aligned}\right\} \quad (24)$$

For (23) and (24), fast neutron absorption has been neglected and Σ_{1c} and Σ_{1r} are the slowing down cross sections for fast neutrons. The absorption cross sections for thermal neutrons are Σ_{2c} and Σ_{2r} for the core and reflector respectively. The number of fast neutrons produced by fission is $k\Sigma_{2c}\Phi_{2c}$ per cm^3 per second. Resonance capture of neutrons can be allowed for by including the resonance escape probability in the calculation of k. If the reflector is infinite, the boundary conditions are simply:

$$\left.\begin{aligned} \Phi_{1c} &= \Phi_{1r} \\ \Phi_{2c} &= \Phi_{2r} \\ D_{1c}\frac{\partial\Phi_{1c}}{\partial_n} &= D_{1r}\frac{\partial\Phi_{1r}}{\partial_n} \\ D_{2c}\frac{\partial\Phi_{2c}}{\partial_n} &= D_{2r}\frac{\partial\Phi_{2r}}{\partial_n} \end{aligned}\right\} \quad (25)$$

at the core reflector interface. Solutions of (23) are obtained [7] by expressing Φ_{1c} and Φ_{2c} as linear combinations of the solutions of:

$$\nabla^2\Phi + B^2\Phi = 0 \tag{26}$$

where B^2 satisfies

$$\left.\begin{aligned}(1 + L^2B^2)(1 + L_s^2B^2) &= k \\ L^2 = \frac{D_{2c}}{\Sigma_{2c}} \qquad L_s^2 &= \frac{D_{1c}}{\Sigma_{1c}}\end{aligned}\right\} \tag{27}$$

These linear combinations involve coupling coefficients, usually denoted by S_1 and S_2 for the core. This conventional two-group analysis has been described in some detail because there is an almost trivial modification of it [12] which is very useful. The above two-group analysis is suitable only for well-moderated systems because it neglects fission in the fast neutron group. If this omission is rectified by including a term $(\eta_1 - 1)\Sigma_1'$* in the fast group equation of (23), the buckling relation (27) becomes:

$$\left.\begin{aligned}(1 + L^2B^2)(1 + L_s^2B^2 - \tau) &= k \\ \text{where} \qquad \tau &= (\eta_2 - 1)\frac{\Sigma'_{1c}}{\Sigma_{1c}}\end{aligned}\right\} \tag{28}$$

The parameter τ is a measure of the hardness of the neutron spectrum; it is about 0.004 in a well-thermalized core (note that $\tau = 0$ yields the conventional two-group equations) and about 1,000 in a pure metal system. The coupling coefficients are almost the same as those of conventional theory; they are:

$$S_1 = \frac{p\Sigma_{1c}}{\mu^2 D_{2c} + \Sigma_{2c}}, \qquad S_2 = \frac{p\Sigma_{1c}}{\Sigma_{2c} - \nu^2 D_{2c}} \tag{29}$$

where p is the resonance escape probability. The coefficient S_2 is difficult to calculate accurately from (29) for other than well-thermalized systems, because its numerator and denominator become very small. It is better to use:

$$\left.\begin{aligned}S_2 &= p\frac{\Sigma_{1c}}{D_{2c}Q} \\ \text{where} \qquad Q &= \frac{\tau - 1}{L_s^2} - (\eta_2 + \tau - 1)/(L_s^2 + (1-\tau)L^2)\end{aligned}\right\} \tag{30}$$

As the spectrum becomes harder the thermal group parameters become insignificant in Thornton's analysis, so that the fact that they are ill-defined is then unimportant. In the case of a solid metal core,

$$\tau \gg \eta_2 - 1, \text{ so that } \tau \text{ appears as } \frac{\tau}{L_s^2}, \text{ i.e. } (\eta_1 - 1)\frac{\Sigma'_{1c}}{D_{1c}};$$

Σ_{1c} has then no effect on the analysis. The value of Thornton's method is that it provides a simple bridge between conventional two-group diffusion theory for well-thermalized systems and the albedo method, which uses in its simplest form one group in the core and two groups in the reflector, for dealing with fast cores and moderating reflectors.

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* η_1, and subsequently η_2, are used for the value of η for fast and thermal neutrons respectively.

Before discussing albedo or 'one-and-a-half group' methods, there are two other simple applications of diffusion theory which are of very considerable importance; Buckling Conversions and Lattice Theory. The latter is so well known as a part of reactor theory that we refer the reader who is interested in criticality problems involving lattices to the standard works [6, 7] and reviews [13, 14].

Buckling Conversions

There is an important result for bare reactors which is often useful when problems of class (*ii*) are considered. The conditions for the result to be valid are discussed in detail by Weinberg and Wigner [6]. The first fundamental theorem of reactor theory can be stated as follows.

The stationary neutron distribution $\Phi(\underset{\sim}{r}, E)$ in a critical bare reactor is separable in space and energy:

$$\Phi(\underset{\sim}{r}, E) = q(E)\psi(\underset{\sim}{r}) \tag{31}$$

the spatial distribution $\psi(\underset{\sim}{r})$ is the fundamental solution of the wave equation:

$$\nabla^2\psi(\underset{\sim}{r}) + B^2\psi(\underset{\sim}{r}) = 0 \tag{32}$$

that is, the solution which is positive throughout the reactor and vanishes on the extrapolated boundary.

The theorem simplifies the calculation of the critical sizes of bare reactors with shapes for which the wave equation can be solved either by the method of separation of variables or by the variational methods described on page 69. Furthermore, if one makes use of the concept of reflector savings [6, 7], changes in the critical size of reflected reactors with changes in shape are easily computed. The accuracy of the method has been discussed by Callihan [15].

Albedo Methods

The albedo method is a simple means of calculating sizes of small fast cores with moderating reflectors which was developed in the first place for fairly large cores, but which gives surprisingly good results for small cores. The theory is tantalizingly simple, and the method in its variational form demands no more than the solution of a simple quadratic equation for the determination of critical size.

As a rule, systems calculated to be critical by the simplest form of albedo method with the aid of diffusion theory are actually subcritical. A modification [16] of the method, however, calculates systems as critical when they are actually supercritical. We will content ourselves here with the form of the albedo method which often gives underestimates for critical size.

The albedo method for fast cores with moderating reflectors. In the simplest form of the albedo method, there are assumed to be fast and thermal groups of neutrons in the moderating reflector and a fast group in the core. The core and reflector are regarded as separated by an infinitesimally thin shell of core material which is black to thermal neutrons. For every thermal neutron absorbed on striking the shell, η fast neutrons are produced, half of which enter the core and half the reflector.

The albedo A for the core is defined as the ratio of the fast neutron current J_{fc}^- entering the core to the fast neutron current J_{fc}^+ leaving the core, i.e.:

$$A = J_{fc}^-/J_{fc}^+ \tag{33}$$

The thin shell and the reflector can be treated together with advantage by introducing the concept of the effective albedo for the reflector $A^\star$. The effective albedo is defined as the ratio of the fast neutron current J_{fu}^- returning to the core from the union of the shell and reflector to the fast neutron current J_{fu}^+ entering the union from the core, i.e.:

$$A^\star = J_{fu}^-/J_{fu}^+ \tag{34}$$

If J_{fr}^+ is the fast neutron current entering the reflector from the shell and J_{fr}^- is the fast neutron current returning to the shell from the reflector, then μ, the probability of a fast neutron entering the reflector to return as a fast neutron, is given by:

$$\mu = J_{fr}^-/J_{fr}^+ \tag{35}$$

Furthermore, if J_{tr}^- is the thermal neutron current returning to the shell from the reflector, then $\mu^\star$, the probability of a fast neutron entering the reflector to return as a thermal neutron, is:

$$\mu^\star = J_{tr}^-/J_{fr}^+ \tag{36}$$

Explicit expressions for μ and $\mu^\star$ in terms of reflector dimensions and composition can be obtained if it is borne in mind that J_{tr}^+, the thermal neutron current entering the reflector, is zero, i.e.:

$$J_{tr}^+ = 0 \tag{37}$$

The effective albedo $A^\star$ can now be expressed in terms of μ, $\mu^\star$ and η by considering the neutron multiplication in the shell. For the outward-bound stream of neutrons across the shell,

$$J_{fr}^+ = J_{fu}^+ + \tfrac{1}{2}\eta J_{tr}^- \tag{38}$$

and for the inward-bound stream,

$$J_{fu}^- = J_{fr}^- + \tfrac{1}{2}\eta J_{tr}^- \tag{39}$$

These equations give, with the definitions of $A^\star$, μ and $\mu^\star$:

$$A^\star = (\mu + \tfrac{1}{2}\eta\mu^\star)/(1 - \tfrac{1}{2}\eta\mu^\star) \tag{40}$$

The criticality condition for the albedo method is given by equating corresponding fast currents for the core and the union, which leads to the condition:

$$A = A^\star \tag{41}$$

The condition for criticality that the albedo for the core should equal the effective albedo of the reflector has been derived without specifying the laws for neutron migration in the core and the reflector. The laws chosen for neutron migration are in keeping with the albedo boundary conditions in that they are simple and reasonably accurate. For large cores, ordinary diffusion theory is assumed to hold for the core and the reflector, whereas for small cores modified diffusion theory is used for the core and ordinary diffusion theory for the reflector.

The fast flux ψ_0 in the core is assumed to satisfy:

$$\nabla^2\psi_0 + \lambda^2\psi_0 = 0 \tag{42}$$

The fast flux ψ and the thermal flux θ in the reflector are taken as obeying

$$\nabla^2\psi - \rho^2\psi = 0 \tag{43}$$

and

$$\nabla^2\theta - \omega^2\theta + \sigma^2\psi = 0 \tag{44}$$

For the present the parameters λ^2, ρ^2, ω^2 and σ^2 may be taken as given. Their expression in terms of the standard parameters of modified and ordinary diffusion theory is deferred until applications are considered.

The current ratios of (33), (35) and (36) can be written for both modified and ordinary diffusion theory as:

$$\left.\begin{aligned} A &= \left(\psi_0 + 2d_{of}\frac{\partial\psi_0}{\partial n}\right)_s \Big/ \left(\psi_0 - 2d_{of}\frac{\partial\psi_0}{\partial n}\right)_s \\ \mu &= \left(\psi + 2d_f\frac{\partial\psi}{\partial n}\right)_s \Big/ \left(\psi - 2d_f\frac{\partial\psi}{\partial n}\right)_s \\ \mu^* &= \left(\theta + 2d_t\frac{\partial\theta}{\partial n}\right)_s \Big/ \left(\psi - 2d_f\frac{\partial\psi}{\partial n}\right)_s \end{aligned}\right\} \tag{45}$$

where S is the surface of the core, and the d's are constants which are related to diffusion coefficients. The boundary condition (37) becomes:

$$\left(\theta - 2d_t\frac{\partial\theta}{\partial n}\right)_s = 0 \tag{46}$$

Granted the nuclear parameters d, the equations (40), (41), (45) and (46) may be used to calculate the critical size. For spheres, cylinders and slabs μ, μ^* are given in Table II below. In preparing the table, ordinary diffusion theory has been assumed to hold for the reflector, i.e.:

$$L\omega = 1, \qquad L_s\rho = 1, \qquad \sigma^2 = l_f/l_t L_s^2 \tag{47}$$

where L is the diffusion length for thermal neutrons in the reflector;
L_s is the slowing down length for the reflector;
l_f is the fast mean free path in the reflector;
l_t is the thermal mean free path in the reflector.

For the core, the Laplacian λ^2 is given according to modified diffusion theory by:

$$\lambda/\alpha = \tan(\lambda/\beta) \tag{48}$$

where α is the inverse mean free path of neutrons in the core and β is the mean number of secondaries produced per unit path.

TABLE II. μ, μ^* AND A FOR SPHERES, CYLINDERS AND SLABS

	μ	μ^*	A
Sphere	$1-2d_f(\rho a+1)/a$	$4r_1 d_f\rho^2/r_2$	$a+2d_{of}(\lambda a\cot\lambda a-1)$
	$1+2d_f(\rho a+1)/a$	$(\rho+\omega)\,[1+2d_f(\rho a+1)/a]\,[1+2d_t(\omega a+1)/a]$	$a-2d_{of}(\lambda a\cot\lambda a-1)$
Cylinder	$1-2d_f\rho\,\dfrac{K_1(\rho a)}{K_0(\rho a)}$	$4\dfrac{r_1}{r_2}d_f\rho^2\left(\omega\dfrac{K_1(\omega a)}{K_0(\omega a)}-\rho\dfrac{K_1(\rho a)}{K_0(\rho a)}\right)$	$1-2d_{of}\lambda\dfrac{J_1(\lambda a)}{J_0(\lambda a)}$
	$1+2d_f\rho\,\dfrac{K_1(\rho a)}{K_0(\rho a)}$	$(\omega^2-\rho^2)\left(1+2d_f\rho\dfrac{K_1(\rho a)}{K_0(\rho a)}\right)\left(1+2d_t\omega\dfrac{K_1(\omega a)}{K_0(\omega a)}\right)$	$1+2d_{of}\lambda\dfrac{J_1(\lambda a)}{J_0(\lambda a)}$
Slab	$\dfrac{1-2d_f\rho}{1+2d_f\rho}$	$\dfrac{4\dfrac{r_1}{r_2}d_f\rho^2}{(\rho+\omega)(1+2d_f\rho)(1+2d_t\omega)}$	$\dfrac{1-2d_{of}\lambda\tan\lambda a}{1+2d_{of}\lambda\tan\lambda a}$

In the table, a denotes either the radius or half-thickness and the d's are coefficients given by:

$$\left.\begin{aligned} r_1 &= \frac{1_f}{3d_f} = 1 \\ r_2 &= \frac{1_t}{3d_t} = \frac{2}{(3 \times 0.71)} \\ r_3 &= \frac{1}{3\alpha d_{of}} = 1 \end{aligned}\right\} \tag{49}$$

As an illustration of the use of the albedo method, consider the following calculation for the critical size of a water-reflected sphere of U 235 metal. The nuclear parameters for the core and reflector are given in Table III below. The parameters for the reflector are fairly standard. The parameters for the core are based on averages over the fission spectrum.

TABLE III. NUCLEAR PARAMETERS USED IN ILLUSTRATION OF ALBEDO METHOD

	$L_s(cm)$	$L(cm)$	$l_f(cm)$	$l_t(cm)$
Water reflector	5.51	2.88	3.26	0.55
Graphite reflector	20.00	50.00	3.60	2.40
Core	$\sigma_{sc} = 5.85$ barns $\sigma_f = 1.31$ barns $\sigma_c = 0.15$ barns		$\alpha = 0.3465$ $\beta = 0.4294$	

The calculated critical radius is 5.88 cm, which gives a calculated critical mass of 16 kg for an infinite water reflector. This is a safe answer and not too conservative considering the simplicity of the method, the experimental figure being not more than 24.7 kg. At this stage it is pertinent to look for possible sources of the discrepancy between theory and experiment within the simple structure of the albedo method.

If the core parameters in Table III are used in calculating the critical mass of a bare sphere of metallic U 235, the albedo method gives, on putting μ and μ^{**} to zero, a figure of 35.25 kg. The experimental critical mass for a bare sphere is 52 kg. This suggests that the nuclear parameters of Table III for the fast group in the core may be inappropriate for the albedo method. This possibility is supported by the following calculation. Given the experimental figure for the critical radius of a bare sphere of metallic U 235, one can calculate by the albedo method the requisite Laplacian λ^2 for agreement between theory and experiment. Since λ^2 is defined in terms of α (the inverse mean free path of neutrons in the core), and β (the mean number of secondaries produced per unit path), it is the same for all reactors of the same composition whether they are bare or reflected. On using the Laplacian derived from the experimental value for the critical radius of a bare sphere of metallic U 235, the albedo method gives critical masses of 22.2 kg and 34.8 kg for unplated

* For the cadmium-plated sphere μ^* is taken to be zero.

and cadmium-plated* spheres of metallic uranium with water reflectors, whereas the experimental figures are 24.7 kg and 35 kg. Agreement between experiment and the albedo method is now fairly good.

Enough has perhaps been said for the present in outlining a useful method for obtaining the Laplacian. A detailed comparison of the predictions of the albedo method with experiment is given later for some U 235 and Pu 239 cores in the forms of cylinders, spheres and slabs (cadmium-plated and unplated) for reflectors of water, graphite and beryllium.

Multi-group version of the albedo method. The accuracy of the albedo method can be improved by taking more energy groups in the core and the reflector than are used in the simple albedo method. In one version [17] of the albedo method there are two groups in the core and four groups in the reflector. Coupling of the groups due to fission and the slowing down of neutrons are indicated in Figure 1 by heavy black and white arrows respectively. The thin shell on the surface of the core is transparent to all fast neutrons, which are contained in groups 0 and 1, but is black to the intermediate and thermal neutrons occupying groups 2 and 3. Neutrons in groups 2 and 3 on being absorbed by the shell produce fast neutrons, and the yields per neutron absorbed are η_2 and η_3 for groups 2 and 3. The groups 0 and 1 are assumed to span the fission spectrum in such a way that a fraction p of fission neutrons are born in group 0. The assumption is made that the remaining fraction of the fission neutrons are, for practical purposes, born in group 1. The diffusion equations and boundary conditions for the multi-group albedo method are shown in Figure 1. The suffices c, r, u stand for the core, reflector and the union of the shell and reflector respectively. In the core $\hat{p}\,\lambda_1^2\,\psi_{1c}$ represents the coupling due to fission of group 1 into group 0. Group 0 is coupled into 1 through the fission term $(1-\hat{p})\lambda_0^2\,\psi_{0c}$ and the slowing down term $\gamma_{01}^2\,\psi_{0c}$. The coupling between energy groups in the reflector should be clear from the diagram. The coefficients in the core equations can be calculated either on the modified or two-group diffusion theory. The distribution of the yields of fission neutrons, which are produced in the shell by neutrons of groups 2 and 3 from the reflector, is indicated in the diagram. Note that no neutrons in groups 2 and 3 enter the reflector from the shell since the shell is assumed to be a perfect absorber for neutrons in these groups.

Regarding the shell and reflector as a black box with inputs J_{0u}^+ and J_{1u}^+ and outputs J_{0u}^- and J_{1u}^- one can write:

$$J_u^- = A^\star J_u^+ \tag{50}$$

where J_u^- denotes the column vector with elements (J_{u0}^-, J_{1u}^-) and J_u^+ is the column vector with elements (J_{0u}^+, J_{1u}^+). The form of the matrix $A^\star$ will be explained later. The matrix operator $A^\star$ will be called the effective albedo for the reflector.

For the core regarded as a black box one can write:

$$J_c^- = A\,J_c^+ \tag{51}$$

where J_c^- denotes the column vector with elements (J_{c0}^-, J_{c1}^-) and J_c^+ is the column vector with elements (J_{0c}^+, J_{c1}^+). The form of the matrix A will be explained later. The matrix operator A will be called the albedo for the core. Note that the inverse of A gives the surface multiplication matrix operator.

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* For the cadmium-plated sphere $\mu^\star$ is taken to be zero.

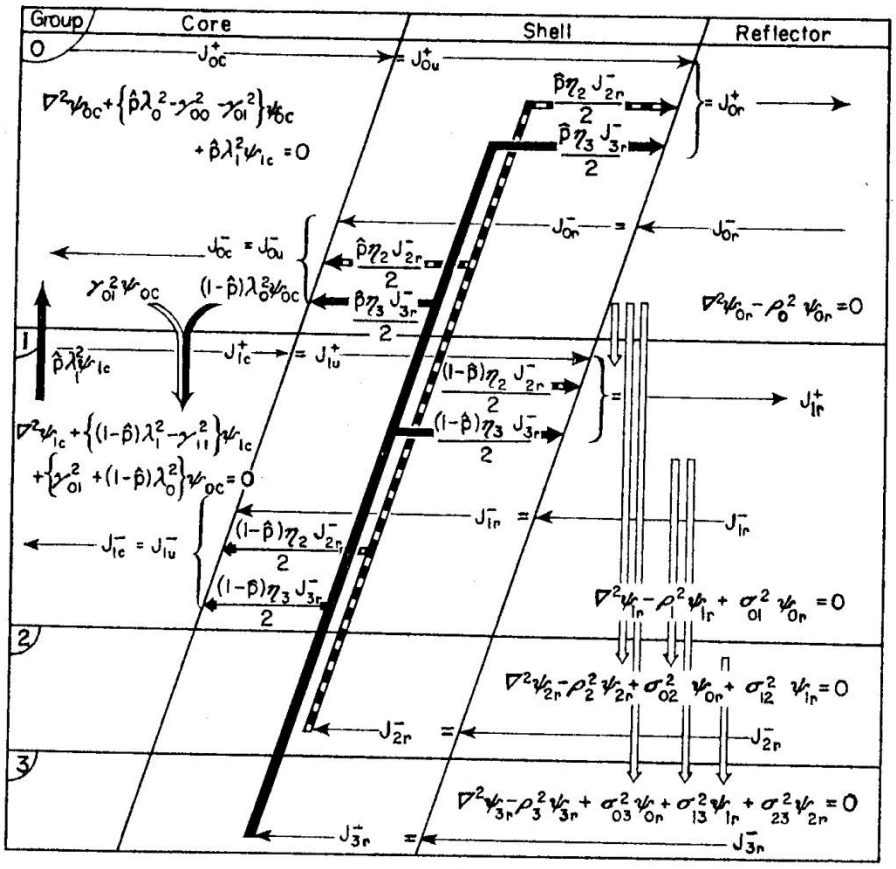


Figure 1. *Multigroup version of the Albedo Method.*

The critical condition occurs when $J^+ = J_u^+$ and $J_c^- = J_u^-$ which gives $A = A^\star$ as the condition for criticality.

If J_r^- is the column vector (J_{0r}^-, J_{1r}^-), and $J_r^{\star-}$ is the column vector (J_{2r}^-, J_{3r}^-) and J_r^+ is the column vector $(J_{0r}^+, J_{1r}^+)^\star$, then the behaviour of the reflector can be expressed by:

$$\left.\begin{aligned} J^- &= M\ J_r^+ \\ J_r^{\star-} &= M^\star J_r^+ \end{aligned}\right\} \tag{52}$$

where M and $M^\star$ are 2×2 matrices whose coefficients depend upon the reflecting material. The shell equations can be simply expressed in the matrix form:

$$\left.\begin{aligned} J_r^+ &= J_u^+ + \frac{\hat{P}H}{2} J_r^{\star-} \\ \text{and} \\ J_u^- &= J_r^- + \frac{\hat{P}H}{2} J_r^{\star-} \end{aligned}\right\} \tag{53}$$

TABLE IV. COMPARISON OF ALBEDO METHODS WITH EXPERIMENTAL RESULTS

Assembly	U 235 Critical Radius or Semi-Thickness *(cm)*				Pu 239 Critical Radius or Semi-Thickness *(cm)*		
	Calculated		Experimental	Monte Carlo	Calculated		Experimental
	Simple Albedo	Multi-group Albedo			Simple Albedo	Multi-group Albedo	
Bare sphere	8.7*	8.33	8.7		6.26*	6.69	6.26
cylinder	6.31	6.24	6.22 5.71 □		4.48	4.45	△
slab	3.67	△	○ 3.05 3.05 □	3.285	2.47	2.46	△
Water-reflected:							
sphere	6.56	6.96	6.79		4.85	4.85	4.95
cylinder	3.88	3.87	3.68 4.06 □		2.73	2.89	△
slab	0.91	△	0.95 0.93 □	0.835	0.49	△	△
Water-reflected Cd-plated:							
sphere	7.62	△	7.63		5.59	△	5.33
cylinder	4.84	△	△		3.5	△	△
slab	2.13	△	△		1.28	△	△
Graphite-reflected:							
sphere	5.85	6.81	6.12		4.47	5.12	4.44
cylinder	2.36◇	3.35	2.92		1.74◇	2.54	△
slab	0.6	△	0.63		0.38	△	△
Graphite-reflected Cd-plated:							
sphere	6.45	△	△		4.88	△	△
cylinder	3.6	△	△		2.55	△	△
slab	0.74	△	△		0.40	△	△

* Tied to experimental bare mass.
△ Not available.
◇ The radius here is sufficiently small for the standard boundary condition of the albedo method to require modification. If the factor 0.71 is replaced by 0.94 the critical radius is 2.41 cm.
○ Guided extrapolation of experiments.
Experimental values have been taken from a report by Paxton[19]. The Monte Carlo results are from Woodcock.
□ Experimental values (private communication) Paxton[20].

where the 2×2 matrices $\hat{\mathrm{P}}$ and H are given by:

$$\hat{\mathrm{P}} = \begin{bmatrix} \hat{p} & 0 \\ (1-\hat{p}) & 0 \end{bmatrix} \tag{54}$$

$$\mathrm{H} = \begin{bmatrix} \eta_2 & \eta_3 \\ 0 & 0 \end{bmatrix} \tag{55}$$

If unity is not an eigenvalue of $\hat{\mathrm{P}}\mathrm{H}\,\mathrm{M}^\star/2$ one can eliminate J_r^+, J_r^- and $\mathrm{J}^\star{}_r^-$ from the equations (52) and (53); comparing the result with (50) gives:

$$\mathrm{A}^\star = \left(\mathrm{M} + \frac{\hat{\mathrm{P}}\mathrm{H}}{2}\,\mathrm{M}^\star\right)\left(\mathrm{I} - \frac{\hat{\mathrm{P}}\mathrm{H}}{2}\,\mathrm{M}^\star\right)^{-1} \tag{56}$$

where I is the unit matrix.

Some results of simple and multi-group albedo calculations are compared in Table IV with the experimental results (where known) for spheres, cylinders and slabs of uranium and plutonium with water and graphite reflectors. Further details are given by Ackroyd and Ball [18]. These results suggest that the use of diffusion theory in conjunction with suitably chosen mathematical models can give estimates of the critical size of fast systems with moderating reflectors rather better than is commonly supposed. The albedo methods are often useful for checking and interpolation work, and they are particularly useful for interaction calculations since they are simple and have a fair accuracy.

Finite Medium Theory

Although classical diffusion theory has been justified only for large systems it gives surprisingly good results for the critical size of some fairly small systems. Of course, flux plots obtained by diffusion theory are sometimes significantly in error near interfaces of regions, since the flux near a region can be shown to have two components: the infinite medium solution of diffusion theory, i.e. modified diffusion theory; and a transient term which dies out in a distance from the interface of a few mean free paths. The great attactions of diffusion theory are its mathematical simplicity and its ability, if skilfully used, to give good answers well outside its operated range of validity. Apart from diffusion theory the only method that has been developed in detail for systems of awkward shape is the Monte Carlo method (see page 92), which unfortunately requires lengthy computations.

A renewed attempt has been made recently by Ackroyd *et al* [18, 21, 22] to extend the justifiable range of diffusion theory by taking into account: (*a*) the size of the system; and (*b*) the effects of interfaces. It is true that the effect (*b*) has been allowed for in a mathematical way by the well-known Serber-Wilson method [1], but this method violates the physical fact that the neutron density is continuous at an interface. The method, moreover, assumes that apart from the discontinuity of the flux at interfaces, the flux is given within the regions by the infinite medium solution, i.e. there is no consideration of fine detail in the flux behaviour at interfaces. The limitation of the Serber-Wilson method is that it is restricted to spherical systems.

In finite medium theory, which applies at present to spherical, slab and finite cylindrical systems, the system is divided into a number of domains

which may be greater than, equal to or less than the number of physical regions. The simplest case arises when each domain corresponds to a physical region. Boundary transients can be treated by assigning two domains to each region; one to the central part of the region, the other to the neighbourhood of the surface of the region. A number of small regions can be homogenised by assigning one domain to cover all the regions. The method is outlined for one-group systems, and the extension of the method to two-group systems is then sketched.

For each domain V_k of a one-group system there is assigned a Laplacian and diffusion coefficient D_k and a coefficient B_k. The inverse mean free path and number of secondaries per collision are denoted by α_k and c_k respectively. The values given to α_k and c_k are those appropriate to the material in V_k (if a domain extends over more than one region, then α_k and c_k vary with the composition of V_k). In V_k the neutron density is assumed to satisfy an equation of the form, either

$$\left.\begin{aligned} &\nabla^2 n + \lambda_k^2 n = 0 \\ \text{or}\quad &\nabla^2 n - \mu_k^2 n = 0 \end{aligned}\right\} \tag{57}$$

The neutron density and the neutron current are assumed to be continuous at the interfaces of V_k. The outward and inward currents are defined only at the interfaces, by the expressions of the form:

$$\left.\begin{aligned} J_k^+ &= v\left(B_k n - \frac{D_k}{2}\frac{\partial n}{\partial u}\right) \\ J_k^- &= v\left(B_k n + \frac{D_k}{2}\frac{\partial n}{\partial u}\right) \end{aligned}\right\} \tag{58}$$

where u is the co-ordinate in the direction of the positive normal to the interface. The component of the angular neutron density for a point on an interface, and for the direction of the inward normal at that point, is assumed to be given in terms of the inward current, by an expression which is satisfied exactly if the angular neutron density is isotropic at the point for those directions inwards to the interface.

Imposing the conditions that:

i) The neutron density, according to finite medium theory, satisfies at the centre the integral equation of transport theory for the neutron density;

ii) The finite medium expression for the current at the centre satisfies the integral relation of transport theory;

iii) The net production of neutrons in V_k equals the net loss across the interfaces;

gives:

$$B_k = \frac{1}{4} \tag{59}$$

$$D_k = \alpha_k(c_k - 1)\lambda_k^2 \text{ or } \alpha_k(1 - c_k)\mu_k^2 \tag{60}$$

and formulae for λ_k and μ_k in terms of α_k, c_k and the *size* of V_k. The formula (60) is of the same form as that obtained by Davison [2] but D_k in (60) depends

on λ_k (or μ_k), and hence is dependent on the size of the system. When the region V_k becomes large the formulae of finite medium theory reduce to those of modified diffusion theory. Typical formulae for the Laplacians are given in Table V.

TABLE V. LAPLACIANS ACCORDING TO FINITE MEDIUM THEORY

System	Laplacian
Absorbing sphere of radius a_1 inverse mean free path α_0, secondaries per collision $c_0 < 1$	Laplacian μ_0 given by $1 = e^{-\alpha_0 a_1}\left\{\frac{\sinh \mu_0 a_1}{\mu_0 a_1}\left(1 + 2\frac{\alpha_0(c_0-1)}{\mu_0^2 a_1}\right) - 2\frac{\alpha_0(c_0-1)}{\mu_0^2 a_1}\cosh \mu_0 a_1\right\} + \alpha_0\left[\log\left(\frac{\alpha_0+\mu_0}{\alpha_0-\mu_0}\right) + E_i\{a_1(\mu_0-\alpha_0)\} - E_i\{-a_1(\mu_0+\alpha_0)\}\right]$ if $\alpha_0 > \mu_0$ $1 = e^{-\alpha_0 a_1}\left\{\frac{\sinh \mu_0 a_1}{\mu_0 a_1}\left(1 + 2\frac{\alpha_0(c_0-1)}{\mu_0^2 a_1}\right) - 2\frac{\alpha_0(c_0-1)}{\mu_0^2 a_1}\cosh \mu_0 a_1\right\} + \frac{\alpha_0 c_0}{2\mu_0}\left[E_i\{a_1(\mu_0-\alpha_0)\} - E_i\{-a_1(\mu_0+\alpha_0)\}\right]$ if $\mu_0 > \alpha_0$.
Multiplying sphere of radius a_1 inverse mean free path α_0 secondaries per collision $c_0 > 1$	Laplacian λ_0 given by $1 = \frac{\alpha_0 c_0}{\lambda_0}\left[\tan^{-1}\left(\frac{\lambda_0}{\alpha_0}\right) + \left[Im\, E_i\{(\alpha_0 + i\lambda_0)a_1\}\right]\right] + e^{-\alpha_0 a_1}\left[\frac{\sin \lambda_0 a_1}{\lambda_0 a_1}\left(1 - 2\frac{\alpha_0(c_0-1)}{\lambda_0^2 a_1}\right) + 2\frac{\cos \lambda_0 a_1}{\lambda_0 a_1}\frac{\alpha_0(c_0-1)}{\lambda_0}\right]$
Infinite absorbing half - space inverse mean free path α_0, secondaries per collision c_0	Laplacian μ_0 given by $\mu_0 < \alpha_0$ $1 = \frac{\alpha_0 c_0}{2\mu_0}\log\left(\frac{\alpha_0+\mu_0}{\alpha-\mu_0}\right)$
Multiplying slab of thickness $2t$ inverse mean free path α_1, and secondaries per collision c_1	$\alpha_1 c_1 \int_0^t e^{\alpha_1 \omega}\cos \lambda_1 \omega d\omega = \frac{\alpha_1 c_1}{\alpha_1^2+\lambda_1^2}\left\{e^{\alpha_1 t}(\alpha_1 \cos \lambda_1 t + \lambda_1 \sin \lambda_1 t) - \alpha_1\right\}$

There is a close analogy between the formulae of finite medium theory and modified diffusion theory for spherical and slab systems. For cylindrical systems the formulae for the Laplacians in finite medium theory involve indefinite integrals of $K_0(t)$ (a modified Bessel function of the second kind) which obscure the drawing of comparisons with the formulae for Laplacians in modified diffusion theory.

A comparison of modified and finite medium theory is made in Table VI. Checks for assessing the accuracy of the diffusion theory have been made

by the Carlson S_4 transport programme and by a Monte Carlo calculation.

Table VI indicates that for both multiplying systems and absorbing systems in the form of spheres, one-group finite medium diffusion theory is better than modified or infinite medium diffusion theory. Figure 2 compares flux plots, obtained by Carlson S_4, classical and finite medium diffusion calculations, for the reflected sphere of Table VI.

TABLE VI. COMPARISON OF ONE-GROUP CALCULATIONS

Critical Systems				
System		Modified Diffusion Theory (c.p. Davison [1])	Size of Core *(cm)* Finite Medium Theory	Carlson S_4 Transport
Bare sphere U 235		7.98 (Davison's extrapolation length used)	7.94 (No extrapolation length required)	7.96
Bare infinite cylinder U 235		5.72	5.66 (No extrapolation length required)	5.59
Sphere of U 235 with natural uranium infinite reflector		6.29	5.76	5.83
Slab of U 235 with natural uranium infinite reflector		1.08	1.05	1.00
Sub-Critical Systems				
Spheres of natural uranium with isotropic surface source of thermal neutrons		Escapes/Entries		
Radius of Sphere		Modified Diffusion Theory	Finite Medium Theory	Monte Carlo (4,000 sample) $\pm$ Standard Deviation
cm	Total Mean Free Paths			
3.35	2.43	0.247	0.346	0.354 ± 0.007
0.76	0.55	0.699	0.719	0.741 ± 0.006
0.38	0.275	0.835	0.841	0.898 ± 0.004

In the two-group version of finite medium theory the assumptions made are similar to those used in the one-group version. If the two-group equations for the neutron densities in a region V_k are written in the form:

$$\left.\begin{aligned} \nabla^2 n_1 + b_{11k} n_1 + b_{12k} n_2 &= 0 \text{ for neutrons of speed } v_1 \\ \Delta^2 n_2 + b_{28k} n_1 + b_{22k} n_2 &= 0 \text{ for neutrons of speed } v_2 \end{aligned}\right\} \qquad (61)$$

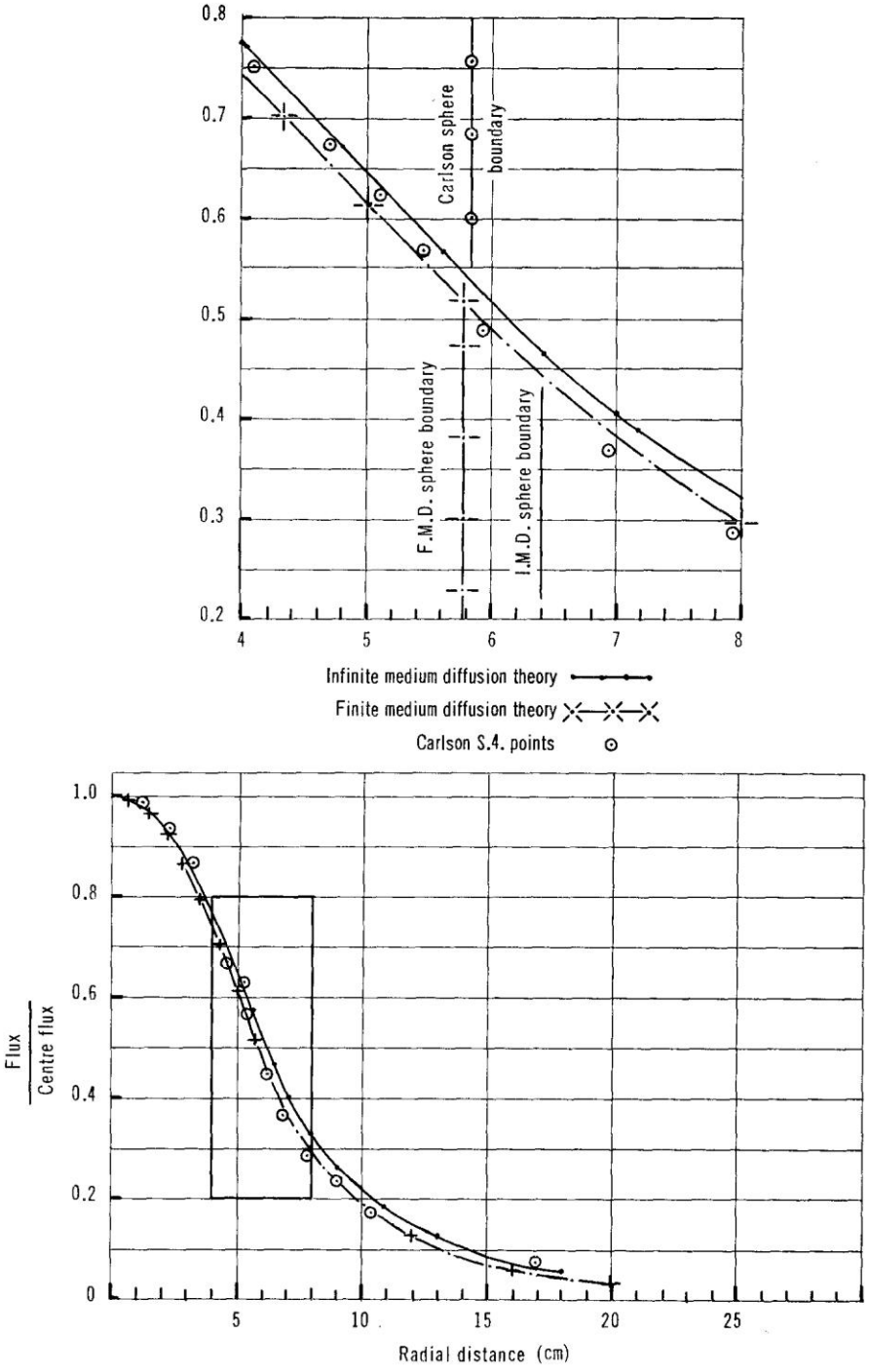


Figure 2. *Flux/centre flux vs. radial distance*

the diffusion coefficients for groups 1 and 2 are given by:

$$\begin{aligned} D_{1k} &= \alpha_{1k}(c_{11k} - 1)/b_{11k} = \nu_2\alpha_{2k}c_{12k}/\nu_1 b_{12k} \\ D_{2k} &= \alpha_{2k}(c_{22k} - 1)/b_{22k} = \nu_1\alpha_{1k}c_{21k}/\nu_2 b_{21k} \end{aligned} \qquad (62)$$

respectively, where α_{1k} is the inverse mean free path for group 1, c_{12k} the secondaries of group 1 per collision in group 1, c_{12k} the secondaries in group 1 per collision in group 2, etc. The expressions for neutron currents are of the same form as (58), and the coefficient of the neutron density term can be shown to be a quarter. Equation (62) shows that the parameters of the diffusion theory are determined when expressions have been found for b_{11k} and b_{22k} in terms of the inverse mean free paths and yields for each energy group. They can be found in the same way as the one-group Laplacians are determined; the procedure is an iterative instead of a direct one, because an *a priori* assumption has to be made about the functional form of the group transfer terms in (61). A simpler procedure, which may be adequate, is to calculate the b_{11k} and b_{22k} as if they were Laplacians of distinct one-group systems.

Variational Methods

Variational methods have been used mainly for problems on energy-independent or one-group systems, because the operators for such systems are self-adjoint and consequently the associated variational principles are simple to apply. The restriction of variational methods to mainly one-group problems should not be taken to imply that the results obtained are of academic significance only. The value of variational methods perhaps may be appreciated the more by considering some of the results that have been obtained. The field of application for variational methods can be divided roughly as follows:

a) The derivation of approximate boundary conditions from transport theory;
b) The determination of the effects of changes of shape on critical size;
c) The checking of the accuracy of some numerical methods.

Variational Principles for Transport Theory

There are two kinds of problems which can be treated by variational methods: (*i*) critical size problems associated with the homogeneous form of the transport equation, and (*ii*) those governed by the inhomogeneous form of the transport equation.

Critical size problems. The one-group integral equation for the neutron density at $\underset{\sim}{r}$ can be written [1]:

$$n(\underset{\sim}{r}) = \frac{\gamma}{4\pi}\int_V \frac{\alpha(\underset{\sim}{r}')c(\underset{\sim}{r}')e^{-\tau(\underset{\sim}{r},\underset{\sim}{r}')}\, n(\underset{\sim}{r}')}{|\underset{\sim}{r} - \underset{\sim}{r}'|^2}\, dV' \qquad (63)$$

where $c(\underset{\sim}{r})$ is the yield of secondary neutrons per primary neutron at $\underset{\sim}{r}$;
$\alpha(\underset{\sim}{r})$ is the inverse mean free path at r;
$\tau(\underset{\sim}{r}, \underset{\sim}{r}')$ is the optical distance between $\underset{\sim}{r}$ and $\underset{\sim}{r}'$;
γ is a variable parameter.

If the system is critical, γ_0, the lowest eigenvalue of (63), must be unity. The variational principle for (63) states that the maximum value of the functional:

$$I(\tilde{n}) = \frac{\int_V \int \alpha(\underset{\sim}{r})\alpha(r')c(\underset{\sim}{r})c(\underset{\sim}{r}')\tilde{n}(\underset{\sim}{r})\tilde{n}(\underset{\sim}{r}')e^{-\tau(\underset{\sim}{r},\underset{\sim}{r}')}\, dV dV'}{4\pi \int_V \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\tilde{n}(\underset{\sim}{r})^2\, dV} \tag{64}$$

is $1/\gamma_0$. The maximum value is attained by the everywhere-positive function $n(\underset{\sim}{r})$ from amongst all admissible trial functions $\tilde{n}(\underset{\sim}{r})$. The valuable features of the variational principle are:

i) A rough trial function can yield a quite accurate value of $1/\gamma_0$;
ii) $1/\gamma_0$ is always underestimated by any trial function, excepting the exact solution.

Pryce [23] in determining the critical size of a bare homogeneous sphere used the trial function:

$$\tilde{n}(\underset{\sim}{r}) = 1 - qr^2 \tag{65}$$

where r denotes the radial co-ordinate and q is a parameter which is chosen to maximize the functional (64). Davison and Fuchs [24] have fitted the formula:

$$\beta a = \frac{1.814}{\sqrt{1 - \alpha/\beta}} + 0.16\sqrt{1 - \alpha/\beta} - 0.697 \tag{66}$$

to Pryce's numerical results over the range $1.5 < \beta/\alpha < \infty$. Fuchs [3] has calculated the critical radius of a homogeneous sphere embedded in an infinite reflector, which is a pure scatterer and has the same total mean free path as the core. The trial function used is given by (65) in the core, and by the diffusion theory solution:

$$\tilde{n}(\underset{\sim}{r}) = A + B/r \tag{67}$$

in the reflector. The boundary condition that $\tilde{n}(\underset{\sim}{r})$ is continuous at $r = a$, the core reflector interface, is imposed. Thus, only two of the three parameters p, A and B can be chosen to maximize (64). Davison and Fuchs [24] give the formula:

$$\beta a = \frac{0.907}{\sqrt{1 - \alpha/\beta}} + 0.370(1 - \alpha/\beta) \tag{68}$$

for the critical radius a. As $\beta/\alpha \to 1$ the formula (68) agrees with diffusion theory; and as $\beta/\alpha \to \infty$ (i.e. the multiplication is very large and hence the reflector has a negligible effect on the critical size) the formula agrees with Peierls' [25] result for a bare sphere. The problem solved by Fuchs highlights the principal difficulties in applying the variational principle (64). The integrations involved in (64) are very difficult to perform unless the total mean free path is constant throughout the volume of the reactor. The choice of the trial functions $\tilde{n}(\underset{\sim}{r})$ is severely restricted for the same reason. Nevertheless, the solutions which have been obtained are useful for checking other methods. Frankel and Nelson [47] give variational calculations for the critical size of the bare slab, cube and cylinder.

Variational Principle for the Inhomogeneous Equation

The integral equation of transport theory, for the neutron density $n(\underset{\sim}{r})$ in the presence of sources $s(\underset{\sim}{r})$ in the region V, can be written:

$$n(\underset{\sim}{r}) = \int_V \left\{ \frac{\alpha(\underset{\sim}{r}')c(\underset{\sim}{r}')n(\underset{\sim}{r}')}{4\pi} + s(\underset{\sim}{r}') \right\} \frac{e^{-\tau\,(\underset{\sim}{r},\underset{\sim}{r}')}}{|\underset{\sim}{r} - \underset{\sim}{r}'|^2}\, dV' \tag{69}$$

The ψariational principle for (69) can be stated thus: if $\tilde{n}(\underset{\sim}{r})$ is a trial function and the finite system is not supercritical, the functional

$$J(\tilde{n}) = \int_V \Bigg[\left\{ \int_V \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\tilde{n}(\underset{\sim}{r}) \left(\frac{\alpha(\underset{\sim}{r}')c(\underset{\sim}{r}')n(\underset{\sim}{r}')}{4\pi} + 2s(\underset{\sim}{r}') \right) \frac{e^{-\tau\,(\underset{\sim}{r},\underset{\sim}{r}')}\, dV'}{|\underset{\sim}{r} - \underset{\sim}{r}'|^2} \right\} - \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\tilde{n}^2(\underset{\sim}{r}) \Bigg] dV \tag{70}$$

has an upper bound equal to:

$$\int_V \int_V \frac{\alpha(\underset{\sim}{r})c(\underset{\sim}{r})n(\underset{\sim}{r})s(\underset{\sim}{r}')e^{-\tau(\underset{\sim}{r},\underset{\sim}{r}')}dV'dV}{|\underset{\sim}{r} - \underset{\sim}{r}'|^2} \tag{71}$$

and $J(\tilde{n})$ furnishes a fair approximation to (71) even for relatively poor trial functions. An extension of this principle to infinite systems is discussed by Davison [1]. Marshak [41] used the extended principle to determine the linear extrapolation length of a black sphere. Kushneriuk and McKay [42] have used a similar principle to determine the linear extrapolation length for composite cylindrical control rods. This work has been extended by B. Montagnini [43] who has made some useful simplifications in the analysis. Despite these efforts, the difficulty of carrying out the integrations involved prevents the general use of the variational method in solving the inhomogeneous equation. The variational method for solving the integral equation of transport theory is successful only when the system is fairly simple, and then a very accurate result can often be obtained.

Variational Principles for Diffusion Theory

There are many reactors for which the neutron spectra are not modified very much by changes in shape. For such reactors the change of critical size due to a change in shape is given with fair accuracy by a one-group diffusion theory. One method for calculating the change of critical size with shape is to use buckling conversions (as outlined on page 51). This approach is restricted to the simpler changes of shape, and some feel for the choice of extrapolation lengths is required if consistently accurate results are to be obtained.

The variational principles given by Ackroyd [26,27] are well suited to deal with critical size-shape problems. Some of the results which have been got are indicated here; they are not difficult to derive from certain isoperimetric inequalities associated with the variational principles, and they are obtained with very much less effort than by other methods. The variational principles have also been used by Ackroyd and Perks [28] to check some

aspects of the accuracy of difference solutions of the diffusion equations. A detailed review of the use of variational principles for one- and two-group diffusion theory is given by Ackroyd [27].

Variational Principle for One-Group Heterogeneous Reactors

The one-group diffusion equations for the flux ψ_1 in the core V_1, and the flux ψ_2 in the reflector V_2 are taken to be:

$$\nabla \cdot \left(\sum_{i=1}^{3} D_{1i} \frac{\underset{\sim}{u}_i}{Q_i} \frac{\partial \psi_1}{\partial q_i} \right) + \gamma^2 \psi_1 = 0 \text{ in } V_1 \tag{72}$$

and:

$$\nabla \cdot \left(\sum_{i=1}^{3} D_{2i} \frac{\underset{\sim}{u}_i}{Q_i} \frac{\partial \psi_2}{\partial q_i} \right) - \omega^2 \psi_2 = 0 \text{ in } V_2 \tag{73}$$

for an orthogonal co-ordinate system (q_1, q_2, q_3). The diffusion coefficients D_{1i} and D_{2i} $(i = 1, 2, 3)$ are in general independent positive functions of position. The coefficients γ^2 and ω^2 are also positive functions of position. The boundary conditions for the fluxes are assumed to be:

$$\left.\begin{aligned} &\psi_1 = \psi_2 \text{ on } S_1 \text{ the interface of } V_1 \text{ and } V_2 \\ &\psi_2 = 0 \quad \text{on } S_2 \text{ the bare surface of } V_2 \end{aligned}\right\} \tag{74}$$

and on S_1, the normal component of the neutron current is continuous, i.e.:

$$\left[\sum_{i=1}^{3} D_{1i} \frac{\underset{\sim}{u}_i}{Q_i} \frac{\partial \psi_1}{\partial q_i} \right]_n = \left[\sum_{i=1}^{3} D_{2i} \frac{\underset{\sim}{u}_i}{Q_i} \frac{\partial \psi_2}{\partial q_i} \right]_n \tag{75}$$

If Φ_1 and Φ_2 are continuous functions, then:

$$\int_{V_1} \left[\sum_{1=i}^{3} \frac{D_{1i}}{{Q_i}^2} \left(\frac{\partial \Phi_1}{\partial q_i} \right)^2 - \gamma^2 {\Phi_1}^2 \right] dV + \int_{V_2} \left[\sum_{1=i}^{3} \frac{D_{2i}}{{Q_i}^2} \left(\frac{\partial \Phi_2}{\partial q_i} \right)^2 + \omega^2 {\Phi_2}^2 \right] dV \geqslant 0 \tag{76}$$

provided that Φ_1 and Φ_2 satisfy the relaxed boundary conditions:

$$\left.\begin{aligned} &\Phi_1 = \Phi_2 \text{ on } S_1 \\ &\Phi_2 = 0 \quad \text{on } S_2 \end{aligned}\right\} \tag{77}$$

The equality sign in (76) holds if, and only if, $\Phi_1 = \psi_1$ and $\Phi_2 = \psi_2$. An important feature of this variational principle is that the boundary conditions are not as restrictive as those for the diffusion equations and those used in other variational principles, e.g. Hassitt's modification of Kohn's variational principle [29].

Direct Application of the Variational Principle to a Homogeneous Reactor

For a homogeneous reactor the variational principle (76) reduces to:

$$\int_{V_1} D_1 \{ (\nabla \Phi_1)^2 - \lambda^2 {\Phi_1}^2 \} dV + \int_{V_2} D_2 \{ (\nabla \Phi_2)^2 + \mu^2 {\Phi_2}^2 \} dV \geqslant 0 \tag{78}$$

This principle has been used directly by Ackroyd and Ball [30] to give upper and lower bounds for the critical Laplacian and critical height of an elliptical

cylinder of finite height and with an infinite reflector. The diffusion coefficients in the core and reflector were assumed to be the same. The upper bound for the Laplacian was obtained by substituting in (78) approximations Φ_1 and Φ_2 for the fluxes ψ_1 and ψ_2. A lower bound was obtained from the variational principle with the aid of the geometrical process called Steiner symmetrization; Ackroyd and Ball [31]. The calculations, which are very simple, can be used also to give bounds for the critical height. For an example of a completely reflected elliptical cylinder with semi-major and minor axes of 38.54 and 19.27 cm, the reasonably close bounds:

$$26.65 \leqslant h \leqslant 27.49 \tag{79}$$

were obtained for the critical height h.

Rayleigh-Ritz Procedure and Optimized Parameters and Coefficients

The variational principle can be used with a systematic approximation process, such as the Rayleigh-Ritz procedure [32, 33, 34], to give a closer upper bound for the critical Laplacian than can be given in many cases by a direct application of the variational principle. The details are given by Ackroyd *et al* [32], and are illustrated by the calculation of the critical Laplacian of an inhomogeneous slab for which the composition is given by a trigonometric series in z, the perpendicular distance into the slab from one face.

Other systematic methods are those of optimized parameters [33, 34] and coefficients, and it is perhaps most easily understood by means of examples. To illustrate the method of optimized parameters, consider a finite circular cylinder with a complete infinite reflector. The approximations for the fluxes are taken to be:

$\Phi_1 = \mathrm{A}\, \mathrm{J}_o(\alpha r) \cos(\beta z)$	in the core V_1, $0 \leqslant r \leqslant a$, $-c \leqslant z \leqslant c$ with diffusion coefficient D_1, and Laplacian λ^2
$\Phi_2 = \mathrm{A}\, \mathrm{J}_o(\alpha a) \dfrac{\mathrm{K}_o(\gamma r)}{\mathrm{K}_o(\gamma a)} \cos(\beta z)$	in the side reflector V_2, $a \leqslant r \leqslant \infty$, $-c \leqslant z \leqslant c$ with diffusion coefficient D_2 and Laplacian $-\mu_2^2$
$\Phi_3 = \mathrm{A}\, \mathrm{J}_o(\alpha r) \exp\{\delta(c-z)\} \cos(\beta c)$	in the top reflector V_3, $0 \leqslant r \leqslant a$, $c \leqslant z \leqslant \infty$ with diffusion coefficient D_3 and Laplacian $-\mu_3^2$
$\Phi_4 = \mathrm{A}\, \mathrm{J}_o(\alpha a) \cos(\beta c) \dfrac{\mathrm{K}_o(\gamma r)}{\mathrm{K}_o(\gamma a)} \exp\{\delta(c-z)\}$	in the top corner reflector V_4, $a \leqslant r \leqslant \infty$, $c \leqslant z \leqslant \infty$ with diffusion coefficient D_4 and Laplacian $-\mu_4^2$

(80)

The coefficient A is chosen so that Φ_1 is normalized, i.e.:

$$\int_{\mathrm{V}_1} \Phi_1^2 \, d\mathrm{V} = 1 \tag{81}$$

The functions (80) are substituted in the variational principle:

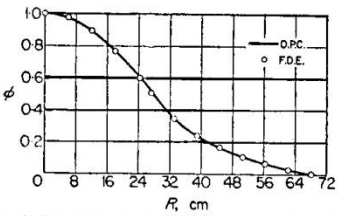
$$\int_{V_1} (\nabla\Phi_i)^2 \, dV + \sum_{i=2}^{4} \frac{D_i}{D_1} \int_{V_i} \{ (\nabla\Phi_i)^2 + \mu_i^2 \Phi_i^2 \} \, dV = \lambda_u^2 \geqslant \lambda^2 \quad (82)$$

to give an upper bound λ_u^2 for λ^2. The parameters α, β, γ and δ are chosen to give the least upper bound for λ_u^2 which is consistent with the choice (80) of approximations for the fluxes. Note that if the properties of the region V_4 are suitably chosen, the expressions are exact ones for the fluxes. Since the method of optimized parameters is known to give the exact solution for the fluxes in special reactors, and one knows roughly that changes in the composition of the corner reflector do not have a great influence on the critical Laplacian for the core and do not affect the flux appreciably, except in the corner and its neighbourhood, one expects to get good results over a wide field. This contention is borne out in Figure 3 (for some of the reactors of Table VII) where variational flux plots are compared with a numerical method of Hassitt [35] which is based on finite difference equations. The method of optimized parameters has been used by Ackroyd and Perks [34] to obtain flux plots for finite elliptical cylinders and hexagonal prisms with complete reflectors.

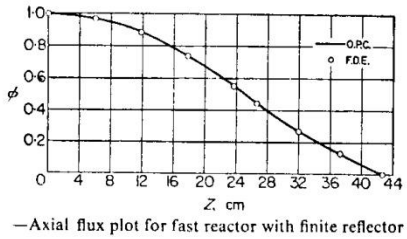
TABLE VII

Reactor No.	No. of Regions per Half Reactor		Laplacian of the Central Region of the Reactor		Percentage Error in Laplacian of Central Region	
	Core	Total	O.P.C.	F.D.E.	O.P.C.	F.D.E.
1	1	4	0.004810	0.00468	—	—
2	2	5	0.005977	0.006044	—	—
3	1	4	0.004957	0.004975	0	+ 0.29
4	2	5	0.007271	0.00736	0	+ 0.89
5	2	5	0.006666	0.006733	0	+ 0.78
6	6	9	0.008270	0.006382	0	— 10.40

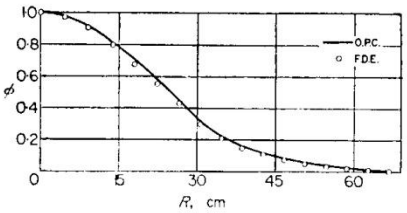
The method of optimized parameters can be used to obtain both an upper bound for the critical Laplacian and an approximate flux plot for reactors with multi-region cores. However, there is a limit to the accuracy that can be achieved in such cases with the method of optimized parameters, because in the examples where exact solutions are known for reactors with multi-region cores, the fluxes in all but the central region are expressed as a linear combination of two independent functions. In the method of optimized coefficients the flux in each region of the core, except the central one, is expressed as a linear combination of two independent functions. The arguments of these functions are determined by the method of optimized parameters, and the coefficients are obtained by the method of optimized coefficients. In Figure 3, flux plots are determined by the method of optimized parameters and coefficients and the difference method is compared for reactor 2. The upper bound for the critical Laplacian is for this example 1.1 % less than the estimate of the difference method for the critical Laplacian. Thus, in this example, the method of optimized parameters and coefficients gives a



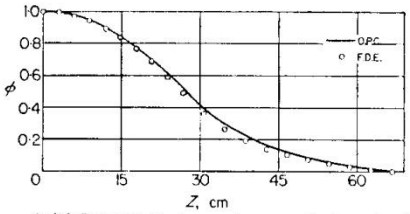
.—Radial flux plot for fast reactor with finite reflector (No. 1).



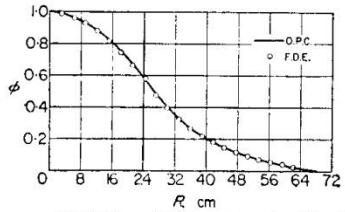
—Axial flux plot for fast reactor with finite reflector (No. 1).



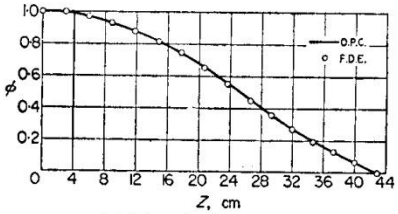
—Radial flux plot for two-region core fast reactor (No. 2).



—Axial flux plot for two-region core fast reactor (No. 2).



—Radial flux plot for test reactor (No. 3).



—Axial flux plot for test reactor (No. 3).

Figure 3. *Flux plots.*

better estimate for the critical Laplacian than the difference method. The method of optimized parameters and coefficients (O.P.C.) can be used in this way to test on a one-group basis finite difference equation methods (F.D.E.) for solving the diffusion equation. In Table VII, the Laplacians for a number of reactors have been obtained by O.P.C. and F.D.E. methods. The reactors are in the form of finite circular cylinders. The critical value of the Laplacian for the central region of the core has been calculated in each case.

The reactors 3, 4, 5 and 6 were chosen so that the critical Laplacian of the central region could be calculated exactly. In the case of reactor 2, the O.P.C. result is more accurate thant that of F.D.E., since O.P.C. always gives an upper bound for the Laplacian. One also expects that for reactors not significantly different from 3, 4, 5 and 6 the O.P.C. results for the Laplacian should be better than those of F.D.E.

First Order Perturbation Theory

In some problems it is necessary to estimate the effect on the criticality of a system when small changes are made, either to the geometry or to the material composition. In such cases it may be preferable to apply perturbation theory rather than to do direct calculations on slightly different systems; if the changes are small, the comparison of results obtained by direct calculation may lose much of its significance due to rounding off errors on the individual results. Also by applying perturbation theory one may obtain the answers to several problems more or less simultaneously. A further use for perturbation theory arises in the adjustment of basic scattering data (cf. page 110).

The perturbation equation which forms the basis of all perturbation calculations gives the change in some suitable index of criticality, in terms of arbitrary changes $\Delta\alpha$ and $\Delta\beta$ in the α and β respectively. The terms in the equation are very similar no matter what parameter is chosen as the index of criticality, though the detailed construction of the equation obviously depends on this. The general first order perturbation theory was given by Pendlebury [36], who derived the perturbation equation from the Boltzmann transport equation. Tait [37] gave a two-group perturbation theory using the integral equations as his starting point, as did Fuchs [38] in his one-group treatment. The general equation of perturbation theory can also be derived from the variational equation.

An outline of the derivation of the equation of general perturbation theory starting from the time-dependent form of the Boltzmann transport equation (1) will now be given, followed by the reduction to multi-group form. The multi-group form in terms of an alternative index of criticality — the collision generation criticality index — is also given.

In the first instance the index of criticality is chosen to be the time constant λ of the system; in terms of the time-dependent neutron flux $\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}, t)$ this is obtained by assuming λ is the *maximum* eigenvalue occurring in the general solution:

$$\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}, t) = \sum_i \Phi_i(\underset{\sim}{r}, v, \underset{\sim}{\Omega},) \exp(\lambda_i t) \tag{83}$$

Since λ is the maximum value of the λ_i it follows that $\Phi_i(\underset{\sim}{r}, v, \underset{\sim}{\Omega}, t)$ is everywhere positive, and the system is subcritical, critical or supercritical dependent on

whether λ is negative, zero or positive. The Boltzmann equation to be solved for λ can be expressed in the form:

$$\left.\begin{aligned}\frac{\lambda}{v}\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) = &- \underset{\sim}{\Omega}.\operatorname{grad}\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) - \alpha(\underset{\sim}{r}, v)\,\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) \\ &+ \frac{1}{4\pi}\iint \beta \,\{\, \underset{\sim}{r}, (v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega}) \,\{\, \Phi(\underset{\sim}{r}, v', \underset{\sim}{\Omega}')\, dv'\, d\Omega'\end{aligned}\right\} \quad (84)$$

where $\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega})$ satisfies the boundary condition that it is continuous across all interfaces, is zero for all incoming directions at a free surface and is positive everywhere. Note that there are slight differences in notation for the integral terms in the Boltzmann equations (1) and (84).

Before proceeding further, one must introduce the adjoint equation (this is essential in order to eliminate terms representing changes in the neutron flux Φ in the final equation). Suppose L is an operator and is a function of variables denoted by ξ, if P and Q are functions of ξ then the adjoint operator of L, which is denoted by L^+, is defined by:

$$\int \mathrm{PLQ}\,d\xi = \int \mathrm{QL^+P}\,d\xi \quad (85)$$

where the integrations are taken over all ξ space. Thus the adjoint of grad is $-$ grad, and of:

$$\iint \beta \,\{\, \underset{\sim}{r}(v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega}) \,\{\, \ldots\, dv'\, d\Omega, \text{ is } \iint \beta \,\{\, \underset{\sim}{r}(v, \underset{\sim}{\Omega} \to v', \underset{\sim}{\Omega}') \,\{\, \ldots\, dv,\, d\Omega'.$$

If the adjoint of $\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega})$ is written as $\Phi^+(\underset{\sim}{r}, v, -\underset{\sim}{\Omega})$ then the equation satisfied by this is:

$$\left.\begin{aligned}\frac{\lambda^+}{v}\Phi^+(\underset{\sim}{r}, v, -\underset{\sim}{\Omega}) = &\ \underset{\sim}{\Omega}.\operatorname{grad}\Phi^+(\underset{\sim}{r}, v, -\underset{\sim}{\Omega}) - \alpha(\underset{\sim}{r}, v)\,\Phi^+(\underset{\sim}{r}, v, -\underset{\sim}{\Omega}) \\ &+ \frac{1}{4\pi}\iint \beta \,\{\, \underset{\sim}{r}, (v, \underset{\sim}{\Omega} \to v'\, \underset{\sim}{\Omega}') \,\{\, \Phi^+(\underset{\sim}{r}, v', -\underset{\sim}{\Omega}')\, dv'\, d\Omega'\end{aligned}\right\} \quad (86)$$

and the boundary conditions to be satisfied by $\Phi^+(\underset{\sim}{r}, v, \underset{\sim}{\Omega})$ are that it is continuous across all boundaries and is zero for all *incoming* directions* at a free surface and is positive everywhere. (N.B. If the adjoint function in the above equation had been written as $\Phi^+(\underset{\sim}{r}, v, +\underset{\sim}{\Omega})$ then one would have had to make this zero at a free surface for all outgoing directions.) It can be shown that $\lambda^+ = \lambda$. To obtain the change $\Delta\lambda$ in λ resulting from (small) changes $\Delta\alpha(\underset{\sim}{r}, v)$ in $\alpha(\underset{\sim}{r}, v)$ and $\Delta\beta[\underset{\sim}{r}, (v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})]$ in $\beta[\underset{\sim}{r}, (v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega})]$ one considers the ordinary equation for the original system and the adjoint equation for the perturbed system (or vice versa) and after some manipulation obtains the perturbation equation:

* This is actually a consequence of the definition of the adjoint operators and of the boundary conditions in Φ^+.

$$\left.\begin{aligned}
&\Delta\lambda \iiint \frac{1}{v}\,\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega})\,\Phi^{+}(\underset{\sim}{r}, v, -\underset{\sim}{\Omega})dvd\Omega d\tau \\
&= -\iiint \Delta\alpha(\underset{\sim}{r}, v)\,\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega})\,\Phi^{+}(\underset{\sim}{r}, v, -\underset{\sim}{\Omega})dvd\Omega d\tau \\
&+ \frac{1}{4\pi}\iiiint\int \Delta\beta \mid \underset{\sim}{r}, (v', \underset{\sim}{\Omega}' \to v, \underset{\sim}{\Omega}) \mid \Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) \\
&\qquad \Phi^{+}(\underset{\sim}{r}, v, -\underset{\sim}{\Omega})dv'dvd\Omega' d\Omega d\tau
\end{aligned}\right\} \tag{87}$$

$\left(\int \ldots d\tau \text{ implies integration over all space.}\right)$

This gives the change $\Delta\lambda$ in λ due to specified changes in the α and β in terms of these changes and the angular flux and its adjoint for the original system. The multi-group form of the equation can be obtained by writing:

$$\left.\begin{aligned}
\Phi(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) &= \sum_i \Phi_i(\underset{\sim}{r}, \underset{\sim}{\Omega})\,f_i(v) \\
\Phi^{+}(\underset{\sim}{r}, v, \underset{\sim}{\Omega}) &= \sum_i \Phi_i{}^{+}(\underset{\sim}{r}, \underset{\sim}{\Omega})\mathrm{C}_i
\end{aligned}\right\} \tag{88}$$

where $\Phi_i(\underset{\sim}{r}, \underset{\sim}{\Omega})$ represents the angular flux of neutrons in group i. $f_i(v)$ defines the spectrum of neutrons in group i, $f_i(v)dv$ being the probability that the neutron speed lies between v and $v + dv$. It is usual to take:

$$\left.\begin{aligned}
&f_i(v) = g(v), \text{ say for } v_i \leqslant v \leqslant v_{i+1}, \text{ and otherwise zero} \\
&\mathrm{C}_i = \mathrm{C}, \text{ a constant, for } v_i \leqslant v \leqslant v_{i+1} \text{ and otherwise zero}
\end{aligned}\right\} \tag{89}$$

The group constants $\alpha_i(\underset{\sim}{r})$ and $\beta_{ij}(\underset{\sim}{r})$ for each material and for the particular case of isotropic scatter are defined by:

$$\left.\begin{aligned}
\alpha_i(\underset{\sim}{r}) &= \int_i \alpha(\underset{\sim}{r}, v)\,f_i(v)dv \\
\beta_{ij}(\underset{\sim}{r}) &= \int_j dv \int_i \beta \mid \underset{\sim}{r}, (v' \to v) \mid f(v')dv'
\end{aligned}\right\} \tag{90}$$

These definitions are discussed by Parker [39]. Hence one obtains:

$$\left.\begin{aligned}
&\Delta\lambda \iint \sum_i \frac{1}{v_i}\,\Phi_i(\underset{\sim}{r}, \underset{\sim}{\Omega})\Phi_1{}^{+}(\underset{\sim}{r}, -\underset{\sim}{\Omega})d\Omega d\tau \\
&= -\iint \sum_i \Delta\alpha_i(\underset{\sim}{r})\Phi_i(\underset{\sim}{r}, \underset{\sim}{\Omega})\Phi_1{}^{+}(\underset{\sim}{r}, -\underset{\sim}{\Omega})d\Omega d\tau \\
&+ \frac{1}{4\pi}\int \sum_i \sum_j \Delta\beta_{ij}(\underset{\sim}{r})\Phi_i(\underset{\sim}{r})\Phi_j{}^{+}(\underset{\sim}{r})d\tau
\end{aligned}\right\} \tag{91}$$

where:

$$\left.\begin{aligned}\Phi_i(\underset{\sim}{r}) &= \int \Phi_i(\underset{\sim}{r}, \underset{\sim}{\Omega})d\Omega \\ \Phi_i^+(\underset{\sim}{r}) &= \int \Phi_i^+(\underset{\sim}{r}, \underset{\sim}{\Omega})d\Omega\end{aligned}\right\} \tag{92}$$

which is the corresponding multi-group form of the perturbation equation. In deciding the relative criticality of two systems it is only necessary to know the sign of the RHS in equation (91). If, however, one requires a measure of the deviation from critical, one then needs to know absolute values and under these circumstances, for example, it may be more convenient to work in terms of the collision generation criticality index A defined on page 83, and to have the perturbation equation expressed in terms of it. The transformation of equation (91) into the corresponding equation involving the generation criticality index A will now be considered.

In column 1 of Table VIII below, two sets of group constants are given and the corresponding time constants for same system.

TABLE VIII

	1		2	
		Time Constant		Time Constant
1	β_{ij} α_i	λ	$\beta_{ij} + \Delta\beta_{ij}$ $\alpha_i + \Delta\alpha_i$	$\lambda + \Delta\lambda$
2	$A\beta_{ij}$ α_i	0	$(A + \Delta A)$ $(\beta_{ij} + \Delta\beta_{ij})$ $\alpha_i + \Delta\alpha_i$	0

The data in cell (2, 1) of Table VIII arises because A is the number by which the β_{ij} have to be multiplied to make the system critical, that is, have a zero time constant. The information listed in column 2 represents the corresponding situations to those listed in column 1 for the same geometrical system when the α's and β's are changed by amounts $\Delta\alpha$ and $\Delta\beta$.

The situation represented by cell (1, 2) of Table VIII can be thought of as a perturbation of the situation represented by cell (1, 1) and this is described by equation (91).

The situation represented by cell (2, 2) of Table VIII can be thought of as a perturbation of the situation represented by cell (2, 1). Equation (91) can be used to derive the corresponding perturbation equation. The $\Delta\lambda = 0$, and $\Delta\alpha$ and $\Delta\beta$ to be substituted in equation (91) are:

$$\left.\begin{aligned}&\Delta\alpha = \Delta\alpha_i \\ &\Delta\beta = (A + \Delta A)(\beta_{ij} + \Delta\beta_{ij}) - A\beta_{ij} = A\Delta\beta_{ij} + \beta_{ij}\Delta A \\ &\qquad\text{(neglecting } \Delta\beta_{ij}\Delta A \text{ term).}\end{aligned}\right\} \tag{93}$$

Whence:

$$\left.\begin{aligned}&\frac{\Delta A}{4\pi}\int\sum_i\sum_j\beta_{ij}(\underset{\sim}{r})\Phi_i(\underset{\sim}{r})\Phi_j^{+}(\underset{\sim}{r})d\tau\\&=\iint\sum_i\Delta\alpha_i(\underset{\sim}{r})\Phi_i(\underset{\sim}{r},\underset{\sim}{\Omega})\Phi_i(\underset{\sim}{r},-\underset{\sim}{\Omega})d\Omega d\tau\\&\quad-\frac{A}{4\pi}\int\sum_{ij}\Delta\beta_{ij}(\underset{\sim}{r})\Phi_i(\underset{\sim}{r})\Phi_j^{+}(\underset{\sim}{r})d\tau\end{aligned}\right\}\qquad(94)$$

One further equation is of particular interest and this obtains when λ is small (hence $A\sim 1$). In this case, cell (2,1) of Table VIII can be thought of as a perturbation of cell (2, 2) with:

$$\left.\begin{aligned}\Delta\lambda&=\lambda\\\Delta\alpha&=0\\\Delta\beta&=(1-A)\beta_{ij}\end{aligned}\right\}\qquad(95)$$

Substitution in equation (91) gives *(for small λ)*,

$$\begin{aligned}&\Delta\lambda\iint\sum_i\frac{1}{v_i}\Phi_i(\underset{\sim}{r},\underset{\sim}{\Omega})\Phi_i^{+}(\underset{\sim}{r},-\underset{\sim}{\Omega})d\Omega d\tau\\&\quad=\frac{(1-A)}{4\pi}\int\sum_{,j}\beta_{ij}(\underset{\sim}{r})\Phi_i(\underset{\sim}{r})\Phi_j^{+}(\underset{\sim}{r})d\tau\end{aligned}\qquad(96)$$

Spherical Harmonics Method

The spherical harmonics method [1] offers a convenient way of obtaining reliable answers to the critical size or multiplication of simple systems in a few-group approximation, in either plane or spherical geometry, when one has only limited computing facilities. It can be generalized to other geometries but its main practical use is in the simple geometries mentioned, in particular, in spherical geometry, and hence only this will be dealt with here.

The main feature of the method is the expansion of the angular neutron flux $\Phi_i(r,\mu)$ in group i, in terms of Legendre Polynomials in μ, thus:

$$\Phi_i(r,\mu)=\frac{1}{4\pi}\sum_{l=0}^{\infty}(2l+1)\psi_{1,\,i}(r)P_1(\mu)\qquad(97)$$

— here, r is the length of the radius vector from a point to the origin and μ is the cosine of the angle between the direction of motion of a neutron and the radius vector. It follows that $\psi_{0,\,i}$ is the scalar flux and $\psi_{1,\,i}$ the vector flux of neutrons in group i. The multi-group transport equation for the case of isotropic scatter* in the stationary state and without source terms, is:

$$\begin{aligned}&\mu\frac{\partial\Phi_i(r,\mu)}{\partial r}+\frac{(1-\mu^2)}{r}\frac{\partial\Phi_i(r,\mu)}{\partial\mu}+\alpha_i(r)\Phi_i(r,\mu)\\&\quad=\frac{1}{2}\sum_j\beta_{ji}(r)\int_{-1}^{+1}\Phi_i(r,\mu)d\mu\end{aligned}\qquad(98)$$

* The method can be generalized to take anisotropic scatter into account but the resulting mathematics is somewhat more involved. It is discussed to some extent by Davison [1].

where $\alpha_i(r)$ is the inverse mean free path of neutrons in group i, and $\beta_{ji}(r)/\alpha_j(r)$ is the average number of neutrons produced in group i at a collision of a neutron in group j.

Substitution of (97) into (98) gives, after some manipulation, the infinite set of differential difference equations for each of the regions in the system:

$$\begin{aligned}(l+1)\left\{\frac{d}{dr}+\frac{(l+2)}{r}\right\}\psi_{i,\,l+1}+(2l+1)\alpha_i\psi_{i,\,l}\\ +\,l\left\{\frac{d}{dr}-\frac{(l-1)}{r}\right\}\psi_{i,\,l-1}=\delta_{0,\,l}\sum_j\beta_{ji}\psi_{j,\,0}\end{aligned}\qquad(99)$$

(constant material density in each of the regions is assumed)

where $\delta_{0,\,l}=0$ if $l\neq 0$, $=1$ if $l=0$. The general solution of these differential difference equations can be expressed in the operational form:

$$\left\{\frac{1}{r}\,\mathrm{D}\right\}^{l}\{r^{l+1}\psi_{i,\,l}\}=\sum_j \mathrm{F}_{j,\,i,\,l}(\mathrm{D}^2)\{r\psi_{j,\,0}\}\qquad(100)$$

where $\mathrm{D}\equiv\dfrac{d}{dr}$ and $\mathrm{F}_{j,\,i,\,l}(\mathrm{D}^2)$ is given by:

$$\begin{aligned}&(l+1)\mathrm{F}_{j,\,i,\,l+1}(\mathrm{D}^2)+(2l+1)\alpha_i\mathrm{F}_{j,\,i,\,l}(\mathrm{D}^2)+l\mathrm{D}^2\mathrm{F}_{j,\,i,\,l-1}(\mathrm{D}^2)=0\\ &\mathrm{F}_{j,\,i,\,0}=\delta_{i,\,j}\qquad \mathrm{F}_{j,\,i,\,1}=\beta_{ji}-\alpha_i\delta_{i,\,j}\end{aligned}\qquad(101)$$

The general solution of (100) embraces the solution of (99); for example, if it is assumed that ψ_{j0} is known, then the solution of ψ_{il} from (100) contains more constants of integration than are required by (99). One must bear this in mind when obtaining solutions by way of equation (100).

To obtain an exact solution, that is, to determine the $n_{i,\,l}$ for all l, involves the solution of the differential equations (100) and in the limit $l\to\infty$ these become infinite order equations. An exact solution is therefore not feasible in practice, but an approximate solution is obtained by setting $\psi_{i,\,\mathrm{M}+1}\equiv 0$ for some particular value of M, and this leads to what is known as the $\mathrm{P_M}$ approximation. *It is interesting to note that when* $\mathrm{M}=1$, *i.e. the* $\mathrm{P_1}$ *approximation, the solution is in fact that of simple diffusion theory.* When it is assumed that $\psi_{i,\,\mathrm{M}+1}\equiv 0$ it follows from equation (100) that:

$$\sum_j \mathrm{F}_{j,\,i,\,\mathrm{M}+1}(\mathrm{D}^2)\{r\psi_{j,\,0}\}=0\qquad(102)$$

whence:

$$\|\mathrm{F}_{j,\,i,\,\mathrm{M}+1}(\mathrm{D}^2)\|\{r\psi_{i,\,0}\}=0\qquad(103)$$

The solutions $r\psi_{i,\,0}$ can then be expressed in the form:

$$r\psi_{i,\,0}=\sum_k \mathrm{A}_k\mathrm{R}_i(\nu_k^2)\exp(\nu_k r)\qquad(104)$$

where ν_k are the roots of the determinant:

$$\|\mathrm{F}_{j,\,i,\,\mathrm{M}+1}(\nu^2)\|=0\qquad(105)$$

Some of the roots may be complex. The roots occur in pairs of the form $\pm\nu$ but these are treated as distinct in (104). $\mathrm{R}_j(\nu_k^2)$ is the co-factor of the term $\mathrm{F}_{j,\,1\,\mathrm{M}+1}(\nu^2)$ in the determinant:

$$\|\mathrm{F}_{j.i,\mathrm{M}+1_1}(\nu^2)\|.$$

A_k are constants to be determined later by the boundary conditions. The solution for $\psi_{i,l}$ can now be obtained from (100) bearing in mind the need to satisfy equation (99) and the assumed condition that $\psi_{j,l,M+1} \equiv 0$.

$$\boxed{\psi_{i,l}(r) = \sum_k A_k S_{i,l}(\nu_k) H_l(\nu_k r)} \tag{106}$$

where:

$$\left.\begin{aligned} H_{l+1}(z) &= H_{l-1}(z) - (2l+1)H_l(z)/z \\ H_0(z) &= e^z/z, \qquad H_1(z) = e^z(1 - 1/z)/z \end{aligned}\right\} \tag{107}$$

and:

$$\left.\begin{aligned} (l+1)S_{i,l+1}(\nu) + \frac{(2l+1)\alpha_i}{\nu} S_{i,l} + l S_{i,l-1} &= 0 \\ S_{i,o} = \nu R_i(\nu), \; S_{i,1} &= \sum_j \beta_{ji} R_j(\nu) - \alpha_i R_i(\nu) \end{aligned}\right\} \tag{108}$$

The above solution applies to a source-free region. Where sources are present an appropriate modification to the solution may be necessary, though sources can often be taken into account by application of suitable boundary conditions. Davison [1] discusses in some detail the effect of various order approximations and the boundary conditions to be applied with the method in the one-group approximation, and the same discussion applies to the multi-group case. Briefly, the approximation used is usually odd order (i.e. M, odd) on the grounds that an even order approximation does not in general give a much better result than the previous odd order approximation and involves more work. The boundary conditions to be applied are:

i) At an interface, $\psi_{i,l}(r)$ continuous for $l = 0,1, \ldots M$. (109)

ii) At a free surface, either:

a) $\Phi_i(r, \mu) = 0$ for all negative roots of $P_{M+1}(\mu) = 0$ (i.e. μ values representing incoming directions) — this is known as Mark's boundary condition [1]

or *b*) $\int_{-1}^{0} \mu^{2n+1} \Phi_i(r, \mu) d\mu = 0, \quad n = 0,1 \ \ldots, \quad (M-1)/2$

— this is known as Marshak's boundary condition. The relative merits of (*a*) and (*b*) are discussed by Davison [1]. (110)

iii) If a source of neutrons enters from the outside, this can be taken into account by setting either:

a) $\Phi_i(r, \mu) =$ some appropriate values, for all negative roots of $P_{M+1}(\mu) = 0$

or *b*) $\int_{-1}^{0} \mu^{2n+1} \Phi_i(r, \mu) d\mu =$ some appropriate values for $n = 0,1, \ldots, (M-1)/2$. (111)

iv) A central cavity with or without source can be taken into account by boundary conditions analogous to (*ii*) and (*iii*):

$$
\left.
\begin{aligned}
&a)\quad \Phi_i(r,\mu) = \Phi_i(r,-\mu) + S(\mu) \text{ for all positive roots of } P_{M+1}(\mu) = 0,\ S(\mu) \text{ source strength in direction } \mu \\
\text{or}\quad &b)\quad \int_0^1 \mu^{2n+1}\Phi_i(r,\mu)d\mu = \int_{-1}^0 \mu^{2n+1}\Phi_i(r,\mu)d\mu \\
&\qquad\qquad + \int_0^1 \mu^{2n+1}S(\mu)d\mu \quad n = 0,1,\ldots,(M-1)/2
\end{aligned}
\right\} \quad (112)
$$

If a neutron population is maintained in the absence of sources, that is, the system is critical, then the application of the boundary conditions leads to a homogeneous set of simultaneous equations for the A_k and elimination of the A_k leads to a determinantal equation for the critical dimension. If the system is subcritical and sources are present, one is able to solve the set of simultaneous equations resulting from the application of the boundary conditions, for the A_k; thereafter the neutron scalar and vector flux distribution are readily determined and hence any multiplication which may be required is obtained.

The spherical harmonics method is not the ideal method of solution for use on high-speed computers. It suffers from the disadvantage that the set of equations (99) is not stable in the sense that their solution — numerical, or semi-analytical as in the present case — leads to an accumulation of error during the computation. It is therefore essential to pay attention to this feature during any numerical work, and when necessary, to use a large number of significant figures. This disadvantage sets a limit to the usefulness of the method and the difficulty increases as the number of groups and number of different regions increases; hence it is desirable to restrict oneself to only a few groups and a few regions.

The S_n Method

The S_n or Carlson method is a multi-group method which is particularly well suited for use on high-speed electronic computers, for solving many of the problems of neutron transport theory. In particular it can be used to determine critical sizes and multiplications of subcritical systems in plane, spherical or cylindrical geometry. The original version of the method, and several of its developed forms, are due to B. G. Carlson and co-workers [44,45,46,47,48] at Los Alamos. Some independent development work has also been carried out by Underhill [49] and his group at Aldermaston, notably in connection with the version of the method which takes anisotropic scattering into proper account; this particular aspect is dealt with separately on page 89. In many problems, anisotropic scattering is adequately taken into account by means of the transport approximation and in this, appropriate modifications are made to the cross sections (cf. page 110) which are then used in conjunction with the assumption of *isotropic* scattering. It is the particular case of isotropic scattering which is considered in this Section though most of the main features of the S_n method are common to both isotropic and anisotropic scattering.

The main features of the original S_n method and the discrete S_n method developed by Carlson and co-workers are now described with particular reference to the problems of critical size and multiplication determinations in spherical geometry. For an extension to cylindrical geometry, reference

should be made to the papers by Carlson and co-workers [44,45,47]. The problems of critical size and multiplication determinations are steady state problems but the S_n method can also be used with appropriate extension to deal with time-dependent problems, though these will not be considered here (cf. Carlson [44,47]). In the special class of time-dependent problems in which the neutron population varies exponentially with time, the problem reduces to a steady state one by making a substitution of the form:

$$\Phi_g(r, \mu, t) = \Phi_g(r, \mu) \exp(\lambda t)$$

(notation here applies to spherical geometry, but the substitution applies generally) where $\Phi_g(r, \mu)$ is the neutron scalar flux per unit change in μ about the direction μ. (μ is the cosine of the angle between the motion of the neutron and the radius vector.) Suffix g indicates the neutron group.

The group approximation in spherical geometry gives the following form of the steady state transport equation:

$$\left\{ \mu \frac{\partial}{\partial \mu} + \frac{(1 - \mu^2)}{r} \frac{\partial}{\partial \mu} + \rho \alpha_g \right\} \Phi_g(r, \mu) = S_g(r) + S_g^{(E)}(r, \mu) \qquad (113)$$

where α_g is the inverse mean free path for neutrons in group g at unit material density, ρ is the material density,

$$\left.\begin{aligned} S_g(r) &= \frac{1}{2} \rho \sum_{g'} \beta_{g'g} \Phi_{g'}(r) \\ \Phi_g(r) &= \int_{-1}^{+1} \Phi_g(r, \mu) d\mu \end{aligned}\right\} \qquad (114)$$

where $\beta_{g'g}$ is the transfer probability for transfer from group g' to group g at unit material density. α_g and $\beta_{g'g}$ can be functions of r due to changes of material composition.

$S_g^{(E)}(r, \mu)$ is an externally imposed neutron source. (Externally does not necessarily mean *outside* the system; it may for instance refer to a body source.) This can often be taken into account by the appropriate application of boundary conditions.

In the critical size calculation the non-zero solution of the homogeneous equation obtained by setting $S_g^{(E)} = 0$, is required. This is the usual eigenvalue problem with the critical dimension as the required eigenvalue.

The S_n method consists of dividing the range $(-1, +1)$ of μ into n intervals and assuming that $\Phi_g(r, \mu)$ is linear function of μ in each interval. It is often assumed that the intervals are equal, but this is not essential. Denoting the intervals by $(\mu_{j-1}, \mu_j) j = 1,2 \ldots, n$; $\mu_o = -1$, $\mu_n = 1$, $\Phi_g(r, \mu)$ is given by:

$$\Phi_g(r, \mu) = \frac{(\mu - \mu_{j-1})}{(\mu_j - \mu_{j-1})} \Phi_g(r, \mu_j) + \frac{(\mu_j - \mu)}{(\mu_j - \mu_{j-1})} \Phi_g(r, \mu_{j-1}) \qquad (115)$$

$$\text{for} \quad \mu_{j-1} \leqslant \mu \leqslant \mu_j.$$

The order of the approximation, n, may be any positive integer and when n equal intervals are used it is usual to refer to it as the S_n approximation.

Substituting (115) into (113) and integrating over each of the intervals (μ_{j-1}, μ_j) in turn, the following set of n ordinary differential equations are obtained:

$$\left(a_j \frac{d}{dr} + \frac{b_j}{r} + \rho\alpha_g\right) \Phi_g(r, \mu_j)$$
$$+ \left(\bar{a}_j \frac{\partial}{\partial r} - \frac{b_j}{r} + \rho\alpha_g\right) \Phi_g(r, \mu_{j-1}) = 2S_g(r) + 2S_{gj}^{(E)}(r) \quad (116)$$

where:

$$a_j = (2\mu_j + \mu_{j-1})/3$$
$$\bar{a}_j = (\mu_j + 2\mu_{j-1})/3$$
$$b_j = \frac{2}{3(\mu_j - \mu_{j-1})} \{3 - \mu_j^2 - \mu_j\mu_{j-1} - \mu^2_{j-1}\} \quad (117)$$
$$S_{gj}^{(E)}(r) = \frac{\left\{\int_{\mu j-1}^{\mu j} S_g^{(E)}(r, \mu) d\mu\right\}}{(\mu_j - \mu_{j-1})}$$

This integral may be approximated to in any suitable way, but whatever approximation is used it is ultimately controlled by the need for neutron conservation.

These equations are then solved numerically by iteration, using a simple finite difference approximation, and for this purpose the radius r is divided into I intervals $(r_{i-1}, r_i) i = 1,2, \ldots I$. An essential condition for stability is that for $(-1 \leqslant \mu \leqslant 0)$ the radial difference equations are integrated from the outer towards the inner parts of the system, and for $1 \geqslant \mu \geqslant 0$, the equations are integrated outwards. In other words, they are integrated in the direction of the neutron motion. This also simplifies the application of boundary conditions. If this procedure is not followed, truncation errors in the numerical integration build up in an uncontrollable way.

In the integration of equations (116), $S_g(r)$ and $S_{gj}^{(E)}(r)$ are assumed to be known. Since the boundary conditions are usually specified at the outer boundary, it is in general preferable to carry out the inward integrations first. This requires another equation for each involving $\Phi(r, -1)$ to supplement equations (116) and this is obtained by setting $\mu = -1$ in the transport equation to give:

$$\left\{-\frac{d}{dr} + \rho\alpha_g\right\} \Phi_g(r, -1) = S_g(r) + S_g^{(E)}(r, -1). \quad (118)$$

In carrying out the iteration, each group is treated separately and it is usual to start with the highest energy group, i.e. with $g = 1$. A pass through all the groups with the subsequent determination of a new estimate $S'_g(r)$ to S_g is known as an *outer* iteration. In some variations of the method, $S_g(r)$ is divided into components, one of which is due to the contributions from elastic and/or inelastic (n, n') scattering, and another component due to fission. The contribution to S_g from elastic and inelastic scattering is of the form (say):

$$\frac{1}{2}\rho \sum_{g'=1}^{g} \beta'_{g'g} \Phi_{g'} = {}_1S_g,$$

i.e. $\beta'_{g'g}$ is a triangular matrix, and in such cases an iterative procedure can be carried out on each group in turn, iterating with respect to Φ_g in ${}_1S_g$ but keeping the contribution from fission fixed. An iteration of this kind is referred to as an *inner* iteration. This is the principle of the fission gener-

ation technique in which the contribution to S_g from fission is kept constant during the complete outer iteration.

For a subcritical system in which an external source is specified either by way of boundary conditions or a distributed source, the Φ_g converge to finite values and the convergence can be assessed, for example, by the evaluation of the ratio $\langle S_g \rangle / \langle S_{g'} \rangle$ where S_g is the value of S_g (or that component arising from fission) at the beginning of an outer iteration and $S_{g'}$ the value at the end. $\langle S_g \rangle$ represent suitable averages taken over space and it is convenient in practice to take them as volume integrals. In the subcritical system with sources, $\langle S_g \rangle / \langle S_{g'} \rangle \to 1$ and it is only necessary to specify the degree of convergence required in order to terminate the iterative procedure. If the system with sources is by chance supercritical, then: $\langle S_g \rangle / \langle S_{g'} \rangle \to A < 1$.

In order to determine a critical size, the solution of (116) is obtained with $S_g^{(E)} \equiv 0$ for some estimated critical dimension of the system. In this case, $\langle S_g \rangle / \langle S_{g'} \rangle$ in general converges to a limit $\neq 1$: if the limit $= 1$ then the estimated dimension is the correct one. If the limit of $\langle S_g \rangle / \langle S_{g'} \rangle = A$ (say*) then the system is subcritical or supercritical, dependent on whether A is greater or less than unity. (A is in fact the inverse of the generation constant (cf. page 92) when S_g includes the source from both scattering and fission processes.) Proceeding by a process of iteration until A is arbitrarily close to unity, the critical dimension can thus be determined.

The accuracy of the S_n method can be increased for certain types of problem by imposing an integral conservancy condition on the solution of the equations (116), so that truncation errors do not lead to an artificial increase or decrease in the neutron flux. The transport equation itself is an equation of conservancy and so also are equations (116), but in view of the numerical approximation, (116) do not conserve neutrons exactly. In other words, if they are integrated in accordance with the approximations used, over any given volume, then the net flux of neutrons out of the volume is not exactly equal to the net rate of generation of neutrons within it. For many problems this leads to negligible error but for some problems, for example those in which the transmission of neutrons through thick layers of material is required, it is important that the overall conservancy of the neutrons should be preserved. In this case it is necessary to introduce into the iterative procedure an integral type of conservancy condition over each volume cell considered; thus if r_{i-1} and r_i represent two consecutive points in the integration mesh, then the integrated net flux out of the volume between r_{i-1} and r_i over the spherical surfaces r_i and r_{i-1} should equal the net rate of generation of neutrons within this volume. This conservation is obtained by altering the definition of the term b_j in (116). The formulation of the conservancy condition is discussed by Carlson [44, 45]. The S_n method (in one dimension) has been programmed for the IBM 704 by Carlson and his group and a description of the code — SNG — is given in Report LAMS-2201 [45]. An isotropic (one-dimensional) S_n code is one of the options in the multi-purpose code SAINT (cf. page 89) written by Underhill and his group at Aldermaston. With any of these one-dimensional codes, problems can be solved in either plane or spherical geometry. The case of plane geometry is obtained from the equations for spherical geometry by setting $b_j = 0$ (cf. equations (116)).

Improved and simplified versions of the S_n method have been developed by Carlson and co-workers and though two different forms exist, both are

* This is actually the inverse of the collision generation constant defined on page 92.

known as the discrete S_n method. The first form of the discrete S_n method was described at the Geneva Conference [46] and in Report LAMS-2201 [45], though in the latter it is referred to as the *modified* S_n method. It is coded as the SNK code (cf. LAMS-2201). The second form is the basis of the one-dimensional code DSN and the two-dimensional code TDC and is described in Report LA-2260 [47]. The main features of these two forms of the discrete S_n method will now be described.

The first discrete (or modified) S_n method, is not as rigorous as the original version of the S_n method but it is considerably simpler and therefore easier to apply. The basis of the method is to replace the S_n equations (116) by equations of the form:

$$\left(\bar{\mu}_j \frac{d}{dr} + \frac{C_j}{r} + \rho\alpha_g\right) \Phi_g(r, \bar{\mu}_j) - \frac{C_j}{r} \Phi_g(r, \mu_{j-1}) = 2S_g(r) \qquad (119)$$

where the C_j are constants to be determined; $\mu_{j-1} \leqslant \bar{\mu}_j \leqslant \mu_j$; $j = 1,2, \ldots n$; $b_o = 0$; $\mu_o = -1$. The assumption of equation (119) is a somewhat arbitrary step to take and its justification lies in the fact that it greatly simplifies the numerical solution and ultimately leads to reliable results.

$\Phi_g(r, \mu_j)$ is defined by extrapolation thus:

$$\Phi_g(r, \mu_j) = \frac{(\mu_j - \mu_{j-1})}{(\bar{\mu}_j - \mu_{j-1})} \Phi_g(r, \bar{\mu}_j) - \frac{(\mu_j - \bar{\mu}_j)}{(\bar{\mu}_j - \mu_{j-1})} \Phi_g(r, \mu_{j-1}) \qquad (120)$$

It is further assumed that:

$$\Phi_g = \sum_j \frac{1}{2} (\mu_j - \mu_{j-1}) \Phi_g(r, \mu_j) \qquad (a)$$

and: (121)

$$J_g(r) = \sum_j \frac{1}{2} (\mu_j - \mu_{j-1}) \mu_j \Phi_g(r, \bar{\mu}_j) \qquad (b)$$

{ Φ_g is the scalar flux of group g neutrons, and
J_g is the net outward flux per unit area of group g neutrons }

Integration of the transport equation over μ requires that:

$$\left(\frac{d}{dr} + \frac{2}{r}\right) J_g(r) + \rho\alpha_g \Phi_g(r) = S_g(r) \qquad (122)$$

which is a necessary condition of neutron conservation. Equations (119) to (121) are therefore conditioned to satisfy this equation, thus leading to the determination of C_j:

$$C_j = 2 \left\{ \bar{\mu}_j - \frac{2}{(\bar{\mu}_j - \mu_{j-1})} \sum_j^j \frac{1}{2} (\mu_k - \mu_{k-1}) \bar{\mu}_k \right\} \qquad (123)$$

The $\bar{\mu}_j$ are determined (somewhat arbitrarily) so that in the diffusion theory approximation (scalar flux represented as a linear function of μ), the component of the vector flux arising from the μ component in the scalar flux Φ is exactly satisfied by the approximations used (this condition is automatically satisfied in the original S_n approximation). Thus:

$$\frac{1}{2} \int_{-1}^{+1} \mu^2 d\mu = \frac{1}{2} \sum_j (\mu_j - \mu_{j-1}) \bar{\mu}_j^2 \equiv \frac{1}{3} \qquad (124)$$

(Carlson refers to this as the diffusion theory condition.)

With equal intervals, $(\mu_j - \mu_{j-1}) = 2/n$ and $\bar{\mu}_j = \frac{1}{2}(\mu_j + \mu_{j-1}) + \varepsilon$; this condition is satisfied provided $\varepsilon = \frac{1}{2}\left[\left(1 + \frac{4}{3n^2}\right)^{\frac{1}{2}} - 1\right]$ sign $(\mu_j + \mu_{j-1})$.

The solution of equations (119) follows by iteration in a similar way as in the original S_n method, and the integral type of conservancy condition can also be included to give improved accuracy.

In view of the approximations made in the modified S_n method, detailed aspects of the results, such as the angular distribution of neutrons at a given radius, are perhaps not as well represented as in the original S_n method but the overall representation in terms of net fluxes, integrated scalar fluxes and eigenvalues, is found to be very good.

In the second form of the discrete S_n method, the difference equations are obtained directly from the differential equation. Thus:

$$\mu \frac{\partial}{\partial r}\Phi_g(r, \mu) + \frac{(1 - \mu^2)}{r}\frac{\partial \Phi_g(r, \mu)}{\partial \mu} + \rho(r)\alpha_g \Phi_g(r, \mu) = S_g \qquad (125)$$

becomes:

$$\left.\begin{aligned} \bar{\mu}_m u_i \{ \Phi_{g,i,m} - \Phi_{g,i-1,m} \} + \gamma_m s_i \{ \Phi_{g,i,m} - \Phi_{g,i,m-1} \} \\ + 2h_i \Phi_{g,i,m} = \Delta_i S_{g,i} \end{aligned}\right\} \qquad (126)$$

where $\Phi_{g,i,m}$ is the angular flux of neutrons in group g at the mesh points r_i, μ_m. $(i = 1,2 \ldots;\ m = 0, 1, 2, \ldots, n)$

$$\left.\begin{aligned} &\Delta_i = r_i - r_{i-1} \\ &\bar{\mu}_m \text{ is an average } \mu \text{ over the interval } (\mu_{m-1}, \mu_m) \\ &s_i = \frac{\Delta_i}{r_i}\frac{(r_i - r_{i-1})}{(r_i + r_{i-1})/2} \\ &h_i = \rho_i \alpha_g \Delta_i/2 \end{aligned}\right\} \qquad (127)$$

ρ_i and $S_{g,i}$ are mid-point values for ρ and S_g.

The γ_m and u_i are chosen to give conservation when the equations are integrated (cf. LA-2260) [47] so that:

$$\left.\begin{aligned} u_i &= 3\Delta_i(r_i^2 + r_{i-1}^2)/2(r_i^3 - r_{i-1}^3) \\ \gamma_m - \gamma_{m-1} &= -\bar{\mu}_m - \bar{\mu}_{m-1};\quad \gamma_1 = -\bar{\mu}_1 \end{aligned}\right\} \qquad (128)$$

and:

$\bar{\Phi}_{g,i,m}$ is the angular flux at the mid-point of the mesh and the relations:

$$\left.\begin{aligned} \Phi_{g,i,m} + \Phi_{g,i-1,m} &= 2\bar{\Phi}_{g,i,m} \\ {}_g\Phi_{,i,m} + \Phi_{g,i,m-1} &= 2\bar{\Phi}_{g,i,m} \end{aligned}\right\} \qquad (129)$$

are assumed to hold. Thus (126) simplifies to:

$$\left.\begin{aligned} \{ \bar{\mu}_m u_i + \gamma_m s_i + h_i \{ \Phi_{g,i,m} + \{ -\bar{\mu}_m u_i + \gamma_m s_i + h_i \{ \Phi_{g,i-1,m} \\ - 2\gamma_m s_i \bar{\Phi}_{g,i,m-1} = \Delta_i S_{g,i} \end{aligned}\right\} \qquad (130)$$

The $\bar{\mu}_m$ are fixed by application of the diffusion condition as in the previous version of the discrete S_n method.

The following relations are also used for the scalar and vector fluxes:

$$\left.\begin{aligned} \Phi_{g,i} &= \frac{1}{2}\sum_m \bar{\Phi}_{g,i,m}\Delta\mu_m \\ J_{g,i} &= \frac{1}{2}\sum_m \bar{\mu}_m\bar{\Phi}_{g,i,m}\Delta\mu_m \end{aligned}\right\} \tag{131}$$

Given $S_{g,i}$ and the boundary conditions, equations (130) are then solved to give $\Phi_{g,i,m}$ for each m in turn, beginning with $m = 0$. Note that for $m = 0$, $\bar{\mu} = -1$ and $\gamma = 0$. In order to reduce round-off errors, equations (130) are solved for $\Phi_{g,i-1,m}$ if μ_m is negative, and for $\Phi_{g,i,m}$ if $\bar{\mu}_m$ is positive. This is equivalent to integrating in the direction of the neutron current as was necessary in the original S_n method. The values obtained for $\Phi_{g,i,m}$ allow a new estimate of $S_{g,i}$ to be made, and the final solution is obtained by iteration in the usual way.

Anisotropic S_n Method

It is worth noting that the simplified form of the DSN equations does not save much machine time, but the full advantage of the simplification is apparent in two dimensions (TDC), where a very considerable saving is made on the time taken to do similar calculations based on a two-dimensional extension of the ordinary S_n equations.

The anisotropic version of the S_n method allows one to take into proper account the effect of anisotropic scattering, and this is particularly important where the transport of neutrons in hydrogenous substances is concerned.

When anisotropic scattering is taken into account the Boltzmann transport equation in multi-group approximation and in spherical co-ordinates can be written:

$$\left.\begin{aligned} &\left\{\mu\frac{\partial}{\partial r} + \frac{(1-\mu^2)}{r}\frac{\partial}{\partial\mu} + \alpha_g\rho(r)\right\}\Phi_g(r,\mu) \\ &= \frac{1}{2\pi}\int_0^{2\pi}\int_{-1}^{+1}\sum_{g'=1}^{G}\beta_{g'g}(\bar{\mu})\rho(r)\Phi_{g'}(r,\mu')d\mu'd\Phi \end{aligned}\right. \tag{132}$$

$\beta_{g'g}(\mu)$ is the transfer cross section representing transfer of neutrons from group g' to group g which are scattered through an angle $\cos^{-1}\bar{\mu}$ for material at 1 g/cm³.

$$\bar{\mu} = \mu\mu' - \sqrt{(1-\mu^2)(1-\mu'^2)}\cos\Phi \tag{133}$$

Other symbols are as defined on page 83.

Expanding $\beta_{g'g}(\bar{\mu})$ in terms of Legendre Polynomials $P_l(\mu)$ and using the relation:

$$P_l(\bar{\mu}) = P_l(\mu)P_l(\mu') + 2\sum_{m=1}^{l}\frac{(1-m)!}{(1+m)!}P_1{}^m(\mu)P_1{}^m(\mu')\cos(m\Phi) \tag{134}$$

where $P_1{}^m$ are the Associated Legendre Polynomials, the R.H.S. of equation (132) becomes, after integration over Φ:

$$\sum_{g'=1}^{G}\sum_{l=0}^{\infty}\frac{(2l+1)}{2}\beta^1{}_{g'g}P_l(\mu)\rho(r)\int_{-1}^{+1}P_l(\mu')\Phi_{g'}(r,\mu')d\mu' \tag{135}$$

where:

$$\beta^{l}{}_{g'g} = \int_{-1}^{+1} \beta_{g'g}(\bar{\mu}) P_l(\bar{\mu}) d\bar{\mu} \tag{136}$$

In the S_n formulation, the anisotropic source term is required to be averaged over an angular interval (μ_{j-1}, μ_j)* and integrated over a radial interval (r_i, r_{i+1}). Denoting this averaged source term by $S(i, j)$ and using the basic assumption of the S_n method that $\Phi(r, \mu)$ is a linear function of μ in an angular interval, $\mu_{j-1} < \mu < \mu_j$, a set of scalar products for all 'i' of the form :

$$S_g(i, j) = \sum_{g'=1}^{G} \sum_{l=0}^{L} \sum_{j'=1}^{J} A_{jl} \beta^{l}{}_{g'g} B_{j'l} F_{g'}(i, j'). \tag{137}$$

can be derived. J and L are the *total* number of angular intervals and Legendre moments respectively.

A_{jl}, $B_{j'l}$ and $F_{g'}(i, j')$ are defined as follows:

$$\begin{aligned}
A_{jl} &= \frac{1}{(\mu_j - \mu_{j-1})} \int_{\mu_{j-1}}^{\mu_j} P_l(\mu) d\mu \quad \text{for } j > 1. \\
A_{1l} &= P_l(-1). \\
B_{j'l} &= \frac{(2l+1)}{2} \left\{ \int_{\mu_{j'}}^{\mu_{j'+1}} \left(\frac{\mu_{j'+1} - \mu}{\mu_{j'+1} - \mu_{j'}} \right) P_l(\mu) d\mu \right. \\
&\left. + \int_{\mu_{j'-1}}^{\mu_{j'}} \left(\frac{\mu - \mu_{j'-1}}{\mu_{j'} - \mu_{j'-1}} \right) P_l(\mu) d\mu \right\} \quad \text{for } 1 < j' < J. \\
B_{1l} &= \frac{(2l+1)}{2} \int_{\mu_1}^{\mu_2} \left(\frac{\mu_2 - \mu}{\mu_2 - \mu_1} \right) P_l(\mu) d\mu. \\
B_{Jl} &= \frac{(2l+1)}{2} \int_{\mu_{J-1}}^{\mu_J} \left(\frac{\mu - \mu_{J-1}}{\mu_J \mu_{J-1}} \right) P_l(\mu) d\mu \\
F_{g'}(i, j') &= \int_{r_i}^{r_{i+1}} \rho(r) \Phi_{g'}(r, \mu_{j'}) dr. \\
&\text{Note: } \rho(r) \text{ is required to be constant between } r_i \text{ and } r_{i+1}. \text{ Hence:} \\
F_{g'}(i, j') &= \frac{1}{2} \rho_{i+\frac{1}{2}} \{ \Phi_{g'}(r_i, \mu_{j'}) + \Phi_{g'}(r_{i+1}, \mu_{j'}) \}.
\end{aligned} \tag{138}$$

The form of equation (137) is the same as given by Carlson in LA 1891 [44].

The summation over 'j' in equation (137) is limited by J, the total number of angular intervals. The truncation of the summation over 'l' to L, however, is purely a matter of expediency, as mathematically the summation should be extended to infinity**. It can be seen from equation (135) that in practice

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* For $\mu_1 (= -1)$, the source term required is the value at $\mu = -1$.

** In practice it is usually adequate to take into account only $l \leqslant 5$, i.e. $L = 5$.

the truncation occurs either from β^l becoming zero for a large enough value of 'l' or from the moment:

$$\Phi^l(r)\left(=\int_{-1}^{+1} P_l(\mu)\Phi(r,\mu)d\mu\right)$$

of neutron scalar flux becoming zero.

A machine code — SAINT — has been developed by Underhill and his group at Aldermaston, to deal with the anisotropic source term given by equation (137) above. SAINT is a multi-purpose, one-dimensional S_n code, with options for isotropic and anisotropic scatter calculation; versions exist for use on 32K IBM 704, 709 and 7090 machines. An account is now given of the salient features of the anisotropic scatter option.

In SAINT, the nuclear data is supplied in the form $\beta^l{}_{g\,g}$ up to any reasonable order of l on binary cards of a standard format. The summation over l in (137) is carried out once and for all during the input stage of a calculation, so during the course of a calculation, the source term is evaluated by:

$$S_g(i,j) = \sum_{g'}\sum_{j'} C(g,g',j,j')\Phi_{g'}(i,j'). \qquad (139)$$

This decreases the amount of computation needed at the expense of increasing the amount of storage required. In particular, calculation time is effectively independent of the number of moments used.

The procedure adopted during the solution is the 'fission generation' iterative technique. Here the fission component of the scattering term is split off from the rest of the scattering terms and it is assumed that fission neutrons are emitted isotropically with an energy spectrum independent of material or incident neutron energy. The contribution by fission is thus handled by a 'fission source term' which is recomputed from the neutron scalar fluxes at the end of each 'outer iteration'. Within each outer iteration the fission source term is unchanging and the neutron scalar fluxes appropriate to the fission source term are calculated. Thus, in equation (139), only slowing down neutron processes occur and so the g' summation is over the range $g' = 1(1)g$ ($g = 1$ denoting the neutron group of highest energy).

For the purposes of calculation within each outer iteration, the R.H.S. of (139) is split up into an off-diagonal and a diagonal component:

$$S_g(i,j) = \sum_{g'=1}^{g-1}\sum_{j'} C(g,g',j,j')\Phi_g\ (i,j') + \sum_{j'} C(g,g,j,j')\Phi_g(i,j'), \qquad (140)$$

and for each outer iteration one starts with $g = 1$. In this case, the first term of the R.H.S. of (140) is zero and a one-group iterative process ('inner iteration') is used to calculate Φ_1 to the required degree of convergence. Proceeding to $g = 2$, the first term on R.H.S. of (140) is evaluated using the already calculated Φ_1, followed by another set of one-group iterations till Φ_2 is calculated to the required degree of convergence.

This process is repeated, always evaluating the first term of the R.H.S. of (140) once for each g in terms of the already calculated Φ's, till the scalar flux for the final group has been calculated. The outer iteration is then completed by the evaluation of a new fission source term.

The whole process is terminated when two successive fission source terms differ by less than some predetermined value.

This iterative scheme is particularly useful for problems where no fission processes are involved; for example, calculating the transmission of neutrons through a shield. Here only one outer iteration is needed to complete the calculation, the inner iteration being taken to finality for each group in turn.

For problems involving a fission process (for example, a critical size calculation) the degree of convergence to which each inner iteration is carried is determined by the degree of convergence attained by the outer iteration. This works reasonably well but a completely satisfactory scheme has not yet been devised.

The code can be used for critical size calculation (no external source present) or an external source can be specified either as an isotropic source distributed over desired energy groups and regions of the system, or as a shell source from which neutrons are emitted into specified groups with specified angular distributions. In the external source problem, vector flux analysis allows one to calculate any multiplication which may be required. In all cases, either plane or spherical (but not cylindrical) geometries can be specified.

In general, an anisotropic calculation takes about four times as long as a similar isotropic calculation. For calculations on systems without fissile material, anisotropic calculations are quite fast, a transmission problem involving 8 neutron groups in S_4 approximation with 50 radial points taking of the order ten minutes on the IBM 709.

The code uses five tapes, distributed for the IBM 709/7090 machines over channels, A, B and C.

The Monte Carlo Method

The interaction of neutrons with matter is basically a stochastic process with the various probability laws which govern the process determined by the nuclear physics. The probability laws can be specified in terms of neutron cross sections and angular and energy distributions of the emergent neutrons from the different types of collision. If such a specification of the basic laws is made, either in tabular or analytic form, then the actual problem of the interaction and transport of neutrons can be simulated by carrying out a process of random sampling from the various laws in a specified order, and accumulating the results obtained. This is the basic principle behind the Monte Carlo method (in its applications to neutronics), and in its simplest form an exact simulation of the system under consideration is aimed at. Any departure from this physical reality is referred to as sophisticated Monte Carlo and the main objective of sophistication is to obtain the most accurate results with the minimum of effort, in other words, to make the most efficient use of the effort put into solving a given problem. Special techniques towards this end are interesting theoretically and they are also very important from a practical point of view in a wide variety of problems. It must, however, be emphasized that straightforward simulation methods do play a significant part in Monte Carlo neutronics studies.

It must also be emphasized that whether straightforward simulation Monte Carlo or whether sophisticated techniques are used, it is in general necessary in practical application to do Monte Carlo calculations on an electronic computer if reliable results are to be obtained to any but the very simplest of problems. The following paragraphs are therefore written with this in mind.

In the Monte Carlo method one is tracking neutrons from one collision to the next, and the results can be accumulated as a function of time, or as a function of the number of collisions, or as a function of any other suitable parameter. In going from one collision to the next one begins, for a particular neutron, with its position at a collision just taken place, its direction of motion and its energy. The problem is then to determine:

i) How far it travels before having its next collision, and hence the location of its next collision;

ii) What type of collision takes place, e.g. fission, elastic scatter, (n,γ) etc.;

iii) The number of emergent neutrons, their energy, angle of scatter and hence direction of motion.

At this stage the process repeats itself and continues until either all the neutrons have escaped from the system or have settled, according to some predetermined criterion, into their eigendistribution. The determination of (*i*), (*ii*) and (*iii*) is carried out by appropriate use of random numbers from the probability laws involved. This is now discussed in more detail.

i) *Determination of path length.* The distance which a neutron travels from one collision to the next is dependent on its mean free path l, which is a function of its energy and of the material in which it is moving. If x is the neutron's path length measured from its previous collision, then the probability $p(x)dx$ that the next collision takes place in x, $x + dx$ is given by:

$$p(x)dx = \exp\left(-\frac{x}{l}\right)\frac{dx}{l}. \tag{141}$$

The problem of determining the distance X which a neutron travels before having its next collision is basically that of mapping the uniform distribution $f(y)dy = dy(0 \leqslant y \leqslant 1)$ onto the distribution (141).
Note that:

$$Pr(x \leqslant X) = 1 - \exp\left(-\frac{X}{l}\right). \tag{142}$$

This is to be identified with a random number, R_1 (say) in (0,1), giving:

$$X = l.ln(1 - R_1) = -l.lnR_2 \tag{143}$$

where R_2 is random in (0,1).

If a determination of this sort takes a neutron across a boundary, then, if the boundary is a free surface, the neutron escapes from the system, otherwise the neutron must be re-located at the boundary with appropriate direction of motion and a re-determination of its path length in the second region made using another random number and the mean free path applicable to that region.

ii) *Type of collision.* The probabilities of the different collision types are all explicit functions of the basic data. Let $a_i(E)$ $(1 \leqslant i \leqslant k)$ be the *relative* probability for collision type i for a neutron of incident energy E. $a_i(E)$ is proportional to the cross section for reaction i. A random number R is chosen and multiplied by:

$$S = \sum_{i=1}^{k} a_i(E)$$

and RS is then tested consecutively against $a_1(E)$, $a_1(E) + a_2(E)$, $a_1(E) + a_2(E) + a_3(E)$, ... etc. Where RS first exceeds one of these partial sums, the collision type is identified; thus, for example, if RS $> a_1(E)$ but $a_1(E) + a_2(E)$, then the collision is of type 2.

iii) *Number, energy and direction of emergent neutrons.* The number of neutrons, where this is different from an integer, as in fission, can be determined by sampling in a way similar to that in (*ii*) above, either using the experimental distribution of the numbers of neutrons per fission, or using a pseudo distribution which gives the correct mean. In most instances this latter procedure is adequate and allows the number of neutrons per fission to be taken as one of two integers. It also has the advantage that by reducing the spread of values which the actual distribution of the number of neutrons per fission allows, it gives a better estimate of the mean value after a given number of fissions.

The energy of the emergent neutrons can also be obtained in a similar way if the possibilities are discrete with given relative probabilities. In cases where the possibilities are not discrete and where the distribution has no particular mathematical form as in the cases of experimentally determined continuous energy spectra, and differential elastic scatter distributions, the first stage is to divide the interval (0,1) from which the random numbers are selected into K equal sub-intervals (generally, for computer convenience, K is a power of 2). A chosen random number must lie in one of these sub-intervals and correspond to a particular value of x, the variate to be sampled from. In other words, the variate x is divided into K equiprobable intervals and a suitable value of x taken to correspond to each interval; in fact the K values of x are precomputed according to the law:

$$\int_{B}^{x_k} f(x)dx = \frac{1 + 2k}{K} \quad (k = 0, 1, 2, \ldots, K - 1), \tag{144}$$

where $f(x)$ is the normalized empirical frequency distribution, B being the lower limit of integration, and is determined by the physics. Thus, the distribution f is approximated to by K delta-functions (linear interpolation between them can be used to improve the representation if required).

It will be appreciated at this stage that one of the first requirements for Monte Carlo calculations is a stock of random numbers. When using a digital computer for Monte Carlo work it is generally inconvenient (on the grounds of storage economy) to input random numbers from a tested source and instead a sequence of pseudo-random numbers is generated by a suitable recurrence relation. A relation of the type:

$$R_{n+1} = AR_n (\text{mod } m) \tag{145}$$

where A and m are constants whose optimum choice depends on the digital computer used, is now almost universal. Many basic studies regarding the periodicity and the 'randomness' (in the sense that a number of statistical tests for randomness should be satisfied) of the numbers generated by such a sequence have been carried out [50].

When tracking neutrons in a Monte Carlo calculation one can do this either in sequence or in parallel. Tracking in sequence involves tracking a neutron and its descendants until they have either all escaped from the system or been absorbed, before selecting a new neutron for study, and this is sometimes of advantage in calculations of central, body or surface multiplications.

In eigenvalue problems (e.g. the determination of the decay rate of a near-critical assembly) it may be preferable to track neutrons in parallel, taking each of a pre-assigned sample from one stage to the next. If the object of study is a function of the eigendistribution, then reliable estimates of the function cannot in general be obtained until this distribution is attained, and hence it is important to select as representative a starting distribution as possible to reduce the amount of tracking during the settling to the eigendistribution. Other ways of reducing this wasted tracking are discussed, for example, by Parker and Woodcock [51].

When studying systems which are close to critical, it is necessary to measure the degree of supercriticality or subcriticality in terms of some suitable parameter. One choice of parameter is the time-constant λ defined by:

$$N(t) = N_0 e^{\lambda t} \tag{146}$$

where N_0 is the eigendistribution population at a time zero and $N(t)$ that at time t. Thus, a possible way of adjudging criticality is to track neutrons, assembling the population figures as a function of time and hence determining λ. When the distribution has settled to its eigendistribution the plot of ln N(E) as a function of t will be linear, with slope λ. Positive λ indicates supercriticality, negative λ subcriticality. In general, however, λ is not an ideal parameter for this purpose. For instance, when there are neutrons of a wide range of energies present, the faster neutrons undergo a large number of collisions while the slowest are settling into their eigen state, and this involves wasteful tracking. It is often much better practice to forget the time dependence and to track neutrons from one collision to the next, each collision giving rise to a new collision generation. Thus, one obtains the neutron population as a function of the number of collision generations. The ratio of the population in generation $(n + 1)$ to that in generation n after the distribution has settled into its eigen state is known as the *collision generation constant* and the system is supercritical or subcritical dependent on whether the generation constant is greater or less than unity.

The use of the collision generation constant in the determination of criticality still suffers from a sensitivity to the degree of settling to the eigen state. Thus, a lack of settling of neutrons in the outer parts of a thick reflector can have a significant effect on the collision generation constant although these neutrons have a negligible effect on the true criticality of the system. There are various ways in which this difficulty can be mitigated: for instance, one may study only the population in the fissile core as a function of collision generation constant in determining the degree of criticality; or alternatively one can determine the fission generation constant, that is the growth from one fission to the next; but one of the best methods is that which leads to the traverse technique [52].

In a system consisting of a fissile core surrounded by a reflector, if, when an eigen state has been reached, M neutrons leave the core for every neutron which enters and for every neutron which leaves the core into the reflector, and a fraction R are reflected back to the core, then the system is supercritical or subcritical according to whether MR is greater or less than unity and the system is just critical when MR is unity.

Neutrons, usually in the fission spectrum, are started isotropically near the centre of the core and tracked until they are either absorbed or lie on the specified boundary. Note that no attempt is made to start neutrons in a simulated eigendistribution. The tracking now switches to the reflector

materials and continues until the neutrons are either absorbed, lost from the system, or have returned to the specified boundary. The procedure is then repeated, picking up the neutrons from the boundary and tracking them in the core. Successive print-outs of inward and outer boundary crossings are produced. This is the traverse technique. The ratio of the outward crossing to the previous inward crossing is an estimate of M and the ratio of the inward crossing to the previous outward crossing an estimate of R. Experience shows that the estimates of M and R tend to their final values more rapidly than the system settles to its eigen state, and that they are not much affected by lack of settling in outer regions of the system which are not important from the criticality point of view. It is found in practice that M and R have settled after only one or two traverses, and so the results for subsequent traverses can be pooled to give a better estimate of M and R.

The traverse technique described above is an example of how the appropriate accumulation of results — in terms of boundary crossings — and the use of a suitable index of criticality — RM — can be used to facilitate a particular type of calculation. In the next Sections a brief account will be given of special techniques which are of practical use in certain other types of problem and which depend on a degree of sophistication in the Monte Carlo tracking.

In the *weighting method* a neutron is made to carry a weight which can change from collision to collision, or at some other appropriate stage, during the tracking. As an example, the neutron's weight immediately after a collision may be obtained by multiplying its previous weight by the expected number of secondary neutrons occurring at the collision, thereby reducing sampling errors occasioned by different numbers of neutrons emerging after a collision. Uncontrolled weighted tracking can on occasion be inefficient, since a large amount of computing time may be spent on particles carrying low weight. This leads naturally to a new concept, namely, splitting neutrons with too great a weight *(track splitting)* and absorbing them when the weight is too low *(' Russian Roulette ')*. The criteria for track splitting and playing Russian Roulette can, and indeed frequently should (otherwise the procedure may revert substantially to simulation Monte Carlo), vary throughout phase-space. This variation is a particular example of *importance sampling* where attention is focussed on classes of particles considered to be particularly important contributors to the desired result, by subdividing the tracks in the appropriate regions of phase-space.

The technique of *antithetic variation* [53] is a very useful one which can, with advantage, be applied also to unweighted tracking Monte Carlos. Where two or more neutrons are to be liberated at, for example, a collision, the emergent particles are emitted not independently with the appropriate frequency distribution, but in a correlated manner. This is equivalent to systematic, as opposed to random, sampling at a collision and involves only one random choice instead of one for each particle. This systematic sampling can be done either in scatter angle or energy, possibly in both simultaneously.

In shielding problems where one is interested in the transmission of neutrons through very thick layers of absorbing material, only a very small fraction (often $e^{-10} - e^{-20}$) of the incident neutrons are actually transmitted, but it is necessary to be able to calculate this fraction. Direct simulation Monte Carlo would require the tracking of far too many neutrons in order to obtain any results, and so one must resort to special techniques. The

weighting method with suitable innovations can be used to solve such problems but there is also another method which essentially involves making a mathematical transformation to the original Boltzmann equation, followed by a Monte Carlo solution of the stochastic problem described by the transformed equation. The transformation is to replace the neutron current $N(x, \mu, E)$, where x is the distance co-ordinate in plane geometry and μ is the cosine of the angle between the direction of the neutron and the x axis:

$$N(x, \mu, E) = \psi(x, \mu, E) \exp(-kx) \tag{147}$$

where k is a constant. By suitable choice of k the variations of:

$$\int \psi(x, \mu, E) dE$$

with x can be controlled to within (say) a factor 10 over the whole range of x, so that direct simulation Monte Carlo can be used to determine $\psi(x, \mu, E)$. Substitution of (147) into the transport equation for $N(x, \mu, E)$ gives a corresponding transport equation for $\psi(x, \mu, E)$ which is the same as the equation for N except that the inverse mean free path is reduced by the quantity μK at all energies and consequently becomes dependent on the direction of the neutrons. Both this method and the weighting method have been satisfactorily used at Aldermaston to solve simple transmission problems.

Traffic and Matrix Methods

In neutron traffic theory a neutron is not free to take any one of the physically possible tracks; it has to take certain selected routes or lanes which are defined geometrically. There are two forms of neutron traffic models; the continuous and the discrete. A sketch of unpublished work on the continuous model is given by Ackroyd and Ball [18] together with an outline of the discrete traffic model for a non-multiplying medium. In the latter model neutrons can travel only along a network of discrete lanes lying in the medium, occupy discrete energy levels, and be changed in direction at the lattice points or junctions of the network. Removal is assumed to take place continuously along a lane between lattice points according to the usual attenuation formula. The redirection of a neutron after a scattering collision in a lane is allowed to take place at lattice points only. The scattering is advanced if the neutron is redirected at the last lattice point it passed on entering the lane, and the scattering is retarded if redirection takes place at the next lattice point along the lane. A method of assigning scattered neutrons to lanes is to determine their physical directions by a Monte Carlo technique [54]; each neutron is then assigned to the lane nearest to its physical direction. Thus, the only positions and directions of neutrons which are of interest, are given by the lattice points.

A convenient system of lanes is obtained with a cubic lattice, by joining a lattice point to the nearest twenty-six lattice points. The number of lanes per lattice point is constant and their directions do not vary with the lattice point. A model similar to the discrete traffic model has been proposed by Schiff [55]. In his model, interactions of neutrons with nuclei are permitted at lattice points, and neutrons can travel between lattice points only in directions parallel to the co-ordinate axes. In the Schiff model, there is no attenuation along a lane itself. The position, energy and direction of a neutron, i.e. its state, are discrete variables which can be represented conveniently

by a vector. If the vector J_I represents a beam of neutrons incident on a slab (say), there correspond a transmission vector J_T and a reflection vector J_R which are related by:

$$\left.\begin{aligned} J_T^+ &= T^+ J_I^+ \\ J_R^+ &= R^+ J_I^+ \end{aligned}\right\} \tag{148}$$

for transmission in the positive sense, left to right; and

$$\left.\begin{aligned} J_T^- &= T^- J_I^- \\ J_R^- &= R^- J_I^- \end{aligned}\right\} \tag{149}$$

for transmission from right to left. The matrices T^+ and T^- represent the effect of transmission from left to right, and from right to left, respectively. Similarly, the effect of reflections is represented by the matrices R^- and R^+. If the block is homogeneous, then of course:

$$T^+ = T^-, \qquad R^+ = R^- \tag{150}$$

One of the attractions of the traffic version of Monte Carlo is that one can solve the problem of transmission and reflection from a thick block by dividing the block into small units, for which transmission and reflection matrices can be calculated conveniently. Since the direction of lanes in the neutron traffic model is invariant to the lattice point, it is possible to couple blocks together and obtain the overall performance of the blocks from the transmission and reflection matrices of the units by matrix algebra.

A similar coupling technique [56] can be used with a straight Monte Carlo for the units in a block. A Monte Carlo calculation is performed for each unit, assuming a discrete input with respect to energy and angular distributions for the incident neutrons, as in the traffic method. The reflected and transmitted neutrons are tabulated and then assigned in each case to the nearest direction and energy level of a system of discrete directions and energy levels. The reflected and transmitted neutrons can then be represented by vectors in the same way as for the traffic model. The overall performance of the block is obtained from the performances of the units by matrix methods exactly as for the traffic model.

Geometrical Methods

Although there are many approximate methods of solving the Boltzmann equations of reactor theory, ranging from diffusion theory to the Carlson S_n method, none of these methods gives a precise upper limit for the error of the approximation. It is true that with processes such as those used in Carlson's S_n and the spherical harmonics method, an indication of the error can be obtained from the trend of a succession of calculations made to higher and higher orders of approximation. A check is also available in the few cases where exact solutions are known. The fact that these methods do not give a precise upper limit for the error of the approximate solutions is usually ignored on the intuitive grounds that the trend of higher and higher order approximations is reliable. Furthermore, it is not always obvious which are good and which are bad approximations (e.g. see Davison's discussion of the odd and even order approximations for the spherical harmonics method [1]). Checks against the few exact solutions may not reveal the weak-

nesses of an approximate method. Even supposing that intuition can be relied upon, there remains the practical problem of performing a succession of calculations to higher and higher orders of approximation, which for some problems leads to much expensive work on a large computer. Some saving in computing time could be achieved if the methods of calculation used gave a precise upper limit for the error made with any order of approximation, for it would be necessary to go only as far as the order of approximation with an acceptable error.

A review is now given of a new method proposed by Ackroyd [57] for obtaining approximate solutions of the one-group Boltzmann equation for absorbing media. A sketch is then given of a similar method which has been proposed for the multi-group Boltzmann equations for a reflected reactor [58]. The method for the one-group Boltzmann equation gives an upper limit for the error made with any order of approximation, and should, therefore, be useful for checking both the neutron density and the slowing-down density for each group in multi-group shielding calculations based on approximate methods such as diffusion theory. The method is flexible, since it is not restricted like analytical methods such as the Wiener-Hopf [1] techniques to bodies of simple shape, and it can be applied with either numerical or analytical techniques. The main purpose of this note is, however, to serve as an introduction to the study of the multi-group Boltzmann equations by similar techniques.

In the classical methods of integrating the Boltzmann equation (151), the first step is to approximate the angular density $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ in some manner, and the complexity of the approximation is indicated by a subscript. Thus, in the application to spherically symmetric systems of the Carlson S_n method, the angular neutron density approximation at radius r, is sectionally linear in μ (the cosine of the angle between the radius vector $\underset{\sim}{r}$ and the direction $\underset{\sim}{\Omega}$ of the angular density), with n sections. Similarly, in the D_n or discrete ordinate method of Wick-Chandrasekhar [59, 60], the angular neutron density is approximated by an interpolating polynomial of degree m over the complete range $-1 \leqslant \mu \leqslant 1$. The well-known spherical harmonics method has been shown by Goertzel [61] to be equivalent to the D_n method as regards the angle approximation used for the angular density. These approximations allow the integration over angles which appears in the Boltzmann equation to be carried out. The Boltzmann equation is reduced by the approximations to a system of coupled linear ordinary equations in the radial co-ordinate r only. The approximate solution of the Boltzmann equation is then obtained by numerical integration of the coupled ordinary differential equations.

Apart from rounding errors, two types of error clearly arise with conventional methods; an error due to angular truncation of the scattering kernel and angular density and the spatial error due to the replacement of the coupled differential equations by difference equations. The angular truncation can be eliminated by restricting the scattering kernels to those with only a finite number of spherical harmonics [62] in the expansion for the kernel. This approach is unsatisfactory in principle, because it restricts the class of scattering kernels, and also in practice, because a spherical harmonic expansion is not necessarily the most convenient representation of a scattering kernel.

The geometrical method reviewed below is outlined for those scattering kernels which can be integrated exactly, when weighted in certain ways. The general case is treated by Ackroyd [57].

The Boltzmann Equation Solved

The angular neutron density $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ in a medium V with inverse mean free path $\alpha(\underset{\sim}{r})$ and a yield of $c(\underset{\sim}{r})$ secondary neutrons per primary neutron removed is given for a distributed source s' by the Boltzmann equation:

$$\underset{\sim}{\Omega}.\ \text{grad}\, n(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha(\underset{\sim}{r}) n(\underset{\sim}{r}, \underset{\sim}{\Omega}) =$$
$$\alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} n(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' + s'(\underset{\sim}{r}, \underset{\sim}{\Omega})/v \tag{151}$$

for neutrons of speed v. Whatever the scattering law,

$$\int_{\Omega'} f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega = 1 \tag{152}$$

and

$$f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) = f(\underset{\sim}{r}, \underset{\sim}{\Omega} \to \underset{\sim}{\Omega}') \tag{153}$$

The scattering kernel is taken to be dependent on position $\underset{\sim}{r}$, for the scattering law is assumed to vary with position in the medium.

The boundary condition for a source of strength $t(\underset{\sim}{r}, \underset{\sim}{\Omega}) v\, \underset{\sim}{\Omega}.\underset{\sim}{\nu}'$ neutrons per unit area, per unit solid angle, per unit time on the surface S of V is:

$$n(\underset{\sim}{r}, \underset{\sim}{\Omega}) = t(\underset{\sim}{r}, \underset{\sim}{\Omega}) \geqslant 0 \tag{154}$$

for all directions $\underset{\sim}{\Omega}$ inward to S at $\underset{\sim}{r}$, i.e. in the same sense as the unit vector $\underset{\sim}{\nu}'$, the inward normal to S at $\underset{\sim}{r}$. Since $s'(\underset{\sim}{r}, \underset{\sim}{\Omega})$ and $t(\underset{\sim}{r}, \underset{\sim}{\Omega})$ are non-negative, $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ is non-negative on physical grounds.
The medium is said to be non-multiplying if:

$$c(\underset{\sim}{r}) \leqslant 1/4\pi \max_{\mathrm{V}} f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) \tag{155}$$

The first method given assumes that (155) holds.

In solving the Boltzmann equation (151) it is convenient to rewrite as:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\, n(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r}) n(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r}) n(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &- \alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} n(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' = s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned}\right\} \tag{156}$$

where:

$$\left.\begin{aligned} \alpha_0(\underset{\sim}{r}) &= \alpha(\underset{\sim}{r}) \{ 1 - 4\pi \hat{f}\, c(\underset{\sim}{r}) \{ \\ \alpha_1(\underset{\sim}{r}) &= 4\pi\, \alpha(\underset{\sim}{r}) c(\underset{\sim}{r})\, \hat{f} \\ \hat{f} &= \max_{\mathrm{V}} f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) \end{aligned}\right\}$$

We then define an approximate solution of (156) and show how it can be obtained from relaxed forms of the Boltzmann equation (156).

Definition of an Approximate Solution

The functions $m_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$, $m_2(\underset{\sim}{r}, \underset{\sim}{\Omega})$ and $m_3(\underset{\sim}{r}, \underset{\sim}{\Omega})$ can be said to approximate to the solution $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ of the Boltzmann equation if ρ^2, the ratio of:

$$\left\{\begin{aligned}&\int_\Omega\int_S |n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_1(\underset{\sim}{r},\underset{\sim}{\Omega})|^2\,|\underset{\sim}{\Omega}.\underset{\sim}{\nu}|\,dS d\Omega\\ &+\int_\Omega\int_V \alpha_0(\underset{\sim}{r})\,(|n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_1(\underset{\sim}{r},\underset{\sim}{\Omega})|^2 + |n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_2(\underset{\sim}{r},\underset{\sim}{\Omega})|^2)\,dV d\Omega\\ &+\int_\Omega\int_V \left\{\begin{aligned}&\alpha_1(\underset{\sim}{r})(|n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_1(\underset{\sim}{r},\underset{\sim}{\Omega})|^2 + |n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_3(\underset{\sim}{r},\underset{\sim}{\Omega})|^2)\\ &-\alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'}\left[\begin{aligned}&-|n(\underset{\sim}{r},\underset{\sim}{\Omega})-m_1(\underset{\sim}{r},\underset{\sim}{\Omega})|\,|n(\underset{\sim}{r},\underset{\sim}{\Omega}')-m_1(r,\Omega')|\\ &+|n(\underset{\sim}{r},\underset{\sim}{\Omega})-m_3(\underset{\sim}{r},\underset{\sim}{\Omega})|\,|n(\underset{\sim}{r},\underset{\sim}{\Omega}')-m_3(r,\Omega')|\end{aligned}\right] f(r,\Omega'\to\Omega)d\Omega'\end{aligned}\right\}\times dVa\end{aligned}\right.$$

to:

$$\left.\begin{aligned}&\int_\Omega\int_S n(\underset{\sim}{r},\underset{\sim}{\Omega})^2\,|\underset{\sim}{\Omega}.\underset{\sim}{\nu}|\,dS d\Omega + 2\int_\Omega\int_V \alpha_0(\underset{\sim}{r})n(\underset{\sim}{r},\underset{\sim}{\Omega})^2\,dV d\Omega\\ &+2\int_\Omega\int_V\left(\alpha_1(\underset{\sim}{r})n(\underset{\sim}{r},\underset{\sim}{\Omega})^2-\alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'} n(\underset{\sim}{r},\underset{\sim}{\Omega})n(\underset{\sim}{r},\underset{\sim}{\Omega}')f(\underset{\sim}{r},\underset{\sim}{\Omega}'\to\underset{\sim}{\Omega})d\Omega'\right)dV d\Omega\end{aligned}\right\}\quad(157)$$

(where ν is the outward normal to S, the surface bounding volume V) can be made smaller than any given positive number ξ. The functions $m_1(\underset{\sim}{r},\underset{\sim}{\Omega})$, $m_2(\underset{\sim}{r},\underset{\sim}{\Omega})$ and $m_3(\underset{\sim}{r},\underset{\sim}{\Omega})$ are assumed to be continuous with respect to $\underset{\sim}{r}$ and piecewise continuous with respect to $\underset{\sim}{\Omega}$ in V, and satisfy the transport equation:

$$\left.\begin{aligned}&\underset{\sim}{\Omega}.\ \mathrm{grad}\, m_1(\underset{\sim}{r},\underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})m_2(\underset{\sim}{r},\underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})m_3(\underset{\sim}{r},\underset{\sim}{\Omega})\\ &-\alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'} m_3(\underset{\sim}{r},\underset{\sim}{\Omega}')f(\underset{\sim}{r},\underset{\sim}{\Omega}'\to\underset{\sim}{\Omega})d\Omega' = s_0(\underset{\sim}{r},\underset{\sim}{\Omega})\end{aligned}\right\}\quad(158)$$

On S the boundary condition:

$$m_1(\underset{\sim}{r},\underset{\sim}{\Omega}) = t(\underset{\sim}{r},\underset{\sim}{\Omega})\quad(159)$$

may be assumed to hold for all directions inward to the surface, but this assumption is not essential. The ratio ρ^2 is non-negative.

In the sequel, a method is given for determining functions $m_1(\underset{\sim}{r},\underset{\sim}{\Omega})$, $m_2(\underset{\sim}{r},\underset{\sim}{\Omega})$ and $m_3(\underset{\sim}{r},\underset{\sim}{\Omega})$ so that ρ^2 can be made less than any assigned ξ, however small. The approximation procedure can be regarded as making $n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_1(\underset{\sim}{r},\underset{\sim}{\Omega})$, $n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_2(\underset{\sim}{r},\underset{\sim}{\Omega})$ and $n(\underset{\sim}{r},\underset{\sim}{\Omega}) - m_3(\underset{\sim}{r},\underset{\sim}{\Omega})$ arbitrarily small over space and all directions in a least squares sense.

The approximate solution defined so far is approximate in the global sense, since it is obtained by making sufficiently small an integral taken over the whole medium and all directions of a positive definite integrand. Although large, localized discrepancies between the true solution $n(\underset{\sim}{r},\underset{\sim}{\Omega})$ and the approximate solution (represented by $m_1(\underset{\sim}{r},\underset{\sim}{\Omega})$, $m_2(\underset{\sim}{r},\underset{\sim}{\Omega})$ and $m_3(\underset{\sim}{r},\underset{\sim}{\Omega})$) cannot offset each other, they may, if sufficiently well localized, have only a small effect on the ratio ρ^2. Bounds for the true solution averaged over a small locality are therefore obtained. The average neutron density $\bar{n}_i$ over an arbitrarily small region V_i,

$$\left.\begin{aligned}&\bar{n}_i = \frac{1}{V_i}\int_{V_i}\int_{\Omega'} n(\underset{\sim}{r},\underset{\sim}{\Omega}')\,d\Omega'\,dV\\ &\text{is bounded by: } \check{\bar{n}}_i \leqslant \bar{n}_i \leqslant \hat{\bar{n}}_i\end{aligned}\right\}\quad(160)$$

and these bounds can be drawn arbitrarily close.

The angular neutron density averaged over the arbitrarily small solid angle ω about the direction $\underset{\sim}{\Omega}$ for all points on an arbitrarily small plate Σ can also be bounded. The direction $\underset{\sim}{\Omega}$ is taken to be normal to the flat plate Σ, (Figure 4). The bounds:

$$\left.\begin{aligned} &\check{n}_\Sigma(\underset{\sim}{\Omega}, \omega) \leqslant n_\Sigma(\underset{\sim}{\Omega}, \omega) \leqslant \hat{n}_\Sigma(\underset{\sim}{\Omega}, \omega) \\ &\text{for the average angular density:} \\ &n_\Sigma(\underset{\sim}{\Omega}, \omega) = \frac{1}{\omega\Sigma} \int_\Sigma \int_\omega n(\underset{\sim}{r}', \underset{\sim}{\Omega}')\, d\Omega'\, dS \end{aligned}\right\} \tag{161}$$

can be made arbitrarily close. The point $\underset{\sim}{r}'$ lies in Σ, and the direction $\underset{\sim}{\Omega}'$ lies in the cone of solid angle ω and axis $\underset{\sim}{\Omega}$.

Relaxed Boltzmann Equations

In solving the Boltzmann equation (156) approximately, in the sense defined above, two relaxed Boltzmann equations are considered, for which exact solutions can be constructed. A linear combination of exact solutions of these relaxed equations is then used to obtain an approximate solution of the Boltzmann equation, which can be made as close as is required to the exact solution.

The first relaxed Boltzmann equation is of the form:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\, u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r}) u_2(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1\, u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &= \alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' + s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned}\right\} \tag{162}$$

and the associated boundary condition is:

$$u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = t(\underset{\sim}{r}, \underset{\sim}{\Omega}) \geqslant 0 \text{ on S} \tag{163}$$

This equation is easily solved by choosing any suitable function $u_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$, which satisfies the boundary condition (163), and then using it in (162) to give $u_2(\underset{\sim}{r}, \underset{\sim}{\Omega})$.

The second relaxed Boltzmann equation is of the form:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\, v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r}) v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r}) v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &= \alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' + s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned}\right\} \tag{164}$$

but there is no boundary condition necessarily associated. This equation can be solved by choosing any continuous function $v_3(\underset{\sim}{r}, \underset{\sim}{\Omega})$ and solving for $v_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$. The general solution is:

$$v_1(\underset{\sim}{r}_0 + \lambda\underset{\sim}{\Omega}, \underset{\sim}{\Omega}) = e^{-\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \lambda)} \int \mathrm{F}(\underset{\sim}{r}_0 + \lambda\underset{\sim}{\Omega}, \underset{\sim}{\Omega}) e^{\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \lambda)} d\lambda \tag{165}$$

where

$$\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \lambda) = \int_0^\lambda \alpha_0(\underset{\sim}{r}_0 + \mu\underset{\sim}{\Omega}) d\mu \tag{166}$$

is the optical distance between $\underset{\sim}{r}$ and $\underset{\sim}{r}_0$ for a medium with inverse total mean free path $\alpha_0(\underset{\sim}{r})$, and

$$\mathrm{F}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = \alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' + s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) - \alpha_1(\underset{\sim}{r}) v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}). \tag{167}$$

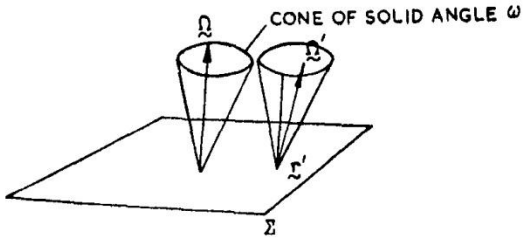

Figure 4. *Proof plane and cones.*

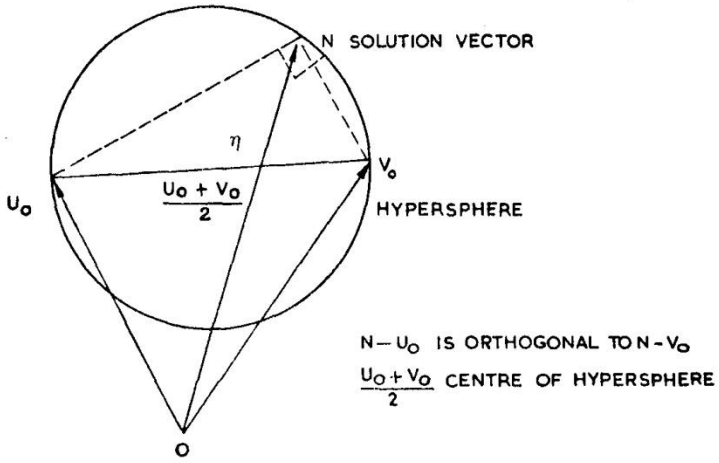

Figure 5. *The hypersphere.*

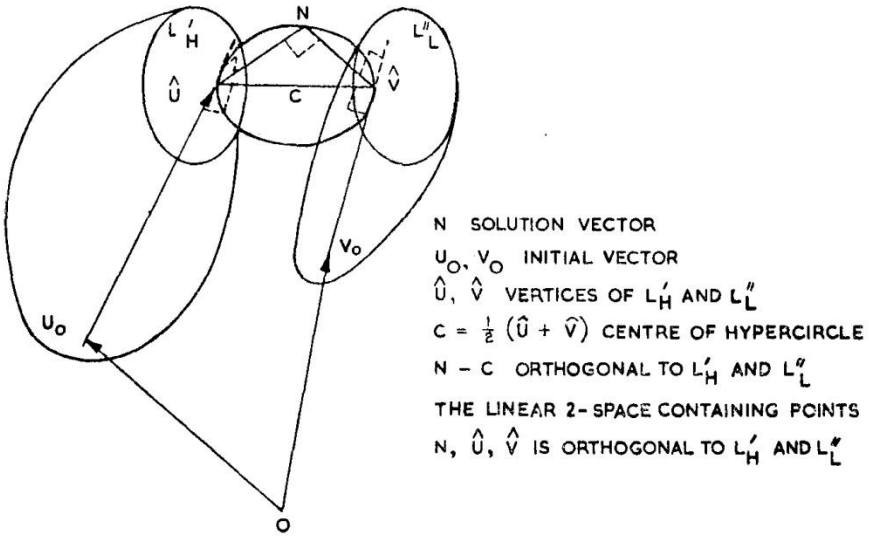

Figure 6. *The hypercircle.*

Although there is no need for v_1 to satisfy a boundary condition, the particular solution:

$$v_1(\underset{\sim}{r}_0 + \lambda\underset{\sim}{\Omega}, \underset{\sim}{\Omega}) = e^{-\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \lambda)} \left\{ \int_0^{\lambda} \mathrm{F}(\underset{\sim}{r}_0 + \mu\underset{\sim}{\Omega}, \underset{\sim}{\Omega}) e^{\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \mu)}\, d\mu + t(\underset{\sim}{r}_0, \underset{\sim}{\Omega}) \right\} \tag{168}$$

satisfies the boundary condition:

$$v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = t(\underset{\sim}{r}, \underset{\sim}{\Omega}) \quad \text{on} \quad \mathrm{S} \tag{169}$$

for the direction $\underset{\sim}{\Omega}$ inwards to S, since $\underset{\sim}{r}_0$ is an arbitrary vector from 0 to a point on S.

A Function Space

Scalar product of vectors. Any vector $\underset{\sim}{U} \longleftrightarrow (u_1, u_2, u_3)$ in the function space has three components, u_1, u_2, and u_3, which are real, continuous functions of $\underset{\sim}{r}$, and $\underset{\sim}{\Omega}$, the independent variables in the transport equation. The component u_1 is also assumed to have continuous derivatives with respect to position $\underset{\sim}{r}$. The scalar product of any two vectors $\underset{\sim}{U} \longleftrightarrow (u_1, u_2, u_3)$ and $\underset{\sim}{V}(v_1, v_2\, v_3)$ of the function space is defined as:

$$\underset{\sim}{U}.\underset{\sim}{V} = \left\{ \begin{aligned} &\int_{\Omega}\int_{\mathrm{S}} u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) |\underset{\sim}{\Omega}.\underset{\sim}{\nu}|\, d\mathrm{S}d\Omega \\ &+ \int_{\Omega}\int_{\mathrm{V}} \alpha_0(\underset{\sim}{r}) \{u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + u_2(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_2(\underset{\sim}{r}, \underset{\sim}{\Omega})\}\, d\mathrm{V}d\Omega \\ &+ \int_{\Omega}\int_{\mathrm{V}} \left\{ \begin{aligned} &\alpha_1(\underset{\sim}{r}) \{u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + u_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_3(\underset{\sim}{r}, \underset{\sim}{\Omega})\} \\ &- \frac{\alpha(\underset{\sim}{r}) c(\underset{\sim}{r})}{2} \int_{\Omega'} \left(\begin{aligned} &u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}') \\ &u_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}') \end{aligned} \right. \\ &\qquad \left. \begin{aligned} &+ u_1(\underset{\sim}{r}, \underset{\sim}{\Omega}') v_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &+ u_3(\underset{\sim}{r}, \underset{\sim}{\Omega}') v_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned} \right) f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' \end{aligned} \right\} d\mathrm{V}d\Omega \end{aligned} \right. \tag{170}$$

where $\underset{\sim}{\nu}$ is the outward drawn normal to S, the surface of the region over which the vectors are defined, the element of volume of this region being dV. The metric associated with this function space is positive definite.

Two Fundamental Classes of Vectors

Two classes of vectors are introduced which have the important property that every vector of one class is orthogonal to every vector of the other class.

Vectors of Class A. The components of the class A vector $\underset{\sim}{U}' \longleftrightarrow (u'_1, u'_2, u'_3)$ satisfy:

$$\left. \begin{aligned} &\underset{\sim}{\Omega}.\ \mathrm{grad}\, u'_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r}) u'_2(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r}) u'_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &\qquad - \alpha(\underset{\sim}{r}) c(\underset{\sim}{r}) \int_{\Omega'} u'_3(\underset{\sim}{r}, \underset{\sim}{\Omega}') f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' = 0 \\ &u'_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = u'_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) \quad \text{in} \quad \mathrm{V} \end{aligned} \right\} \tag{171}$$

and the boundary condition on S for inward directions $\underset{\sim}{\Omega}$

$$u'_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = 0 \tag{172}$$

Vectors of Class B. The components of the class B vector $\underset{\sim}{V}'' \leftrightarrow (v_1'', v_2'', v_3'')$ satisfy:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\ v''_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})v''_2(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})v''_3(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &\qquad - \alpha(\underset{\sim}{r})c(\underset{\sim}{r}) \int_{\Omega'} v''_3(\underset{\sim}{r}, \underset{\sim}{\Omega}')f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega})d\Omega' = 0 \\ &v''_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = v''_2(\underset{\sim}{r}, \underset{\sim}{\Omega}) \quad \text{in} \quad V \end{aligned}\right\} \quad (173)$$

The components are not, however, subject to any boundary condition.

In the sequel, class A and B vectors are distinguished by the use of primed and double-primed letters respectively. A class A vector is easily constructed: choose $u'_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$ in V satisfying the boundary condition (172) and then use (171) to calculate $u'_3(\underset{\sim}{r}, \underset{\sim}{\Omega})$ and $u'_2(\underset{\sim}{r}, \underset{\sim}{\Omega})$ in this order. A class B vector V″ is not difficult to construct: choose $v''_3(\underset{\sim}{r}, \underset{\sim}{\Omega})$ over V, then $v''_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$ is given by:

$$v''_1(\underset{\sim}{r}, \underset{\sim}{\Omega}) = e^{-\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, |\underset{\sim}{r}-\underset{\sim}{r}_0|)} \left\{ \int_0^{|\underset{\sim}{r}-\underset{\sim}{r}_0|} F''(\underset{\sim}{r}_0 + \mu\underset{\sim}{\Omega}, \underset{\sim}{\Omega})e^{\sigma_0(\underset{\sim}{r}_0, \underset{\sim}{\Omega}, \mu)}\, d\mu + v''_1(\underset{\sim}{r}_0, \underset{\sim}{\Omega}) \right\} \quad (174)$$

Solution by the Method of the Hypercircle

The solution of the Boltzmann equation which is obtained in this Section by an application of Synge's method of the hypercircle [63] is an approximate one in the sense of (157). Let

$$N \leftrightarrow \{n(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ n(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ n(\underset{\sim}{r}, \underset{\sim}{\Omega})\}$$

denote the vector in the function space corresponding to the exact solution $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ of the Boltzmann equation, and let

$$M \leftrightarrow \{m_1(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ m_2(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ m_3(\underset{\sim}{r}, \underset{\sim}{\Omega})\}$$

be the vector corresponding to the approximate solution which is defined by $m_1(\underset{\sim}{r}, \underset{\sim}{\Omega})$, $m_2(\underset{\sim}{r}, \underset{\sim}{\Omega})$ and $m_3(\underset{\sim}{r}, \underset{\sim}{\Omega})$, then (157) can be written:

$$\rho^2 = (N - M)^2/N^2 \quad (175)$$

Since the function space is positive definite, it is found that M can be made to approach N as closely as one pleases by making $(N - M)^2$ arbitrarily small.

In applying the method of the hypercircle, two initial vectors U_0 and V_0 are used together with sets of A and B vectors. The components of:

$$U_0 \leftrightarrow \{u_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ u_{02}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ u_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega})\}$$

are chosen to satisfy:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\ u_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})u_{02}(r., \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})u_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &= \alpha(\underset{\sim}{r})c(\underset{\sim}{r}) \int_{\Omega'} u_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega}')f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega})d\underset{\sim}{\Omega}' + s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &u_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = u_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned}\right\} \quad (176)$$

and the boundary condition:

$$u_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = t(\underset{\sim}{r}, \underset{\sim}{\Omega}) \geqslant \text{on S for inward directions } \underset{\sim}{\Omega} \quad (177)$$

The vector U_0 is easily constructed by choosing $u_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega})$ to satisfy the boundary condition (177), and then using (176) to determine $u_{02}(\underset{\sim}{r}, \underset{\sim}{\Omega})$. The components of:

$$V_0 \leftrightarrow \{v_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ v_{02}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ v_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega})\}$$

are given by:

$$\left.\begin{aligned} &\underset{\sim}{\Omega}.\ \text{grad}\ v_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})v_{02}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})v_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &= \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'} v_{03}(\underset{\sim}{r}, \underset{\sim}{\Omega}')f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega})d\Omega' + s_0(\underset{\sim}{r}, \underset{\sim}{\Omega}) \\ &v_{01}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = v_{02}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \end{aligned}\right\} \quad (178)$$

but are not subject to any boundary condition. Since $N - U_0$ is an A vector, and $N - V_0$ is a B vector,

$$(N - U_0),\ (N - V_0) = 0 \quad (179)$$

which shows that the unknown point N (Figure 5), lies on a hypersphere with the points U_0 and V_0 as ends of a diameter, and centre $C = (U_0 + V_0)/2$.

The above procedure for obtaining an approximate solution for $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ by locating the vector N on a hypersphere of known centre and radius can be improved by using the fact that N lies on a hypercircle of arbitrarily small radius. An approximate solution for N can be obtained by finding the vertices of two linear subspaces, one lying in the subspace of A vectors and the other in the subspace of B vectors. The mid-point of the line joining these vertices is then taken to be an approximation for the point N. The point N lies on a hypercircle with the mid-point of the vertices as its centre. Since the diameter of the hypercircle is the distance between the vertices, and these can be brought arbitrarily close together, the unknown point N can be approached arbitrarily close by a known point, the centre of the hypercircle C.

The sets of A and B vectors required for the method of the hypercircle are defined as follows.

A typical vector of the set of A vectors is denoted by:

$U'_h \longleftrightarrow \{u'_{1h}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ u'_{2h}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ u'_{3h}(\underset{\sim}{r}, \underset{\sim}{\Omega})\}$. The components of U'_h satisfy

$$\underset{\sim}{\Omega}.\ \text{grad}\ u'_{1h}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})u'_{2h}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})u'_{3h}(\underset{\sim}{r}, \underset{\sim}{\Omega})$$

$$- \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'} u'_{3h}(\underset{\sim}{r}, \underset{\sim}{\Omega}')_3(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega})d\Omega' = 0 \quad (180)$$

and

$$u'_{1h}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = u'_{3h}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \text{ in V}$$

and the boundary condition on S for inward directions $\underset{\sim}{\Omega}$:

$$u'_{1h}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = 0 \quad (181)$$

A typical vector of the set of B vectors is denoted by:

$$V''_l \longleftrightarrow \{v''_{1l}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ v''_{2l}(\underset{\sim}{r}, \underset{\sim}{\Omega}),\ v''_{3l}(\underset{\sim}{r}, \underset{\sim}{\Omega})\}.$$

The components of V''_l satisfy:

$$\underset{\sim}{\Omega}.\ \text{grad}\ v''_{1l}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_0(\underset{\sim}{r})v''_{2l}(\underset{\sim}{r}, \underset{\sim}{\Omega}) + \alpha_1(\underset{\sim}{r})v''_{3l}(\underset{\sim}{r}, \underset{\sim}{\Omega})$$

$$- \alpha(\underset{\sim}{r})c(\underset{\sim}{r})\int_{\Omega'} v_3''_l(\underset{\sim}{r}, \underset{\sim}{\Omega}')f(\underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega})d\Omega' = 0 \quad (182)$$

and

$$v''_{1l}(\underset{\sim}{r}, \underset{\sim}{\Omega}) = v''_{2l}(\underset{\sim}{r}, \underset{\sim}{\Omega}) \text{ in V}$$

but the components do not have to satisfy any boundary condition.

Consider the linear subspaces:

$$L'_H,\ U = U_0 + a'_h U'_h\ (h = 1, 2, \ldots H) \quad (183)$$

and:

$$L''_L,\ V = V_0 + a''_l V''_l\ (l = 1, 2, \ldots L) \quad (184)$$

where the U'_h and V''_l are A and B vectors respectively. The initial vectors

U_0 and V_0 are defined by (176), (177) and (178). The vertices or closest points of approach of these subspaces are given by:

$$\hat{U} = U_0 + a'_h U'_h \tag{185}$$

$$\hat{V} = V_0 + a''_l V''_l \tag{186}$$

for the a'_h and a''_l, having the particular values given by:

$$a'_h U'_h . U'_k = (V_0 - U_0) . U'_k \quad (h, k = 1, \ldots H) \tag{187}$$

$$a''_l V''_l . V''_j = (U_0 - V_0) . V''_j \quad (l, j = 1, \ldots L) \tag{188}$$

where summation with respect to a repeated suffix is understood. If the U'_h and V''_l are each linearly independent sets of vectors, these equations can always be solved for a'_h, a''_l since the Grammian determinants $|U'_h . U'_k|$ and $|V''_l . V''_j|$ do not vanish.

Since the components of N satisfy (156), and the components of U_0 and U'_h satisfy (176), (177) and (180), (181) respectively, the vector $N - \hat{U}$ is an A vector. Similarly, the vector $N - \hat{V}$ is a B vector. Thus, the orthogonality of A and B vectors gives:

$$(N - \hat{U}) . (N - \hat{V}) = 0 \tag{189}$$

i.e. N lies on the hypersphere $(N - C)^2 = R^2$

$$C = \frac{1}{2}(\hat{U} + \hat{V}), \qquad R^2 = \frac{1}{4}(\hat{U} - \hat{V})^2 \tag{190}$$

The orthogonality of A vector $N - \hat{U}$ and the B vectors V''_l gives:

$$(N - \hat{U}) . V''_l = 0, \qquad (l = 1, 2, \ldots L) \tag{191}$$

Similarly,

$$(N - \hat{V}) . U'_h = 0, \qquad (h = 1, 2, \ldots H) \tag{192}$$

Thus the point N lies on the hypercircle (Figure 6) which is the intersection of the hypersphere (190) and the hyperplane of class H + L defined by (191) and (192). The centre of the hypercircle is given by:

$$C = (\hat{U} + \hat{V})/2 \tag{193}$$

and its radius:

$$R = \frac{1}{2}\sqrt{(\hat{U} - \hat{V})^2}$$

using (185) to (186) inclusive, or:

$$\left.\begin{aligned} R &= +\frac{1}{2}\sqrt{(U_0 - V_0)^2 - a'_h a'_k \, U'_h . U'_k - a''_l a''_j V''_l . V''_j} \\ &= \frac{1}{2} + \sqrt{(\hat{U} - \hat{V})^2} \end{aligned}\right\} \tag{194}$$

If both sets of vectors U'_h, $(h = 1, \ldots H)$ and V''_l, $(l = 1, 2, \ldots L)$ are chosen to be linearly independent sets, then increases in H and L decrease R. If a complete set of vectors U'_h, and a complete set of vectors V''_j are chosen, then R vanishes.

Since the point N lies on the hypercircle:

$$(N - C)^2 = R^2 \tag{195}$$

i.e. C is an approximation to N in the global sense. The global error R^2/N^2 can be made arbitrarily small by a sufficiently large number of vectors in the independent sets U'_h and V''_l. Furthermore, the parametric equations of the hypercircle are:

$$\left.\begin{aligned} X &= C + RJ \\ J^2 &= 1 \\ J.U'_h &= 0 \qquad (h = 1, 2, \ldots H) \\ J.V''_l &= 0 \qquad (l = 1, 2, \ldots L) \end{aligned}\right\} \tag{196}$$

from (185), (186), (187), (192).

Since N ranges on the hypercircle (196),

$$-2R|C_H| \leqslant N^2 - C^2 - R^2 \leqslant 2R|C_H| \tag{197}$$

where C_H is the component of C lying on the hyperplane defined by the last two members of (196), the magnitude of C_H defined by:

$$C - c'_h U'_h - c''_l V''_l$$

is given by:

$$\left.\begin{aligned} C^2_H &= C^2 - C(c'_h U'_h + c''_l V''_l) \\ &= C^2 - (c_h' U'_h)^2 - (c''_l V''_l)^2 \\ \text{where:} \qquad c'_h U'_h . U'_k &= C.U'_k \\ c''_l V''_l . V''_j &= C.V''_j \end{aligned}\right\} \tag{198}$$

Thus, (195) and (197) give:

$$\frac{R^2}{C^2 + R^2 + 2R|C_H|} \leqslant \frac{(N - C)^2}{N^2} \leqslant \frac{R^2}{C^2 + R^2 - 2R|C_H|} \tag{199}$$

Since the radius of the hypercircle, R, can be made arbitrarily small, $\frac{(N - C)^2}{N^2}$ can be made arbitrarily small.

Bounds for the Neutron Density Averaged over a Locality

The solution that has been obtained so far for N is approximate in the global sense. Bounds for the neutron density averaged over an arbitrary locality can be obtained through the use of a Green's vector. (This vector is analogous in some ways to a Green's function, but there is an important distinction. The components of a Green's vector are everywhere finite, and thus it is possible to evaluate numerically the scalar product of a Green's vector, and any other vector with finite components.)

Bounds of the following type have been obtained for:

$$\bar{\bar{n}}_i = \int_{\Omega} \int_{V_i} \alpha(\underset{\sim}{r}) |1 - c(\underset{\sim}{r})| n(\underset{\sim}{r}, \underset{\sim}{\Omega}) dV d\Omega \Big/ \int_{V_i} \alpha(\underset{\sim}{r}) |1 - c(\underset{\sim}{r})| dV$$

a kind of average for the neutron density over any (small) volume V_i

$$0 \leqslant (k^+ - C.G^+ - R|G_0^+|)\beta_i \leqslant \bar{\bar{n}}_i \leqslant (k^+ + R|G_0^+| - C.G^+)\beta_i \tag{200}$$

where k^+ is expressed in terms of a Green's vector G^+, the surface and volume sources for the reactor; C is the centre of the hypercircle of radius R; and

G_0^+ is the component of G^+ lying in the hyperplane of class H + L which is used in defining the hypercircle. The coefficient β_i is given by:

$$\beta_i = V_i \Big/ \int_{V_i} \alpha(\underset{\sim}{r}) |1 - c(\underset{\sim}{r})| dV \tag{201}$$

Since the span $2R|G_0^+|$ of the inequality for $\bar{n}_i$ can be made arbitrarily small by making the radius R of the hypercircle as small as one pleases, $\bar{\bar{n}}_i$ can be determined to any required accuracy. Since the volume V_i can be taken as small as one pleases, the inequality (200) gives an index of the pointwise accuracy of the hypercircle solution.

Bounds for the Angular Neutron Density Averaged over a Locality

The following bounds can be obtained with the aid of a suitable Green's vector G^x, once the solution vector N has been located on the hypercircle (195) of radius R and centre C:

$$-k^x + C.G^x - R|G_0^x| \leqslant \frac{1}{\omega\Sigma} \int_{\omega} \int_{\Sigma} n(\underset{\sim}{r}, \underset{\sim}{\Omega}) \underset{\sim}{\Omega}.\underset{\sim}{\nu} d\mathrm{S} d\Omega \leqslant -k^x + C.G^x + R|G_0^x| \tag{202}$$

for the $n(\underset{\sim}{r}, \underset{\sim}{\Omega})$ averaged with weight $\underset{\sim}{\Omega}.\underset{\sim}{\nu}$ over the plate Σ (Figure 4) for cones of solid angle ω. Since the span of the inequality (202) is $2R|G_0^x|$ (where G_0^x is the component of G^x lying in the hyperplane defining the hypercircle (195)), it can be made arbitrarily small by making the radius of the hypercircle as small as one pleases; and

$$\frac{1}{\omega\Sigma} \int_{\omega} \int_{\Sigma} n(\underset{\sim}{r}, \underset{\sim}{\Omega}) \underset{\sim}{\Omega}.\underset{\sim}{\nu} d\mathrm{S} d\Omega$$

can be approximated to any requisite degree of accuracy. The solid angle ω and the area Σ in Figure 4 can be chosen arbitrarily small.

The Multi-group Boltzmann Equations

The multi-group Boltzmann equations are written with the aid of the summation conversion as:

$$\left.\begin{aligned} \underset{\sim}{\Omega}.\,\mathrm{grad}\,\psi_m(i, \underset{\sim}{r}, \underset{\sim}{\Omega}) + \{a_{mn}(i, \underset{\sim}{r}) + b_{mn}(i, \underset{\sim}{r}) + c_{mn}(i, \underset{\sim}{r})\}\,\psi_n(i, \underset{\sim}{r}, \underset{\sim}{\Omega}) \\ = K_c(i) \int_{\Omega'} a_{nl}(i, \underset{\sim}{r}) \psi_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') t_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' \\ + \int_{\Omega'} b_{nl}(i, \underset{\sim}{r}) \psi_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') s_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' \end{aligned}\right\} \tag{203}$$

for $\psi_m(i, \underset{\sim}{r}, \underset{\sim}{\Omega})$ the angular neutron density for direction $\underset{\sim}{\Omega}$ and position $\underset{\sim}{r}$, for neutrons in the energy group m and region i of the reactor. The first term of:

$$\{a_{mn}(i, \underset{\sim}{r}) + b_{mn}(i, \underset{\sim}{r}) + c_{mn}(i, \underset{\sim}{r})\}\,\psi_n(i, \underset{\sim}{r}, \underset{\sim}{\Omega})$$

relates to productive capture, the second to removal by scattering, and the last term to unproductive capture of neutrons. The first and second integrals represent the sources of secondary neutrons created by fission and scattering respectively. The matrices $a_{mn}(i)$, $b_{mn}(i)$ and $c_{mn}(i)$ are diagonal with non-negative elements.

To solve (203), the following equations are solved by the method of the hypercircle. The angular neutron density $\Phi_m(i, \underset{\sim}{r}, \underset{\sim}{\Omega})$ due to the mimic fission sources:

$$\mathrm{K}(i) \int_{-\Omega'} a_{nl}(i, \underset{\sim}{r}) f_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') t_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega'$$

and the mimic scattering sources:

$$\int_{\Omega'} b_{nl}(i, \underset{\sim}{r}) f_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') s_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega',$$

satisfies

$$\begin{aligned} \underset{\sim}{\Omega} \cdot \operatorname{grad} \Phi_m(i, \underset{\sim}{r}, \underset{\sim}{\Omega}) + \{a_{mn}(i, \underset{\sim}{r}) + b_{mn}(i, \underset{\sim}{r}) + c_{mn}(i, \underset{\sim}{r})\} \Phi_n(i, \underset{\sim}{r}, \underset{\sim}{\Omega}) \\ = \mathrm{K}(i) \int_{\Omega'} a_{nl}(i, \underset{\sim}{r}) f_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') t_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' \\ + \int_{\Omega'} b_{nl}(i, \underset{\sim}{r}) f_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega}') s_{mn}(i, \underset{\sim}{r}, \underset{\sim}{\Omega}' \to \underset{\sim}{\Omega}) d\Omega' \end{aligned} \qquad (204)$$

where the $f_l(i, \underset{\sim}{r}, \underset{\sim}{\Omega})$ can be arbitrary, except that they must satisfy the same homogeneous boundary conditions as the ψ_m and Φ_m. The equations (204) can be solved for the appropriate boundary conditions to any required degree of accuracy by introducing a certain function space with a positive definite metric. The equations (203) are then solved by matching the vector Φ, which represents the Φ_m in the function space, to the vector F representing the f_l in the function space. The match can be accomplished for only certain eigenvalues of $\mathrm{K}(i)$, one of which is $\mathrm{K}_c(i)$. In this way, the multi-group Boltzmann equations are solved.

Data Requirements

Basic Needs

In order to put the various methods described above to *practical* use, it is necessary to have reliable basic data. These may be in the form of group constants as required by most of the methods, or they may constitute a detailed set of basic neutron cross sections and scatter laws which can be used almost directly with the Monte Carlo method. The group constants must, of course, also be calculated in the first instance from the detailed basic cross section data, but one has a certain amount of choice in deciding whether to use other people's group constants or to calculate them oneself. (K. Parker [39], for example, gives a list of references to group constants which are generally available.) One also has the same kind of choice where detailed cross section data are concerned, but in this case the range from which to choose is much more limited. Detailed tabulations of scattering data — cross sections and scatter laws — for a number of elements and isotopes are given by Kalos and Troubetzky [65], Lustig [66], Buckingham, Parker and Pendlebury [67] also give detailed data for a number of elements and isotopes, mainly in graphical form. These compilations are complete in the sense that they provide all the basic data necessary for criticality calculations on certain types of system. The report BNL 325 [68] and Supplement is a most valuable compilation of total cross-sectional data and some partial cross sections, but it

only gives experimental values and also does not contain information on scattering laws. It is therefore incomplete in the sense described above, as also is the data given by R.J. Howerton [69] which aims to give complete sets of cross sections but does not contain any information on angular and energy distributions of secondary neutrons.

If it is decided to use other people's data, then one merely has to satisfy oneself that the data are sufficiently reliable for the use to which they are to be put and that in particular they give correct critical sizes to within a tolerable uncertainty for the types of system to be considered.

Consider now the problem of obtaining satisfactory neutron scattering data. This is a non-trivial problem of fundamental importance. The *first stage* is to do a literature search and to assemble all the available experimental data — cross sections and angular and energy distributions of secondary neutrons over the appropriate range of incident neutron energy, for the elements or isotopes of interest. The experimental data are usually incomplete and it is necessary to fill the various gaps in accordance with the predictions of current nuclear theory. (A literature survey of the data for beryllium, leading to a set of recommended data, is given, for example, by K. Parker [70]). The detailed information so obtained could in principle be used with the Monte Carlo method after only slight preliminary manipulation, though in practice a conversion to a standard form is usually preferred. To obtain group constants it is necessary to average the detailed data, using an appropriate spectrum weighting, over the chosen energy groups. These manipulations can all be carried out on desk calculating machines but they are very tedious and very time-consuming. The obvious way to proceed is to mechanize the process and to use the electronic computer which is to be used in the neutron transport calculation, to calculate the group constants and to do any preliminary manipulation for Monte Carlo use. The output — card or tape — can be made suitable for direct insertion as part of the machine input for the neutron transport calculation. In this way, the whole process of data handling is greatly speeded up.

At Aldermaston a system of this kind has been in use for several years and is described in detail in A.W.R.E. Reports 0-20/60 [71]* and 0-1/61[39]**. Briefly, all the basic scattering data — cross sections, angular and energy distributions for the various possible reactions — are written on to punched cards suitable for use with IBM 704, 709 and 7090 machines. Machine programmes convert this into a form which is more convenient for Monte Carlo use; it involves in the first instance the conversion of cross sections to histogram form***, and the data for the various elements and isotopes are written on to magnetic tape. If now one requires the data for any combination of the elements and isotopes, another programme takes the magnetic tape as part of its input and calculates the total cross section for the mixture and the probabilities of the various partial reactions for the different elements or isotopes per collision in the mixture, all as a function of incident neutron energy. The output from this is on punched cards suitable for direct insertion

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* Describes the basic system, giving full details of card formats, and the processing of the data for Monte Carlo use.

** Gives some account of the basic philosophy of group constant calculation and describes in detail the group constant programme CANDY.

*** On machines of smaller capacity than the IBM 704, 709 and 7090 it might be preferable to represent the cross section in terms of a finite series of suitable orthogonal polynomials.

with the Monte Carlo neutron transport programmes. The angular and energy distributions are also converted to forms which are of particular convenience for sampling purposes.

The group constant machine programme — CANDY — calculates group constants directly from the cards containing the detailed basic data, for any specified group boundaries and spectrum weighting within the groups. In particular, the spectrum weighting may be of the form $1/\sigma_{tot}$ as may be required in energy regions where resonances occur. From the use of CANDY one obtains punched cards containing the appropriate group constants (suitable for direct use with the Carlson S_n programme or with the anisotropic programme) and also a printed list of the group constants. The use of CANDY involves only a minute or two of IBM 7090 time, followed by off-line printing. Although the system used for this data handling at Aldermaston has been developed for particular use on the IBM 704, 709 and 7090 machines, the principles involved are of great generality and the system can be used with appropriate modification on any high-speed computer.

Having obtained what appears, from the nuclear physics aspect, to be the best set of neutron scattering data for elements and isotopes of interest, one must then examine it to find out to what extent it is satisfactory for practical use; for this purpose one must use a reliable method of calculation to obtain certain integral results for comparison with experimental values. In this connection, the comparison of calculated and experimental critical masses is of particular importance. When making such a comparison, even for a simple system like GODIVA (bare sphere of high enriched U 235), one is very likely to obtain a calculated critical mass which is significantly at variance with the measured value. Such discrepancies are due almost entirely to errors and uncertainties in the basic data, and the error on the calculated critical mass may be as great as 20 % (often the calculated value is less than the experimental value for fast and intermediate systems). It is worth noting that different groups working independently in different parts of the world (e.g. Yiftah, Okrent and Moldauer [73] in the U.S.A. and Parker and Pendlebury in the United Kingdom) come to the same general conclusion in this respect. Scattering data which give an underestimate to critical sizes may of course be adequate for safety considerations but one usually requires to know accurate critical masses. In this case it is necessary to make adjustments to the basic scattering data so that they give the correct critical masses, using *accurate* methods of calculation for a suitable range of systems. In carrying out such adjustments it is necessary to bear in mind the uncertainties in the basic data, and, if possible, to make all adjustments within experimental error of the original values. It is also necessary to ensure that the adjusted data are consistent with nuclear theory. If in making adjustments to the data, the basic nuclear physics is incorrectly represented, then, whilst the adjusted data may give the right critical sizes to some systems, they may give quite wrong answers to others, and this is where the real danger lies.

The difficulties mentioned above, and the adjustment of the basic data which is required, apply to all systems, including the relatively simple fast systems, i.e., systems in which the neutron spectrum is concentrated in the region above 1 keV. When dealing with the more complicated moderated systems one may experience difficulty in dealing with the resonances between about 1 and 1,000 eV in the fissile elements. The ideal way of dealing with these is to represent them in terms of the level parameters using a multi-level

scattering formula. (The mechanization of this is at present under consideration at Aldermaston.) In this way, apart from getting a better representation of the cross section, temperature effects (Doppler broadening) can readily be taken into account if this is necessary — as, for example, in finding the temperature coefficient of reactivity for a given reactor system. In calculating group constants over the resonance region it is necessary to pay particular attention to the spectrum weighting which is used. One must allow for the flux depression — the so-called self-shielding effect — at energies corresponding to a resonance in the cross section. This leads one to use a spectrum weighting $\propto 1/\sigma_{tot}$ over resonance regions in the way outlined by Roach [74].

The interaction of thermal neutrons (energy < 0.1 eV at 300° k) with matter is complicated by atomic motion and molecular and crystal effects. A certain amount of experimental work is in progress at the present time to obtain scattering data for low-energy neutrons within the thermal distribution, but much remains to be done. For some purposes it is adequate to treat all neutrons below a certain energy, say 0.4 eV, as belonging to a thermal group and to use suitable one-group constants for this. Such constants are obtained either by averaging the appropriate cross sections over a Maxwellian velocity distribution in the case of simple substances, or by using experimentally determined diffusion lengths obtained by measurements with thermal neutrons. This latter procedure is particularly desirable when hydrogenous substances are involved, because the effectiveness of hydrogen in compounds varies considerably with incident neutron energy at these low energies.

The reader is reminded of the remark made earlier in this Section, that the problem of obtaining reliable scattering data is not a trivial one.

Few-Group Data

For some purposes it may only be necessary to have few-group constants and these can always be obtained in principle by averaging the detailed basic data over a suitable spectrum; or they may, in some instances, be deduced from direct experimental measurements. As with multi-group constants they will have to be adjusted to give agreement with some experimental critical sizes, but clearly in this case one can only fit a very limited number of critical sizes. Thereafter, the constants should, in general, only be used to interpolate between the systems to which they have been fixed, since any extrapolation may give calculated critical sizes which are grossly in error. For well-moderated systems, two groups are usually sufficient to obtain accurate interpolations for critical masses. It is usually essential only to use one-group methods, even for purposes of interpolation between experimental values, for systems in which the effect of moderation is not important, though even in a moderated system, if the neutron spectrum does not vary much over the system, a one-group approximation can suffice for critical size determination.

A good method of estimating one-group cross sections for fast systems containing fissile and fertile material has been given recently by B. Segal [75]. The method is based upon multi-group analyses of twenty-one reflected fast cores of widely different size and composition, but with similar reflectors of heavy elements. It is found that the microscopic one-group parameters (denoted by bracketed letters) for the kth constituent in the core:

$$\langle\sigma_{tr}\rangle^k \ , \ \langle\sigma'_{tr}\rangle^k \ , \ \langle\sigma_f\rangle^k \ , \ \langle\nu\sigma_f\rangle^k \ , \ \langle\sigma_c\rangle^k \qquad (205)$$

fall on smooth curves if the reactors are arranged in a certain order according to the relative hardness or softness of their core spectra. Segal introduces two microscopic transport cross sections:

$$\langle\sigma_{tr}\rangle^k \quad \text{and} \quad \langle\sigma_{tr}\rangle^k$$

The first is used to calculate the macroscopic transport cross section Σ_{tr}, and the second is used to calculate the diffusion coefficient. The softness of the spectrum is defined as:

$$S = \frac{\Sigma_{tr}}{\nu\Sigma_f - \Sigma_c} \qquad (206)$$

where the macroscopic one-group cross sections are calculated in the usual way from the one-group microscopic cross sections.

The one-group cross sections Σ'_{tr}, Σ_{tr}, Σ_f and Σ_c for a proposed reactor of given size and of known composition, apart from the critical ratio R of fissile to fertile material, are obtained by an iterative procedure, which converges usually after the second iteration. A trial value S_1 for S is taken, which may be guessed from experience to facilitate convergence, and then the first estimate $\langle\sigma\rangle^k$ of the cross sections (205) are read off the curves for a softness S_1. A first approximation R_2 to the critical ratio can then be obtained from a one-group diffusion theory calculation. This calculation gives the first approximation to the core composition, and one can now form the macroscopic parameters Σ_{tr}, $\nu\Sigma_f$ and Σ_c to find a second approximation S_2 to S. The second cycle of the iteration can then be commenced.

The Treatment of Anisotropic Scattering

In many problems, anisotropic scattering is adequately taken account of by means of the transport approximation, and in this there are basically two forms of the approximation: diagonal transport correction and full transport correction. In both cases anisotropic scattering is taken into account by an appropriate modification of the cross sections, and with these modified values scattering is taken to be isotropic.

The diagonal and full transport correction. In one-group diffusion theory, it is easily shown that the anisotropy of scattering can be allowed for by using effective values $\alpha^\star$ and $\beta^\star$ of α and β in a transport equation of isotropic form. Here, α is the inverse mean free path at unit material density and β/α is the average number of neutrons per collision. The effective values $\alpha^\star$ and $\beta^\star$ are given by:

$$\left.\begin{aligned} \alpha^\star &= \alpha - \beta' \\ \beta^\star &= \beta^0 - \beta' \end{aligned}\right\} \qquad (207)$$

where:

$$\beta^0 = \int_{-1}^{+1} \beta(\bar{\mu})d\bar{\mu}$$

and:

$$\beta^1 = \int_{-1}^{+1} \bar{\mu}\beta(\bar{\mu})d\bar{\mu}$$

(The notation is that of the anisotropic S*n* method-see page 89).

Within the limits of the P_1 spherical harmonics approximation, the substitution is exact. Proceeding to a one-group transport equation, the substitution is no longer exact, but experience has shown it to be good in many cases. With a multi-group equation, however, the situation becomes more difficult. The above modifications generalize [76] to:

$$\left.\begin{aligned}\alpha^{\star}_{g'g} &= \delta_{g'g}\alpha_g - \beta^1_{g'g}\\ \beta^{\star}_{g'g} &= \beta^0_{g'g} - \beta^1_{g'g}\end{aligned}\right\} \tag{208}$$

where the term $\alpha^{\star}_{g'g}$ is used by substituting the expression:

$$\Sigma\alpha^{\star}_{g'g}\Phi_{g'} \quad \text{for} \quad \alpha_g\Phi_g$$

in the transport equation (cf. equation (113)).

These off-diagonal absorption terms may occasion some surprise but they are easily interpreted by noting that in general they are negative. They can thus be interpreted as a source due to neutrons changing group without changing direction. This is in line with ' transport correction ' which in general can be regarded as replacing an anisotropic scattering distribution with an isotropic component and a δ-function forward scattering component.

The modification (208) thus alters the form of the transport equation, but if the only anisotropic process is elastic scattering from heavy nuclei,

$$\beta'_{g'g} = \delta_{g'g}\beta^1_{gg} \tag{209}$$

and (208) reduce to:

$$\left.\begin{aligned}\alpha^{\star}_g &= \alpha_g - \beta^1_{gg}\\ \beta^{\star}_{g'g} &= \beta^0_{g'g} - \delta_{g'g}\beta^1_{gg}\end{aligned}\right\} \tag{210}$$

Approximation (210), which leaves the transport equation in normal form, is known as the diagonal transport correction since it only affects the diagonal terms in the scattering matrix.

The full transport correction uses equations (208) and the extra terms in the transport equation can easily be allowed for as long as only the slowing-down processes are anisotropic.

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REFERENCES

1. B. Davison, *Transport Theory of Neutrons*, Oxford University Press, London, 1956.
2. B. Davison, " Transport Theory of Neutrons ", *LT-18*, Chalk River (1947).
3. K. Fuchs, " Effect of a Scattering Container on the Critical Radius and Time Constant ", U.K. Report *MS,85.*
4. S. Frankel and G. Goldberg, " The Mathematical Development of the End-Point Method ", *AECD-2056 (LADC-76)* (1948).
5. K.M. Case, F. de Hoffman and G. Placzek, *Introduction to the Theory of Neutron Diffusion*, Vol. 1, Govt. Printing Office, Washington, D.C., 1953.

6. A.M. WEINBERG and E.P. WIGNER, *The Physical Theory of Neutron Chain Reactors*, University of Chicago Press, 1958.
7. S. GLASSTONE and M.C. EDLUND, *The Elements of Nuclear Reactor Theory*, Van Nostrand, New York, 1952.
8. M.E. MANDL and J. HOWLETT, " A Method for Calculating the Critical Mass of an Intermediate Reactor " in *Proc. Second U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 5*, 433.
9. H. HURWITZ and R. EHRLICH, " Comparison of Theory and Experiment for Intermediate Assemblies " in *Proc. Second U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 5*, 423.
10. M.E. MANDL, " The Spherical Harmonics Method in Plane and Spherically Symmetric Geometry in Multi-Velocity-Group Theory, and its Application in the Two-Velocity-Group P_3 Approximation ", *AERE T/R 1295.*
11. M.E. MANDL, " Multi-Group Theory with an Application to Inelastic Scattering in Uranium ", *AERE T/R 1500.*
12. D.J. THORNTON, Unpublished.
13. J.B. SAMPSON and J. CHERNICK, " Resonance Escape Probability in Thermal Reactors " in *Progr. Nucl. Energy, Series 1, Physics and Mathematics*, Vol. 2, p. 223, Pergamon Press, London, 1958.
14. A. HASSITT, " Methods of Calculation for Heterogeneous Reactors: Part B " in *Progr. Nucl. Energy, Series I, Physics and Mathematics*, Vol. 2, p. 290, Pergamon Press, London, 1958.
15. D. CALLIHAN, " Homogeneous Critical Assemblies " in *Progr. Nucl. Energy, Series I, Physics and Mathematics*, Vol. 1, p. 227, Pergamon Press, London, 1956.
16. R.T. ACKROYD and J.D. MCCULLEN. " Albedo Calculation Methods " in *Proc, Second U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 38.
17. R.T. ACKROYD and J.M. BALL, " Multi-Group Albedo Methods " (to be published).
18. R.T. ACKROYD and J.M. BALL, " Some Simple Mathematical Models for Criticality Problems " in *Progr. Nucl. Energy, Series IV*, Vol. 3, p. 292-323, Pergamon Press, London.
19. H.C. PAXTON and G.A. GRAVES, " Critical Masses of Fissionable Metals as Basic Nuclear Safety Data ", *LA-1958 (Del)* (1956).
20. H.C. PAXTON, Private communication, 1959.
21. R.T. ACKROYD, A. CAMPBELL and H. HASKEY, " A Finite-Medium Diffusion Theory, Part I, Bare Spheres ", *DEG Report 270 (R)* (1960),
22. R.T. ACKROYD, " A Finite-Medium Diffusion Theory for One-Group Systems " (to be published).
23. M.H.L. PRYCE, " Critical Conditions in Neutron Multiplication 1941 ", *U.K. Report MSP-2A.*
24. B. DAVISON and K. FUCHS, " The Critical Radius and the Time Constant of a Sphere Embedded in a Spherical Scattering Container ", *U.K. Report MS-97* (1946).
25. R.E. PEIERLS, " Critical Conditions in Neutron Multiplication " in *Proc. Camb. Phil. Soc. 35*, 611 (1939).
26. R.T. ACKROYD. " Critical Size, One-Group Diffusion Theory " in *Proc. First U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1955, 5*, 454 .
27. R.T. ACKROYD, " Variational Principles in Critical Size Calculation " in *Proc. Second U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 48.
28. R.T. ACKROYD and M.A. PERKS, " Accuracy and Scope of Variational and Difference Solutions of the Diffusion Equations of Reactor Theory ". *J. Nucl. Energy 10.* 141-146 (1959).
29. A. HASSITT, " An Improved Variational Method for the Solution of the One-Group Diffusion Equations ", *J. Nucl. Energy 2*, 112 (1955).
30. R.T. ACKROYD and J.M. BALL, " Upper and Lower Bounds for Critical Laplacian and Critical Size of some Finite Elliptical Cylinders ", *IGR-R/R-219* (1957).
31. R.T. ACKROYD and J.M. BALL, " On the Conjecture that Steiner Symmetrization Reduces Critical Mass ", *RD(R) 8135*, Risley (1955).

32. R.T. ACKROYD, A. CAMPBELL and H. HASKEY " A Rayleigh-Ritz Procedure for Determining the Flux and the Critical Laplacian of One-Group Diffusion Theory for Heterogeneous Reactors ", *IGR-R/R-210* (1957).
33. R.T. ACKROYD and M.A. PERKS, " Applications of the Rayleigh-Ritz Approximations, Part I, Upper Bounds for the Critical Laplacians of Some Simple Solids ", *IGR-R/R-220* (1957).
34. R.T. ACKROYD and M.A. PERKS, " Application of the Rayleigh-Ritz Approximations, Part II, Upper Bounds for the Critical Laplacians of Some Complex Solids ", *IGR-R/R-221* (1957).
35. A. HASSITT, " A Programme for Solving the Two-Group Diffusion Equations of Cylindrical Reactors ", *AERE-T/M-148* (1957).
36. E.D. PENDLEBURY, " General Perturbation Theory in Neutronics ", *Proc. Phys. Soc. 68A*, 474 (1958).
37. J.H. TAIT, " Two-Group Theory in Neutron Transport Theory ", *Proc. Phys. Soc, 67A*, 615 (1954).
38. K. FUCHS, " Perturbation Theory in Neutron Multiplication Problems ", *Proc. Phys. Soc. 62A*, 791 (1949).
39. K. PARKER, " Machine Production of Group Cross Section for Use in Carlson S_n and other Multi-Group Neutronics Calculations on High-Speed Computers ", *AWRE 0-1/61* (1961).
40. S. FRANKEL and E. NELSON," Methods of Treatment of Displacement Theory ", *AECD-3497 (LADC-79)* (1953).
41. R.E. MARSHAK, " The Variational Method for Asymptotic Neutron Densities ", *Phys. Rev. 71*, 688 (1947).
42. S.A. KUSHNERIUK and D.C. MCKAY, " Neutron Density in an Infinite, Non-Capturing Medium Surrounding a Long Cylindrical Body which Scatters and Captures Neutrons ", *CRT-566* (1954).
43. B. MONTAGNINI, " An Extension of a Variational Calculation by Kushneriuk and McKay to Evaluate the Extrapolation Length of Fuel and Absorber Elements ", *Energia Nucleare 7*, 724 (1960).
44. B.G. CARLSON, " Solution of the Transport Equation by S_n Approximations ", *LA-1891* (1955).
45. B.G. CARLSON ," The S_n Method and SNG Code ", *LAMS-2201* (1958).
46. B.G. CARLSON and G.I. BELL, " Solution of the Transport Equation by the S_n Method" in *Proc. Second U.N. Int. Conf. Peaceful Uses Atomic Energy. Geneva, 1958, 16*, 535.
47. B.G. CARLSON, " Numerical Solution of Transient and Steady-State Neutron Transport Problems ", *LA-2260* (1959).
48. B.G. CARLSON. C. LEE and J. WORLTON, " The DSN and TDC Neutron Transport Codes ", *LAMS-2346* (1959).
49. L.H. UNDERHILL, Unpublished.
50. M.A. MEYER (ed), *Symposium on Monte Carlo Methods*, J. Wiley, New York, 1956.
51. J.B. PARKER and E.R. WOODCOCK, (to be published in *Progr. Nucl. Energy*, Pergamon Press, London).
52. E.R. WOODCOCK and J.B. PARKER, "Infinite Slab Criticality Calculations ", *AWRE 0-14/60* (1960).
53. J.M. HAMMERSLEY and K.W. MORTON, " A New Monte Carlo Technique; Antithetic Variates ", *Proc. Camb. Phil. Soc. 52*, 449 (1956).
54. R.T. ACKROYD, H. HASKEY and F. CHAPMAN, " Neutron Traffic Model for a Non-Multiplying Medium " (to be published).
55. D. SCHIFF, " Multi-Group Integral Network Method ", *WAPD-108* (1954).
56. H. HASKEY, " Calculation of Neutron Transport through Adjoining Media by a Matrix Method ", (1960) (Limited circulation).
57. R.T. ACKROYD, " Geometrical Methods for Determining the Accuracy of Approximate Solutions of the Boltzmann Equation ", (1960) (Limited circulation).
58. R.T. ACKROYD, Addendum to Reference 57 (to be published).
59. G.C. WICK, " Plane Diffusion Problems ", *Z. Phys. 121*, 702 (1943).
60. S. CHANDRASEKHAR, *Radiative Transfer*, Clarendon Press, Oxford, 1950.

61. G. GOERTZEL, " The Method of Discrete Ordinates ", *Nucl. Sci. Eng. 4.* 581 (1958).
62. H.A. BETHE, L. TONKS and H. HURWITZ, *Phys. Rev. 80,* 11 (1950).
63. J.L. SYNGE, *The Hypercircle in Mathematical Physics,* Cambridge University Press (1957).
64. K. PARKER, See Reference 39.
65. M.H. KALOS and E. TROUBETZKY, " Fast Neutron Cross Sections... 1960 ", *NDA-2134-2.*
66. H. LUSTIG, " Final Report Covering the Period Dec. 17 1958 to Sept. 30 1959 ", *NDA-2111-3* (1959).
67. B.R.S. BUCKINGHAM, K. PARKER, and E.D. PENDLEBURY, " Neutron Cross Sections.... 1961 ", *AWRE 0.28/60.*
68. D.J. HUGUES and R.B. SCHWARTZ, " Neutron Cross Sections ", *BNL-325* (1968) and Supplement 1 (1960).
69. R.J. HOWERTON,, " Semi-Empirical Neutron Cross Sections ", *UCRL-5351* (1958).
70. K. PARKER, " Neutron Cross Sections of Beryllium ", *AWRE 0-27/60* (1960).
71. J.B. PARKER and K. PARKER," The Preparation of Nuclear Data for Use in Neutronics Calculations on High-Speed Computers ", *AWRE 0-20/60* (1960).
72. K. PARKER, See Reference 39.
73. S. YIFTAH, D. OKRENT and P.A. MOLDAUER, *Fast Reactor Cross-Sections,* Pergamon Press, London, 1960.
74. W.H. ROACH, " Computational Survey of Idealized Fast Breeder Reactors ", *Nucl. Sci. Eng. 8,* 621 (1960).
75. B.M. SEGAL, " A Method for Estimating Fast Reactor One-Group Cross Sections ", *IG Report 209 (RD/R)* (1959).
76. J.B. TAYLOR and L.H. UNDERHILL, (1956) (Unpublished).

MÉTHODES THÉORIQUES UTILISÉES DANS LES CALCULS DE CRITICALITÉ

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INTRODUCTION

Les méthodes théoriques utilisées dans les calculs de criticalité ne diffèrent pas essentiellement des méthodes employées pour le calcul des conditions critiques d'une pile. Toutefois, les études de criticalité s'appliquant à des domaines très divers (usines de séparation d'uranium, usines de plutonium, stockages solides ou liquides, etc.), on conçoit que le champ couvert par ces études pourra être extrêmement vaste, tant par la géométrie que par la composition. Il est donc assez difficile d'effectuer un classement dans ce domaine et de relier entre elles des études destinées à des appareils très différents. D'autre part, l'optique dans laquelle doivent être abordés les calculs de criticalité diffère sensiblement de celle qui est envisagée dans la recherche des conditions critiques d'une pile : dans ceux-ci, on s'attache à la plus grande précision absolue, dans ceux-là, la précision a moins d'importance que la garantie d'une inégalité sur le facteur de multiplication ; l'intuition permet alors souvent de ramener un système complexe à un système géométriquement ou énergétiquement plus simple, de facteur de multiplication plus élevé ; encore faut-il que la garantie offerte par cette schématisation ne soit pas trop large, sous peine de perdre tout intérêt pratique. On reviendra plus loin sur ce point.

MÉTHODES DE CALCUL

Au lieu de chercher à diviser le domaine des études de criticalité en différentes rubriques correspondant chacune à leur application à un type d'appareil, il nous a paru préférable, dans l'état actuel de nos connaissances, de préciser le domaine d'utilisation des différentes méthodes théoriques dont nous disposons :

Domaine où l'on peut éviter l'emploi des théories multigroupes

Domaine des solutions aqueuses thermiques

a) *Constantes utilisées*

On peut en général se limiter à une théorie à deux groupes. On pourrait éventuellement utiliser des constantes déduites de mesures fondamentales,

mais il nous semble préférable, lorsque la chose est possible, de faire usage de paramètres ajustés sur des expériences globales.

On utilise à Saclay le jeu de constantes publiées en 1956 par Thomas [1] jusqu'à une valeur du rapport H/U 235 de l'ordre de 50 à 100, bien que dans la région des systèmes sous-modérés les résultats fournis par une théorie à deux groupes soient peut-être sujets à caution. Les constantes de Thomas sont ajustées sur quatre expériences critiques sphériques portant sur des solutions de UO_2F_2 réfléchies à l'eau. On utilise ce jeu de constantes dans le cas de solutions contenant un sel quelconque d'uranium, dans des géométries variées, avec différents types de réflecteurs. L'emploi, dans un domaine aussi étendu, d'un système de constantes reposant sur un aussi petit nombre d'expériences constitue évidemment une extrapolation abusive. Une étude reste à faire pour établir un jeu de constantes reposant sur des bases plus solides. Cette étude comporterait essentiellement trois étapes :

— Compilation des résultats expérimentaux parus jusqu'à ce jour dans le domaine des solutions aqueuses thermiques ;
— Mise au point d'un jeu de formules comportant un certain nombre de perfectionnements, en particulier en ce qui concerne la définition des sections efficaces dans des milieux contenant une forte proportion de neutrons épithermiques (sections efficaces du type Westcott [2]) ;
— Ajustement de paramètres convenablement choisis sur les résultats expérimentaux.

La difficulté d'une telle étude, par comparaison avec les études analogues déjà effectuées dans le domaine des piles classiques, réside dans le fait que les systèmes envisagés sont souvent de faibles dimensions devant les longueurs caractéristiques. Il devient alors hasardé d'attribuer à un milieu donné des propriétés intrinsèques indépendantes de celles des milieux voisins. Ceux-ci agiront en effet sur le spectre neutronique du milieu initial, qui n'aura de plus aucune raison d'être indépendant des variables d'espace. D'autre part les problèmes de conditions aux limites prendront ici une grande importance.

b) *Programmes disponibles*

Quels sont les programmes susceptibles d'être utilisés en théorie à deux groupes?

i) *Théorie de la diffusion*

Programmes à une dimension :

— Programme RIFIFI (Saclay) [3] pour calculateur Ferranti Mercury. C'est un programme analytique permettant de traiter très rapidement des problèmes à symétrie plane, cylindrique ou sphérique. Il repose sur une légère approximation qui limite son emploi au cas où l'épaisseur d'un milieu donné n'est pas trop mince en regard de son libre parcours (toutefois, les gaines minces peuvent être traitées comme une modification des conditions aux limites à la frontière de deux milieux). Le programme permet de déterminer soit la dimension critique d'un milieu donné ou du système global, soit le $k\infty$ critique d'un milieu.

Pour donner une idée du temps de calcul on peut dire qu'il faut environ une minute pour traiter un problème à 4 milieux, en géométrie cylindrique. Ce temps est plus court en géométrie plane ou sphérique.

— Programme de Linde [4] pour calculateur Ferranti-Mercury. C'est un programme numérique permettant de traiter des cas à symétrie plane, cylindrique ou sphérique. Il permet la détermination du ν critique, mais non, comme le précédent, la détermination directe de la dimension critique.

Programmes à deux dimensions :

— Programme PDQ [5] pour IBM-704. C'est un programme numérique permettant un quadrillage en X — Y ou en R — Z. En utilisant un quadrillage assez serré, on peut traiter des configurations assez compliquées, au prix d'une augmentation du temps machine. Ce programme qui fournit le k effectif d'un système, peut être utilisé jusqu'à 4 groupes.
— Programme CURE [6] pour IBM-704. C'est un programme numérique permettant un quadrillage en X — Y, R — Z ou R — 0.

ii) *Théorie du transport*

Dans certains cas spéciaux, en particulier lorsque les dimensions du système deviennent faibles en regard du libre parcours, ou lorsqu'il existe des zones vides à l'intérieur de celui-ci, il peut être nécessaire d'abandonner la théorie de la diffusion au profit de la théorie du transport*. On a recours dans ce cas à la méthode S_n. On dispose de trois programmes reposant sur cette méthode.

Programmes à une dimension :

— Programme S_n Saclay pour calculateur Ferranti-Mercury. Ce programme, utilisable en géométrie sphérique, permet la détermination du rayon critique ou celle du ν critique, dans les approximations S_4 et S_8. Les spécifications de ce programme sont données dans [7].
— Programmes DSN [8] pour IBM-704. Ce programme, utilisable en géométrie plane et sphérique dans les approximations S_2, S_4, S_6, S_8 et S_{16}, et en géométrie cylindrique dans les approximations S_2, S_4 S_6 et S_8, permet, soit la détermination du k effectif d'un système, soit celle de la dimension critique d'un milieu donné ou du système global, soit encore celle de la concentration critique d'un ou plusieurs isotopes. Il est très souple et permet d'utiliser un très grand nombre de points. Pour fixer les idées, un calcul à deux groupes dans l'approximation S_4, avec 2 milieux et 40 points prend 7 à 8 minutes.

Programme à deux dimensions :

— Programme TDC [8] pour IBM-704. Ce programme permet un quadrillage en R — Z. Par rapport au programme DSN correspondant utilisé en géométrie cylindrique, il multiplie approximativement le temps de calcul par 2 m, où m est le nombre de points utilisé sur l'axe Z.

Domaine rapide pur

Dans le cas d'un système rapide pur, formé d'uranium enrichi ou de plutonium contenant des éléments suffisamment lourds pour que le spectre

* J. Gunther a montré dans une étude récente que les conditions aux limites utilisées en théorie de diffusion multigroupe sont erronées lorsque le rapport des coefficients de diffusion de deux milieux adjacents varie beaucoup d'un groupe à l'autre. On devrait donc dans ce cas utiliser la théorie du transport.

soit peu dégradé par rapport au spectre de fission, on peut se limiter à un petit nombre de groupes, et utiliser les constantes à un, deux ou trois groupes données dans l'ouvrage " Reactor Physics Constants " [9] (p. 421). On peut utiliser les programmes énoncés ci-dessus.

Système rapide réfléchi par un matériau hydrogéné

Il s'agit là d'un système intermédiaire dont le traitement nécessite une théorie multigroupe. Toutefois, une théorie à un petit nombre de groupes peut être utilisée, en adjoignant un groupe thermique aux groupes rapides donnés dans le " Reactor Physics Constants ". Par ce procédé on ignore l'existence des neutrons intermédiaires, mais les résultats obtenus dans quelques cas particuliers semblent concorder de façon satisfaisante avec les résultats expérimentaux. L'utilisation des programmes énoncés plus haut peut soulever dans certains cas quelques difficultés. En particulier le programme analytique de diffusion à deux groupes RIFIFI ne permet pas de traiter ce problème, car, dans le cœur, la présence de fissions rapides et l'absence de ralentissement conduisent à attribuer une valeur négative au paramètre qui, dans un calcul à deux groupes ordinaire, représente la section efficace de ralentissement ; ceci n'est pas prévu dans ce programme. En ce qui concerne les programmes numériques, tant de diffusion que de transport, il est important de noter que la très rapide décroissance du flux thermique dans le cœur, dès qu'on s'éloigne de sa frontière, oblige à choisir des mailles très serrées dans cette région, sous peine d'obtenir des résultats erronés. Signalons que, pour les forts enrichissements, les faibles dimensions du cœur rendent nécessaire l'utilisation des programmes S_n.

Les systèmes formés d'un cœur rapide réfléchi à l'eau peuvent être calculés d'une manière très simple par une théorie étudiée par MM. R. Caizergues et C. Clouet d'Orval [10]. Il s'agit de la théorie à un groupe et demi. Cette théorie attribue un groupe de neutrons au cœur et deux groupes au réflecteur d'eau ; le cœur est supposé noir pour les neutrons thermiques et la condition aux limites à laquelle satisfait le groupe thermique dans le réflecteur est donnée par la longueur d'extrapolation du corps noir ; les neutrons thermiques qui pénètrent dans le cœur produisent des fissions dans une couche supposée infiniment mince à la frontière. On conçoit que cette théorie sera d'autant meilleure que le parcours des neutrons thermiques dans le cœur sera plus faible.

Domaine rapide ou intermédiaire — Systèmes contenant un ralentisseur

a) *Constantes utilisées*

L'emploi des théories multigroupes est alors inévitable. Nous utilisons l'un ou l'autre des jeux de sections efficaces suivants :

— Sections efficaces de Kiehn [11] à 10 groupes ;
— Sections efficaces de Hansen et Roach [12] à 16 groupes.

L'inconvénient du jeu de constantes de Kiehn, destiné à l'étude de réacteurs rapides peu dégradés, est que les constantes de l'hydrogène n'y figurent pas. Il serait cependant facile de les y adjoindre, en pondérant les courbes de sections efficaces sur le spectre utilisé par Kiehn. Il faudrait cependant admettre que la présence d'hydrogène ne modifie pas ce spectre.

b) *Programmes disponibles*

Les programmes de diffusion multigroupes dont nous disposons sont :

i) *Théorie de la diffusion*

Programmes à une dimension :

— Programme PROD II, pour IBM-650, établi par Habetler et Walbran [13], en application de la méthode de Ehrlich et Hurwitz [14]. Ce programme est utilisable en géométrie plane, cylindrique ou sphérique pour un nombre de groupes inférieur à 50. Lorsque le système contient de l'hydrogène, ce programme repose sur l'approximation de Selengut-Goertzel. Nous avons rencontré des difficultés dans l'application de ce programme, en particulier en présence d'hydrogène.

Programme à deux dimensions :

— Programme CURE [6], déjà cité, pour IBM-704, permettant un quadrillage en X — Y, en R — Z ou en R — θ. Ce programme, utilisable pour un nombre de groupes inférieurs à 50, présente le grave inconvénient de ne pas permettre d'inclure les diffusions inélastiques.

ii) *Théorie du transport*

Ce sont les trois programmes cités plus haut. Le programme S_n Saclay [7], utilisable jusqu'à 20 groupes, demande, pour un problème sphérique, dans l'approximation S_4, avec 10 groupes, 3 milieux et 30 points de division, un temps de calcul approximatif de 20 minutes pour une recherche de ν critique et de 1 heure pour une recherche de rayon critique.

Réduction à un petit nombre de groupes :

Il existe certains programmes à grand nombre de groupes permettant, à partir des courbes de sections efficaces déduites des mesures fondamentales, de calculer des paramètres destinés à être introduits dans un calcul simple à deux (ou quatre) groupes ; ces paramètres sont obtenus par une moyenne des courbes de sections efficaces sur un spectre approprié ; ce spectre est calculé par le programme lui-même, compte tenu de la composition du milieu et, dans une certaine mesure, de sa géométrie ; dans le programme MUFT [19], ce spectre est calculé dans l'hypothèse d'un système nu, pour une valeur quelconque du laplacien B^2, en assimilant les fuites à chaque énergie comme une capture fictive ; dans le programme P_1MG [20], le calcul du spectre est plus élaboré, puisqu'il permet de tenir compte d'une géométrie à plusieurs milieux, à une dimension. Nous n'avons pas jusqu'ici utilisé ces programmes pour les études de criticalité, mais nous pensons qu'ils devraient permettre une extension du domaine d'emploi de la théorie simple à deux groupes à des systèmes intermédiaires, en évaluant une fois pour toutes des paramètres pouvant être raisonnablement considérés comme valables dans des régions plus ou moins étendues de modération.

Cas d'un système non réfléchi

Dans le cas d'un système non réfléchi, de forme quelconque, la résolution des équations de diffusion multigroupe peut se ramener à un calcul simple, si l'on peut faire l'hypothèse que la longueur d'extrapolation, et donc le la-

placien B^2, sont les mêmes dans chaque groupe. Les fuites peuvent alors être traitées comme une capture fictive d'intensité D_i, B^2, où D_i est le coefficient de diffusion du groupe *i*. On est alors ramené au cas du milieu infini. Il existe à Saclay un programme établi en autocode sur calculateur Ferranti-Mercury permettant de résoudre rapidement ce problème.

Nous avons en particulier [15] utilisé cette méthode pour le calcul de systèmes concentrés en U 235 et pauvres en hydrogène. Les résultats obtenus avec le jeu de constantes de Hansen et Roach [12] se sont trouvés en accord acceptable avec les seuls résultats de mesure connus concernant de tels systèmes [16].

Mais il est évident qu'en pratique l'hypothèse d'une longueur d'extrapolation indépendante du numéro du groupe est erronée puisque cette longueur est liée au libre parcours de transport λ tri dans chaque groupe *i*, et que ce libre parcours peut varier beaucoup lorsqu'on passe de l'énergie de fission à l'énergie thermique. Cependant une bonne approximation consiste à utiliser comme longueur d'extrapolation moyenne Λ, celle donnée par le théorème d'Inonu [17], qui définit la pondération à adopter : en première approximation, la longueur d'extrapolation du groupe *i* doit être pondérée par les fuites de neutrons correspondant à ce groupe *i*, ce qui conduit dans le cas d'une surface plane ou peu incurvée à l'expression :

$$\Lambda = 0{,}71 \frac{\sum_i \lambda_{tri} \Phi_i \Delta u_i}{\sum_i \lambda_{tri} \Phi_i \Delta u_i}$$

où Φ_i est le flux moyen dans le groupe *i* et Δu_i l'intervalle de léthargie de ce groupe.

Majoration des conditions critiques

Un procédé très souvent employé dans les études de criticalité lorsqu'on veut garantir la sécurité d'un système consiste à remplacer celui-ci par un système plus simple et plus réactif. Par exemple, on remplacera souvent un système de dimensions finies par un système infini de même composition.

Cette garantie, évidente pour un système nu, ne l'est pas pour un système formé d'un cœur entouré d'un réflecteur. En effet, le remplacement du réflecteur par un milieu fissile de même composition que le cœur n'augmente pas nécessairement la réactivité puisque les neutrons renvoyés par le réflecteur, bien que moins nombreux, peuvent éventuellement, en toute généralité, avoir un spectre tel qu'ils possèdent une importance neutronique plus grande que les neutrons renvoyés par un milieu fissile de même composition que le cœur ; le cas pourrait se présenter pour un système formé d'un cœur rapide ou intermédiaire muni d'un réflecteur hydrogéné. Si toutefois on peut admettre que le produit ηf cans le cœur est une fonction non décroissante de l'énergie, ce qui, à condition de faire abstraction des résonances, est en pratique le cas, on peut, dans la pratique, être certain que la réactivité d'un système réfléchi est inférieure à celle d'un milieu infini de même composition que le cœur, puisque le réflecteur ne peut renvoyer que des neutrons dégradés en énergie, donc d'une importance moindre.

Si la garantie offerte par le remplacement d'un système fini par un système infini est trop large, c'est-à-dire, si le système infini diverge, on sera conduit, dans le cas où le système fini présente une forme trop compliquée

pour pouvoir être traité directement, à remplacer celui-ci par un système de dimensions plus grandes, mais de forme plus simple. On pourra par exemple l'englober dans une sphère ou un cylindre.

Si la garantie est encore trop large on pourra, dans certains problèmes, tirer profit du procédé de symétrisation de Steiner. Ackroyd et Ball [18] ont montré, au moins dans certains cas particuliers (cœur fissile avec réflecteur, en théorie à un groupe), qu'une telle déformation réduit la masse critique, donc conduit à un système plus sûr du point de vue de la criticalité.

Problèmes d'interaction

Un programme d'expérience est en cours d'élaboration. Nous allons essayer de mettre au point des méthodes de calcul destinées à l'interprétation de ces expériences.

Nous serions très intéressés par les résultats expérimentaux concernant des géométries complexes, qui pourraient nous être communiqués. Ces résultats nous permettraient d'éprouver la valeur de nos méthodes de calcul.

REFERENCES

1. J.T. Thomas, " Parameters for Two Group Analysis of Critical Experiments with Water-Reflected Spheres of UO_2F_2-Aqueous Solutions ", *ORNL-2081*, p. 81 (Nov. 1956).
2. C.H. Westcott, " Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra ", *AECL-670* (Aug. 1958).
3. A. Amouyal, P. Bacher, B. Lago, F.L. Mengin et E. Parker, « RIFIFI, Méthode de calcul analytique de la condition critique et des flux d'une pile à régions variées en théorie à deux groupes et à une dimension, programmée pour le calculateur électronique Mercury (Ferranti) », *CEA 1398* (1960).
4. S. Linde, " The Multigroup Neutron Diffusion Equation: 1 Space Dimension ", *RFN-3*, Stockholm (1960).
5. G.G. Bilodeau, W.R. Cadwell, J.P. Dorsey, J.G. Fairey and R.S. Varga, " PDQ - An IBM-704 Code to Solve the Two-Dimensional Far Group Neutron Diffusion Equation ", *WAPD-TN-70* (Aug. 1957).
6. Wachspress, " CURE: Generalized Two-Spaced Dimensions Multigroup Coding for the IBM-704 ", *KAPL-1724* (1957).
7. J. Guerin et B. Lago. « Programme S_n », *SACE-15*, Saclay (1959).
8. C.E. Lee, " The DSN and TDC Discrete S_n Multigroup Neutron Transport Theory Codes ", *Trans. Am. Nucl. Soc., 3*, No. 2, 371 (1960).
9. " Reactor Physics Constants ", *ANL-5800* (July 1958).
10. R. Caizergues et C. Clouet d'Orval, « Milieux faiblement modérés réfléchis par un réflecteur fortement hydrogéné, Théorie à un groupe et demi », *S.E.C./S No. 55 - PRO/CRIT 16*, Saclay (1960).
11. R.M. Kiehn, " Some Applications of the S_n Method ", *Nucl. Sci. Eng. 4*, No. 12, 166 (1958).
12. G.E. Hansen and W.H. Roach, " Sixteen Group Cross Sections ", *N-2-753*, Los Alamos.
13. G.J. Habetler, " One-Space-Dimensional Multigroup for the IBM-650 - Part I: Equations ", *KAPL-1415* (1955).
13. V.A. Walbran, " One-Space-Dimensional Multigroup for the IBM-650 - Part II: Machine Program ", *KAPL-1531* (1956).
14. R. Ehrlich and H. Hurwitz Jr., " Multigroup Methods for Neutron Diffusion Problems ", *Nucleonics 12*, No. 2, 23 (Feb. 1954).

15. R. CAIZERGUES, C. CLOUET D'ORVAL, E. DEILGAT et Mlle C. LOMBARD, « Calculs de criticalité sur des milieux très concentrés en U 235 et pauvres en hydrogène », *EC/S 46 - PRO/CRIT 8.*

16. C.K. BECK, D. CALLIHAN and R.L. MURRAY, " Critical Mass Studies ". Part I. A-4716 (June 1947) and Part II, K-126 (Jan. 1948).

17. E. INONU, à paraître - cité par A.M. Weinberg and E.P. Wigner, *The Physical Theory of Neutron Chain Reactors*, p. 475, University of Chicago Press, 1958.

18. R.T. ACKROYD and J.M. BALL, " On the Conjecture that Steiner Symmetrization Reduces Critical Mass ", *RD(R) 8135*, Risley (1955).

19. R.L. HELLENS, R.W. LONG and B.H. MOUNT, " Multigroup Fourier Transform Calculation - Description of MUFT-III Code ", *WAPD-TM-4* (July 1956).

19. H. BOHL Jr., E.M. GELBARD and G.H. RYAN, " MUFT-IV - Fast Neutron Spectrum Code for the IBM-704 ", *WAPD-TM-72* (July 1957).

19. A.F. HENRY, " 54 Group Library for P-1 Programs ", *WAPD-TM-224* (April 1960).

20. BOHL, et al., " Programme P_1MG ", *WAPD-TM-35.*

ANNEXE

Introduction

Les problèmes de criticalité posés par le traitement des matières fissiles amènent souvent à considérer des cœurs formés de matériaux fissiles à faible teneur en modérateur, entourés d'un réflecteur fortement modéré.

Nous avons essayé d'appliquer à ce genre de milieu une théorie à deux groupes d'énergie, lorsque le cœur absorbe peu les neutrons thermiques, à un groupe d'énergie dans le cœur et deux groupes dans le réflecteur, lorsque le cœur absorbe fortement les neutrons thermiques. Exemple : sphère de Pu métal.

Les résultats ainsi obtenus ont été comparés à ceux obtenus par une méthode Sn à deux groupes et une dimension, programmée sur IBM-7090 : la méthode DSN.

Position du problème

Considérons un réacteur formé d'une sphère d'uranium à divers enrichissements, réfléchie par un réflecteur infini d'eau.

Les neutrons, produits par fission dans le cœur, vont être ralentis dans le réflecteur. Ils ont une certaine probabilité de revenir dans le cœur, à l'état épithermique ou thermique.

On peut donc dire que tous les neutrons rapides sont produits dans le cœur, et la plupart des neutrons thermiques dans le réflecteur.

Nous ferons donc une hypothèse simplificatrice :

— Les neutrons rapides sont produits dans le cœur par fission des neutrons thermiques venant du réflecteur, ainsi que par fissions provoquées par des neutrons rapides ;

— Les neutrons ainsi produits sont supposés d'énergie moyenne voisine de 0,5 MeV ;

— Le réflecteur d'eau reçoit les neutrons rapides produits dans le cœur. Nous prendrons pour valeur des constantes de celui-ci, celles données dans [1].

Ces hypothèses sont évidemment très simplificatrices. Mais nous avons surtout cherché, jusqu'à présent, à obtenir une valeur minimum de la masse critique dans les divers cas étudiés.

Nous négligeons la présence des neutrons épithermiques dans le cœur, et les fissions qu'ils peuvent engendrer. Nous augmentons légèrement la fuite des neutrons dans le réflecteur et le nombre de neutrons revenant thermiques. La diminution du nombre de fissions dues aux neutrons épithermiques est compensée par l'augmentation de fissions thermiques.

THÉORIE A DEUX GROUPES

Les équations utilisées sont les équations classiques de la théorie de la diffusion :

On a :

Milieu cœur :

$$D_1\nabla^2\Phi_1 + [(\nu\Sigma_f)_1 - \Sigma_{a1}]\Phi_1 + (\nu\Sigma_f)_2\Phi_2 = 0$$
$$D_2\nabla^2\Phi_2 - \Sigma_{a2}\Phi_2 = 0$$

Milieu réflecteur :

$$D'_1\nabla^2\Phi'_1 - \Sigma'_1\Phi'_1 = 0$$
$$D'_2\nabla^2\Phi'_2 - \Sigma'_{a2}\Phi'_2 + \Sigma'_1\Phi'_1 = 0$$

Avec :

Φ_1 = flux de neutrons rapides dans le cœur
Φ_2 = flux de neutrons thermiques dans le cœur
D_1 = coefficient de diffusion des neutrons rapides
D_2 = coefficient de diffusion des neutrons thermiques
Σ_{a1} = section efficace d'absorption pour les neutrons du groupe rapide
Σ_{a2} = section efficace d'absorption pour les neutrons du groupe thermique
$(\nu.\Sigma_f)_1$ = nombre de neutrons de fission émis par cm³, pour un flux rapide de 1 n/cm² s
$(\nu.\Sigma_f)_2$ = nombre de neutrons de fission émis par cm³, pour un flux thermique de 1 n/cm² s

De même, dans le réflecteur :

Φ'_1 = flux de neutrons rapides
Φ'_2 = flux de neutrons thermiques
D'_1 = coefficient de diffusion des neutrons rapides
D'_2 = coefficient de diffusion des neutrons thermiques
Σ'_1 = section efficace de transfert du groupe rapide au groupe thermique
Σ'_{a2} = section efficace d'absorption du groupe thermique.

Constantes utilisées

Cœur

a) *Groupe rapide*

Ce sont les constantes données dans [2] :

	ν	σ_{tr}	σ_f	σ_c
Pu 239...	2,98	6,8	1,85	0,26
U 235 ..	2,6	6,8	1,4	0,25
U 238 ..	2,6	6,9	0,095	0,16

Avec :

σ_{tr} = section efficace microscopique de transport
σ_f = section efficace microscopique de fission

σ_c = section efficace microscopique de capture
ν = nombre moyen de neutrons émis par fission.

b) *Groupe thermique*

Ce sont les valeurs tirées de [3] et [4] :

	ν	σ_{tr}	σ_f	σ_c
U 235 ..	2,5	615	511	91
U 238 ..	0	11,44	0	2,44

Conditions aux limites

Ce sont les conditions classiques :

$\Phi_1 = \Phi'_1$ Egalité des flux à la limite cœur-réflecteur
$\Phi_2 = \Phi'_2$

$$D_1 \frac{\partial \Phi_1}{\partial r} = D'_1 \frac{\partial \Phi'_1}{\partial r}$$

Egalité des courants à la limite cœur-réflecteur.

$$D_2 \frac{\partial \Phi_2}{\partial r} = D'_2 \frac{\partial \Phi'_2}{\partial r}$$

Application de la théorie

On a appliqué cette théorie aux sphères de U à divers enrichissements, réfléchies par un réflecteur infini d'eau. Les résultats sont présentés dans les courbes des figures 1 et 2.

THÉORIE A UN GROUPE/DEUX GROUPES (théorie à un groupe et demi)

Lorsque le cœur est très absorbant pour les neutrons thermiques (sphère Pu ou U 235 métal), on peut admettre que les neutrons thermiques sont absorbés dans une couche très mince (à 99 % dans une couche d'épaisseur 2 mm).

Nous admettrons dans le cœur un groupe de neutrons rapides, d'énergie moyenne 0,5 MeV, et deux groupes de neutrons dans le réflecteur.

Equations utilisées :

Cœur :

$$D_1 \nabla^2 \Phi_1 + [(\nu \Sigma_f)_1 - \Sigma_{a1}] \Phi_1 = 0$$

Réflecteur :

$$D'_1 \nabla^2 \Phi'_1 - \Sigma'_1 . \Phi'_1 = 0$$
$$D'_2 \nabla^2 \Phi'_2 - \Sigma'_{a2} \Phi'_2 + \Sigma'_1 \Phi'_1 = 0$$

Conditions aux limites

Les neutrons thermiques venant du réflecteur sont absorbés à la périphérie, et donnent des neutrons rapides. Tout se passe comme s'il existait, à la limite cœur-réflecteur, une source de neutrons rapides, liés au niveau de puissance.

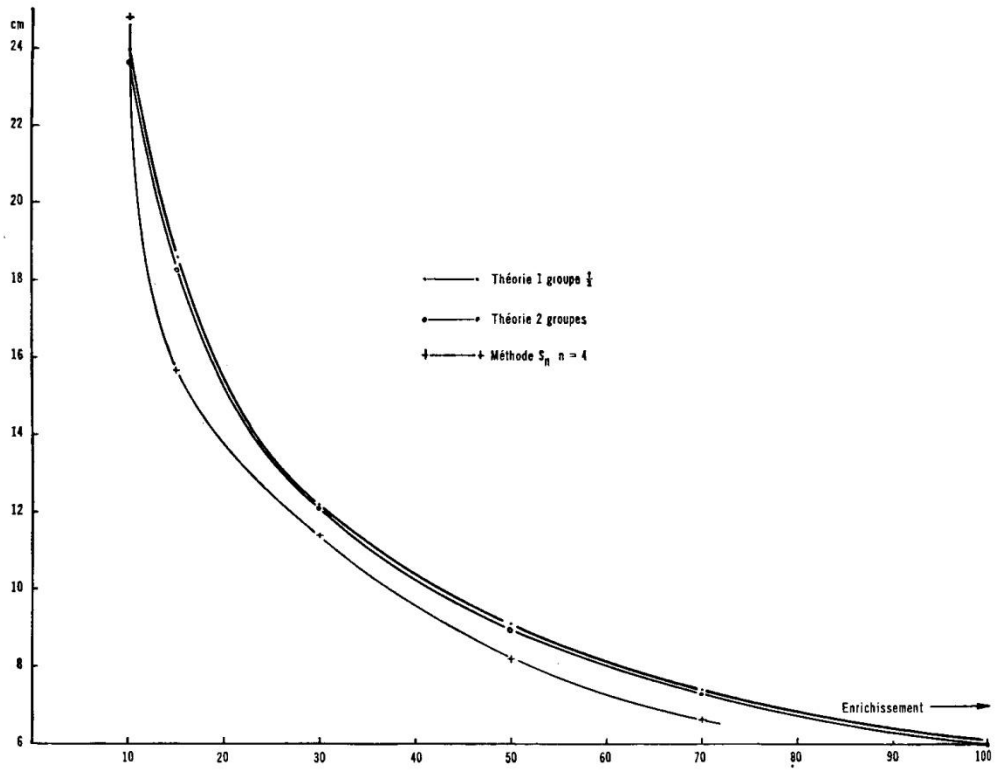

Figure 1. *Rayons critiques en fonction de l'enrichissement.*

Soit :

e = l'épaisseur de la couche mince

S = la surface de la sphère

Le volume ΔV de la couche est tel que :

$$\Delta V \simeq eS$$

Le nombre de neutrons rapides présents dans la couche se conserve à l'état juste critique.

On aura donc :

$$(\nu\Sigma_f)_1\Phi_1\Delta V + \frac{(\nu\Sigma_f)_2}{\Sigma_{a2}}.J'_2.S - \Sigma_{a1}\Phi_1\Delta V - S(I_1 - I'_1) = 0$$

J'_2 = courant de neutrons thermiques entrant dans le cœur par unité de surface ; ce courant vaut $D'_2\left(\frac{\partial\Phi'_2}{\partial r}\right)_{r=R}$ puisque le courant de neutrons thermiques sortant du cœur est nul.

On a, d'autre part :

$$I_1 = -D_1\frac{\partial\Phi_1}{\partial r}\ (r = R)$$

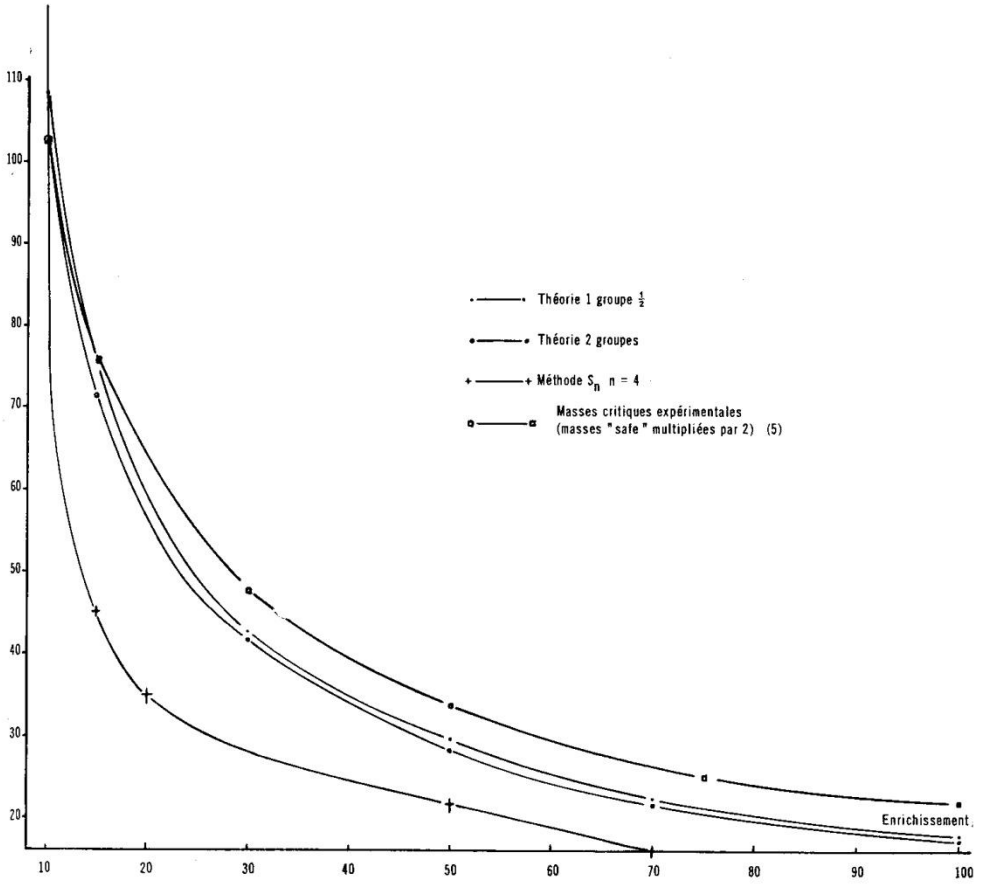

Figure 2. *Masses critiques en kg d'U 235 (en fonction de l'enrichissement).*

$$I'_1 = - D'_1 \frac{\partial \Phi'_1}{\partial r} \quad (r = R)$$

Le nombre de neutrons rapides produits par les neutrons thermiques est égal à :

$$\frac{(\nu \Sigma_f)_2}{\Sigma_{a2}} - SD'_2 \left(\frac{\partial \Phi'_2}{\partial r} \right) r = R = \eta . SD'_2 \left(\frac{\partial \Phi'_2}{\partial r} \right) r = R$$

par seconde.

On a donc finalement la condition :

$$D_1 \left(\frac{\partial \Phi'_1}{\partial r} \right) r = R - D'_1 \left(\frac{\partial \Phi'_1}{\partial r} \right) r = R = \eta D'_2 \left(\frac{\partial \Phi'_2}{\partial r} \right) r = R$$

si e est suffisamment petit.

Les autres conditions aux limites s'écrivent, pour $r = R$:

$$\Phi_1 = \Phi'_1 \qquad \frac{\Phi_2}{\left(\frac{\partial \Phi_2}{\partial r}\right) r = R} = 3D_2\lambda$$

λ (sans dimension) : longueur d'extrapolation du corps noir que l'on prendra égale à 0,71.

Application a une sphère de métal réfléchie par un réflecteur infini d'eau

La résolution de ces équations conduit à l'équation critique :

$$D_1\left(\mu \operatorname{cotg}(\mu R) - \frac{1}{R}\right) + D'_1\left(K_1 + \frac{1}{R}\right) = \frac{\eta . \Sigma'_1}{(K_1 + K_2)\left[1 + 3D_2\lambda\left(K_2 + \frac{1}{R}\right)\right]}$$

avec :

$$\mu^2 = \frac{(\nu\Sigma_f)_1 - \Sigma_{a1}}{D_1}$$

$$K_1^2 = \frac{1}{\tau} = \frac{\Sigma'_1}{D'_1}$$

$$K_2^2 = \frac{1}{L^2} = \frac{\Sigma'_{a2}}{D'_2}$$

Application numérique

On a fait une application numérique aux sphères de Pu et de U enrichi réfléchies par un réflecteur d'eau d'épaisseur infinie.

Les courbes de la figure 1 permettent de comparer les résultats obtenus par les deux théories entre eux, et avec ceux obtenus en multipliant les résultats donnés dans [5] par un facteur 2.

Méthode S_n a deux groupes d'énergie

La théorie de la diffusion n'est valable que lorsque le milieu considéré n'est pas trop absorbant. Ce n'est pas le cas dans la réalité. Les constantes données dans les différents déclassés sont ajustées sur un type de réacteur bien déterminé.

La théorie du transport est plus exacte et peut s'appliquer dans le cas de milieux très absorbants. Il est donc intéressant de comparer les résultats obtenus par la théorie de la diffusion à ceux obtenus par la méthode S*n* à une dimension, programmée sur IBM-7090 (méthode DSN). On a utilisé l'approximation S_4.

Les calculs ont été faits avec des constantes identiques à celles utilisées précédemment. Ils sont condensés dans le tableau suivant :

SPHERES D'URANIUM ENRICHI, RÉFLÉCHIES PAR UN RÉFLECTEUR INFINI D'EAU

Rayon R *(en cm)* en fonction de l'enrichissement e $\left(e = \frac{U\,235}{U\,235 + U\,238}\right)$						
e	0,1	0,15	0,3	0,5	0,7	1
(théorie un groupe et demi) :						
R	24	18,6	12,2	9,1	7,4	6,10
(théorie deux groupes) :						
R	23,65	18,25	12,10	8,95	7,37	6
(méthode S_n) :						
$n = 4$	24,81	15,67	11,38	8,20	6,63	

CONCLUSION

Nous avons essayé, dans cette annexe, d'obtenir, avec des constantes connues, une limite minimum de la masse critique dans des cas où les théories habituelles sont difficiles à appliquer.

Notre but immédiat n'est donc pas d'ajuster des constantes de façon à obtenir des résultats conformes aux résultats pratiques, mais plutôt de mettre au point une théorie permettant d'avoir une limite inférieure des masses critiques dans des systèmes formés d'un cœur faiblement modéré, entourés d'un réflecteur hydrogéné.

REFERENCES

1. J.T. THOMAS, " Parameters for Two Group Analysis of Critical Experiments with Water-Reflected Spheres of UO_2F_2 Aqueous Solutions " in " Applied Nuclear Physics Division Annual Report for Period Ending September 10, 1956 ", p. 81-85, *ORNL-2081* (Nov. 1956).
2. " Reactor Physics Constants ", Fast Reactors, *ANL 5800* (1958).
3. C. PONTIS, " Table des Sections Efficaces ", *CEA 238* (Mars 1958).
4. G.E. HANSEN and W.H. ROACH, " Sixteen Group Cross Sections ", *N-2-753 (Rev. 1)* (1960),
5. H.F. HENRY (Ed.), " Guide to Shipment of U 235 Enriched Uranium Materials " *TID 7019* (1959).

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DISCUSSION

G. BLÄSSER, Euratom :

Dans son exposé, M. Benoist mentionne certaines difficultés dans l'application du programme PROD II. Quelle était la nature exacte de ces dif-

ficultés ? S'agissait-il de difficultés de précision ou de convergence ? Nous avons également rencontré quelques difficultés, et notamment des difficultés de convergence, dans l'application du programme PROD II.

J. GUNTHER, France :

J'aurais voulu donner une précision sur la contribution que j'ai apportée à la communication de M. Benoist. Il s'agit de la validité de l'équation de la diffusion comparée à l'équation du transport dans un cas très précis et très intéressant pour les études de criticalité, celui des réacteurs aqueux. En effet, les réacteurs contenant de l'eau présentent la particularité d'avoir des libres parcours moyens de transport dont le rapport entre celui du groupe rapide et celui du groupe thermique dans une méthode à deux groupes est extrêmement élevé, puisqu'il est de l'ordre de 7. J'ai exploité, pour examiner la validité de la méthode de la diffusion, une remarque qui se trouve dans le livre de Davison : « Neutron Transport Theory ». D'après cette remarque, qui figure dans le chapitre relatif à la théorie multigroupe, les solutions en milieu infini peuvent ne pas être dans la théorie de la diffusion des approximations valables de ce qu'elles sont dans la théorie du transport, lorsque le rapport des libres parcours moyens de transport entre les différents groupes est grand. On sait que l'équation de la diffusion donne des solutions en milieu infini, qui sont des exponentielles dont l'une est de module imaginaire, c'est à-dire correspondant à la solution pour un réacteur nu, et dont les autres sont de module réel, correspondant à des termes transitoires régnant au voisinage des limites. Les modules de ces exponentielles doivent normalement être en nombre égal au nombre de groupes utilisés, de manière à ce que l'on puisse utiliser toutes les conditions limites qui existent. Or, Davison a montré dans son ouvrage que de telles solutions peuvent, dans certains cas, être en nombre inférieur au nombre de groupes, précisément lorsque le rapport des libres parcours de transport est grand. Je me suis alors demandé si, dans le cas des réacteurs contenant de l'eau, on se trouvait dans une telle situation. J'ai trouvé effectivement que c'était le cas, de sorte qu'on ne peut absolument pas dire, lorsqu'on utilise la théorie à deux groupes, que la solution exponentielle à module réel, désigné par « ν » dans l'ouvrage de Glasstone, telle qu'on l'obtient généralement, est une approximation valable de ce qui devrait réellement exister si l'on résolvait l'équation du transport. Aussi ai-je l'impression que, lorsqu'on utilise une théorie à deux groupes avec deux milieux au moins et que l'on a des conditions limites à faire intervenir, la théorie de la diffusion est une beaucoup plus mauvaise approximation de la théorie du transport que lorsqu'on a affaire à un réacteur nu dans lequel seule la solution désignée par « μ » dans l'ouvrage de Glasstone intervient.

R. T. ACKROYD, United Kingdom :

I think the previous speaker is referring to the case of unusable roots, in multi-group diffusion theory, which arises in fitting a solution of the wave equation to that of the integral equation of an infinite medium, that is one way of getting a diffusion theory. You can overcome this if you use the method we have employed for the finite medium theory, where we don't expect to fit a solution of the wave equation everywhere to the integral equation. All we ask is that the neutron economy shall be right and the currents be right. We don't ask for the final detail of agreement of fluxes everywhere.

If you are only asking for critical sizes, then I think that the above is sufficient information to request from a simple theory. The other way to do it would be to get a generalized form of diffusion theory (as Davison has done, by taking into account further spherical harmonics), but I think that if you are going to take all this trouble, it would be preferable, from the point of view of cost, to go to a Carlson programme for the transport equation.

H. LUTZ, Suisse :

M. Benoist a fait remarquer que, dans des systèmes rapides réfléchis par des matériaux hydrogénés, le programme RIFIFI ne convient pas pour le calcul, car il n'est pas prévu de donner une valeur négative au paramètre qui tient compte du ralentissement. Ne pourrait-on pas y remédier ou serait-ce que la théorie n'est pas bonne dans ce cas?

P. BENOIST, France :

La théorie n'est absolument pas en cause sur ce point car il s'agit simplement d'une limitation du programme. Cela tient au fait que l'équation de la diffusion à deux groupes, lorsqu'on l'applique à des systèmes rapides et à ce système en particulier, n'a pas exactement la même forme que l'équation de la diffusion habituelle à deux groupes que l'on applique à des systèmes thermiques. En effet, le paramètre en question change de signe et devient négatif au lieu d'être positif par suite de l'existence des fissions rapides. Il n'est pas prévu d'introduire une section efficace de ralentissement négative dans le programme RIFIFI, qui a été établi dans le but de traiter des problèmes thermiques. C'est là une simple question d'adaptation, mais la théorie n'est pas en cause.

Session II

FUNDAMENTALS
PRINCIPES FONDAMENTAUX (Suite)

Chairman - President :
M. H. DOPCHIE
(Belgique)

A REVIEW OF CRITICALITY DATA OBTAINED BY EXPERIMENTAL METHODS

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Oak Ridge National Laboratory, United States

INTRODUCTION

The design of chemical and metallurgical processes and, indeed, all operations with fissionable isotopes outside of reactor environments must recognize the hazards associated with potentially chain-reacting materials. The method of assuring safety will be determined largely by the process itself, and the magnitude of the controlling variables will depend upon the physical and chemical properties of the process material. This means, in general, that limits must be imposed upon the mass or the volume of the material within a particular operation, or upon its distribution through control of its density, or chemical concentration, or its shape. If a container is cylindrical, for example, a maximum diameter can be specified which will insure against chain reactions. Under certain circumstances isotopic composition is an equally effective control.

The question considered here is, " What is the source of these specifications? "

It has been the practice, at least in the United States, to establish these specifications upon the results of experiments. It is the purpose of this paper to summarize some of these results as illustrations of the information which is available. In general, the presentation will not enable precise evaluation of the individual quantities and the reader is referred to original sources for these details.

Following brief comment on experimental methods, the three more readily fissionable isotopes, U 235, U 233 and Pu 239, are considered with emphasis on U 235 since the greatest experience has been with this most common of the three. An attempt has been made to supplement information generally descriptive of each isotope with more detailed results of particular experiments, some of recent origin, as examples of possible solutions of nuclear safety problems.

A prodigious quantity of data describing critical conditions, gleaned over the past fifteen years from almost illimitable experiments is presented. The results appear in detail in many documents.

DISCUSSION OF EXPERIMENTAL DATA AND THEIR ORIGIN

Perhaps critical experiments constitute the best known source of information applicable to criticality problems. In these, material is arranged

in a chain-reacting configuration of interest, usually with power generation less than one watt. In the most simple of these experiments, as an example, the height of a critical aqueous solution of uranyl nitrate, containing uranium enriched in U 235, would be measured in cylindrical vessels of several diameters; or the critical concentration of the solution would be adjusted to make a selected spherical volume critical. In contrast, limited information useful in transporting and processing reactor fuel elements may be derived from the critical experiments usually required to support the design of the reactor itself.

Valuable information may be obtained from accumulations of fissionable material which are subcritical. In one type of subcritical experiment the source-neutron multiplication by the material, measured as a function of a controlling parameter, is a guide to operational practices which may be safely tolerated. In another type, the neutron-multiplying characteristics and the critical dimensions of a material may be ascertained from the steady-state neutron flux distribution established in a subcritical volume placed adjacent to a large distributed source. Since the neutron flux in the material decreases exponentially with distance from the source, these are known as exponential experiments.

Still another type of experiment utilizes an established low-power reactor. Interpretation of the effect on this reactor's reactivity of a sample of a material of interest centrally placed in the assembly yields knowledge of the material's nuclear properties and estimates of critical quantities.

Experiments of these kinds, performed in the United Kingdom, in Europe, and in the United States, have furnished many data applicable to the safe design and operation of services supporting nuclear reactor programmes. A subsequent paper is intended to describe laboratories, experimental techniques and programmes.

It is impossible to do more here than review the results of these experiments, illustrating the type of information available. Reference is made to an epitome of data of interest in problems of nuclear safety which has been compiled by Paxton [1] and to a summary, including both experimental results and some extrapolations, compiled in the United Kingdom [2]. The comprehensive bibliography of the experimental papers from the United States included in the Nuclear Safety Guide [3] and in Reference [1] will identify the source of many of the data presented in this paper. Results which have appeared in less well-distributed documents will be referenced as completely as possible.

Each value quoted in this summary is either a direct measurement or a short extrapolation of a measurement. The measured quantities were usually corrected at their origin for any known perturbation introduced by undesirable but necessary components of the apparatus and by the environment. The principal difference between that which was measured and that which is included here, however, arises from the necessity of presenting the data in a generalized and interpretable form but yet identifiable with the experiments themselves. Most of the experiments have been performed with the test material in cylindrical or cubical geometry. The results have been transformed to unidimensional geometry — spheres, cylinders of infinite length and slabs of infinite area — by equating geometric bucklings and utilizing an appropriate extrapolation distance. The critical dimensions

have not been altered in an attempt to adjust for physical or chemical differences between test materials. There are included, for example, two sets of data describing uranium enriched to 4.89 % in U 235, one set derived from solutions of UO_2F_2 and the other from a packed powder containing U_3O_8. The densities of these materials differed significantly at the same H/U 235 atomic ratio and, consequently, the critical dimensions are quite dissimilar. The results are reported in this manner because there is no established general method of converting them from one density to another. This point may appear to be unduly stressed but there do exist in the literature values of critical dimensions referred to as data which have been adjusted for gross deviations from the experimental conditions under which measurements were made. It is recognized that, on occasion, conservative extrapolation must be of data for application to areas not covered by experiment.

It is difficult to assign limits of error to data gleaned from the large number of experiments, performed in different laboratories, reported here. A generous uncertainty in the original measurements of critical mass and volume, arising from errors in dimensions, chemical analyses, and the like, is probably no more than $\pm$ 5 % and is considerably less in many cases. The other major error arises from the extrapolation distance used in geometric transformations, although in most cases the value was obtained from a measured neutron flux distribution or was found empirically to fit best the results from experiments with a particular material in a number of different configurations. Since, however, it does depend upon the nuclear properties and the shape of the test material, and upon the neutron reflector conditions, it is very likely that some of the values used are in error. On the other hand the extrapolation distance is additive to the linear dimensions, so errors in it become less important with increasing dimensions. Fortunately those associated with the smaller dimensions, of more highly enriched uranium for example, are better known. It is believed that an overall limit of $\pm$ 5 % will cover the uncertainty in the linear dimensions reported here with proportionately larger values for volumes and masses.

Presentation of Data: U 235-Enriched Uranium

Homogeneous Materials

The properties of neutron multiplying media depend upon the characteristics of the fissionable nucleus and upon the presence of other nuclei, particularly those capable of reducing the energies of fission neutrons to those more favourable to the establishment of a chain reaction. An appropriate unit characterizing this neutron moderating property is the ratio of the number of moderating nuclei present to the number of fissionable nuclei. Since this discussion will be concerned primarily with problems of storage and processing fissionable materials, the most common moderating nucleus is hydrogen present in water and, perhaps, in plastics which may be associated with the processes. The work reported here describes almost exclusively the critical dimensions of hydrogen-moderated media. The H/U 235 atomic ratio is descriptive of the moderation. It is clearly related to the chemical concentration of an aqueous solution. It is related in a different way to the mass of U 235 per unit volume of mixtures of solids or, as an extreme example, of a uranium-aluminium alloy dispersed in water. Figure 1 gives this rela-

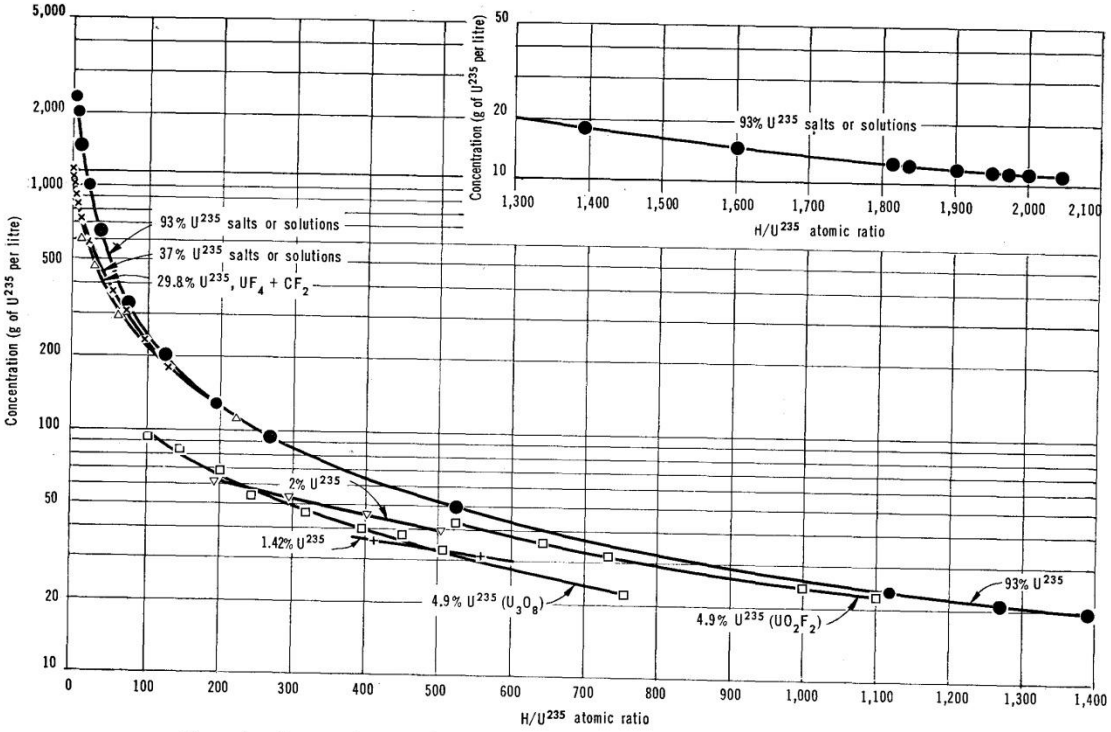

Figure 1. *Concentration as a function of H/U 235 atomic ratio for various U 235 enrichments.*

tion for a number of the test materials used in the experiments. The H/U 235 atomic ratio is plotted as a function of the more common expression, mass of U 235 per unit volume. Heterogeneous lattices of solids in water are commonly described in terms of the volume ratio of these components.

Greater than 90 % *enrichment.* Materials in which fissionable elements are homogeneously distributed have been more widely studied than any other and, of these, aqueous solutions and powder compacts of uranium salts enriched to greater than 93 % in U 235 have been subjected to more extensive investigation. In a great many of these experiments the solutions were contained in cylinders and in a few representative spheres. The assemblies of low density solids were parallelepipedal in shape and, in the limit, the uranium metal was spherical. A generalization of the data from these experiments is plotted as curves A and A′ of Figure 2 exhibiting the critical mass of U 235 in water-moderated spheres as a function of their moderation. Curve A′ represents experiments with no reflector, except the container; curve A represents experiments with an hydrogenous reflector of effectively infinite thickness. The temperatures ranged between 20° and 25°C. The data extend from solutions too dilute to be critical in any quantity, because of

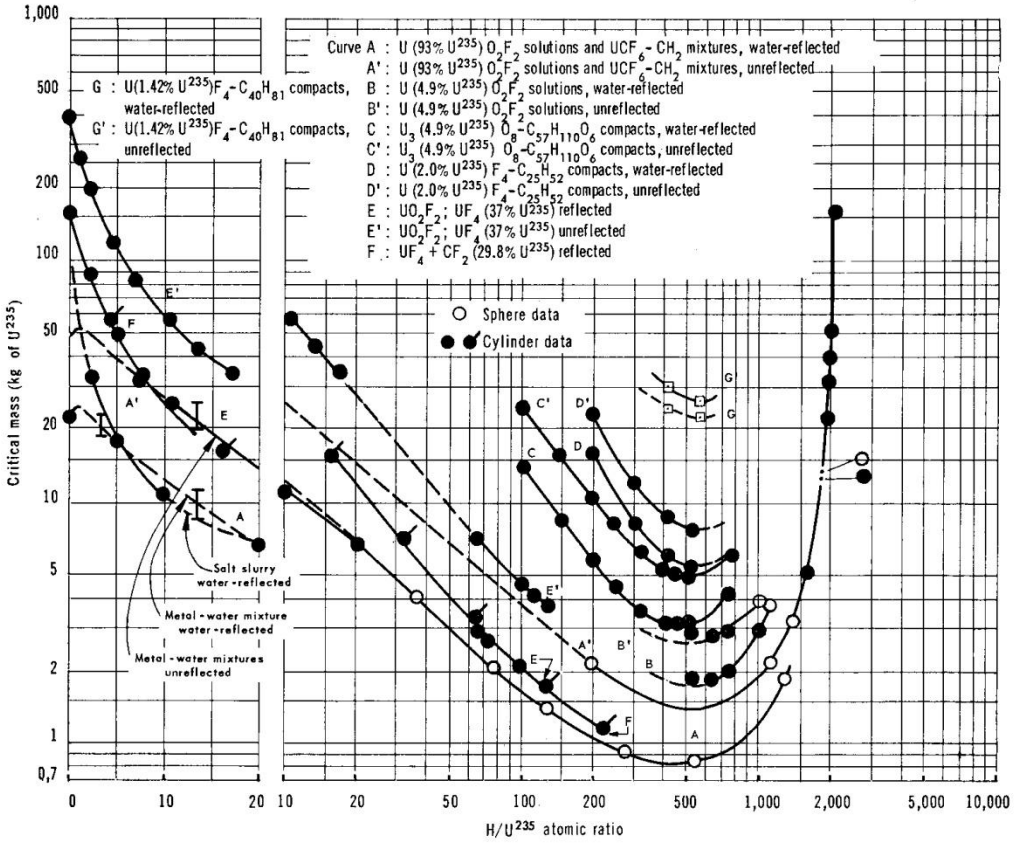

Figure 2. *Critical mass of U 235-enriched uranium in spherical geometry as a function of H/U 235 atomic ratio.*

both the low concentration of uranium and the absorption of neutrons by hydrogen, to uranium metal and to anhydrous uranium salts. The experiments reported as representing the moderation range H/U 235 = 25 to 1,500 were performed with spherical volumes of aqueous solutions of uranyl fluoride, which has high solubility and low parasitic neutron absorption. Within this range, cylinders of solution of many combinations of diameter, height and chemical concentration also have been made critical and the resulting data, although not shown, substantiate curves A and A′. Solubility limits the H/U 235 ratio of solutions to about 20, approximately 1 kg U 235/ litre, if the solute is UO_2F_2.

The region of lower moderation is represented by materials of two types. One is a mixture of UF_4 and plastics having as its limit the hydrogen-free mixture UCF_6, and the other is a suspension of water and metal particulates having as its limit a metal sphere. The critical masses of the limiting materials differ strikingly because of their respective densities and, although the mass of a metal sphere is well known [4], the value for anhydrous UF_4 has not been established. The mass as a function of H/U 235 is shown by two curves over this range of moderation where the intermediate points on the metal-water curve, at H/U 235 of 3.14 and 13.5, were derived from experiments with uranium hydride [5] performed at Los Alamos with metal reflectors. The reduction of these data to the unreflected and water-reflected values is subject to some uncertainty, so the points should be used with no great confidence. The intermediate points on the salt-slurry curves were obtained by introducing additional hydrogenous plastic to the basic material.

Experiments in the high moderation range (H/U 235 $>$ 1,000) examined solutions of uranyl nitrate. The neutron absorption by nitrogen in these dilute solutions is small compared to that by hydrogen, making the results comparable with those from UO_2F_2.

The volumes associated with the masses given in Figure 2 are plotted in Figure 3 (curves A and A′) as a function of the relative hydrogen content, both reflected and water-reflected. Equating geometric bucklings, with appropriate extrapolation distances*, yields the diameter of infinitely long cylinders and the thickness of slabs of infinite area reported as a function of moderation in curves A and A′ of Figures 4 and 5. These results, too, are supported by the extrapolation, to infinite dimensions, of many data from cylindrical and slab-shaped volumes.

The critical thickness of a slab of 93 % U 235-enriched uranium metal as a function of area and of hydrogenous reflector thickness has been recently reported [6]. These results extrapolate to 1.52 and 6.10 cm as the thickness of infinite slabs completely reflected and unreflected, respectively.

An interesting result of the experiments with large volumes of dilute solutions is the determination of the minimum critical chemical concentration of aqueous uranium solutions enriched to greater than 93 % in U 235.

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* The following extrapolation distances were used in the buckling transformations yielding the dimensions appearing in Figures 2-5. Curve A and A′ ($\sim$ 93 % U 235 enrichment, values from Reference 1): 5.9 to 6.6 cm for reflected solutions, 3.8 to 4.5 cm for reflected metal; 2.1 to 2.7 cm for unreflected solutions, 2.1 to 2.3 cm for unreflected metal. Curves B and B′ ($\sim$ 5 % U 235); 4 cm reflected and 1 cm unreflected. Curves C and C′ ($\sim$ 5 %); 7.5 cm reflected and 1 cm unreflected. Curves D and D′ (2 %); 2.7 cm unreflected, reflected dimensions from measured reflector savings. Curves E and E′ ($\sim$ 37 %); 5 cm reflected and 2.5 cm unreflected. Curve F (29 per cent); 5 cm reflected.

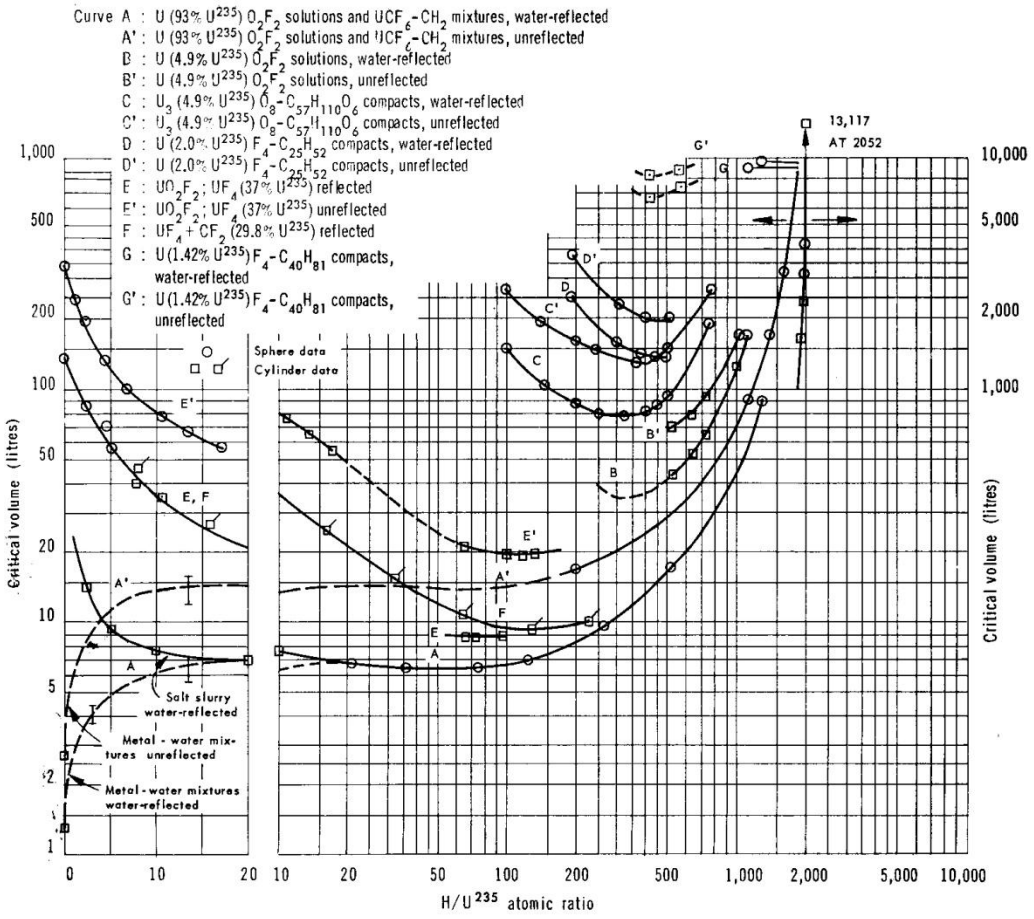

Figure 3. *Critical volume of U 235-enriched uranium in spherical geometry as a function of H/U 235 atomic ratio.*

Extrapolation of a plot of the critical concentration as a function of the geometric buckling of the various volumes to zero buckling [7] shown in Figure 6, gives 12.20 g U 235/litre. Since these experiments were performed with uranyl nitrate solutions the observed concentrations should be decreased by about 1 % to correct for the neutron absorption by the nitrogen.

10 *to* 50 % *enrichment.* Uranium of lower than 90 % U 235 isotropic content has been studied less extensively. Measurements have been made with cylindrical volumes of aqueous uranyl fluoride solutions of 44.6 % U 235 at three chemical concentrations [8]. The cylinders were both unreflected and laterally reflected by water. The combinations of dimensions and concentrations which were made critical at 15°-18° C have been converted to spherical geometry using 5.0 and 2.2 cm, respectively, for the reflected and unreflected extrapolation distances. The results are in Table I. No correction has been made for the 0.3 cm thick stainless steel vessel wall or for the temperature.

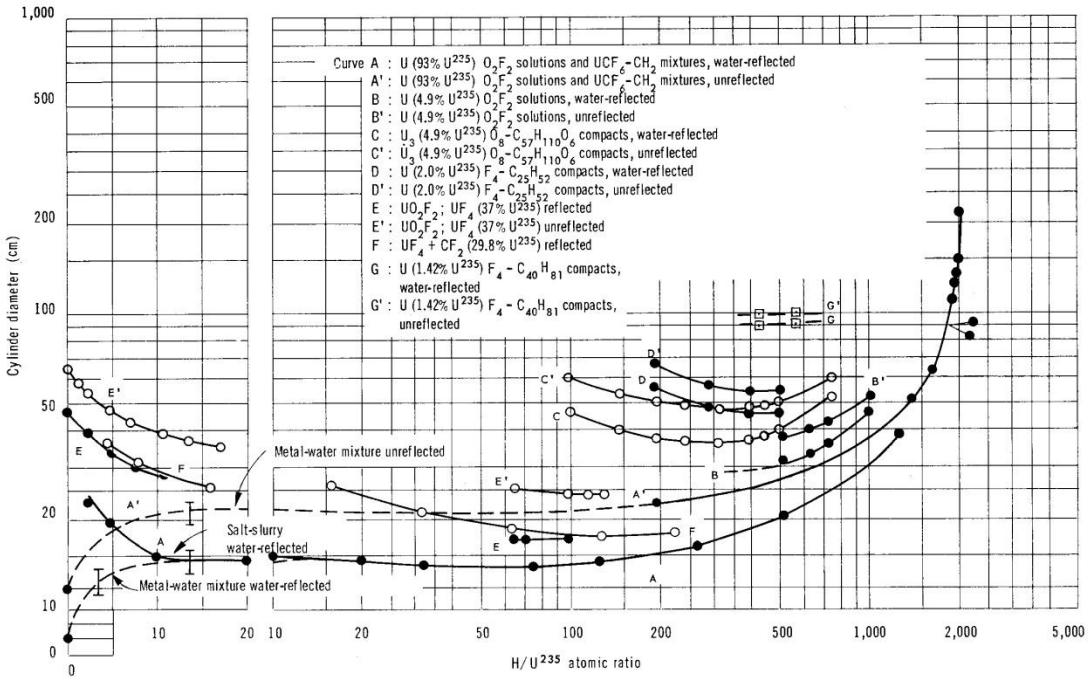

Figure 4. *Critical diameter of infinite cylinders as a function of H/U 235 atomic ratio.*

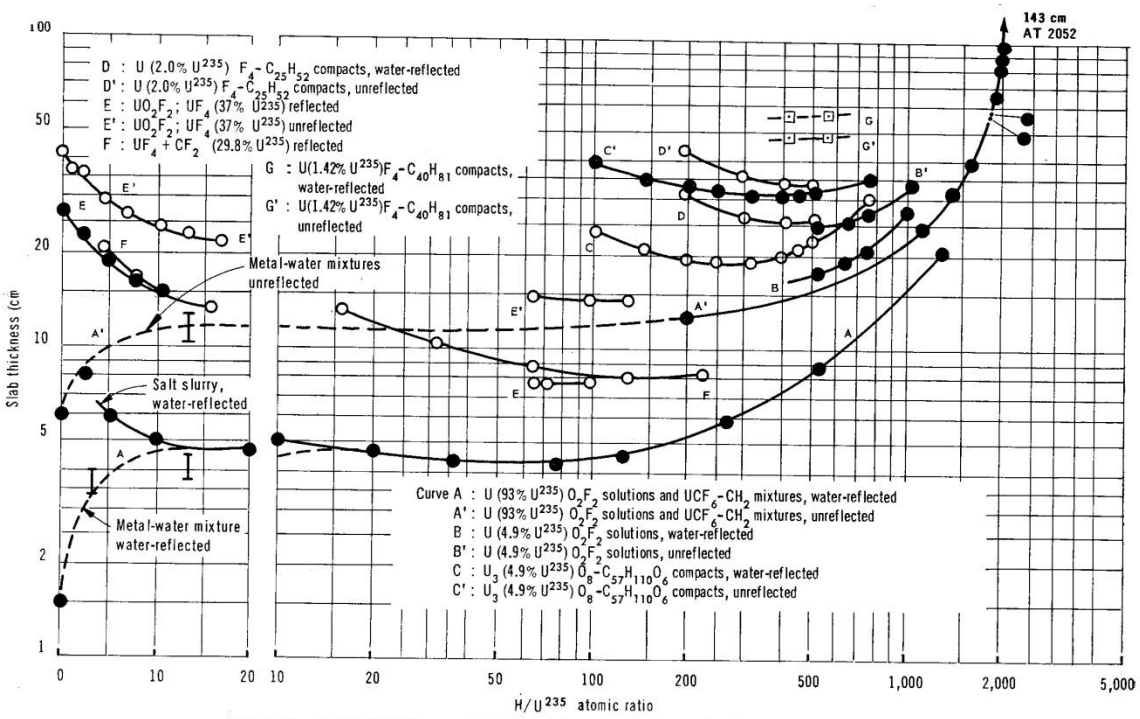

Figure 5. *Critical thickness of infinite slabs as a function of H/U 235 atomic ratio.*

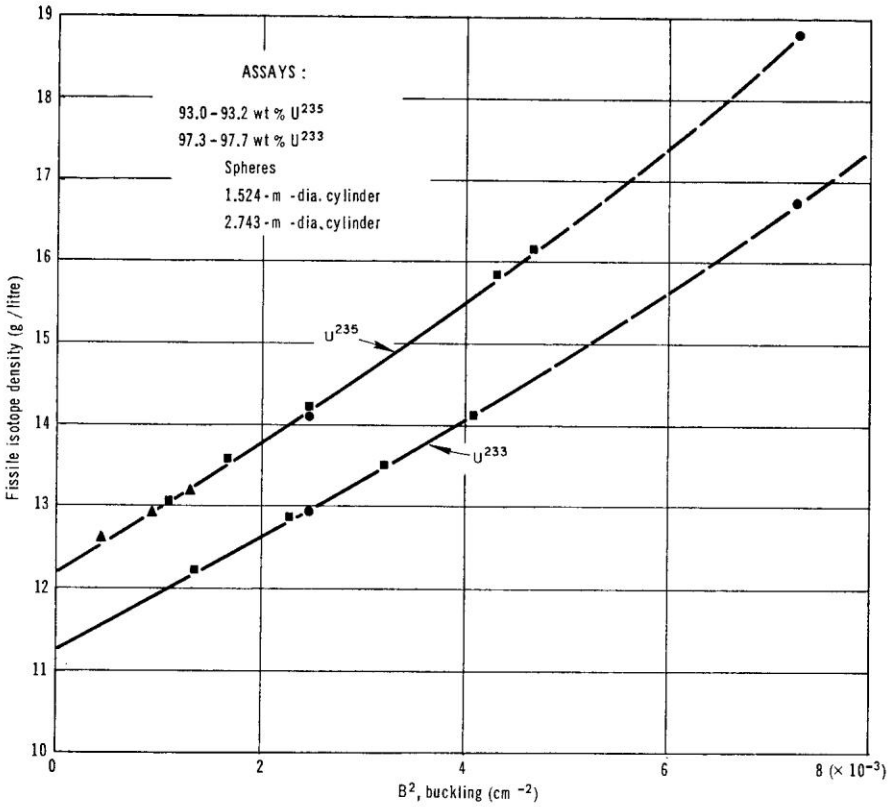

Figure 6. *Fissile isotope densities as a function of buckling in critical aqueous solutions of the nitrates of U 233 and U 235.*

TABLE I. SPHERICAL CRITICAL MASS AND VOLUME OF WATER-MODERATED 44.6 PER CENT U 235-ENRICHED URANIUM

H/U 235	U 235 Concentration (*g/litre*)	Volume (*litre*)		Mass (*kg U 235*)	
		Unreflected	Water-Reflected	Unreflected	Water-Reflected
258	97.4	17.6	11.5	1.71	1.12
493	51.8	—	19.5	—	1.01
678	37.9	—	29.5	—	1.12

There have been moderately extensive measures of the critical dimensions of uranium materials in the 30-40 % U 235 enrichment range. A few data were obtained from uranium enriched to 29.8 % and to ~ 37 % in U 235. The first of these studies was made in 1946, has been published [9] and is included here only to supplement later results. The test material was a physical mixture having a composition UCF_6 formed into blocks. Moderator was added rather heterogeneously by interspersed plastic blocks. A correction for the effect of this heterogeneity was made empirically in the preparation

of curve F of Figures 2 to 5. The inaccuracy of these results may be somewhat large because of this heterogeneity.

More recent experiments have used uranium enriched to 37.5 % in UCF_6 plastic compacts, and to 37.1 % in UO_2F_2 solutions. The former has established the critical dimensions of the unmoderated salt, neglecting the small effect of the carbon, which appear on curves E and E′ of Figures 2 to 5. Here too, moderation was introduced up to H/U 235 = 17 by adding hydrogenous plastic. The results are, therefore, less certain than most and should be considered indicative only.

A comprehensive series of critical experiments, performed at 15°-17° C, in partly-reflected and unreflected cylinders with UO_2F_2 solutions of 30.3 % U 235 isotopic content have been reported from the United Kingdom by Smith and his colleagues [10]. The dimensions of representative unreflected cylinders, 40.6 and 30.5 cm in diameter, have been transformed to spheres using 5.0 and 2.2 cm as the reflected and unreflected extrapolation distances, respectively. No corrections have been applied for the vessel structure or for the temperature. The results appear in Table II.

TABLE II. SPHERICAL CRITICAL MASS AND VOLUME OF WATER-MODERATED 30.3 PER CENT U 235-ENRICHED URANIUM SOLUTIONS

H/U 235	U 235 Concentration *(g/litre)*	Reflected		Unreflected	
		Volume *(litre)*	Mass *(kg U225)*	Volume *(litre)*	Mass *(kg U235)*
76.7	288.0	11.3	3.26	19.5	5.62
106	220.0	11.6	2.54	20.0	4.38
167	146.0	11.6	1.70	20.0	2.93
257	97.8	13.0	1.28	22.1	2.16
378	67.5	16.1	1.08	26.3	1.77
439	58.4	17.1	1.00	27.7	1.62
657	39.4	27.8	1.10	42.1	1.66
815	31.7	38.1	1.24	55.5	1.76

The agreement between the above unreflected results and those at comparable enrichment plotted on Figures 2 and 3 is reasonably good considering the difference in the temperatures of measurement. The reflected-experiment results compare less favourably due, probably, to an improper selection of extrapolation distances. These UK results are believed to be superior to those discussed above because the arrays of 29.8 % enriched uranium were severely latticed in this moderation range and the measurements with 37.5 % enriched solutions are too sparse for good transformation to spherical geometry.

A reported measurement made at a U 235 enrichment of 14.7 %, an early Los Alamos water boiler [11], yielded a reflected critical mass and volume of about 1.2 kg U 235 in 14.8 litres. The mass was determined from an extrapolation of neutron multiplication with a maximum loading of 0.7 kg in the sphere.

Less than 5 % *enrichment.* Figures 2 and 3 also record spherical masses and volumes of U 235 in uranium enriched to 4.9 % from experiments per-

formed with two test materials. In one set the uranium-containing material was U_3O_8 and the hydrogen was introduced by the addition of an organic substance, glycerol tristearate, which has a hydrogen density and nuclear properties not strikingly different from those of water. The two powders were well blended and packed in aluminum containers which were, in turn, stacked to critical in parallelepipedal arrays of various over-all dimensions both with and without water reflector. The procedure was repeated for each of a number of blends differing in hydrogen content and the results are shown as curves C and C′. The second series of experiments at this enrichment, over a more narrow range of moderation, were performed with aqueous solutions of UO_2F_2. At this enrichment the solubility of UO_2F_2 in water establishes a minimum H/U 235 ratio of about 500; the upper H/U 235 limit of these experiments was about 1,000. It may be noted in Figure 1 that the U 235 concentration in the solution at a particular moderation is appreciably greater than that in the powder pack at the same moderation. The critical dimensions of spherical geometries are shown in curves B and B′ to be significantly lower than those of the less dense U_3O_8 compacts. An essentially spherical volume of the solution was made critical at H/U 235 = 1,099 requiring 3.77 kg U 235 in 170.5 litres. The dimensions of infinitely long cylinders and slabs of infinite area are given in Figures 4 and 5.

A summary of the results to date from experiments [12] with 2.00 % U 235 enriched uranium are shown as curves D and D′ of Figures 2, 3, 4 and 5. For these experiments a well homogenized mixture of UF_4 in paraffin ($C_{25}H_{52}$) was fabricated into blocks which could be arranged in closely stacked arrays. The paraffin mixture was necessary to achieve desirable values of the moderation. The reflector was paraffin. This work is at present in progress, with increasing hydrogen content, and it is impossible, at this time, to state with certainty that the minimum critical mass has been established although such a minimum is expected at an H/U 235 of about 500, the composition of the material in the most recent experiments. It is believed that the moderation range thus far studied does include the minimum critical volume.

Arrays of water-reflected UF_4-paraffin compacts containing 1.42 % U 235-enriched uranium have been studied at Dounreay [13], and the results at H/U 235 ratios of 418 and 562 are shown as curves G and G′ in Figures 2 to 5. The minimum critical mass and minimum critical volume for uranium of this enrichment occur within or very close to this concentration range.

The minimum U 235 isotopic enrichment which can be made critical is of interest in nuclear safety practices. Consider first homogeneous mixtures of uranium and hydrogenous materials. Some experiments [14] done at the Hanford Laboratories utilizing the Physical Constants Testing Reactor have yielded 1.02 $\pm$ 0.02 % U 235 as this limiting enrichment. The value is important in that it permits unrestricted processing of uranium of this enrichment in solutions and slurries, provided, of course, allowance is made for uncertainties in the enrichment itself and for the possibility of heterogeneity occurring. That this limiting enrichment does not apply to lattices of uranium in a moderator is strongly emphasized.

The corresponding limiting critical value for uranium metal completely unmoderated has been determined at Los Alamos [4] to be between 5 and 6 % U 235 enrichment. These experiments also describe the variation of the critical mass of unmoderated uranium metal as a function of enrichment.

Heavy water solutions : a special case. Although problems of nuclear safety involving D_2O are encountered infrequently, they being primarily associated with processing certain homogeneous reactor fuels, it is appropriate to refer to experiments which may provide guidance. Early experiments were done in Oak Ridge [15] and Los Alamos [16]. In the latter, the solutions were contained in D_2O-reflected spheres and in unreflected cylinders. The cylinder dimensions were transformed to unreflected spheres of dimensions ranging from 21.6 kg U 235 in 197.7 litres at D/U 235 = 230 to 4.2 kg/U 235 in 340.1 litres at D/U 235 = 2,081. The U 235 enrichment of the uranium was greater than 90 %.

More recent experiments [17] have been done with uranium of 45.5 % U 235 enrichment in concentrations ranging in D/U 235 ratio from 1939 to 6,722 (13.5 to 3.9 g U 235/litre). The solutions were in 61.0 cm diameter cylinders laterally reflected by a 46.5 cm thick layer of graphite. The minimum critical mass was 1.768 kg U 235 occurring at D/U 235 = 4,687. Extrapolation of the data indicated that the cylinder would be critical, infinitely long, at D/U 235 = 9,510.

Heterogeneous Materials

Heterogeneity of nuclear reactor components occurs in two principal arrangements. One of these, in which a neutron reflector surrounds a neutron multiplying medium, has already been considered and is exemplified by a cooling-water jacket encasing a process vessel or by the wall adjacent to an array of units in storage. In the other arrangement, the fissionable material is lumped or latticed in a moderator with the physical dimensions of the components of the unit cell of this complex large compared to the distances describing the behaviour of neutrons. The most outstanding examples of this kind of heterogeneity are, of course, nuclear reactors of natural uranium and graphite where, because of the spacing of the uranium metal rods in the lattice, the effective resonance absorption by U 238 is decreased and chain reactions can occur.

There is still another non-uniformity of components, a variation on either of the principal ones, in which the chemical concentration of the multiplying medium varies more or less uniformly throughout the volume. Although brief mention will be made of experimental measures of this effect later in this Section, the first consideration is of lumped uranium in graphite or water.

Natural uranium. That natural uranium homogenized in graphite or water cannot be made critical has been established. The consequences, however, of optimizing the size of rods or slugs of natural uranium and their spacing in water are not known as definitively. Indeed several early experiments [18] indicated that such an arrangement can be made critical, albeit with only slight excess reactivity. The problem has been studied again recently at Hanford [19] in a series of exponential experiments with 2.35 cm diameter rods about 1 m long. Interpolation of the data from assemblies of three lattice spacings does give a positive value of the buckling, a requirement for a critical system, although the bounds of accuracy of the results leave some uncertainty in the possibility of establishing a chain reaction. Granted that natural uranium can be arranged in water as a critical lattice, the topic is still of academic interest to problems of nuclear safety, since the quantity required

is large and the probability of accidentally achieving the required distribution is small.

U 235-*enriched uranium.* Many experiments have been performed with lattices of uranium rods enriched in U 235 up to about 3 %. Much of the early work was reported at the Second Geneva Conference [20]. Many exponential and neutron multiplication (approach to critical) experiments have been performed more recently at the Hanford Laboratories with material of U 235 enrichment ranging from 0.925 to 3.063 % U 235 and the rod diameters from 0.45 to 4.22 cm. In other experiments the rods were cylindrical annuli. In still another variation, the neutron reflecting liquid was a solution of a uranium salt. The data from several hundred such experiments together with a bibliography of the detailed reports has been compiled by Lloyd [19]. Only a few typical results are included here.

One set of neutron multiplication experiments with 3.063 % U 235-enriched uranium utilized rods of four diameters, 2.34, 1.52, 0.76, and 0.45 cm and of various lengths. A series of measurements at several lattice spacings (water-to-uranium volume ratios) established the minimum critical mass and the maximum buckling for each size rod. The results, summarized in Figure 7, are useful in establishing mass and volume limits for processing metal of this enrichment.

Similar measures are at present underway with uranium rods of 2.00 % U 235 enrichment latticed in water. The first results [21], from rods 2.35 cm in diameter, show a minimum (spherical) critical mass of 6.3 kg U 235, occurring at a water-to-uranium volume ratio of 2.8, and a maximum buckling of about 11,000 10^{-6} cm^{-2} at an H_2O/U volume ratio of 2.2. In more recent unpublished experiments by Lloyd and Clayton [22] with 1.53 cm diameter rods of the same enrichment, the minimum critical mass was found to be 4.64 kg U 235 at an H_2O/U volume ratio of 3.8. The maximum buckling is 11,200 10^{-6} cm^{-2} at an H_2O/U volume ratio of 2.9.

An interesting exponential experiment has been performed at Hanford by Powell [22] with lattices of uranium dioxide enriched in U 235 to 2.6 %. The oxide was made into annular cylindrical pellets, 1,791 cm outside diameter and 0.820 cm inside diameter, having a density of 10.43 g cm^3. The pellets were assembled into rods, by mounting them in plastic tubes, in such a way that the central cavity could be empty or could be filled with water. The results from three lattice spacings in water are given in Table III.

The results of this experiment are typical of those required for establishing safe procedures for reprocessing some power reactor fuels.

In another experiment [19] with rods of 1.007 % U 235, 2.34 cm diameter and 112 cm length, the neutron moderating and reflecting liquid was a solution of boric acid (H_3BO_3). The variables were the lattice spacing and the boron concentration. From a series of measurements it is concluded that an H_3BO_3 concentration of about 4 g/litre will prohibit a nuclear chain reaction in arrays of these materials if the lattice spacing of the rods is in the range 3.56 to 4.06 cm.

Probably the best summary of experimental minimum critical masses and minimum critical volumes of slightly enriched uranium latticed in water is that of Clayton [23]. The values are shown by the first curve on the left of both Figures 8 and 9. The U 235 content of natural uranium is assumed as the asymptote.

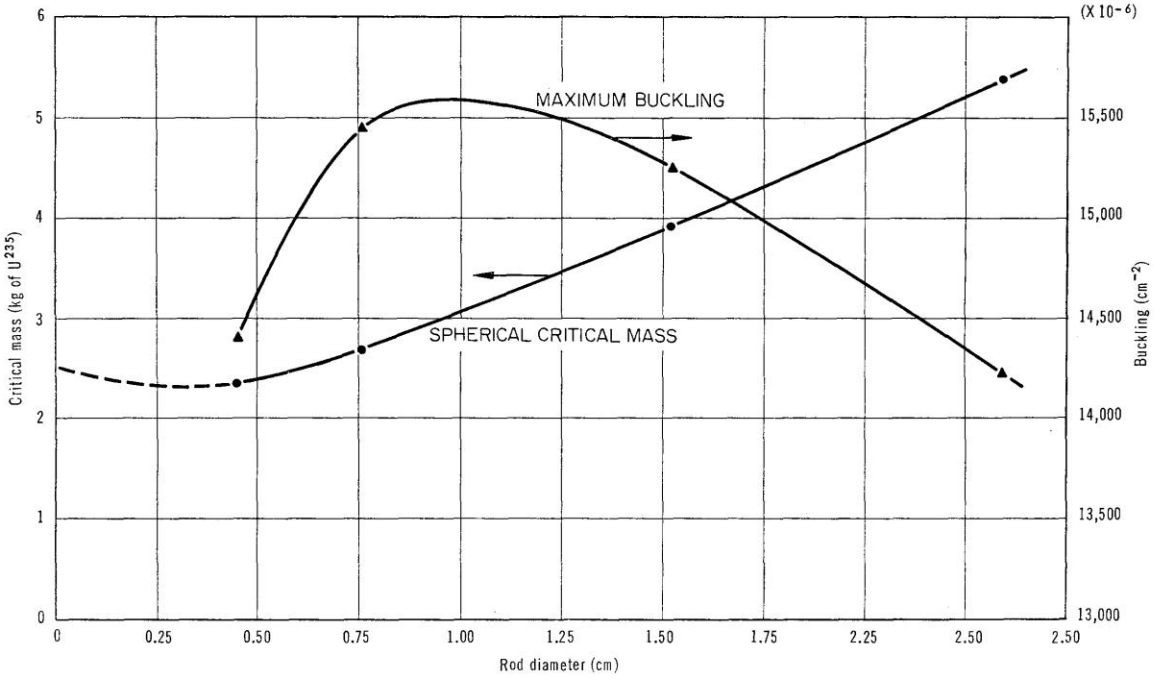

Figure 7. *Maximum buckling and minimum critical mass as a function of rod diameter, for lattices of 3.06 wt % U 235-enriched uranium in water.*

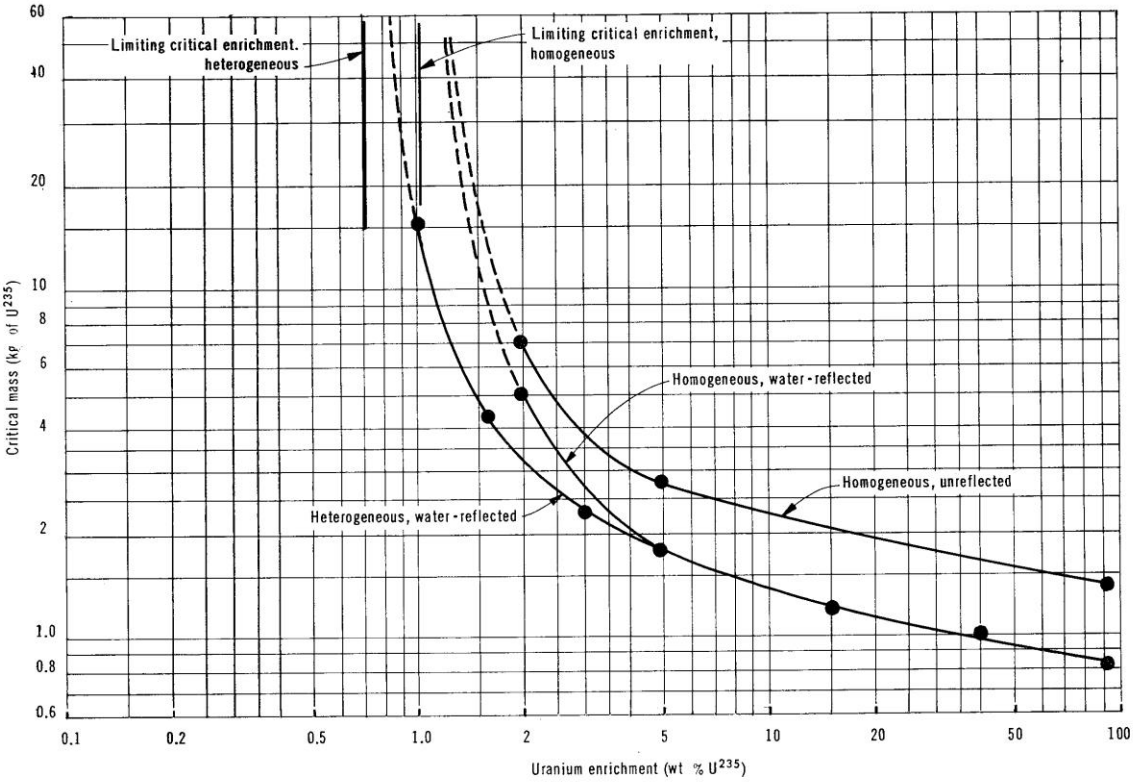

Figure 8. *Minimum critical mass as a function of U 235 enrichment.*

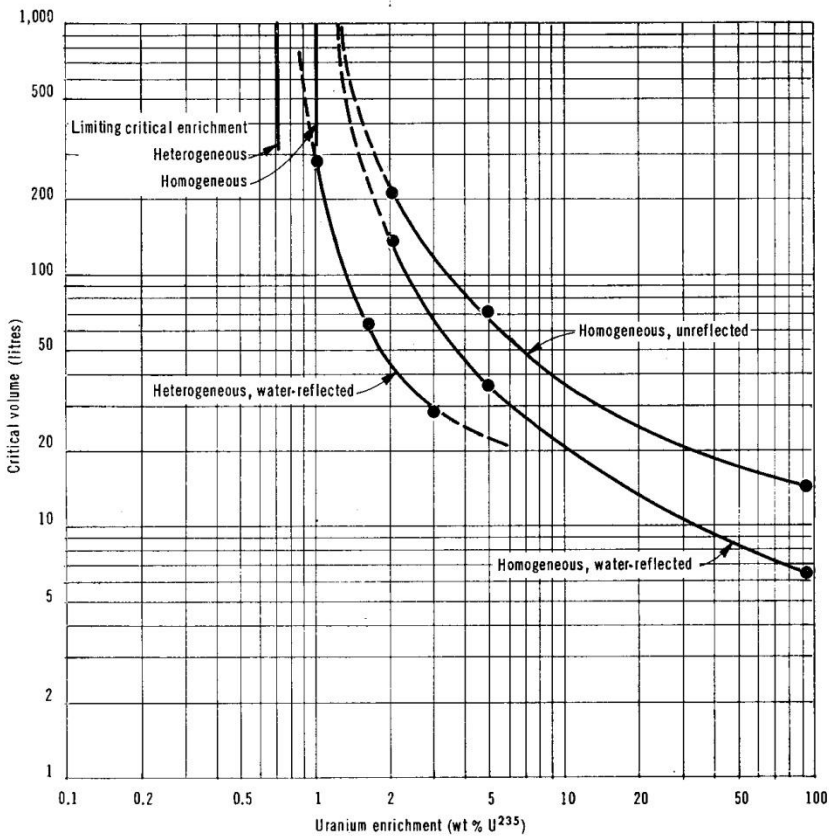

Figure 9. *Minimum critical volume, hydrogen-moderated, as a function of U 235 enrichment.*

TABLE III. BUCKLING AND SPHERICAL CRITICAL MASS OF 2.6 % U 235-ENRICHED URANIUM OXIDE RODS LATTICED IN WATER

Outside diameter of rod: 1.791 cm
Inside diameter of rod: 0.820 cm

Lattice Spacing (*cm*)	H_2O/UO_2 Volume Ratio	Material Buckling 10^{-6} cm^{-2}	Spherical Critical Mass (*kg U 235*)
Central Cavity Water-Filled			
2.540	1.808	10,131	4.79
3.048	3.043	10,421	3.40
3.556	4.504	8,559	3.72
Central Cavity Empty			
2.540	1.542	9,644	5.12
3.048	2.778	10,084	3.59
3.556	4.237	8,467	3.79

The effect of latticing 93 % U 235-enriched uranium is an increase in critical mass. The results of the Los Alamos experiments are well known [24]

and will only be summarized here. The mass and diameter of a water-reflected, unmoderated sphere are about 22.5 kg U 235 and 13.5 cm, respectively. As this piece is successively fractured in water, first into two hemispheres for example, the critical mass, based on indirect experiments, is believed to increase by perhaps as much as 10 %. Of the competing effects — the decrease in density and the increase in moderation — the former dominates. A distribution of the sphere into smaller pieces, of course, decreases the critical mass. The system approaches, as a limit, a suspension of particulates, metal chips or thin machine turnings in water, for example, which behaves like a solution of the same hydrogen and uranium composition.

Discussion of Heterogeneous and Homogeneous Systems

From what has been said it is obvious that the critical quantity of U 235 is strongly dependent upon the distribution of uranium within the water moderator and, further, that the U 235 enrichment determines whether the effect is an increase or decrease in the critical mass. It has been shown that at a U 235 enrichment of 3 % the critical mass is decreased by latticing the uranium in water. The maximum enrichment at which this decrease in mass occurs is problematic but various estimates place the value in the neighbourhood of 5 %. Figures 8 and 9 record the variation of the minimum critical mass and minimum critical (spherical) volume as a function of U 235 enrichment with values for both heterogeneous and homogeneous systems shown at the lower enrichments. Although the curves describing the variation in mass (Figure 8) are not inconsistent with the point of divergence — 5 % enrichment, mentioned above — the curves of Figure 9 do not clearly describe a corresponding value for critical volumes.

Estimates can be made of the dimensions of critical cylinders infinitely long and critical slabs infinite in area by equating bucklings.

Brief comment will now be made on the effect of non-uniform distribution of fissionable material within a moderator, a mild form of inhomogeneity. As an example consider a volume of a solution in which the chemical concentration varies from point to point. It was, indeed, with this type of inhomogeneity that an exemplary experiment was performed [25]. The test material was a uranium solution, enriched to about 93 % in U 235, contained in a cylinder (more exactly, in a series of concentric cylindrical annuli surrounding a central cylinder) with its concentration adjusted to give a stepwise decrease in the mass of U 235 per unit volume along the radius, the greater concentration being in the central region. That this distribution yields a lower critical mass than would a single volume of solution having the same shape and dimensions in which the concentration is uniform may be seen from the following qualitative argument. If, in such a uniform critical volume, a small mass of U 235 is transferred from the periphery to the centre, that is, to a region of greater importance, the reactivity will increase and a reduction in the U 235 content of the vessel must be made to return the system to critical, thereby lowering the critical mass. The analytical prediction [26] that continuing this mass transfer until the critical mass is minimized would yield a uniform radial distribution of the thermal neutron flux was confirmed by experiment. The reduction in the critical mass was about 9 % in cylindrical geometry and would be greater were the gradient to exist in a sphere. This interesting observation serves

as an introduction to another experiment which, in turn, is cause for caution in processes in which precipitation occurs. This second experiment [27] may be briefly described as follows. The test material was finely-divided UO_3, of 93 % U 235 enrichment, normally suspended in water by the action of a conventional mechanical stirrer. As long as the suspension was maintained the critical properties of the slurry were only slightly different from those of a solution of the same relative water content. As this well-stirred pattern was disturbed by interrupting the agitation, reactivity excursions of the order of 10 % occurred. A similar behaviour was sometimes observed as agitation of the settled slurry was begun. A possible explanation of the reactivity changes is a short-lived redistribution of the uranium during the transition period resulting in a change in critical parameters. The effect may be due in part to a distribution of delayed neutron precursors to regions of greater importance. Although the experiments were not completely definitive and were never carried to a satisfactory conclusion, the observations may imply the need for an additional safety factor in the controlling variable of certain operations. It is interesting to note some qualitative agreement exists between those observations and those of de Jong [28].

Miscellaneous Experiments

The results from many experiments performed primarily in the study of particular safety problems can be used for general guidance in the field. A few of these experiments, some as yet unpublished, will be described briefly.

Neutron absorbers. The use of neutron absorbers to enhance nuclear safety is the topic of subsequent papers of this series and at least one [29] of them reviews experiments supporting the practice. One typical example, reported by Bidinger, Schuske and Smith [30] is, however, included here. The material under test in this experiment was a borosilicate glass formed into Raschig rings 3.8 cm long and 3.8 cm in diameter. The wall thickness was 0.20 cm. These glass rings contained 12.5 % B_2O_3. They were packed in a 107 cm diameter tank, occupying 17.8 % of the volume. The lateral surface of the tank was reflected by concrete. The experiment consisted of adding an aqueous solution of ~ 90 % U 235-enriched $UO_2(NO_3)_2$ to the tank and observing the resulting neutron multiplication. Results indicate that the tank could be filled to an infinite height with a solution containing 325 g U 235/litre maximum, without being critical. Assuming that such a 107 cm diameter cylinder, infinitely long, is critical, transformation of its dimensions gives 70 and 67.5 cm respectively as the critical sphere radius and the infinite slab thickness.

Cylindrical annuli. The safe capacity of an annular cylindrical vessel has been shown to be significantly greater, per unit length, than that of a conventional cylinder provided the core of the annulus is a hydrogenous material surrounded by a neutron absorber. The hydrogenous core reduces the energy of the neutrons, which tend to link opposite sides of the annulus, so they may be captured by the absorber. The advantage of this geometry is illustrated by the following experiments [31]. The critical dimensions of cylindrical annuli of a solution of uranium, enriched to 93.2 % in U 235, were measured over a range of both annular thickness and outside diameter. The H/U 235 ratio of the solution was 50.4, near that of minimum critical volume. The curves of Figure 10 show the variation in critical height as

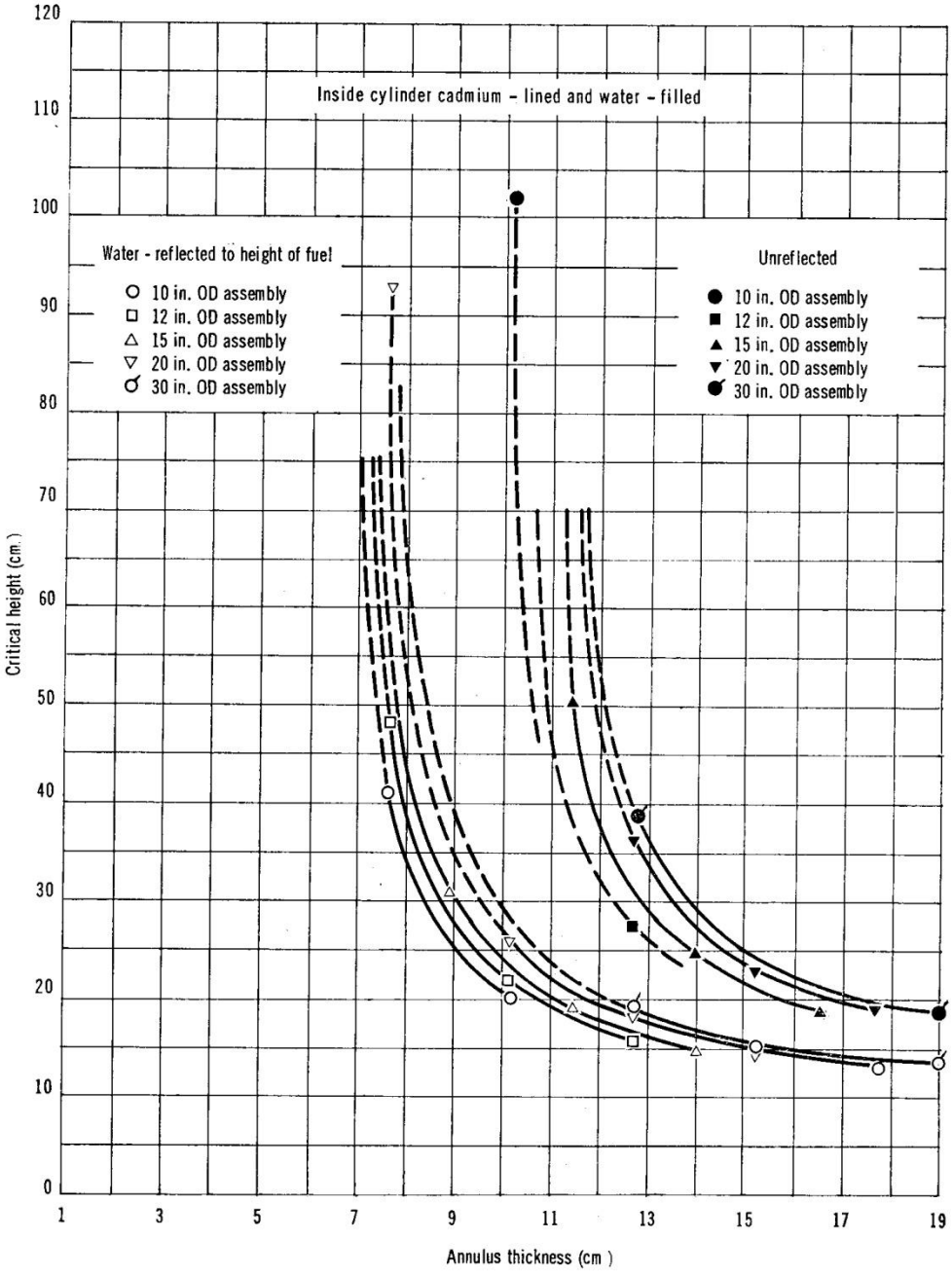

Figure 10. *Critical heights of cylindrical annuli containing aqueous solutions of 93.2% U 235-enriched uranyl fluoride, as a function of the thickness of the annuli. H/U 235 atomic ratio = 50.4.*

the thickness was varied, for several sized vessels. The core of the annulus was lined with 0.05 cm thick cadmium sheet and was filled with water. These data show that an annulus of about 6 cm width will be subcritical when infinitely long and reflected by water. Additional experiments with a less concentrated solution, H/U 235 = 309, required a thicker annulus for criticality. The curves obtained with this more dilute solution would be displaced about 1.2 cm to the right of those shown in Figure 10. It is expected that larger annuli would also be required for solutions more concentrated than those of H/U 235 = 50.4.

Enriched uranium metal submerged in enriched uranium solution. Some work of interest in the design of leaching baths, where both the material being leached and the leaching solution contained enriched uranium, has been reported [32]. This experiment determined the critical thickness of a 12.7 × 20.4 cm slab of ~90 % U 235-enriched uranium metal, surrounded by an aqueous solution of ~90 % U 235-enriched uranyl nitrate, as a function of the chemical concentration of the solution. The solution was in an unreflected vertical cylinder 24 cm in diameter and 40.7 cm high. The slab was centered in the solution with its plane vertical. The results are tabulated:

Solution Concentration (*g U 235/litre*)	Critical Slab Thickness (*cm*)
0	6.9
47.2	4.1
92.6	3.2
156.6	2.5

Urania-thoria pellets. The reprocessing of some reactor fuels entails the removal of high-density-oxide pellets from the fuel elements prior to dissolution, resulting in random arrays of the pellets in a dissolver. As a guide to the design of a dissolver, several criticality experiments have been performed with unclad pellets, consisting of a mixture of ThO_2 and UO_2, randomly packed in cylinders [33]. Water was added to the cylinders to serve as a neutron moderator, and the cylinders were surrounded by a water reflector for all experiments.

The pellets were nominally 0.66 cm in diameter and 1.32 cm long. Their density was 8.3 g/cm³, with a uranium content of 5.94 % and a U 235 content of 5.54 %. A random array resulted in an average concentration of 0.29 g U 235/cm³ of vessel volume. When the pellets were covered with water, the resulting H/U 235 atomic ratio was 34, severely less than optimum moderation. Approximately 900 kg of pellet material (50 kg U 235) was available for the experiments. The results of measurements in cylinders of three diameters are shown in the following table:

Cylinder Material	Cylinder Diameter (*cm*)	Pellet Depth (*cm*)	Pellet Mass (*kg U 235*)	*k*, Multiplication Constant*
Stainless steel...	38.1	153.0	50.0	$\ll 1$
Aluminium	44.2	58.2	25.6	~ 1
Aluminium	50.8	45.7	26.6	1

* The value of *k* is unity in a critical system.

The critical dimensions of random assemblies of these pellets are extremely sensitive to the degree of moderation.

Pool-type reactor fuel elements. The results of a study of pool-type reactor fuel elements were reported some time ago [34]. It is believed, however, that their repetition may assist in the problems of storage and shipment of fuel elements of this type. In general, these elements were constructed of plates containing a uranium-aluminium alloy supported in a frame or box about 7.5 cm^2 and 50-60 cm long. The uranium was enriched to 93.2 % in U 235. Most of the elements available for these experiments contained 168 g U 235, a few had 200 g while still others were loaded with 140 g. These elements were of the type used in the Oak Ridge Research Reactor and in the Bulk Shielding Reactor. The following observations were made in the experiments.

The number of elements in a critical array submerged in water is a strong function of the spacing between the elements. With this spacing optimum, about 1.2 cm between adjacent fuel sections, the number of elements required for criticality contained approximately 2.6 kg U 235, a quantity essentially independent of the mass per element within the above range. Increasing this separation to 3.2 cm increased the critical number to 31, about 5.2 kg U 235, while an array of an infinite number at a 5.7 cm separation would probably not be critical. Each array was a single element high.

No appreciable source neutron multiplication was produced by a submerged, close-packed, 11- × 12-element array in which adjacent rows of elements, in one direction, were separated by a 0.05 cm thick cadmium sheet. The average loading of the 132 elements was 160 g U 235 each. There was no appreciable multiplication when both the cadmium and the water were removed and the array was surrounded by a 30 cm thick paraffin reflector.

An infinite number of elements arranged in a plane in water will be subcritical. Two rows of 26 elements each (a total of twenty-four 200 g elements and 28 of 168 g) submerged at optimum spacing were subcritical.

Beryllium and graphite-moderated U 235*-enriched uranium.* The following information may provide guidance in nuclear safety problems arising in the production and processing of reactor fuel elements containing enriched uranium and special moderating materials such as beryllium and graphite.

Experiments with lattices of 10 % U 235-enriched uranium in beryllium to which graphite and water could be added have been reported by Krasin *et al* [35].

The critical dimensions of BeO-moderated assemblies of 93 %-enriched uranium foils have been measured at the Lawrence Radiation Laboratory [36]. Estimates were made of the dimensions of unreflected homogeneous spheres of BeO and uranium. These values ranged from 16.6 kg U 235 in a 66.2 cm diameter sphere at a Be/U 235 ratio of 247 to 2.0 kg U 235 in a 103 cm diameter sphere at a Be/U 235 ratio of 7,660.

A number of critical assemblies of 93 % U 235-enriched uranium moderated by graphite have been reported by Reynolds [37]. In these experiments the uranium foils were only 0.002 to 0.004 cm thick and permitted some estimation of the dimensions of homogeneous uranium-graphite systems. The C/U 235 atomic ratio ranged from the order of 10^2 to 10^4. The small-

est mass measured was 9.77 kg U 235 in a parallelepiped, approximately 1.3 × 1.5 × 1.5 m, at a C/U 235 ratio of 10,500.

A recent experiment [38] at Los Alamos consisted of a near-cubical block of graphite and thin 93.3 % enriched uranium foils with properties similar to a homogeneous mixture having a C/U 235 ratio of 61.2. The dimensions of the block were 81.28 × 81.28 × 91.16 cm and it contained 324.75 kg U (302.99 kg U 235). The composition was 1.573 g graphite and 0.539 g uranium/cm³. Reduction to spherical dimensions gives a critical mass of 217.0 kg U 235 in a 93.76 cm diameter sphere.

U 235-*enriched uranium with non-hydrogenous diluents.* Measurements have recently been made at Los Alamos [38] of the critical sizes of unreflected 93.3 % enriched uranium metal diluted with various metals. The critical assemblies were constructed as 38.2 cm diameter cylinders of adjustable height, being formed of alternating circular plates of uranium and of the diluent. The results have been reduced to spherical dimensions by equating bucklings assuming an extrapolation distance of 2 cm. Although natural uranium, tungsten, tantalum, copper, zirconium, aluminium and carbon were the diluents investigated in the experiments, only the data describing the effect of the last two are given in Table IV as representative of the programme.

TABLE IV. UNREFLECTED SPHERICAL CRITICAL MASSES OF 93 % U 235-ENRICHED URANIUM DILUTED WITH VARIOUS MATERIALS

Uranium component density..... 18.75 g/cm³
Uranium enrichment ..... 93.3 per cent U 235

Diluent:U Atomic Ratio	Composition		Uranium Volume (%)	Critical Sphere	
	Uranium (*g/cm³*)	Diluent (*g/cm³*)		Radius (*cm*)	Mass (*kg/U 235*)
0	17.98*	0	100	8.61	46.76*
Diluent: aluminium; density = 2.70 g/cm³					
0.339	14.18	0.55	78.7	10.36	64.08
0.678	11.82	0.92	64.9	12.10	84.18
1.024	10.02	1.18	55.1	13.77	105.47
1.359	8.77	1.37	48.0	15.28	125.56
1.691	7.82	1.52	42.6	16.85	149.34
Diluent: carbon; density = 1.65 g/cm³					
0.278	15.58	0.221	86.1	9.65	56.59
0.372	14.80	0.281	82.2	9.96	59.43
0.450	14.30	0.329	79.2	10.20	61.67
0.561	13.97	0.400	75.5	10.70	67.71
0.748	12.71	0.485	69.7	11.21	71.93
1.120	11.07	0.631	60.7	12.30	82.76
1.490	9.97	0.759	53.6	13.44	95.45
2.240	8.07	0.922	43.5	15.47	118.04
3.380	6.23	1.075	33.8	17.97	143.46
4.420	5.26	1.188	28.1	20.39	174.29

* The undiluted uranium density and critical mass of the assembly of a number of uranium plates are less and greater, respectively, than the corresponding values for a single sphere of uranium.

The effect of these nonhydrogenous homogeneous diluents on the critical mass of U 235 can be expressed by the relation $m_c = m_o(F)^{-n}$, where m_c and m_o are, respectively, the critical masses with and without the diluent, F is the volume fraction of the enriched uranium and n is the dilution exponent, a property of the diluent. The values of n listed below from these experiments supplement or revise those recently reported [39] for unreflected U 235-enriched uranium.

Arrays of subcritical units. Placement of individually subcritical units within an array is probably the most frequent operation for which there is the least supporting data. Early critical and neutron multiplication experiments describing arrays of cylindrical volumes of solution and spherical metal units have been reported [40] and are summarized in Reference [1]. The topic is being investigated at the present for the primary purpose of acquiring data to support analytical treatments. Preliminary results from two investigations at present under way at Oak Ridge are included in this Section.

Element	Dilution Exponent: n
Al	1.373
C	1.188
Cu	1.045
Ta	1.294
U 238	0.590
W	1.035
Zr	1.180

In one of these experiments by Mihalczo and Lynn [41], up to 125 slab-shaped pieces of 93.4 % U 235-enriched uranium were arranged with their centres in cubic lattices of over-all parallelepipedal shape. The dimensions of a slab were 2.5 × 20.3 × 25.4 cm and each contained 22.9 kg U 235. The lattices were surrounded by a 2.5 cm thick layer of methacrylate plastic (Plexiglas). Plastic also separated the units in some tests. The observed source neutron multiplication was extrapolated to the estimated critical number of units in each of several arrangements shown in Table V. Although the extrapolations were quite long, from 125 units to more than 300 in one case, the estimates given are believed to be conservative. Included in the table is the ratio of the volume occupied by a unit to the volume of its associated cell in the lattice at each spacing. This quantity is a measure of the metal density within the array.

The second experiment utilized up to 100 cylindrical units of aqueous solution which were arranged in critical configurations [42]. Each unit consisted of an aqueous solution of $UO_2(NO_3)_2$ having a concentration of 410 g U/litre and a specific gravity of 1.55. The U 235 enrichment was 92.6 %. The solution was contained in 13-litre Polyethylene $(CH_2)_n$ bottles having an outside diameter of 13.7 cm and a wall thickness of about 1 cm. The variables studied included the volume content of the bottles and their spacing and geometric pattern within an array. The bottles were arranged with their axes vertical and their bases in a square or triangular pattern. Most experiments were performed with a single tier of bottles with all bases in a

TABLE V. ESTIMATED CRITICAL NUMBER OF 22.9 kg U 235 METAL SLABS LATTICED WITHIN A 2.5 cm-THICK PLASTIC REFLECTOR

Centre-to-Centre Separation of Slabs (*cm*)	Vol. of Unit / Vol. of Cell	Critical Number of Units	
		Unmoderated	Separated by 2.5 cm-thick Plastic Moderator
38.1	0.023	350 ± 30	75^{+5}_{-2}
30.5	0.046	185 ± 10	36^{+5}_{-2}
27.9	0.060	145 ± 5	$27^{+0.5}_{-0}$

horizontal plane, although in one series two such tiers were arranged, one above the other, with the solution separated 14.2 cm vertically.

The number of units in a critical array as a function of their surface-to-surface separation is shown in Figure 11 for each of three different solution volumes. The results from the experiments with the double tier are plotted with ordinates equal to the number of units in one of the tiers, that is, one-half of the total number at a particular spacing. In this manner a double tier is shown to have characteristics similar to a single tier of units but with extended solution height.

PRESENTATION OF DATA: URANIUM 233

The quantity of data available as bases for safe practices in U 233 processes is much less than that for U 235. Critical experiments have been performed in a few laboratories with aqueous solutions and, more recently, with metallic U 233. Generalization of the data is published in Reference [1].

Aqueous Solutions

From the early work of Fox and associates [43] with solutions at concentrations in the 50-500 g/litre range it was possible to establish minimum critical quantities as 590 g U 233 and 3.3 litres. These measurements permit an estimate of 11.2 cm as the minimum diameter of an infinite cylinder and 3.0 cm as the thickness of the corresponding infinite slab.

Clarke and his co-workers [8] have reported experiments with almost isotopically pure U 233 in aqueous UO_2F_2 solutions of intermediate concentration. They were performed in a 30.5 cm diameter steel cylinder reflected on its lateral surface by water. The minimum critical mass observed in the concentration range studied, H/U 233 = 250 to 860, was 917 g in a cylinder 23.8 cm high. This minimum occurred at a concentration of 52.9 g U 233/litre corresponding to H/U 233 = 483.

Still more recent results have been reported by Fox [44] from experiments with dilute solutions of $UO_2(NO_3)_2$ in unreflected cylindrical geometry. These results have been reduced to spherical dimensions by equating buck-

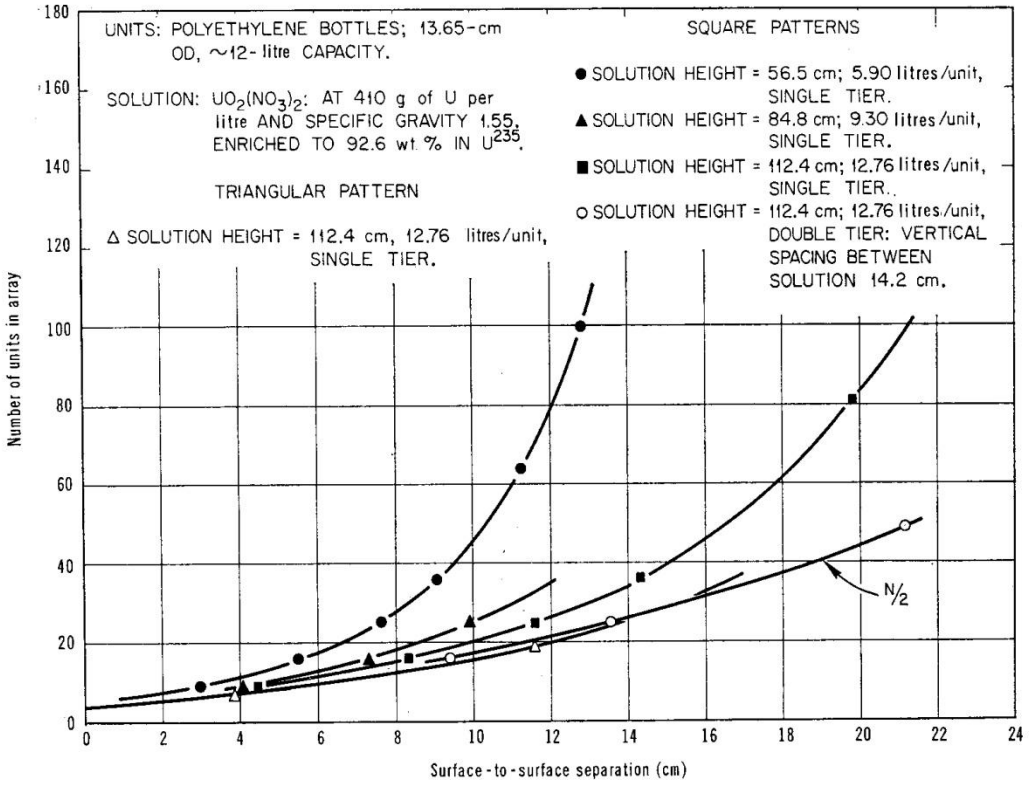

Figure 11. *Unreflected and unmoderated critical arrays of units containing aqueous solution of uranyl nitrate.*

lings, using an assumed extrapolation distance of 3.3 cm, a value which may be too high by about 1 cm. The error introduced, however, in these large volumes is small. The results are summarized in Table VI:

TABLE VI. SPHERICAL CRITICAL DIMENSIONS OF UNREFLECTED SOLUTIONS OF U 233 $O_2(NO_3)_2$

U 233 enrichment .... 97.4 %
Solution container.... 154.7 cm dia. stainless steel cylinder

Concentration		Spherical Volume (*litres*)	Spherical Mass (*kg U 233*)
H/U 233 Atomic Ratio	U 233 (*g/litre*)		
1,819	14.12	402.4	5.68
1,900	13.53	595.8	8.06
1,996	12.88	1,012.8	13.04
2,106	12.21	2,334.9	28.50

A limiting critical chemical concentration of 11.25 g U 233/litre has been obtained in Figure 6 from a plot of critical concentration as a function of the buckling. The correction for the neutron absorption by the impurities of the solution, including the nitrogen, is negligible.

Metal

Data from direct measures on U 233 metal are limited to those made recently at Los Alamos [45] which gives 15.8 kg as the critical mass of an unreflected sphere. The isotopic purity of the sample was 98.15 % and its density was 18.25 g U 233/cm³. A slightly less pure sample, 97.94 % U 233, of the same density had a spherical critical mass of 5.4 kg when enclosed in a 19.7 cm thick natural uranium reflector.

PRESENTATION OF DATA: PLUTONIUM

The experimental results describing the critical parameters of plutonium with water moderation and reflection, the conditions in which it will be encountered in chemical processing, are extremely limited and those applicable to metallurgical operations, although somewhat more abundant, are difficult to generalize. The Pu 240 content of the plutonium varied among the samples, causing uncertainties in the interpretation of the results since its effect on critical dimensions is a function of moderation.

Aqueous Solutions

Critical experiments with aqueous solutions have been reported by Kreusi [46] and by Horton and McCullen [47] at the First Geneva Conference. Although the range of chemical concentration and the geometry studied were narrow, they did allow indication of some of the minimum critical parameters. The minimum critical mass and the minimum volume, in water-reflected aqueous solution, are 510 g and 4.5 litres, respectively. The minimal infinite cylinder diameter and infinite slab thickness are estimated to be 12.4 and 3.3 cm, respectively.

An interesting experiment reported by Bertrand [48] at the Second Geneva Conference, gives 257 g as the critical mass of hydrogen-moderated plutonium in a beryllium oxide-graphite reflected cylinder. The solution concentration was about 26 g plutonium/litre.

Metal

It is recalled that two metallurgical phases of plutonium exist and that their densities, and hence their critical dimensions, are different. Caution must, therefore, be exercised to assure correct application of the data.

The critical mass of an unreflected sphere of δ-phase plutonium, 15.66 g/cm³, 4.5 % Pu 240, has been reported by Jarvis [49] to be 16.28 kg. The estimated value for an unreflected sphere of α-phase plutonium [1], 19.5 g/cm³, is 10 kg. The corresponding values for a water-reflected sphere are 7.6 kg of δ-phase plutonium and 5.6 kg of α-phase [1].

The critical mass of δ-phase plutonium in cylinders reflected by water, graphite and other materials, recently reported by Hansen [50], may be applied

to particular practical problems. Of use in certain applications are the results describing the effects of a number of diluents added to δ-phase plutonium [51].

Studies of neutron reflection of plutonium by a number of metals have also been reported [52].

Plutonium moderated and reflected by plastic and graphite. A number of neutron multiplication experiments, reported by Schuske and his colleagues [53], define practical safety limits which can be applied to heterogeneous assemblies of plutonium, an hydrogenous plastic and graphite. It is inappropriate to transform the results of these experiments into a generalized form — the mass of spheres for example — because of the heterogeneity of the materials and uncertainties in the properties of the arrays. The data are valuable in their application to problems under conditions similar to those of the experiments.

Plutonium-aluminium alloy. The critical mass of plutonium in 1.28 cm diameter rods of a Pu-Al alloy, latticed in water, has been reported by Neeley [54]. The alloy contained 5 % Pu of which, in turn, 5 % was Pu 240. The minimum spherical critical mass, about 1.52 kg Pu, was observed at an H/Pu atomic ratio of approximately 650.

Concluding Remarks

Nuclear safety specifications are predicated primarily upon the results of critical neutron multiplication, sample replacement and exponential experiments from which, collectively, an impressive amount of information has been derived. This paper has attempted to review this information, in an endeavour to show what data are available, rather than to present it in

	Minimum Critical Quantities		
	U 235	U 233	Pu 239
Mass *(kg)*			
Solution	0.82	0.59	0.51
Metal	22.80	7.5	5.6* α-phase 7.6 δ-phase
Diameter of infinite cylinder *(cm)*			
Solution	13.7	11.2	12.4
Metal	7.8	4.8	4.3 α-phase 5.3 δ-phase
Thickness of infinite slab *(cm)*.			
Solution	4.3	3.0	3.3
Metal	1.5	0.7	0.6 α-phase 0.7 δ-phase
Solution volume *(litres)*	6.3	3.3	4.5
Chemical concentration of aqueous solution *g* (of isotope) *per litre*	12.1	11.2	9,371.0
U 235 enrichment, weight %			
Metal, unmoderated	~ 5	—	—
Homogeneous, Hydrogen-moderated	1.0	—	—

* Values are given for both α-phase plutonium, density 19.6 g/cm³ and for δ-phase, density 15.6 g/cm³.

handbook form. For this presentation some of the data have been generalized and have lost accuracy in the transformation.

A preponderance of the data has been derived from experiments with the U 235 isotope, mostly when present as greater than 90 % of the uranium. A sizable amount of information describes uranium enriched to less than 5 % in U 235, both latticed in water and homogeneously mixed with hydrogenous compounds.

The information describing Pu 239 and U 233 is limited to metallic units and aqueous solutions within a narrow range of chemical concentration.

The following summary is included for the information and guidance of those with limited fissionable-material inventory. The values presented are the best estimates of minimum critical quantities under conditions expected in chemical and metallurgical operations. The unit described is spherical, completely reflected by water and, in the case of solutions, optimumly moderated. The metal is unmoderated.

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Recognition is made of the efforts of all the people who have contributed to the many experiments necessary to a firm basis for nuclear safety. Omissions from this unpretentious survey are not intentional; it is hoped that those experiments included are properly credited.

REFERENCES

1. H.C. PAXTON, " Critical Data for Nuclear Safety Guidance ", *LAMS-2415* (1960) See also a series of papers on fast-neutron critical experiments edited by H.C. Paxton *Nucl. Sci. Eng. 8*, 523-719 (1960).
2. " Handbook No. 1 ", Health and Safety Branch, U.K.A.E.A.
3. D. CALLIHAN, W.J. OZEROFF, H.C. PAXTON and C.L. SCHUSKE, " Nuclear Safety Guide ", *TID-7016 (Rev. 1)* (1961).
4. H.C. PAXTON and G.A. GRAVES ," Critical Masses of Fissionable Metals as Basic Nuclear Safety Data ", *LA-1958 (Del)* (1956).
5. G.A LINENBERGER, J.D. ORNDOFF and H.C. PAXTON, " Enriched-Uranium Hydride Critical Assemblies ", *Nucl. Sci. Eng.*, 7,44 (1960).
6. J.T. MIHALCZO and J.J. LYNN, " Neutron Physics Annual Progress Report ", *ORNL-3016*, p. 73 (Sept. 1960).
7. R. GWIN and D.W. MAGNUSON, " Critical Experiments for Reactor Physics Studies ", *ORNL-CF 60-4-12* (1960).
8. W.G. CLARKE, C.C. HORTON and M.F. SMITH, " Critical Assemblies of Aqueous Uranyl Fluoride Solutions ", *AERE R/R-2051* (1956).
9. C.K. BECK, A.D. CALLIHAN and R.L. MURRAY, " Critical Mass Studies, Part II ", *K-126* (1946).
10. J.C. SMITH et al., " Criticality of 30 % Enriched Uranium Solutions in Cylindrical Geometry ", *DEG-Memo-663* (1960).
11. R.E. CARTER et al., " Water Tamper Measurements ", *LA-241* (1945).
12. See p. 71 of Reference 6.

13. J.G. WALFORD. Private communication,

14. H.E. HANDLER, " Nuclear Physics Research Quarterly Report ", *HW-54591*, p. 41 (Dec. 1957).

15. A.H. SNELL, " Criticality Studies on Enriched Uranium — Heavy Water Systems ", *Mon-P-454* (1947).

16. R.N. OLCOTT, " Homogeneous Heavy Water Moderated Critical Assemblies, Part I, Experimental ", *Nucl. Sci. Eng. 1*, 327 (1956).

17. J.R. HARRISON et al., " Critical Assemblies with Heavy Water Solutions of Uranyl Fluoride (HAZEL) ", *AERE R/R-2703* (1958),

18. G. BRANCH et al., " Water Lattice Experiments ", *CP-2842* (1945).

K. DOWNES, " Buckling of a Natural Uranium Light Water Moderated Lattice ", *BNL-2015* (1954).

19. R.C. LLOYD, " Summary Listing of Subcritical Measurements of Heterogeneous Water-Uranium Lattices made at Hanford ", *HW-65552* (1960).

20. H. KOUTS, R. SHER, J.R. BROWN, D. KLEIN, S. STEIN, R. L. HELLENS, H. ARNOLD, R. M. BALL and P. W. DAVISON, " Physics of Slightly Enriched, Normal Water Lattices (Theory and Experiment) " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, 1958, 12*, 446.

21. R.C. LLOYD, E.D. CLAYTON and B.L. JONES, " Nuclear Physics Research Quarterly Report ", *HW-66215*, p. 30 (June 1960).

22. " Nuclear Physics Research Quarterly Report ", HW-67219 (Sept. 1960),

23. E.D. CLAYTON, " Nuclearly Safe Mass Limits, Volume Limits, Infinite Cylinder Diameters and Slab Thicknesses for Slightly-Enriched Uranium Rods in Light Water ", *HW-65328* (1960).

24. J.C. HOOGTERP, G.E. HANSEN, H.C. PAXTON and D.P. WOOD, " Critical Masses of Oralloy Lattices Immersed in Water ", *LA-2026* (1955). See also Reference 4.

25. D. CALLIHAN, " Homogeneous Critical Assemblies " in *Progr. Nucl. Energy, Series I, Physics and Mathematics*, Vol. 1, p. 244 (reporting the work of Morfitt), Pergamon Press, London, 1956; and

J.W. MORFITT, " Minimum Critical Mass and Uniform Thermal Neutron Core Flux in an Experimental Reactor ", *Y-1023* (1953).

26. G. GOERTZEL " Minimum Critical Mass and Flat Flux ", *J. Nucl. Energy, 2*, 193 (1956).

27. D.F. CRONIN and A.D. CALLIHAN, " Critical Mass Studies: Part VII, Aqueous Uranium Slurries ", *ORNL-1726* (1954).

28. K.J. DE JONG, J.A.H. KERSTEN, J.J. WENT, H.H. WOLDRINGH and J.J. van ZOLINGEN, " Subcritical Circulating Suspension Reactor " in *Proc. Second U.N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 525.

29. J.W. WACHTER, " Homogeneous and Heterogeneous Poisoning as Potential Methods for Controlling Criticality ", This Conference, p. 289.

30. G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Nuclear Safety Experiments on Plutonium and Enriched Uranium Hydrogen-Moderated Assemblies containing Boron ", *RFP-201* (1960).

31. J.K. FOX and L.W. GILLEY, " Neutron Physics Annual Progress Report ", *ORNL-2609* p. 31 (Sept. 1958).

32. C.L. SCHUSKE, M.G. ARTHUR and D.F. SMITH, " Neutron Multiplication Measurements on Oralloy Slabs Immersed in Solutions, Part II ", *RFP-69* (1956).

33. J.K. FOX and L.W. GILLEY, " Neutron Physics Annual Progress Report ", *ORNL-3016*, p. 72 (Sept. 1960).

34. See p. 34 of Reference 31.

35. A.K. KRASIN, B.G. DUBOVSKY, M.N. LANTSOV, Y.Y. GLAZKOV, R.K. GONCHAROV, A.V. KAMAYEV, L.A. GERASEVA, V.V. VAVILOV, E.I. INYUTIN and A.P. SENCHENKOV, " Physical Characteristics of Beryllium Moderated Reactor " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 571.

36. F.A. KLOVERSTROM, R.M.R. DECK and A.J. REYENGA, " Critical Measurements on Near-Homogeneous BeO-Moderated Enriched Uranium Fueled Systems ", *Nucl. Sci. Eng. 8*. 221 (1960).

37. H.L. REYNOLDS, " Critical Measurements and Calculations for Enriched-uranium Graphite-moderated Systems " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 632.

38. G.A. JARVIS. Private communication.

39. L.B. ENGLE, G.E. HANSEN and H.C. PAXTON, " Reactivity Contributions of Various Materials in Topsy, Godiva and Jezebel ", *Nucl. Sci. Eng, 8*, 543 (1960).

40. J.K. FOX, L.W. GILLEY and D. CALLIHAN, " Critical Mass Studies, Part IX, Aqueous U 235 Solutions ", *ORNL-2367* (1958): and

E.C. MALLARY, H.C. PAXTON and R.H. WHITE, " Safety Tests for the Storage of Fissile Units ", *LA-1875* (Del) (1958).

41. J.T. MIHALCZO and J.J. LYNN, " Neutron Physics Annual Progress Report ", *ORNL-2842*, p. 67 (Sept. 1959).

42. L.W. GILLEY and J.T. THOMAS. Private communication.

43. J.K. FOX, L.W. GILLEY and E.R. ROHRER, " Critical Mass Studies, Part VIII, Aqueous Solutions of U 233 ", *ORNL-2143* (1956): and

J.T. THOMAS, J.K. FOX and D. CALLIHAN, " A Direct Comparison of Some Nuclear Properties of U 233 and U 235 ", *Nucl. Sci. Eng. 1*, 20 (1956).

44. J.K. FOX, L.W. GILLEY, R. GWIN and D.W. MAGNUSON, " Neutron Physics Annual Progress Report ", *ORNL-2842*, p. 76 (Sept. 1959).

45. H.C. PAXTON. Private communication.

46. F.E. KRUESI, J.O. ERKMAN and D.D. LANNING, " Critical Mass Studies of Plutonium Solutions ", *HW-24515 (Del)* (1952).

47. C.C. HORTON and J.D. McCULLEN, " Plutonium-Water Critical Assemblies " in *Proc. First U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1955, 5*, 156.

48. J. BERTRAND, P. BONNAURE, C.CLOUET D'ORVAL, J. CORPEL, J. DE LAMARE. P. LECOUSTEY, I. PREVOT, R. ROCHE, M. SAUVE, J. TACHON and G. VENDRYES, " ' Proserpine ', a Homogeneous Critical Experiment with Plutonium " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 539.

49. G.A. JARVIS, G.A. LINENBERGER, J.D. ORNDOFF and H.C. PAXTON, " Two Plutonium-Metal Critical Assemblies ", *Nucl. Sci. Eng. 8*, 525 (1960).

50. G.E. HANSEN, H.C. PAXTON and D.P. WOOD, " Critical Plutonium and Enriched-Uranium-Metal Cylinders of Extreme Shape ", *Nucl. Sci. Eng. 8*, 570 (1960).

51. D.P. WOOD, C.C. BYERS and L.C. OSBORN, " Critical Masses of Cylinders of Plutonium Diluted with other Metals ", *Nucl. Sci. Eng. 8*, 578 (1960).

52. E.A. PLASSMANN and D.P. WOOD, " Critical Reflector Thicknesses for Spherical U 233 and Pu 239 Systems ", *Nucl. Sci. Eng. 8*, 615 (1960) :

F.A. KLOVERSTROM," Spherical and Cylindrical Plutonium Critical Masses ", *UCRL-4957* (1957): and

H.R. RALSTON, " Critical Parameters of Spherical Systems of Alpha-Phase Plutonium Reflected by Beryllium ", *UCRL-5349* (1958).

53. A. GOODWIN Jr. and C.L. SCHUSKE, " Plutonium Graphite Assemblies, *RFP-123* (1958) :

A. GOODWIN Jr. and C.L. SCHUSKE, " Plutonium Graphite Assemblies, Part II ", *RFP-158* (1959) :

C.L. SCHUSKE, G.H. BIDINGER. A. GOODWIN Jr. and D.F. SMITH, "Plutonium Plexiglas Assemblies ", *RFP-178* (1960) :

G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Plutonium Plexiglas Assemblies, Part II ", *RFP-190* (July 1960) : and

C.L. SCHUSKE, D.F. SMITH and C.L. BELL, " Plexiglas Reflected Assemblies of Plutonium ", *RFP-213* (1961).

54. V.I. NEELEY, R.C. LLOYD and E.D. CLAYTON, " Neutron Multiplication Measurements with Pu-Al Alloy Rods in Light Water ", *Trans. Am. Nucl. Soc., 3*, No 2, 292 (June 1960).

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DISCUSSION

H. DOPCHIE, Belgium:

You mentioned the minimum critical mass of 2.5 kg for MTR type fuel elements in water. I should like to compare this number with the number we have obtained at our critical experiment with the BR 02 reactor which uses elements similar in composition to MTR type fuel elements. These elements are embedded in a beryllium matrix. We have in this experiment obtained a critical mass of 1.3 kg U 235, which shows the importance of the beryllium reflection as has been well demonstrated by Mr. Woodcock.

E.D. CLAYTON, United States:

With regard to the k_∞ measurements with the 3 % UO_3, personnel at Hanford are currently undertaking a Monte Carlo calculation of k_∞ for the case of no moderation.

J.G. WALFORD, United Kingdom:

Dr. Callihan made several very generous remarks about the United Kingdom work and for these we thank him. I think it is unnecessary for me to point out that the British experimental data amounts to no more than a drop in the ocean when compared with the vast amount of United States work which we have heard described this afternoon. There are, however two small regions in which our experiments may be of interest. Firstly, Dr. Callihan has mentioned that we are working on uranium systems of 1.4 % enrichment; these are perhaps worthy of mention because this enrichment is almost certainly the lowest at which complete assemblies of the homogeneous hydrogen-moderated kind have ever been made critical. We have carried out experiments which have given curves similar to those shown in Figures 2, 3, 4 and 5 of Dr. Callihan's paper, and we have found that the minimum critical mass occurs at about the same H/U 235 ratio as in the United States experiment at 2 % enrichment, that is, in the region of 500-600 H/U 235 atomic ratio; we have found that the minimum critical mass itself at 1.4 % enrichment is about 21 kg U 235. The minimum critical volume is in the region of 700 litres, so these low-enrichment systems are obviously very large, often approaching a cubic metre in size. I am speaking of systems which were actually measured as cubes fully reflected by polythene, but which have been converted to spheres to give the above data*. The fuel material was very similar to that used at Oak Ridge, i.e. a compressed homogeneous mixture of uranium tetrafluoride powder and paraffin wax moderator. The second region in which the British experiments may be of interest occurs at 30 % U 235 enrichment; I think Mr. Thomas can better comment on this.

A.F. THOMAS, United Kingdom:

We have been doing some work with lightly moderated homogeneous mixtures of UO_2 in paraffin wax. We have done some critical cubes, bare and reflected with Perspex, and with Polythene and we have done it at two

(*Editor's note:* The data quoted here have since been added by Dr. Callihan to the graphs of his paper " A Review of Criticality Data obtained by Experimental Methods ".)

core densities. We have tried to avoid the troubles caused by lumping, by making the mixture in 1-in. cubes and pressing it with holes through the centre to reduce the density to three-quarters. We did subsidiary experiments to check that this did not cause any streaming effects. The full results will be published later, but the first results show that the calculation of the variation of critical mass by a simple relation, e.g. for reflector density, can be misleading. The Perspex results with Perspex density 1.2 g/cm^3 yield a critical mass some 15 % less than for Polythene with density 0.92 g/cm^3. One might say that this is fair enough: one has a measured density exponent for a core of about 1.5. If you take the square root of 1.3 — i.e. the total density ratio — you would expect a variation of about 15 %. If this is so, the water-reflected critical mass should be intermediate between the two: in fact, it is greater than the value for both Polythene and Perspex. The next thing we looked at was the hydrogen density; this is in the inverse order, so this does not work. Thus you have to be very careful when you change the chemical form of your reflector and moderator (especially hydrocarbon) to ensure that you get the right density relation. One might be successful with hydrogen density alone for water and polythene perhaps, certainly not for things like Perspex.

W. SCHÜLLER, Eurochemic:

Mr. Thomas' comments stimulate me to ask whether in measuring the critical masses of low-moderated systems with hydrocarbon moderation, it is necessary to correct for the moderating effect of the carbon?

A.F. THOMAS:

We have had a great deal of argument as to whether the CH_2 in our wax is the same thing as water. I don't think that a slight difference in chemical binding is going to make any difference to the rise in the hydrogen cross section at low energies. But I think the presence of the carbon as opposed to oxygen as a scatterer will be significant and we hope to do some tests by adding graphite at the end of the experiments.

W. SCHÜLLER:

I have another question for Dr. Callihan. You were referring to experiments with MTR type assemblies in which you measured the number of assemblies required for criticality. I wonder whether experiments have been made on other types of fuel elements, because this would be extremely interesting for our work. In taking the safe geometric dimensions for optimum heterogeneously-moderated lattices one is rather restricted, whereas for actual fuel elements, due to the poisonous effect of the cladding, one might in practice allow a larger number of assemblies to be accumulated. But results could only be derived from experiments or from rather lengthy calculations.

D. CALLIHAN, United States:

To my knowledge, the only experiments in this category are those done by the critical experiment teams whose function is to support the reactor

development itself. This is an encouragement that is being given to reactor design teams in the United States. When the experiments supporting the reactor design are done, some few measures of the critical conditions of elements in non-reactor environments, in water one might say, with various spacings, should be incorporated in the programme.

CORRELATIONS OF EXPERIMENTAL AND THEORETICAL CRITICAL DATA

Comparative Reliability, Safety Factors for Criticality Control

Hugh C. PAXTON,**
Los Alamos Scientific Laboratory, United States

INTRODUCTION

For the experimental part, this discussion will concentrate on data that are susceptible to generalization. Practically, the scope will be limited to fissile systems that may be approximated by simple descriptions: elementary geometry and generally homogeneous fuel regions. This type of information has been termed 'basic' nuclear safety data. Thus, we ignore the specialized experiment such as that intended to demonstrate safe operating conditions for a specific processing unit.

Similarly, we will not consider specialized calculations such as those designed to give effects of minor changes in a given type of heterogeneous fissile assembly. Rather, most attention will be given to computation which hopefully may substitute for basic experimental data. Before considering this class of calculation, however, let us discuss a somewhat less demanding application of theory.

THE ADJUSTMENT OF EMPIRICAL DATA

As experimental data seldom apply to ideal systems which may be described in similar terms, empirical correlation of the data generally requires conversion to uniform conditions. This process, in turn, makes use of data correlations which may be strictly empirical or guided by theoretical models.

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* Supported by the United States Atomic Energy Commission.

** Drawing freely from theoretical work of:
Bengt G. Carlson [1]
Aurel Goodwin, Jr. [2]
Gordon E. Hansen [1]
Carroll B. Mills [1]
William H. Roach [1]
George Safonov [3]
William R. Stratton [1].

[1] The Los Alamos Scientific Laboratory, Los Alamos, New Mexico.
[2] The Dow Chemical Company, Rocky Flats Plant, Denver, Colorado.
[3] The Rand Corporation, Santa Monica, California.

(It is apparent that successive approximations may be involved if there is a relationship between input and output correlations.) As, for example, corrections on critical mass are frequently within 10 % the ultimate in precision is often not required of the model used for correction. Figures 1 and 2 are examples of two empirical representations of the same data, the first without regard to a theoretical model and the other in a form suggested by such a model [1]. In each case the data are critical volumes of families of cylinders of $UO_2F_2 - H_2O$ solutions in which the uranium is enriched to about 93 % of U 235 by weight: we shall symbolize this uranium as U(93).

Figure 1 shows, for cylinders of various height/diameter values, ratios of critical volumes to the spherical value for the same solution. It may be noted that there is no obvious distinction between curves for solutions at two widely different uranium concentrations. In Figure 2 are represented effective extrapolation distances of critical cylinders, δ_c, which satisfy the buckling relationship:

$$\left(\frac{2.405}{\frac{d}{2}+\delta_c}\right)^2+\left(\frac{\pi}{h+2\delta_c}\right)^2=\left(\frac{\pi}{r_s+\delta_s}\right)^2,$$

where d and h are diameter and height of the critical cylinder and r_s is the radius of the corresponding sphere. The sphere extrapolation distance δ_s is the sum of an experimental reflector savings and an assumed or computed bare-sphere extrapolation distance. (The latter value, of course, remains an inherent part of the correlation.) If simple theory applied precisely, δ_c would be independent of h/d. As it is, deviations from a constant value are sufficiently small to allow of some confidence in extrapolations to extreme values of h/d. Such extrapolations are impossible with the more direct representation of Figure 1.

Another example of a simple calculation that is useful for adjusting or extending experimental data gives critical masses of modestly diluted metal systems in terms of effective absorption and transport cross sections of the diluent. These cross sections, in turn, are obtainable from reactivity coefficients of diluent and undiluted fissile material. Figure 3 compares directly-observed critical masses of diluted enriched uranium metal with results from integrated reactivity coefficients that were measured in an undiluted sphere. The primary critical data are from measurements by Jarvis (unpublished). The computed relationships are expected to hold through a range in which fixed one-group cross sections apply — up to about 50 % by volume dilution for graphite and perhaps to 70 % for most medium-Z and high-Z elements. This sort of correlation provides the bases for corrections of critical data to standard densities and concentrations.

A step further from detailed theory is a scheme for correlating cubic lattices of air-spaced fissile units, which is suggested by the power relationships of Figure 3. In Figure 4 data from Oak Ridge National Laboratory [2] (the best available at present for cubic arrays) follows the form and critical number of given units = (lattice density)$^{-s}$, where s is constant over the range of measurement. Figure 5, which shows the way in which s depends upon type of unit and effectiveness of reflector about the array [3], is then a basis for generalization to unmeasured systems.

The picture of relationship between the critical mass of a plane array of discrete units and that of a uniform slab of the same material leads to the

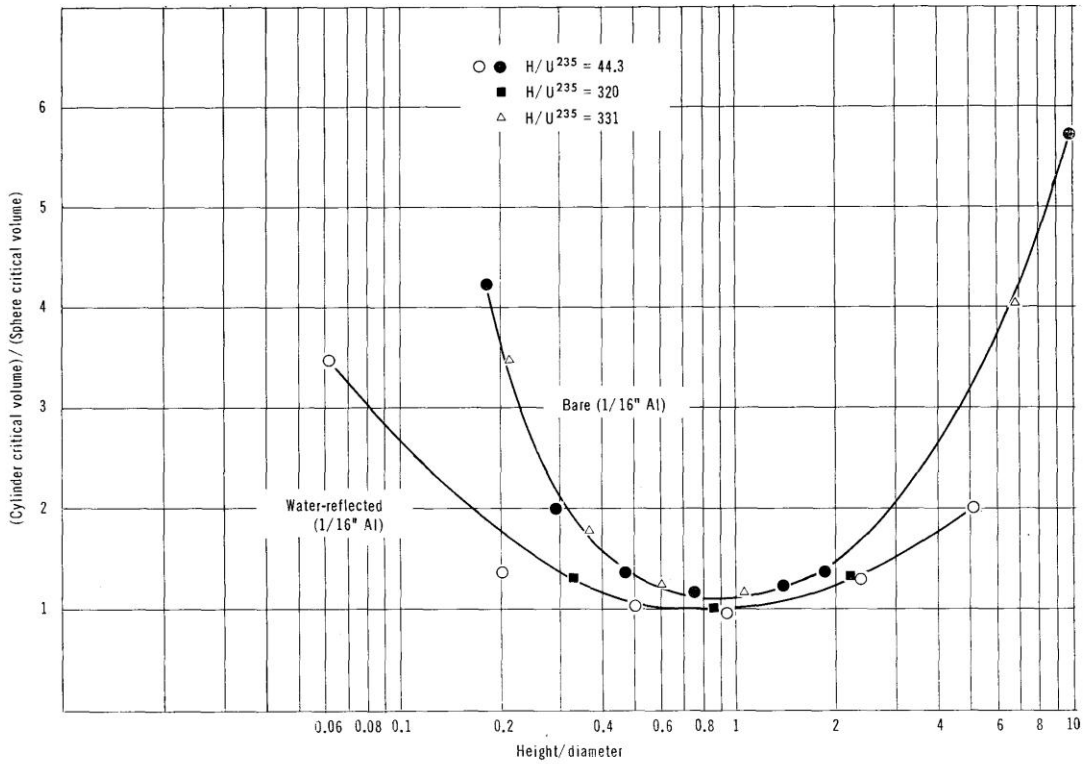

Figure 1. *Critical volumes of cylinders of* $U(93)O_2F_2$ *solution relative to sphere values.*

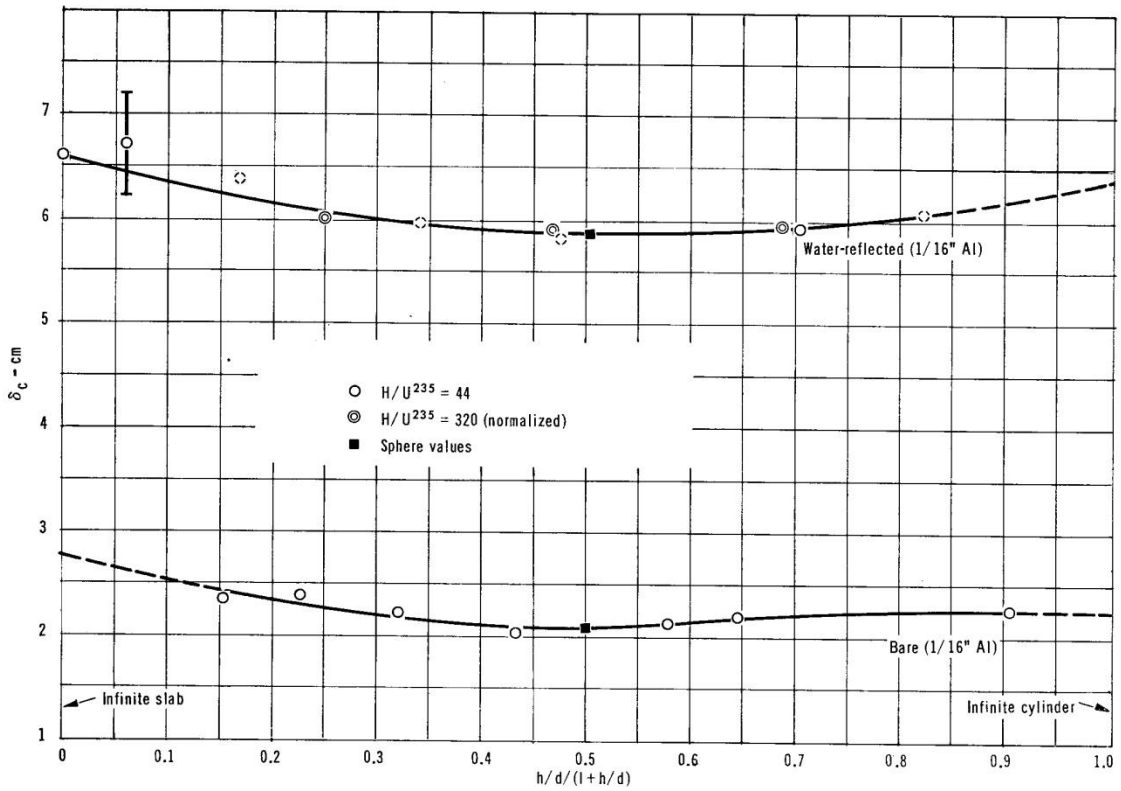

Figure 2. *Effective extrapolation distances for cylinders of U(93)O_2F_2 solutions.*

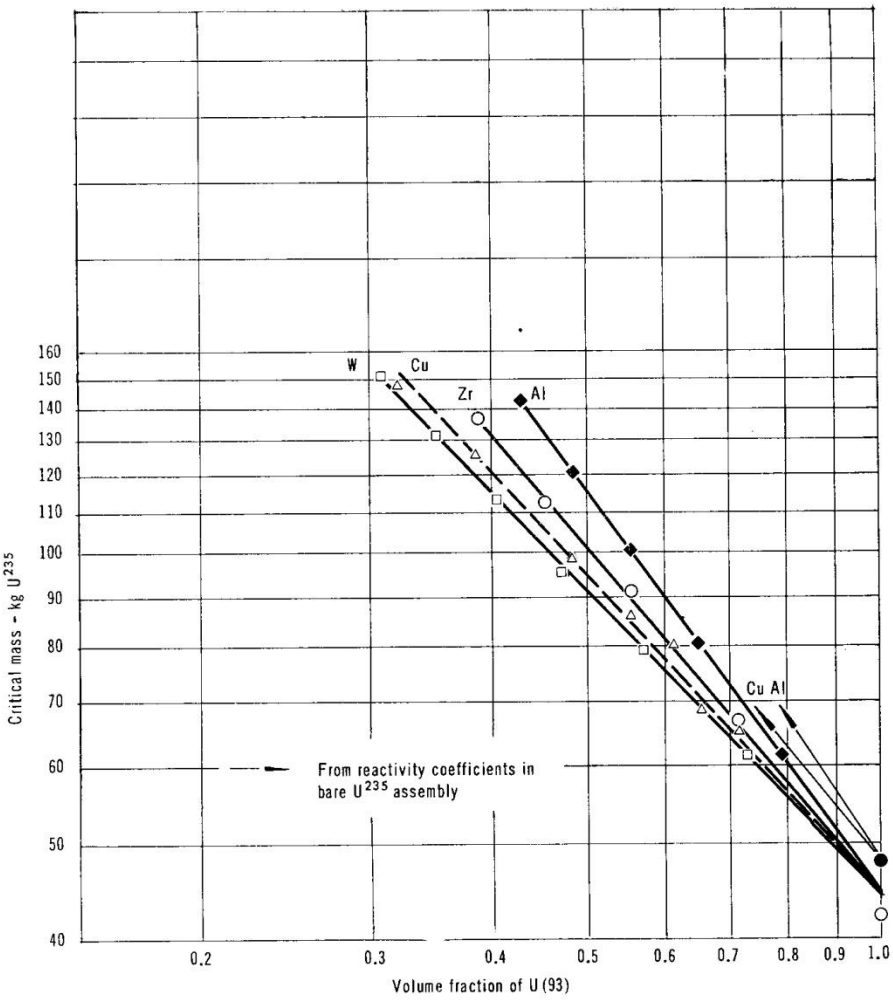

Figure 3. *Critical masses of bare spheres of U(93) diluted with other metals.*

correlations of Figure 6. Here, ORNL data on arrays of solution cylinders [4] are represented in terms of thicknesses of critical solution if spread over the base area of the array. It is obvious that this representation breaks down at a finite height if the cylinder diameter is such that an individual unit can be made critical. Limited data on plane arrays of massive metal spheres appear to fall into a similar pattern, which suggests the possibility of useful generalization.

One feels less comfortable about the generalization of purely empirical forms for which the reason is not clear. An example is the relatively close clustering of ratios of water-reflected to bare critical volumes for a considerable variety of systems with similar moderation, as represented in Figure 7 [5]. Though this correlation is highly suggestive and establishes reasonable limits, we do not expect it to be reliable in detail. In this case, there happens to be

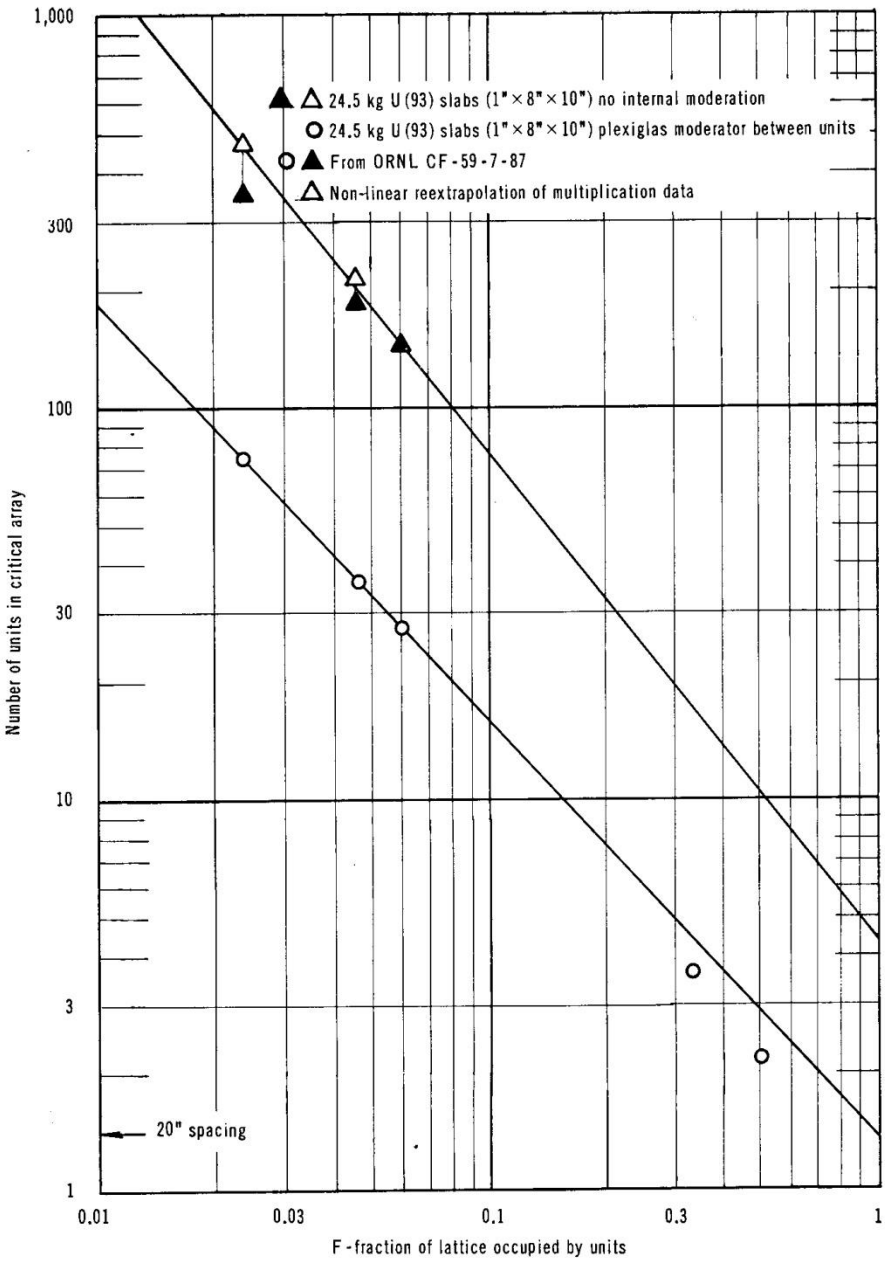

Figure 4. *Critical cubic arrays of metal slabs with and without Plexiglas between units.*

hope for a simple guiding model, because a reasonable trend is from the observed minimum volume ratio of ~ 0.4 (for small, efficient systems) toward unity as neutron leakage becomes successively less important in fission economy (or as k_∞ decreases). Thus, the greatest problem might not be how

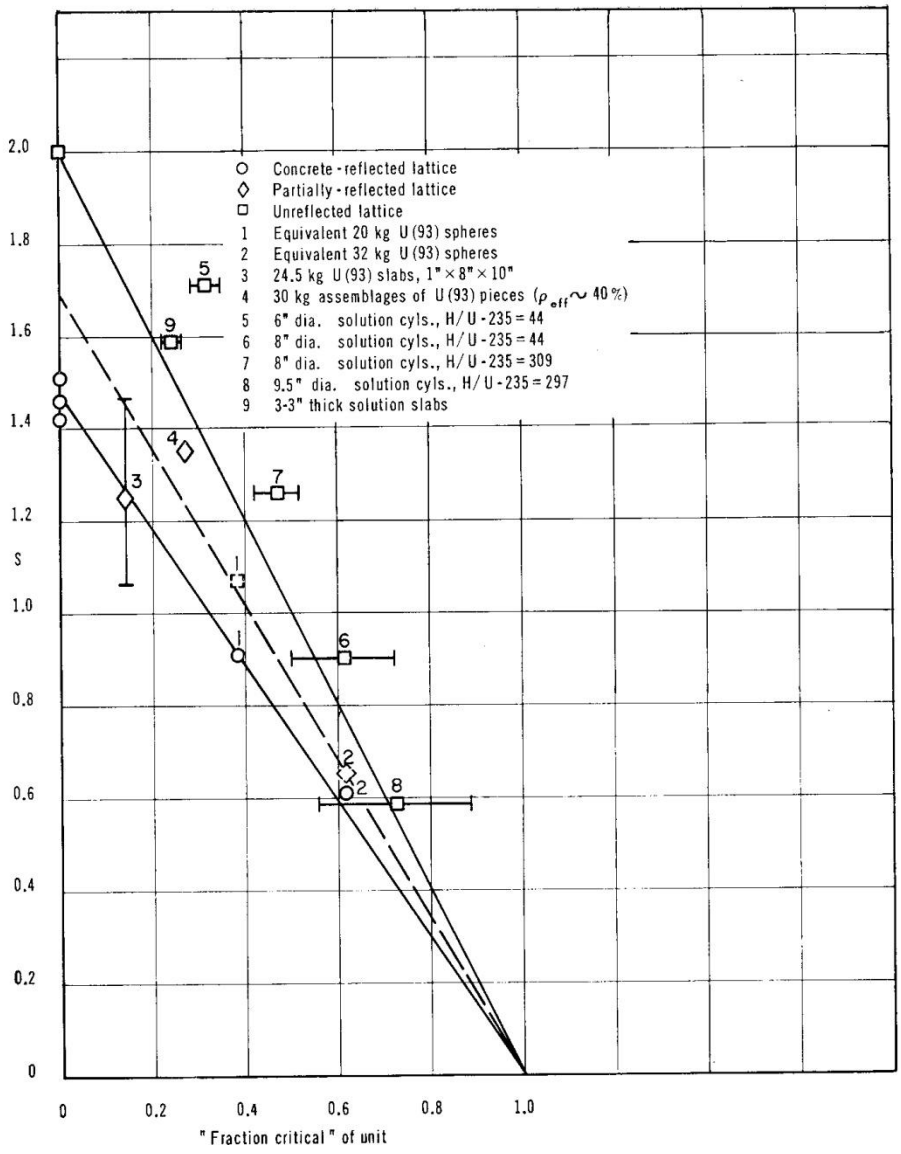

Figure 5. *Lattice-density exponent vs. "fraction critical" of an isolated unit for various degrees of external reflection.*

to improve the correlation, but rather, how to find a better representation in practical engineering terms. Though volume ratios at standardized densities may form a nearly unique function of k_∞, the latter is not a convenient process parameter.

These illustrations, intended primarily to show the utility of crude theoretical models for data adjustment, will serve a second purpose. They give some basis for judging the uncertainties added to primary data when prepared for correlations.

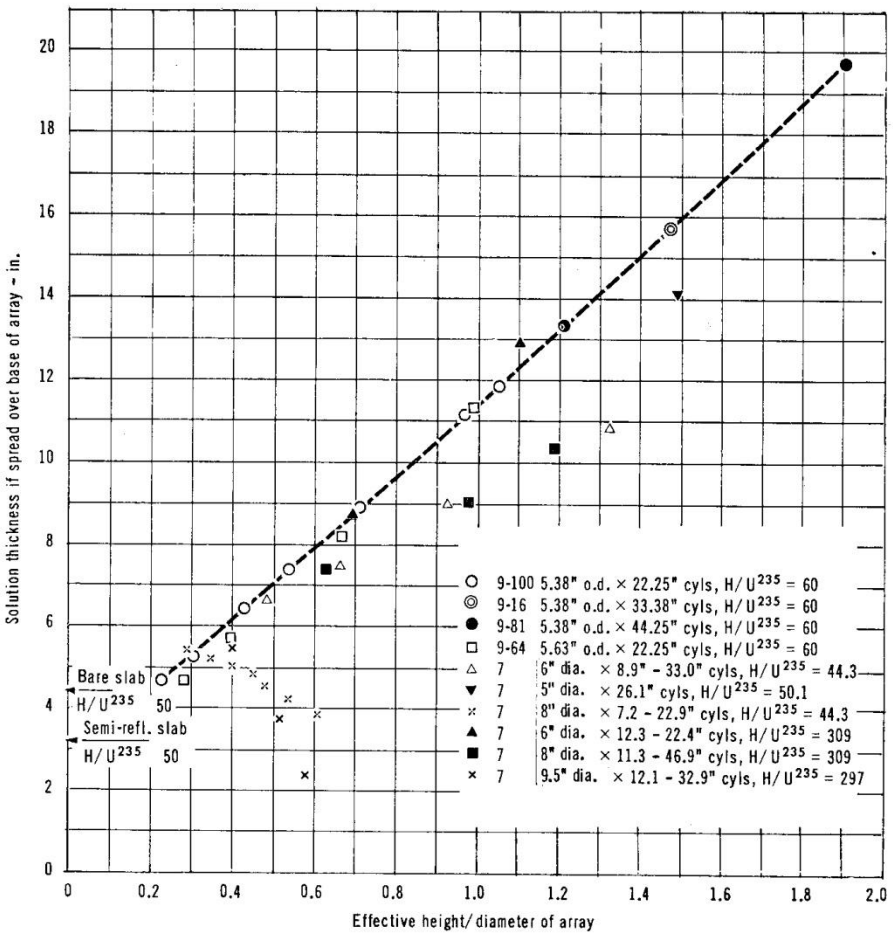

Figure 6. *Correlations of critical arrays of U 235 solution cylinders.*

Computation as a Substitute for Experiment

Attention now will be turned to a realm in which we contend that highly-refined calculation is required. This is where computed values are intended to substitute for experimental data in nuclear safety guidance. Just as experimental values require some index of reliability, computations which replace them should leave the user with a feel for probable error. Practically, there may be even more stringent requirements on computation than on experiment because of doubters who remember the days when a factor-of-two error in calculated critical mass was customary unless nearby experimental data were available for normalization. In spite of vastly improved input data, computational facilities and techniques, some of the stigma remains; hence our opinion that calculation for the purpose under discussion should be so good as to sell itself to the more sceptical. (With the advent of reasonably complete high-quality computational surveys, there may be a place for short cuts " which seem to work ".) The requirements implied above are: realistic

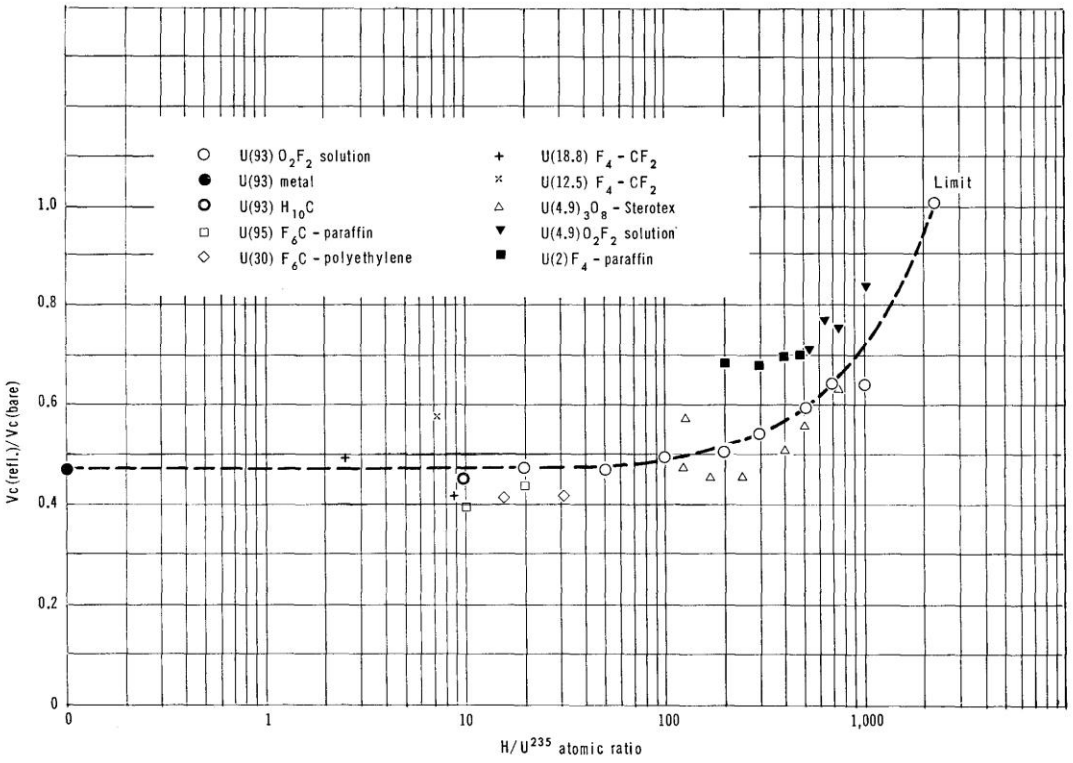

Figure 7. *Ratio of water-reflected-sphere to bare-sphere critical volumes for hydrogen-moderated U 235.*

input, convincing method and a wide enough range of checks against experiment to establish error patterns.

The most useful form in which data might emerge is a computational survey. The classic of this type is the series of surveys of bare, spherical critical systems undertaken by Safonov a number of years ago [6]. For U 233, U 235 or Pu 239, diluted by H_2O, D_2O, Be, BeO or C, he presented critical sizes and certain spectral indices as functions of moderator fuel ratio. Other series cover the ternary systems of U 235–U 238–C, U 235–Be–C, and, at elevated temperature, U 235–U 238–H_2O. Parameters for his multi-group diffusion method were chosen carefully, and reasonably represent processes including inelastic scattering and resonance region effects. As the diffusion approximation cannot apply precisely to a small system for which radius and mean free path for fission are of the same order, it was necessary to adjust high-energy transport cross sections for agreement with critical data from fast-neutron assemblies.

Recently, Mills [7] has applied more modern techniques to the binary systems surveyed by Safonov, and, for H_2O-moderated U 233, U 235 or Pu 239, he included, with spheres, critical cylinders of infinite length and slabs of infinite extent, and added H_2O reflection. Also covered is a portion of the U 235–U 238–H_2O series for bare spheres. The DSN method that Mills employed [8] has the advantage of applying realistically to fast-neutron systems, and input data presumably have been improved since the time of Safonov's work, particularly a specific coded scheme of Bell's for averaging cross sections throughout resonance regions [9]. The high-energy cross section set is that of Hansen and Roach [10] in which parameters have been adjusted within their ranges of uncertainty to fit a wide variety of integral fast-neutron data. Other DSN surveys of interest to us were conducted by Roach for water-reflected U(93)–H_2O and Pu 239–Pu 240–H_2O spheres at normal and reduced densities, and by Stratton for bare and water-reflected U(93)–H_2O–C spheres (both sets unpublished, but cross sections appear in Reference [10]). A difference with Mills' procedure which is expected to be of minor influence is an alternative hand method of averaging resonance cross sections. With the exception of points on Figure 24, anisotropic scattering cross sections for hydrogen were incorporated in all DSN calculations.

It is appropriate to mention that detailed computation, like good experiment, is expensive. The DSN method requires a modern, high-capacity computer, and the necessary time increases with degree of moderation, where advantages over the less-demanding multi-group diffusion method largely disappear. For this reason, Goodwin uses multi-group diffusion calculations for extensive nuclear safety surveys, supplemented by fewer DSN results to provide check points and to cover neutronic complexities that are beyond the simpler method (Reference [11] for example). We shall have occasion to use some of Goodwin's results, while concentrating on those of Safonov, Mills and Roach.

Comparisons of Experiment and Computation

Experimental data for *bare spheres of highly-enriched uranium metal* diluted in various degrees by D_2O, Be, BeO and C (graphite) are represented in Figure 8. Water-moderated systems, which will be considered in more detail, are included only for comparison of form. Errors in data interpretation (shape conversion, unspecified impurities, inhomogeneities and inci-

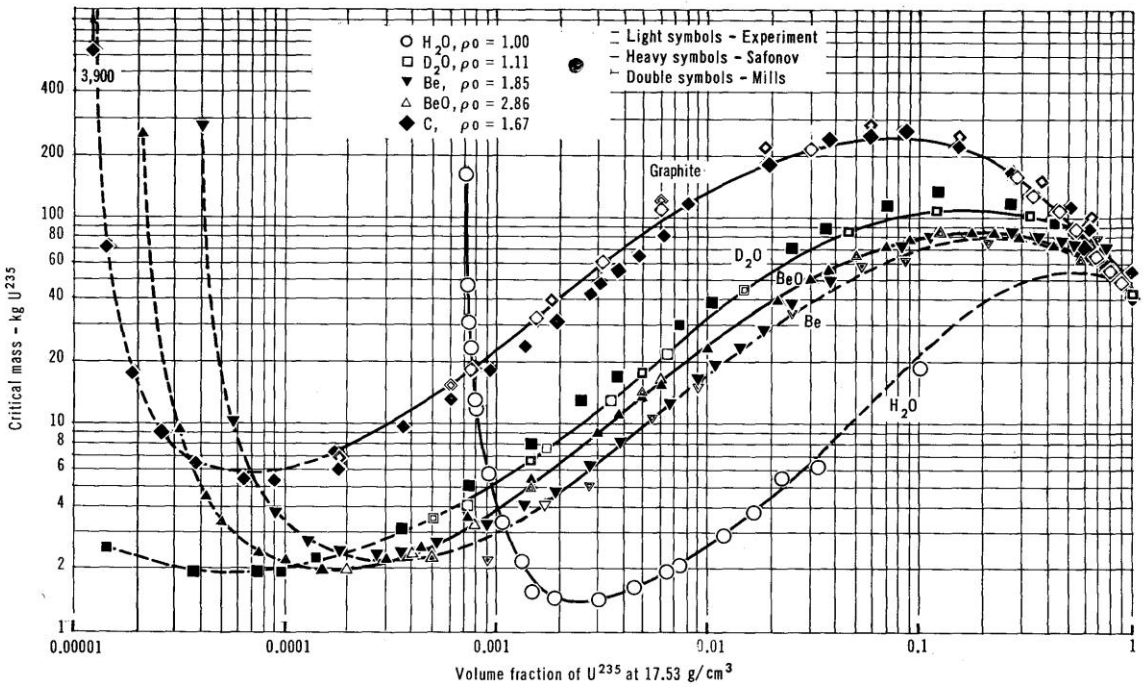

Figure 8. *Critical masses of bare spheres of U(93) with various moderators.*

dental reflection) should be generally within 10 %, with appreciably smaller limits for H_2O and D_2O solutions. Exceptions are the lower point that applies to undiluted metal (the shape was extreme and density uncertain), and possibly the point for ~ 90 % H_2O by volume, where there was a compositional correction. Density corrections, according to critical mass = const (density)$^{-2}$, are quantitative. Calculations by both Safonov and Mills agree reasonably with experiment, deviations being generally within 20 %. In Safonov's case, this is particularly significant because most of the critical experiments followed his work.

Experimental critical masses for *water-moderated, highly enriched uranium spheres*, both bare and water-reflected, are presented in Figure 9. The importance of changes in U 235 density at constant H/U 235 atomic ratio is illustrated by the dual representation of data for concentrated solutions, first at normal densities then at the larger densities corresponding to idealized homogeneous metal-water mixtures. Even though such mixtures cannot be attained in practice, they represent an upper limit to density at a given value of H/U 235, hence a lower limit to critical size. From the corresponding critical volume curves (Figure 10), we see that the minima for solutions disappear for metal-

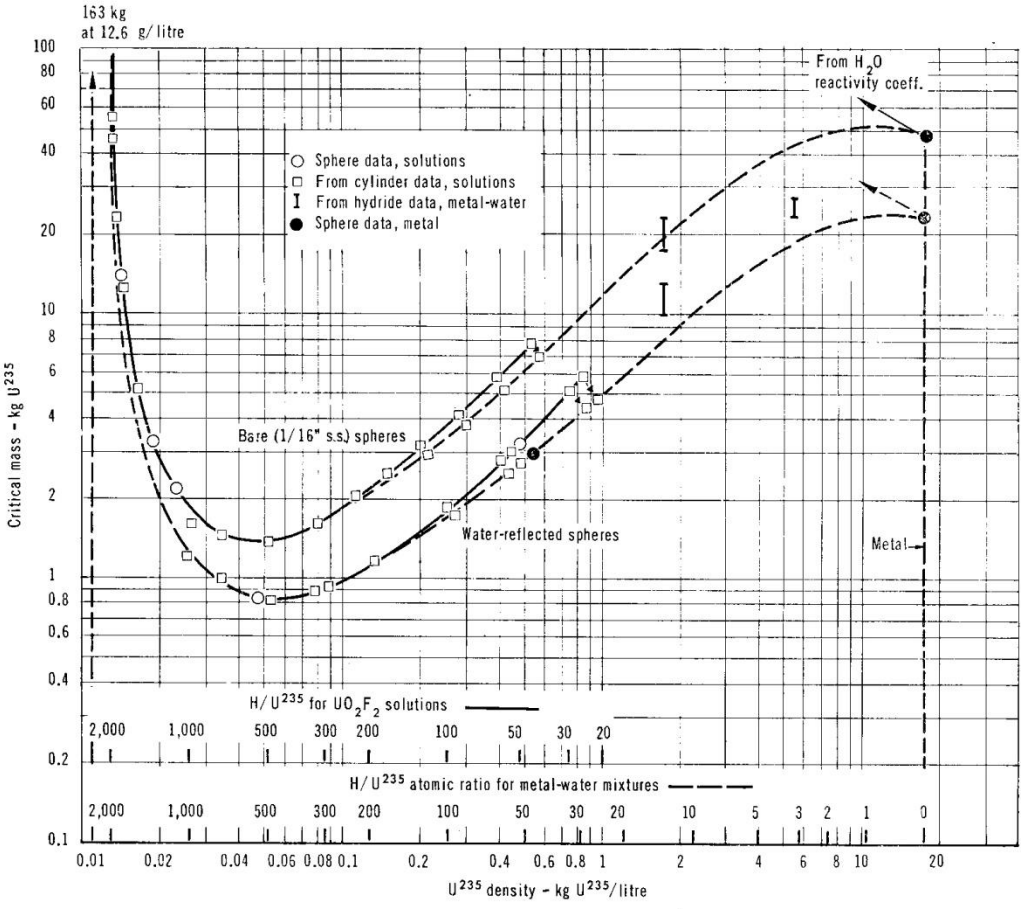

Figure 9. *Experimental critical masses of homogeneous water-moderated U(93.2) spheres.*

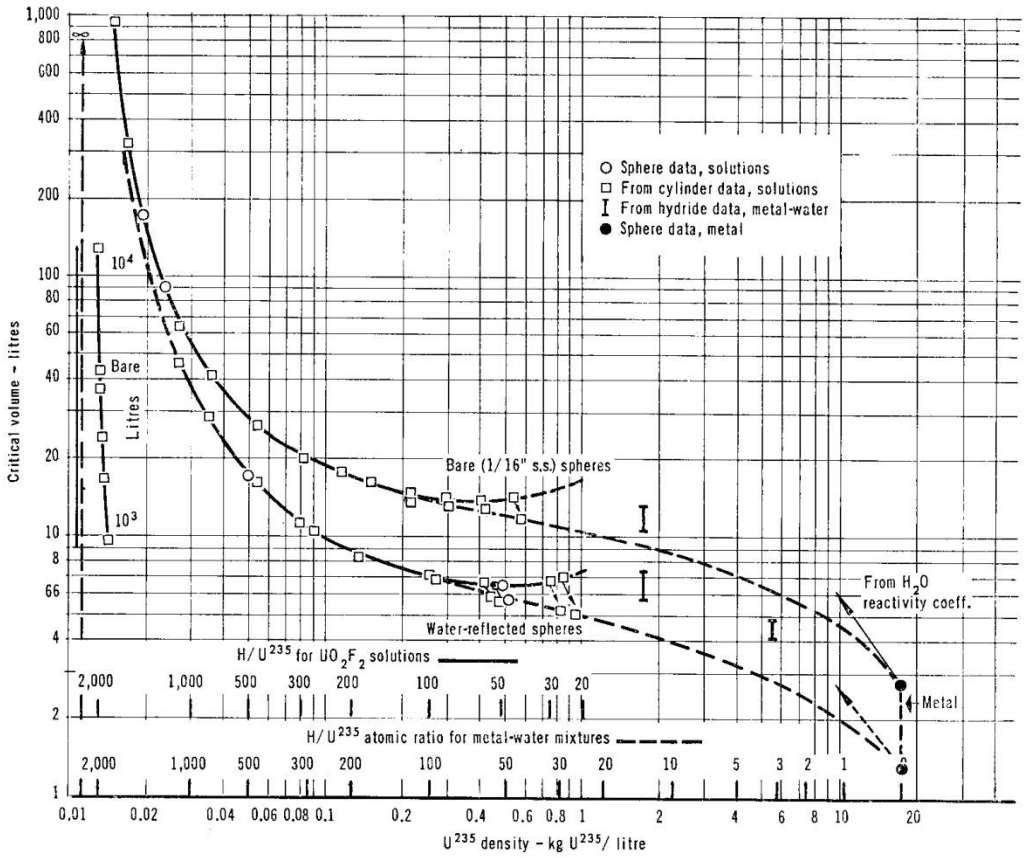

Figure 10. *Experimental critical volumes of homogeneous, water-moderated U (93.2) spheres.*

water mixtures. As has been noted, corrections, to densities of mixtures of uranium compounds and water, for example, are straightforward for unreflected systems. When water reflection is added, however, the influence of a core density change is reduced, as indicated by Figure 11 for water-moderated, water-reflected spheres. The core-density exponents shown, which are from critical calculations by Roach at two sets of densities differing by 20 %, are used here at face value.

With Figure 12 we return to the U 235 critical mass curves with density-corrected computations superposed. Only Mills has values for both bare and reflected spheres. Though experimental points for solutions appear to be accurate to within 5 %, the two water-reflected points that are derived from ' hydride ' assemblies should be discounted because of serious uncertainties in composition and reflector-savings corrections (recently- ' improved ' corrections led to alarming shifts). Direct checks of computation against the undistorted hydride data would serve a better purpose than inclusion on these curves. It should be noted that experimental ' unreflected ' points are biased downward (perhaps 2 % for concentrated solutions) by the nominal 1/16 in.

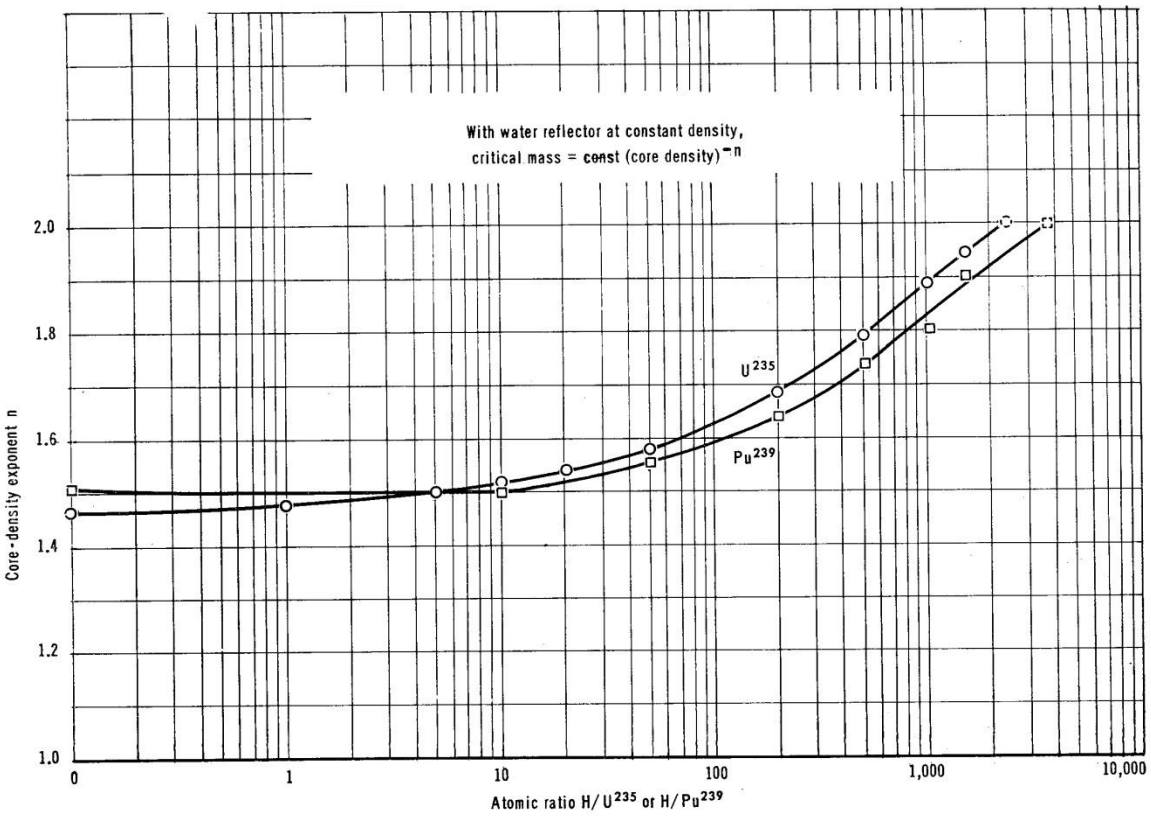

Figure 11. *Core-density exponents for water-reflected, water-moderated U 235 or Pu 239 spheres.*

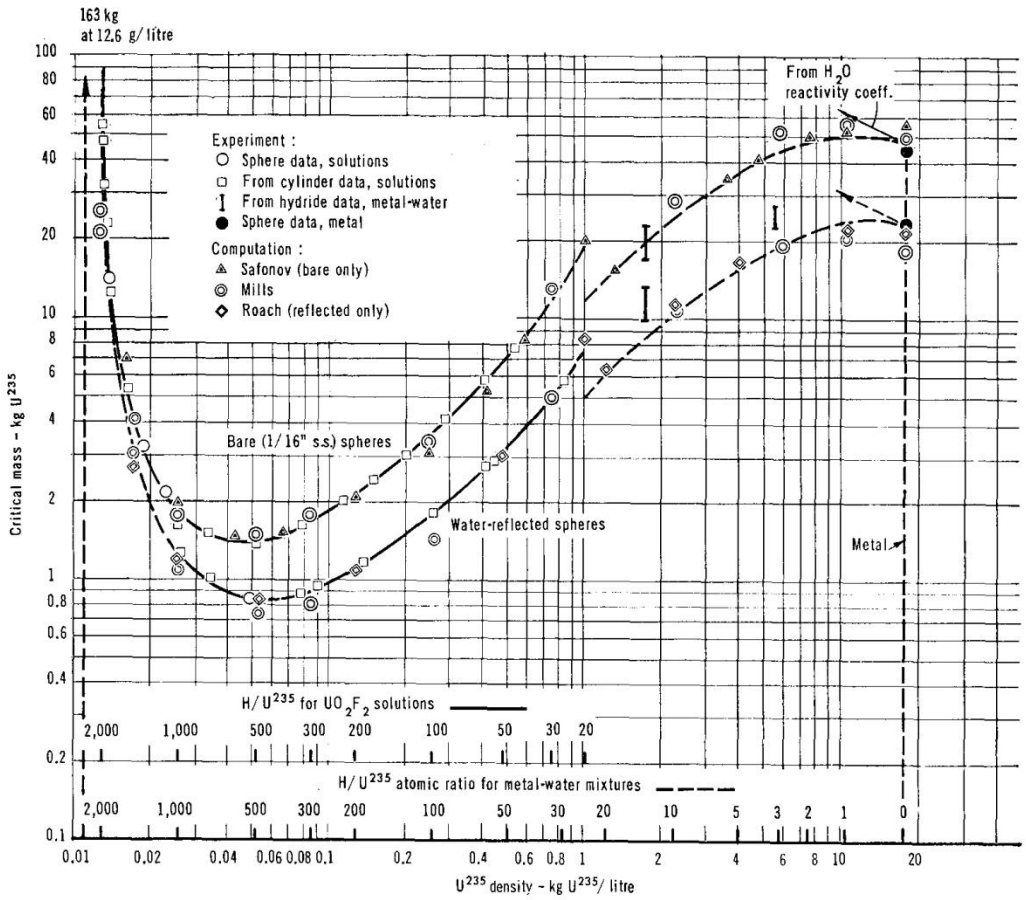

Figure 12. *Experimental and computed critical masses of homogeneous water-moderated U(93-2) spheres.*

stainless steel container which was not included in calculations. Safonov's values, which may be a few percent high due to a thermal base of 500° F, have been chosen as the latest and best of several sets. Generally, the agreement between solutions, data and calculation is appreciably better than the maximum deviation of $\sim 20\,\%$.

Empirical data for *infinitely-long cylinders and slabs of infinite extent* (Figures 13 and 14), of course, are simply based on observation. For undiluted metals and solutions near the critical minima, measurements on long cylinders and broad slabs lead to reasonably convincing values for the infinite systems. In other regions, estimates of slab thicknesses are considerably more sensitive to conversion errors than are the cylinder diameters. In spite of this, it appears that Mills' slab values are systematically high, as contrasted with his calculations for cylinders.

Information about *water-moderated plutonium assemblies* is not as clean as that for U 235. Existing critical data, for metal and for dilute solutions only, has recently been supplemented by subcritical measurements of Schuske

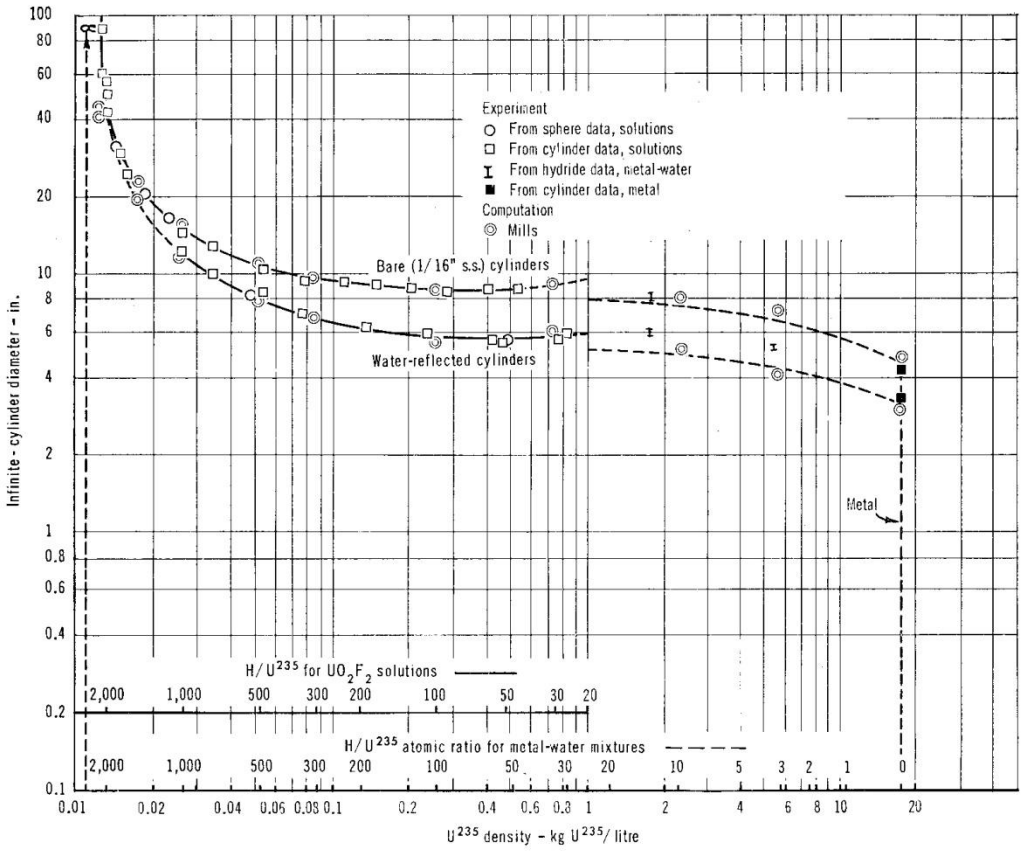

Figure 13. *Estimated critical diameters of infinite cylinders of homogeneous, water-moderated U(93.2).*

and his co-workers at Rocky Flats on plutonium metal sheet with Plexiglas as moderator and, in some cases, reflector [12]. But data on the influence of ever-present Pu 240 and on effects of variation in moderator composition are scanty. For this reason, it is desirable to use detailed computation for adjusting the Rocky Flats observations. Relative *critical masses of Plexiglas-moderated plutonium* (bare or Plexiglas-reflected) and of water-moderated plutonium (bare or water-reflected) from such calculations appear in Figure 15. The curve for bare spheres was obtained by Goodwin and that for reflected spheres by Roach. Figure 16, also due to Goodwin and Roach, gives the percent increase in sphere critical mass of total Pu per percent Pu 240 content of the plutonium.

Making use of the last two sets of curves and density exponents from Figure 11, critical masses from the Plexiglas-moderated assemblies were joined with the older data for solutions and unmoderated metal as in Figure 17. The 'idealized solutions' represented in addition to metal-water mixtures have a relationship between fuel density and hydrogen-fuel ratio which is analogous to that of UO_2F_2 solutions. Practical densities generally would be less because

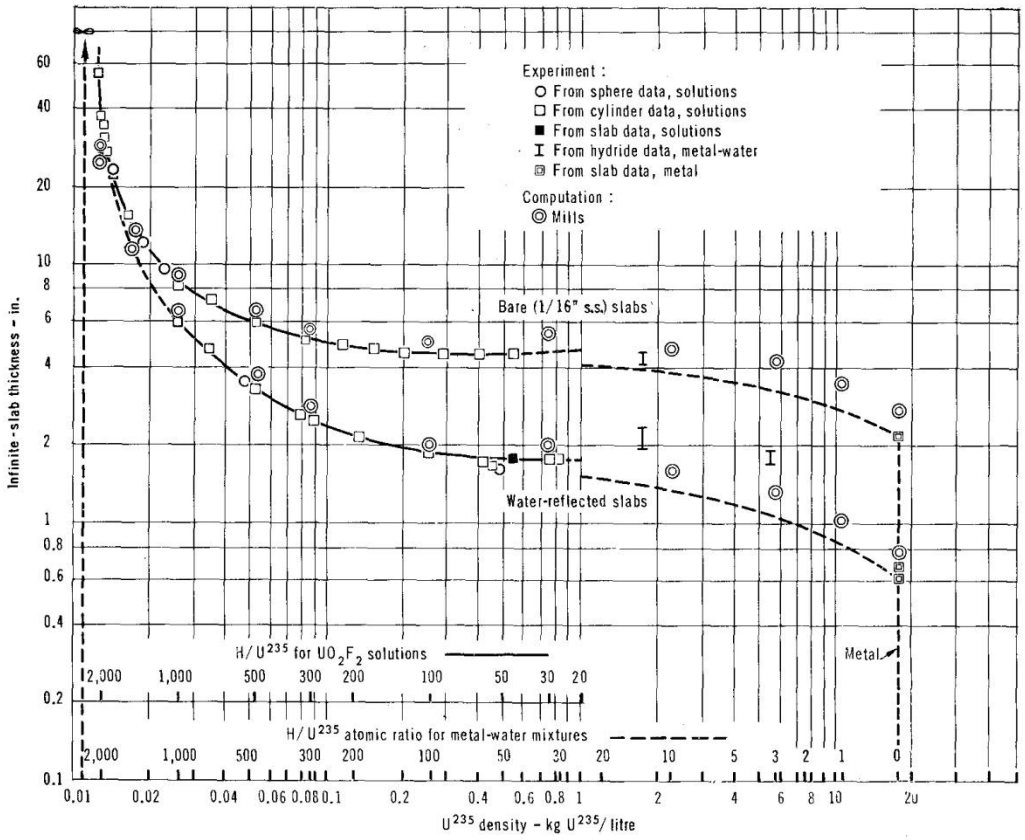

Figure 14. *Estimated critical thicknesses of infinite slabs of homogeneous, water-moderated U(93.2).*

of the excess acid required for stable Pu solutions. In Figure 18, where computed results are added to the experimental curves, the reasonable clustering is particularly significant, as only Safonov used the points applying to moderated assemblies for cross section adjustment (he used just dilute-solution data for this purpose). Here it is difficult to judge whether values from experiment or from calculation are the more reliable. If consistence is a valid criterion, it appears that critical masses are tied down to within 20 % over most of the range of moderation.

Except for water-reflected metal, observations on critical cylinders and slabs containing Pu are so limited that conversions to the infinite forms must be based on models which apply to U 235 (Figures 19 and 20). In spite of this added uncertainty, differences between points based on experiment and those computed by Mills are similar to the uranium cases.

For completeness, corresponding sphere critical masses, cylinder diameters and slab thicknesses for H_2O-moderated U 233 are included in Figures 21, 22 and 23.

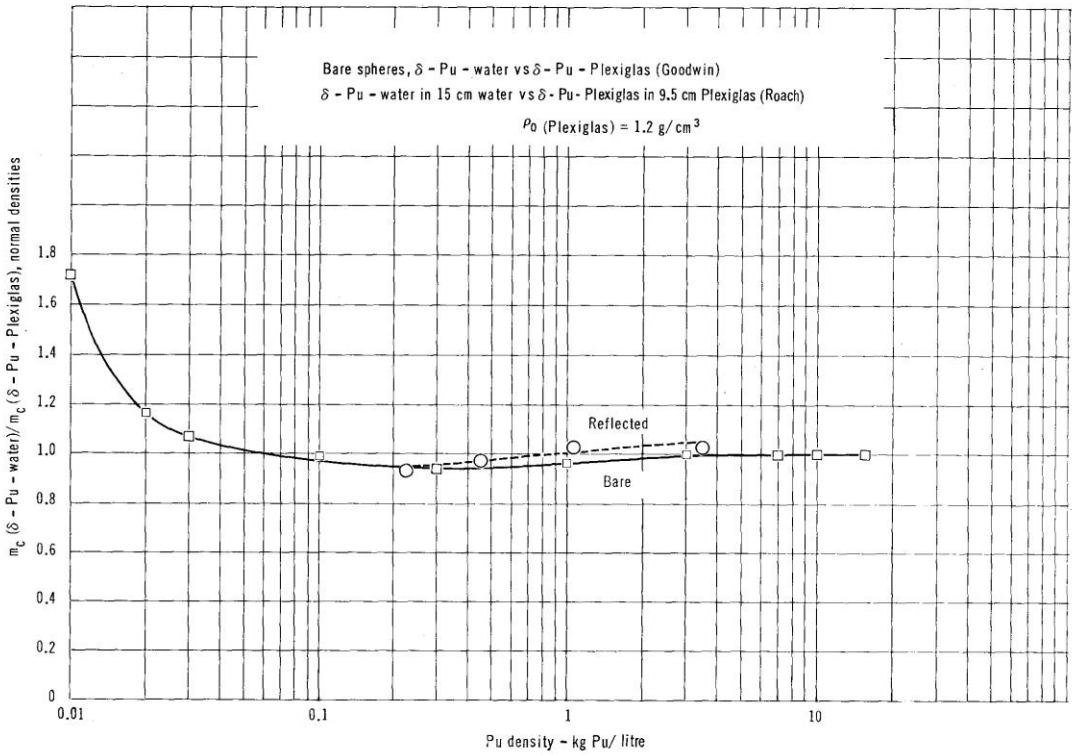

Figure 15. *Computed relationships between critical masses of delta-phase Pu mixtures with water and with Plexiglas.*

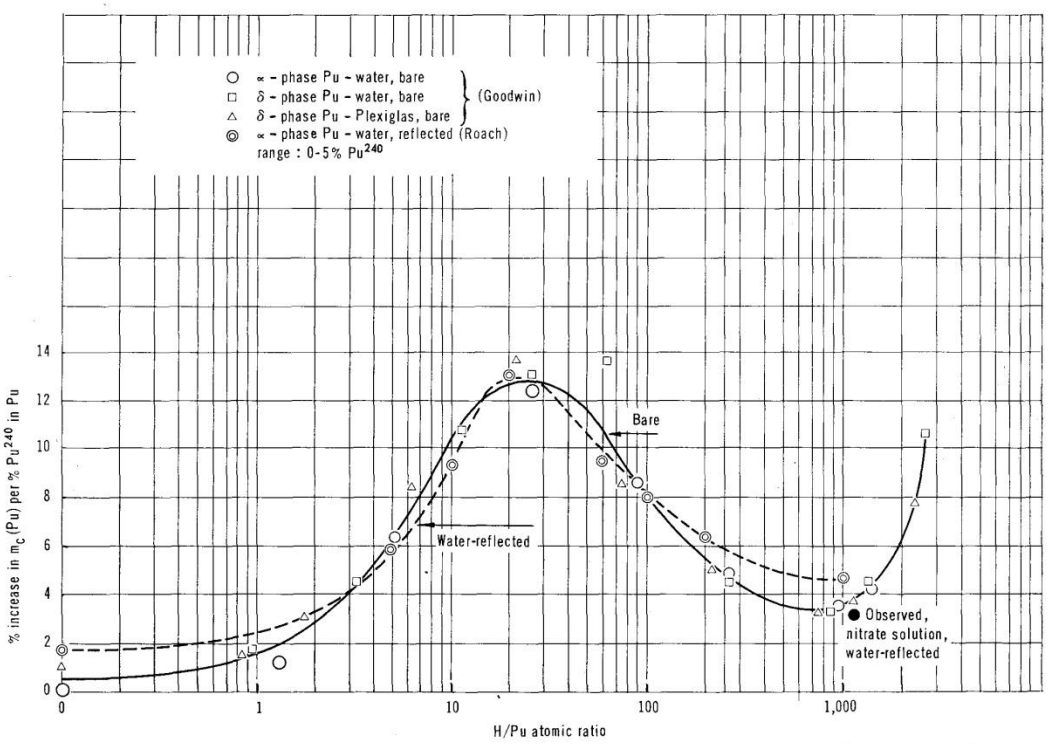

Figure 16. *Computed influence of Pu 240 on critical mass of water-moderated Pu spheres.*

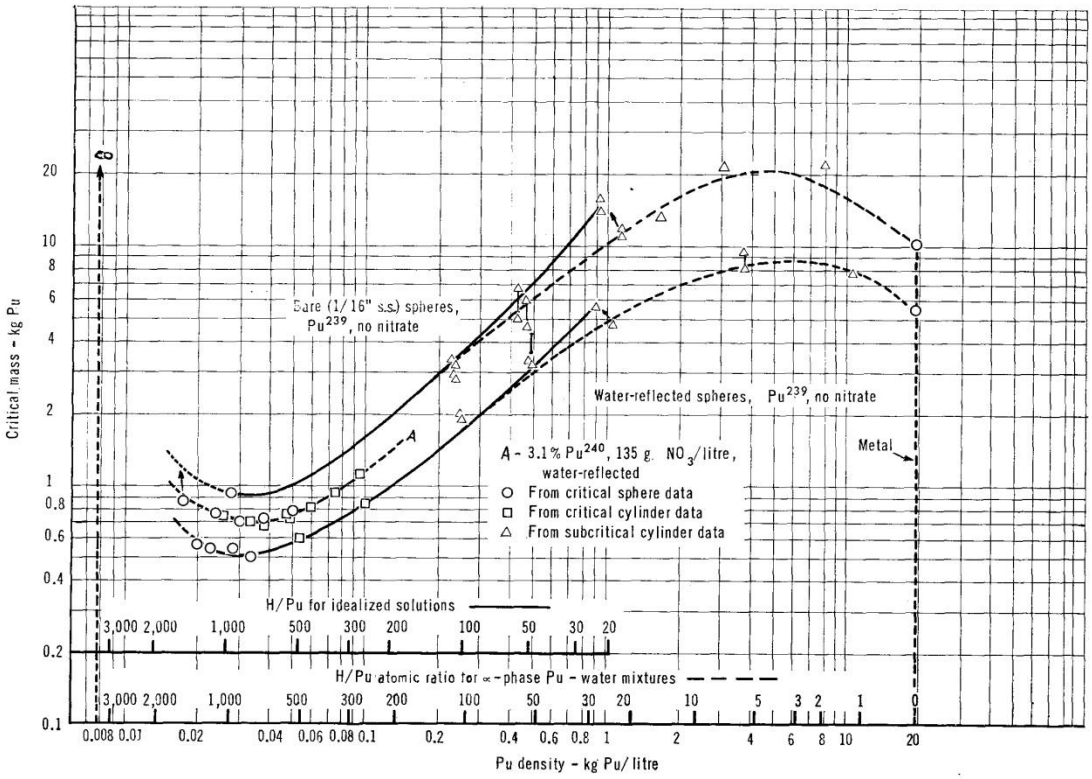

Figure 17. *Reduced experimental critical masses of homogeneous, water-moderated Pu spheres.*

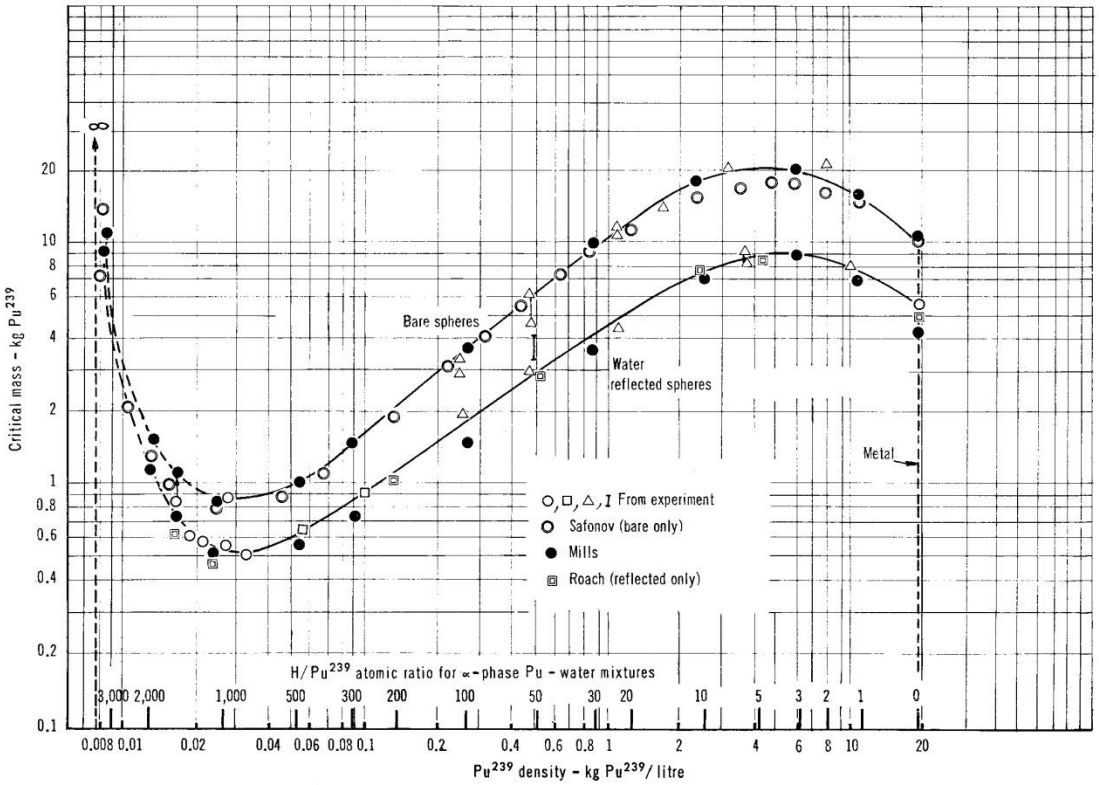

Figure 18. *Computed and experimental critical masses of homogeneous, water-moderated Pu 239 spheres.*

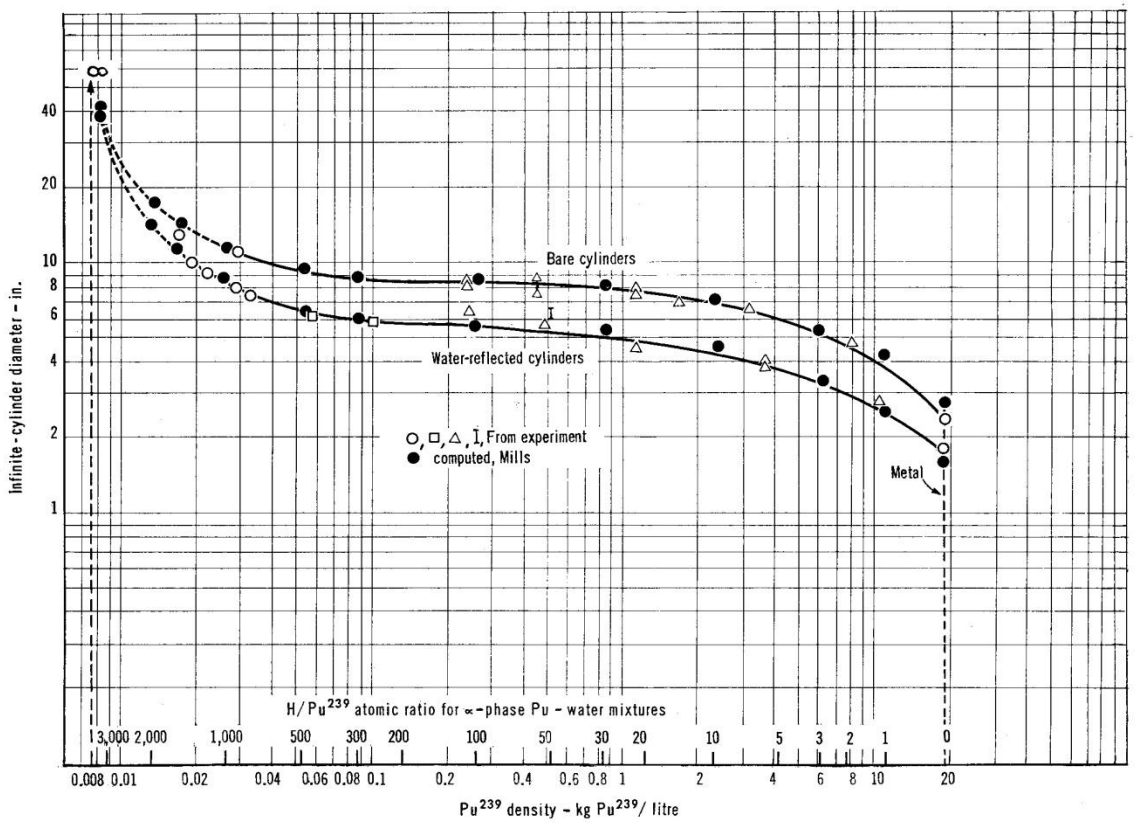

Figure 19. *Computed and experimentally-estimated critical diameters of infinitely-long cylinders of homogeneous, water-moderated Pu 239.*

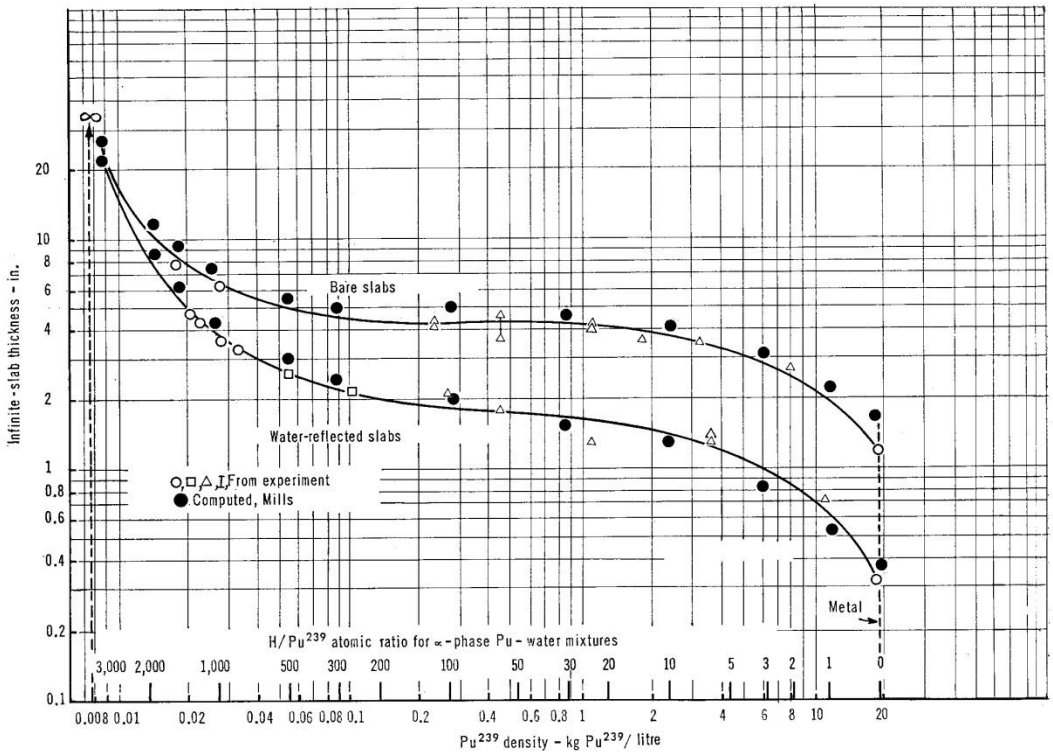

Figure 20. *Computed and experimentally-estimated critical thicknesses of infinite slabs of homogeneous, water-moderated Pu 239.*

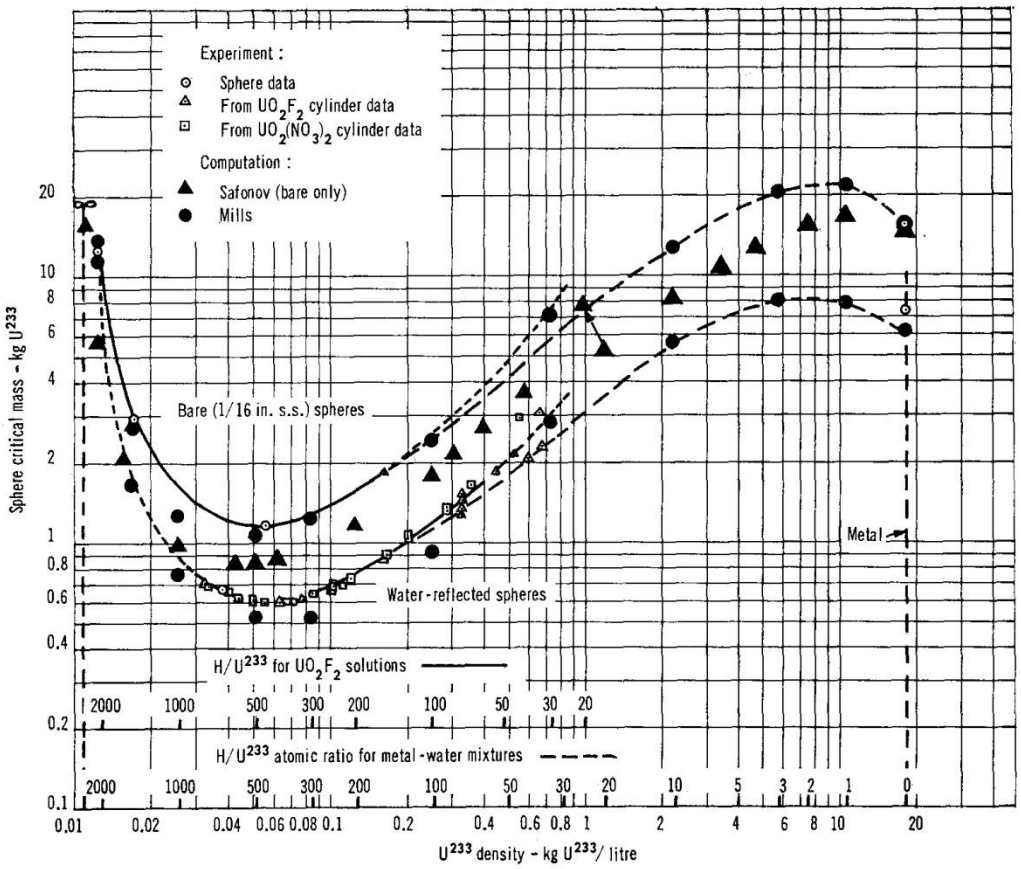

Figure 21. *Computed and experimental critical masses of homogeneous, water-moderated U 233 spheres.*

Computation with Limited Experimental Confirmation

Fissile systems that are sensitive to more than two constituents demand computational guidance to a greater extent than the essentially two-component mixtures upon which we have concentrated. The simple reason is that the new dimension (or more) makes complete experimental coverage prohibitive. Examples of practical interest are U 235-U238–H_2O combinations for partially-enriched fuel element materials, U235–C–H_2O for impregnated graphite elements, and U 235–Be–H_2O for impregnated beryllium.

As the transportation and general handling of partially-enriched uranium compounds (which may become moist) is a current problem which requires attention, let us see what is known about critical masses of water-moderated U 235–U 238 combinations. The principal family of curves in Figure 24 are from Safonov for a temperature of 260° C (500° F) but corrected to normal metal-water densities. To indicate the order of neutronic changes when the temperature is dropped to 20° C, Mills calculates a 25 % decrease in the minimum critical mass for U(2 % U 235) versus a decrease of just 5 % for U(93).

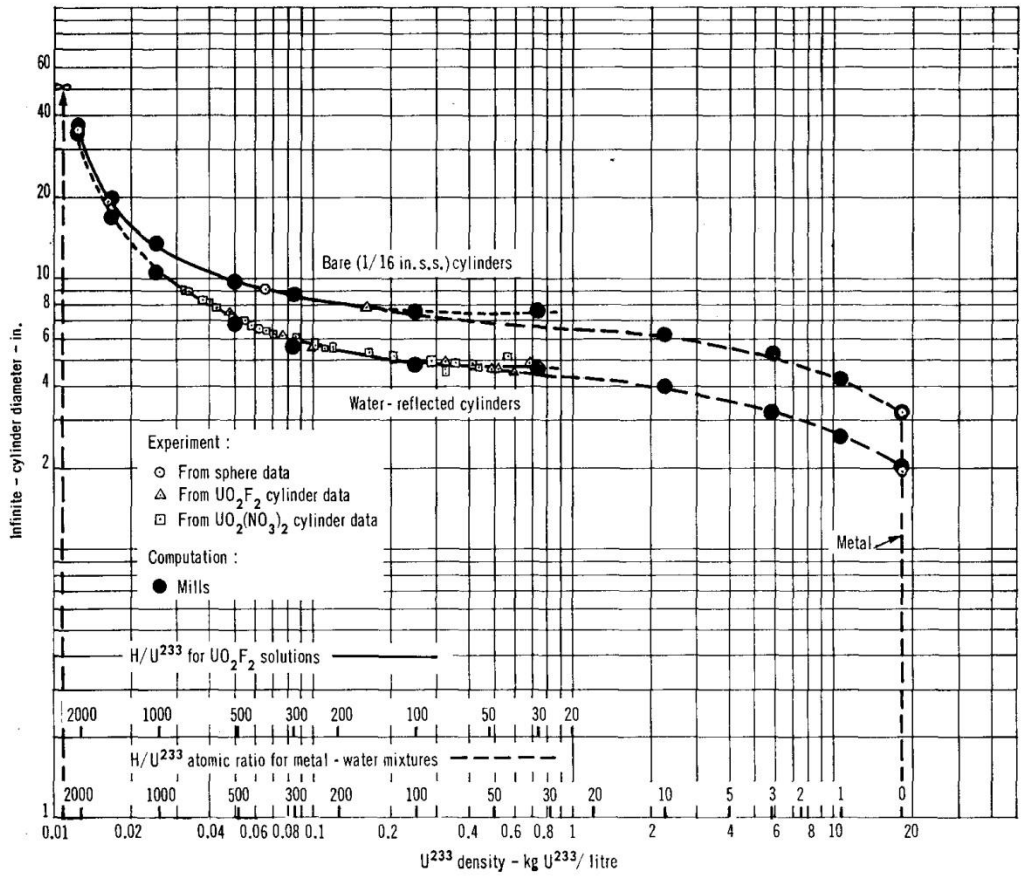

Figure 22. *Computed and experimentally-estimated critical diameters of infinitely-long cylinders of homogeneous, water-moderated U 233.*

The gas model which he used may overestimate the influence of temperature change, and we do not know what differences Safonov might have found. So it is with apology that we have added to Figure 24 room-temperature points from experiment and from a preliminary survey by Mills (for this survey, convergence requirements were $\sim 1\,\%$ on radius instead of the customary factor of 10 less). Though temperature corrections would bring data of the three general types more or less together, limitations on experiment, uncertainties in Mills' calculations and the inappropriate temperature of Safonov seem to combine to call for a thorough computational survey. Mills has undertaken this for U 235–U 238 metal-water spheres of which preliminary results appear in Figure 25 (Though reduced to metal-water densities, the experimental points are still influenced by compositional differences.) Figure 26 in the form of Figure 7 includes points based on further results for water-reflected systems.

The next illustration is a set of critical masses for U(93)–C–H_2O which was computed by Stratton specifically for nuclear safety (Figure 27). As method and cross section sets are identical to those used by Roach, comparisons

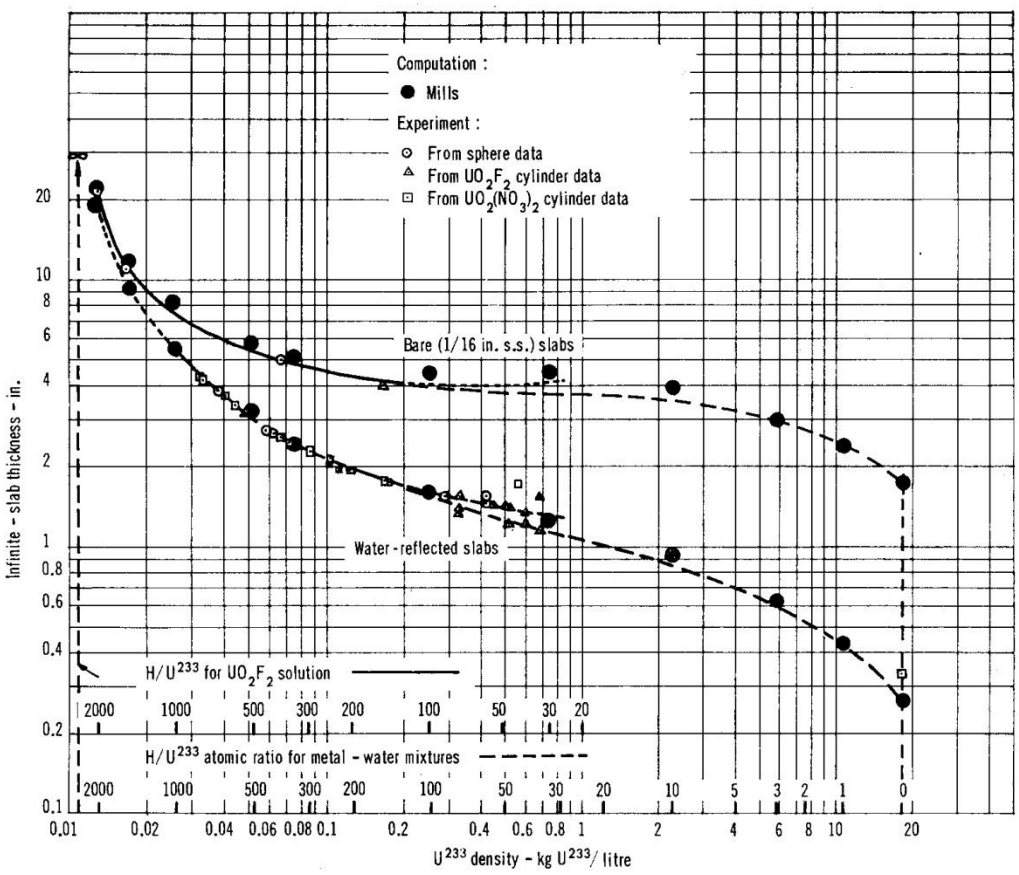

Figure 23. *Computed and experimentally-estimated critical thicknesses of infinite slabs of homogeneous, water-moderated U 233.*

of Roach's results and experiment for straight water moderation (Figure 9) are pertinent. Computed critical masses for graphite-U 235 systems fall closely with those of Mills on Figure 8. The few experimental values are from subcritical measurements and all but the left-hand point refer to systems with inhomogeneous hydrogen distribution. Errors of interpretation could be as great as departures from computed values. For our purposes, limits were established by scaling down computed curves to pass through the lower experimental points, then applying a factor of safety of at least two. From the apparent reliability of calculations with each moderating component separately and by comparison with the crude points for ternary assemblies, it is expected that most of this safety factor will be preserved.

Comments on Reliability and Safety Factors

The correlations which we have seen should leave us with a feeling as to the potential reliability of both experimental and computed critical parameters. In general, experimental data fall into two categories: precision

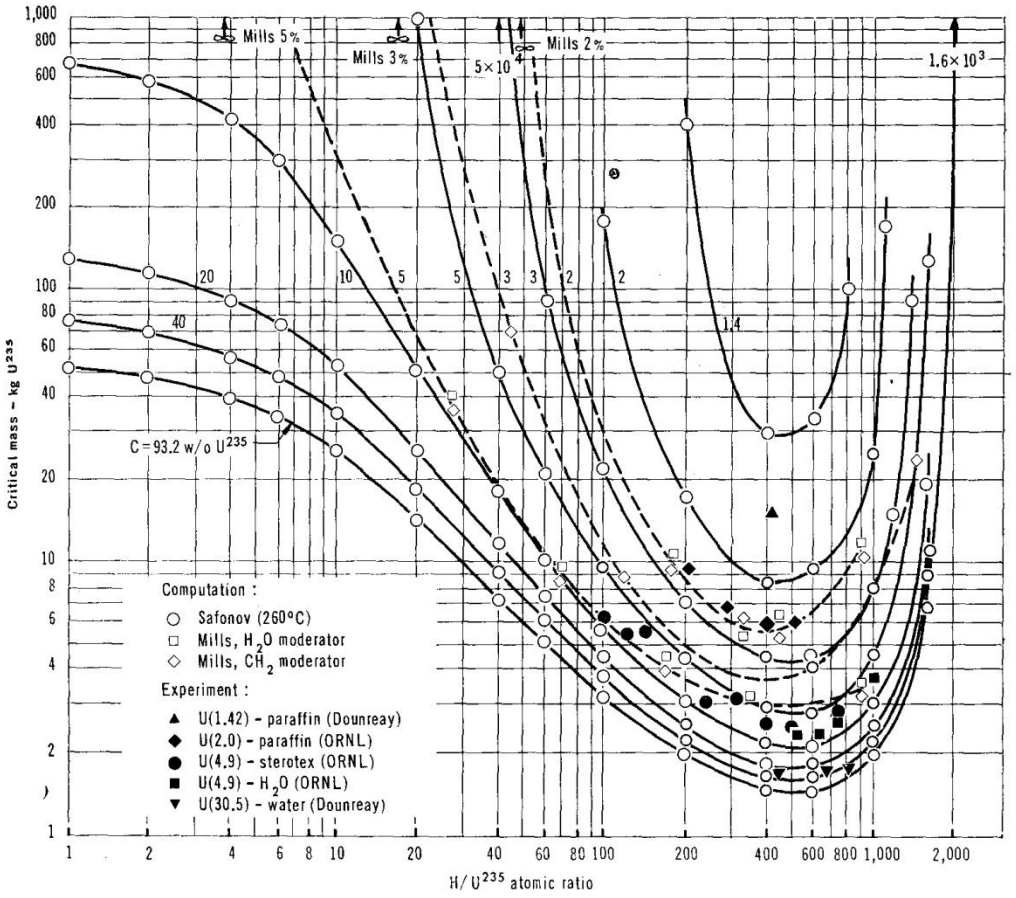

Figure 24. *Critical masses of bare U 235-U 238-H_2O spheres at metal-water densities.*

measurements (critical or near subcritical) which require only well-defined corrections to standard conditions; and information which, because of imprecise critical determination, unclean assembly or a degree of inappropriateness, serves only as a crude guide. Fortunately, there are reasonably abundant (though never sufficient) data of the former class.

The ideas that we have developed about the precision of detailed computation may be supplemented by what can be judged from certain controlling input parameters and their reliability. To illustrate this point, Figure 28 shows results of DSN computation (R. Aamodt, unpublished) which extend the very limited experimental values of Figure 3. The answer to the question as to whether the calculations for dilute ranges may be used for nuclear safety guidance is not a clear " yes " or " no ". Within regions to the right of upturns toward asymptotes, departures from the important anchor-point for undiluted uranium are controlled by relatively straightforward scattering properties of the diluent. Critical mass values will not be high by a factor of two if capture by the diluent has less than 10 % influence on $\bar{\eta}$, the effective excess of neutrons produced per absorption. This criterion will be satisfied over

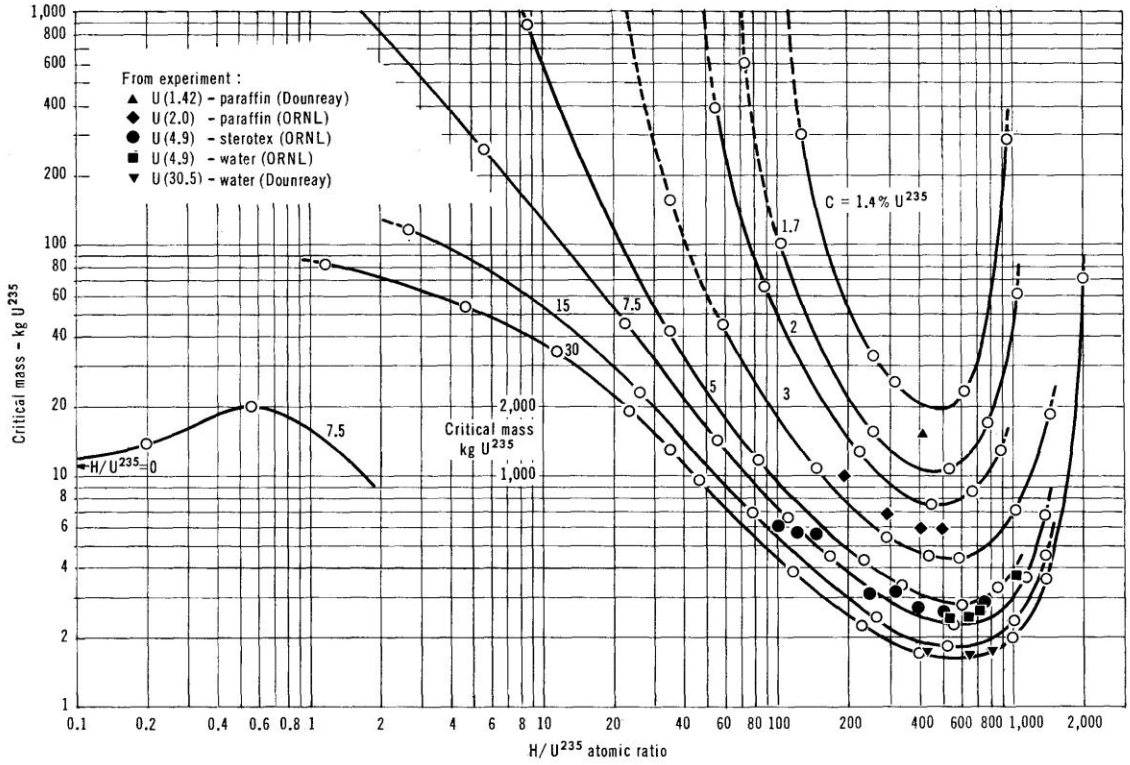

Figure 25. *Critical masses of bare U 235-U 238-H_2O spheres at metal-water densities as computed by Mills.*

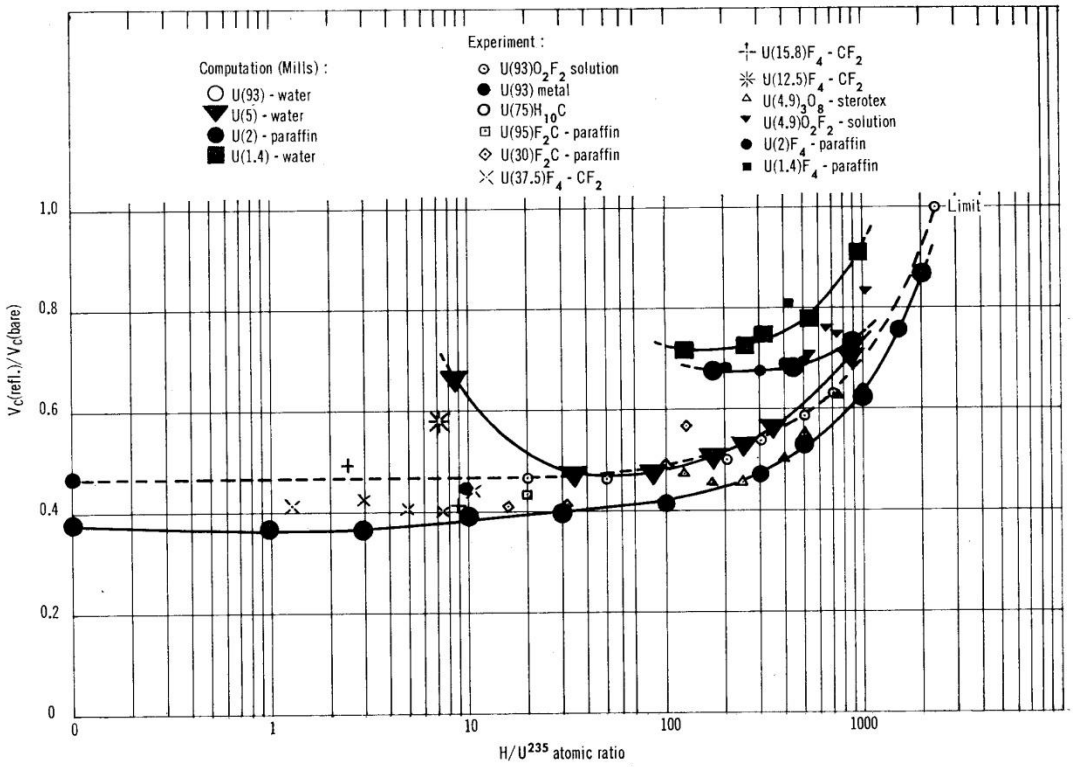

Figure 26. *Ratio of water-reflected-sphere to bare-sphere critical volumes for hydrogen-moderated U 235.*

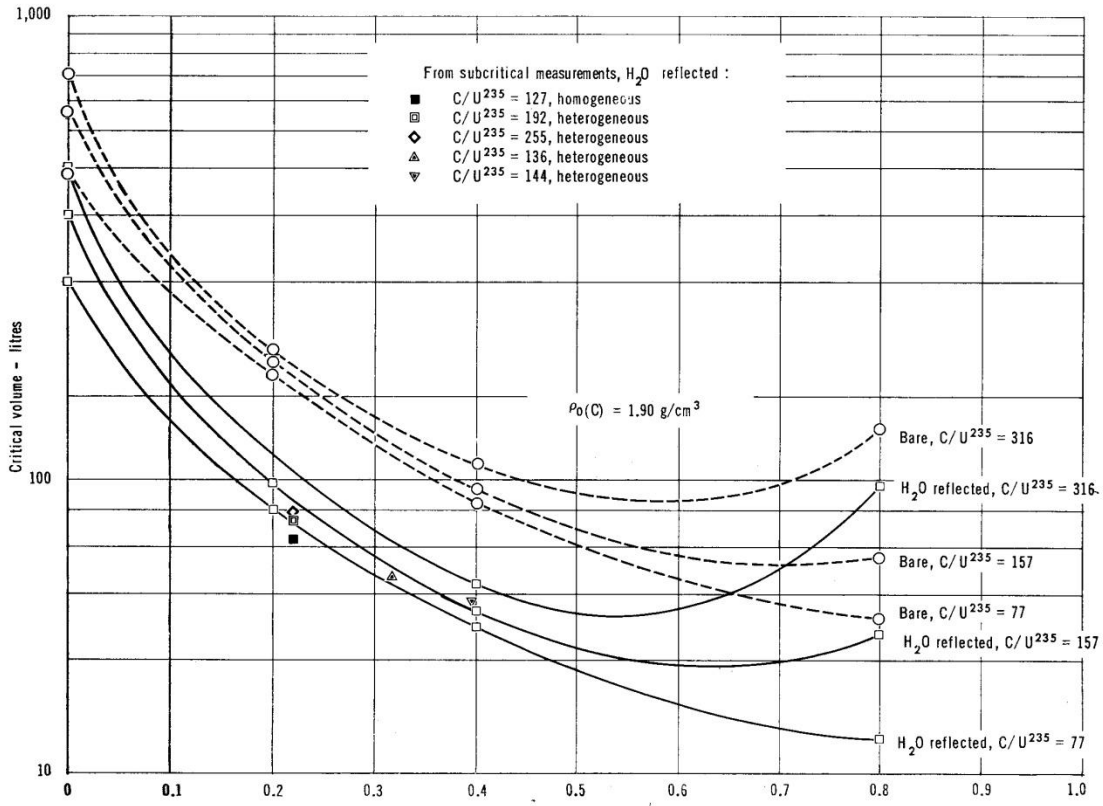

Figure 27. *Critical volumes of U(93)-C-H_2O systems, as computed by Stratton.*

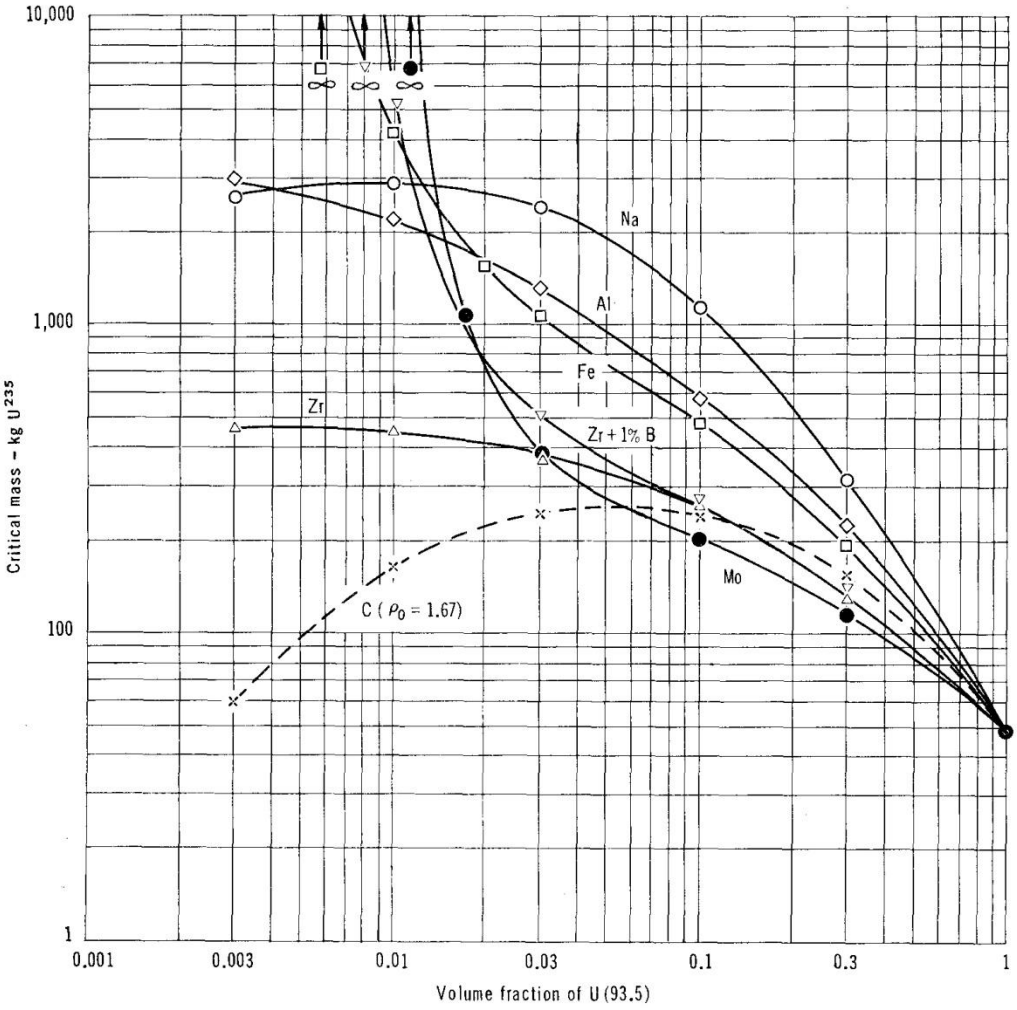

Figure 28. *Critical masses of bare spheres of U(93.5) diluted with other metals (and graphite), as computed by Aamodt.*

most of the range that we are considering. Where curves break away toward asymptotes, however, critical masses (and the limiting critical compositions) become extremely sensitive to capture cross sections, for which 50 % errors have been common. In view of this I would not use, for nuclear safety purposes, either critical masses above the scattering base or asymptote locations unless there were experimental confirmation. (Actually the need for such guidance would imply the existence of enough material to make possible some sort of experimental check.)

Before expressing an opinion about the general reliability of detailed computation, I wish to make a side-line observation about non-input errors which are not uncommon. Any detailed calculation such as DSN is similar to a complex experiment that requires special procedures to verify the proper

functioning of equipment and guard against extraneous effects. With the ever-present possibility of transcription error or machine malfunction, there should be a careful consistency check of any isolated problem before the result is accepted. The advantage of a parametric survey is that difficulties of this type are likely to result in inconsistencies that are obvious. Provided calculations have been properly checked, I believe that a probable error in critical mass of $\pm$ 10 — 15 % is about right for existing DSN techniques with proven cross section sets. This does not apply to regions in which critical masses change precipitously with composition.

A complete discussion of factors that should be applied to critical data for establishing nuclear safety limits would require a full description of the operations for which the limits are intended and of the nature of controls. I do not know how to generalize beyond the recommendations, and added precautions, which appear in the Nuclear Safety Guide [13]. When the necessarily arbitrary safety factors which appear there are translated to new situations, it is obvious that there should be added allowance for uncertainties in data. The importance of such an allowance diminishes if the limit applies to an idealized situation that is actually unapproachable in practice. Examples would be the application to normal vessels of limits for fully-reflected systems, and storage-shelf limits that are based on a cubic array of maximum size units at the minimum allowed spacing. Hidden, but generally large, factors of safety in these cases greatly relieve the demands on guiding data. Information specific to a plant layout, in contrast to that which appears in our surveys, would, of course, eliminate many of the hidden safety factors that are now necessary.

Concluding Remarks

Any worker in the nuclear safety field, when faced by gaps in critical data, feels that his efforts are tentative. In exactly this sense the present review is recognized as describing just the beginning of an effort that should be carried on by many. A continiung, possibly co-ordinated, computational programme is needed — and let us hope that it will be of the best.

In representing detailed computational methods by the DSN and multigroup diffusion techniques, it has not been the intention to discriminate against alternatives such as Monte Carlo. This method could make its appearance when correlations are extended beyond the simple systems that we have considered.

REFERENCES*

1. H.C. Paxton, " Critical Data for Nuclear Safety Guidance ", *LAMS-2415* (1960).
2. J.T. Mihalczo and J.J. Lynn, " Multiplication Measurements with Highly Enriched Uranium Metal Slabs ", *CF 59-7-87* (1959).
3. E.R. Woodcock and H.C. Paxton, " The Criticality Aspects of Transportation of Fissile Materials ", in *Progr. Nucl. Energy, Technology, Engineering and Safety, Series IV*, Vol, 4. Pergamon Press. London, 1961,
4. D. Callihan, " A Review of Criticality Data obtained by Experimental Methods ", see this Conference, p. 139.
5. D.F. Cronin, " Critical Mass Studies, Part X: Uranium of Intermediate Enrichment " *ORNL-2968* (1960) (class.) : and

   J.G. Walford *et al.*, " Criticality of 30 % U 235 Solutions in Cylindrical Geometry ", *DEG Memorandum 663* (1960); also private communication.

6. G. SAFONOV, " Notes on Multi-Group Techniques for the Investigation of Neutron Diffusion ", *R-233* (1952):
G. SAFONOV, " Survey of Reacting Mixtures Employing U 235, Pu 239, and U 233 for Fuel and H_2O, D_2O, C, Be, and BeO for Moderator ", *R-259* (1954): and
G. SAFONOV, " Critical Mixtures of Uranium and 500° F Light Water ", *R-279* (1955).
7. C.B. MILLS, " Minimum Critical Dimensions for Water Solutions ", to appear in *Nucl. Sci. Eng.* (1961).
8. B.G. CARLSON, " The S_n Method and the SNG and SNK Codes ", *AECU-3691* (1958) :
B.G. CARLSON, " The DSN and TDC Neutron Transport Codes ", *LAMS-2346* (1959): and
B.G. CARLSON, " The DSN Code ", *LAMS-2346* (Appendix) (1959).
9. G. BELL, " The Theory of Effective Cross Sections ", *LA-2322* (1959).
10. G.E. HANSEN and W.H. ROACH, " Sixteen-Group Cross Sections ", *N-2-753* (Rev. 1), Los Alamos (1960) : and
W.H. ROACH, " Computational Survey of Idealized Fast Breeder Reactors ", *Nucl. Sci. Eng. 8*, 621 (1960).
11. A. GOODWIN Jr. and C.L. SCHUSKE, " Plexiglas and Graphite-Moderated Plutonium Assemblies ", to appear in *J. Nucl. Energy* (1961).
12. C.L. SCHUSKE, G.H. BIDINGER, A. GOODWIN Jr. and D.F. SMITH, " Plutonium Plexiglas Assemblies ", *RFP-178* (1960) : and
G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Plutonium Plexiglas Assemblies, Part II ", *RFP-190* (July 1960).
13. D. CALLIHAN, W. J. OZEROFF, H.C. PAXTON and C.L. SCHUSKE, " Nuclear Safety Guide ", *TID-7016 (Rev. 1)* (1961).

* Most references to experiments are contained in References [1] and [13].

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DISCUSSION

D. BRETON, France:

J'aimerais demander à M. Paxton s'il peut faire quelques commentaires sur la courbe Figure 16 qui montre l'influence du Pu 240 sur la taille critique de sphères modérées à l'eau.

H. C. PAXTON, United States:

I did not quite understand the point to be discussed here, but I imagine it is this relatively sharp, almost resonance-like influence of Pu 240, and this is largely the effect of the capture resonance of lowest energy (at about 1 eV). For the method in which the cross sections through the resonance region are averaged — and actually for the entire cross section sets that were used for these calculations — I might give a reference that will appear shortly, a Los Alamos report, designated LAMS-2543. This report gives the Hansen-Roach cross sections and includes the method of averaging the cross sections through the resonance range.

R. J. ACKROYD, United Kingdom:

I have a remark from my colleague, Mr. Pendlebury : " In connection with the use of the Carlson method for slab geometry and the need to go to a high S_n approximation, referred to by Dr. Paxton, it is worth noting that $\Phi(x, \mu)$ the neutron current in direction μ, where $\mu = \text{Cos } \theta$ (θ being the angle between the x axis and the neutron direction) is a discontinuous function of μ at $\mu = 0$ on the boundary. The direct application of the Carlson method does not give this discontinuity, but the approximation to it is better when a high S_n approximation is used, and this may possibly account for the need for a high S_n approximation in slab geometry. "

Session III

APPLICATION OF CRITICALITY CONTROL PRINCIPLES TO PLANT DESIGN

APPLICATION DES PRINCIPES DE CONTROLE DE LA CRITICALITÉ DANS LA CONCEPTION DES INSTALLATIONS

Chairman - Président:

Prof. M. ZIFFERERO

(Italie)

SAFE-BY-SHAPE CHEMICAL PLANTS

K. M. HILL AND H. E. PARKER
United Kingdom Atomic Energy Authority

INTRODUCTION

Safe-by-shape chemical plants are plants utilizing unique safety precautions and designed to handle fissile materials arising from reactor and weapons programmes. In processing fissile materials the designer has a choice of five concepts for criticality control:

1. *Mass limitation.* A mass-limited plant is safe by virtue of the operator's having an exact knowledge of the location of all fissile material within the plant and by his exercise of control to ensure that the mass at any location does not exceed the minimum critical mass for the conditions applying there.
2. *Concentration limitation.* A concentration-limited plant is safe by virtue of the operator's maintaining the fissile material in a specified form and at concentration less than that determined to be safe for the particular location within the plant.
3. *Safe by shape.* A safe-by-shape plant is safe by virtue of the fact that its physical dimensions and shape are such that a critical state can never be achieved under any conceivable conditions of concentration, chemical form, homogeneity and degree of moderation applicable to the plant and process.
4. *The addition of nuclear poisons.* Such a plant is safe because the poisons added are present in such amounts and at such locations that the neutron multiplication factor is always less than unity. Poisons may be present in the soluble or insoluble forms.
5. *A combination of* (1) — (5). The concept is not in favour with operating staffs for obvious reasons but is nevertheless frequently involved to some degree.

Before undertaking the design of a plant, the concepts should be compared for safety economy. At the present state of development of the atomic energy industry safety has the predominant rôle, and here the safe-by-shape concept is undoubtedly an easy winner since all other concepts have a continuing risk of human error. Concentration limitation is a sound policy for plants extracting plutonium from irradiated natural uranium where throughputs are high and the concentration of fissile material, at least until the point of separation from uranium is reached, is extremely low. The addition of soluble nuclear poisons looks attractive but safety considerations demand costly instrumentation and the method is chemically very limited in application.

Permanently fixed insoluble nuclear poisons, i.e. absorbers, may be considered as an extension of the safe-by-shape concept.

For high-enrichment plants, therefore, the choice for the designer is usually between mass limitation or safe-by-shape. The mass-limited plant is usually a batch or a semi-continuous process suitable for small throughput. Where throughputs amount to a large number of critical masses per annum a safe-by-shape continuous process shows considerable savings, particularly in operating costs, in addition to enhanced safety. The purpose of this paper is to discuss the problems confronting the designer of safe-by-shape processing plants and to show how some of these problems have been solved.

Limited-geometry plants are frequently thought of in terms of squeezing very large amounts of material through plant of narrow cross section. In many cases this problem does not arise on anything like the scale that might be supposed. Because of the intrinsically small quantities of fissile material involved in any programmes utilizing atomic energy, the throughputs of many plants are frequently much the same as more conventional plants in terms of throughput per unit cross section. This, however, is not so likely to be the case in the future. For example, a 15,000 MWe fast reactor programme could well entail processing in the order of 100 kg plutonium/day. This is far in excess of present throughputs available from safe-geometry equipment.

General Conception

High throughput, safe-geometry plant requires to be truly chemically engineered from start to finish and the selection of the optimum processing steps can ease the whole problem of design and operation. For this reason continuous processes tend to be favoured, these generally possessing the attribute of high throughput.

The operating costs of fissile material plants are very high compared with conventional chemical plants, largely because of the small scale of operation. The staffing of such plants is related to the number of plant units and has little relation to the actual throughput through an individual unit. In many cases the same staff could operate a non-fissile plant many times the size. Capital costs of fissile material plants are high, the greatest proportion of the cost going into health and safety measures associated with alpha hazards and building space required for criticality reasons. Much of this money is devoted to making plants relatively foolproof.

There is little point in designing safe-by-shape chemical plants unless they are safe under all conditions of fault operation, otherwise a false sense of security may be generated in the operating staff. These conditions are often difficult to define, particularly as the development work aimed at producing a new plant has usually been concentrated on the correct process conditions up to the time when the plant design commences, and the true chemistry involved in the fault conditions is frequently determined only towards the close of the development programme. These problems usually boil down to the definition of maximum fissile material densities and degrees of hydrogen moderation. A particularly important step in the design of many plants is the determination of those sections in which the presence of metal and high-density compounds cannot be envisaged; in these sections the problems of specifying plant geometry are relatively simple. In the sections where moderated metal or high-density compounds can occur, the geometry

is much more restrictive. The clearest possible thinking is required at this stage of design to ensure that these restrictions are only applied where strictly necessary since their effect on plant cost is great.

Interpretation of Criticality Data

Although many data have been produced on unreflected and partially-reflected conditions, it is rarely possible to make use of these conditions. The degree of reflection provided by shielding walls is frequently indeterminable at the early stages of design and it is usually necessary to proceed on the basis of fully-reflected conditions. The correctness of this approach is borne out in practice when on the final criticality clearance of the plant it is generally apparent that in very few cases could less reflected conditions have been specified.

In that section of the plant handling solutions of fissile materials the interest is mainly centred on vessels of which one or more of the dimensions may be considered infinite, from the criticality viewpoint. Fortunately, with U 235 the critical diameters of cylinders and thicknesses of slabs do not alter very much over a considerable range up to densities in excess of those likely to be achieved by precipitation from an aqueous solution (see Figure 1).

The position with Pu 239 is not so favourable, but in large-scale power programmes a mitigating factor arises in that high-irradiation plutonium contains considerable proportions of the higher isotopes and this tends to produce curves of critical dimensions *versus* density similar in nature to those of U 235 (see Figure 2).

The determination of the safety factors to apply to the critical dimensions is best left to the specialist criticality expert since these are dependent principally on the accuracy of the original experiments and the calculations which have succeeded them. From the nominal safe dimensions so determined, the engineer usually selects suitable nominal sizes close to them. As the throughput of plants increases, this approach will have to be modified and sizes of equipment much closer to the safe size will have to be used rather than nominal pipe sizes.

The section of plant handling aqueous solutions is typified by solvent-extraction equipment, evaporation and suchlike operations; this type of plant frequently requires many units in succession and the optimum utilization of space requires that these should not be arranged in a single line. A useful approach here is to consider the plant as a lattice of infinite cylinders and to determine the optimum diameter/spacing arrangement. This approach is exemplified by the experimental fast reactor chemical separation plant at Dounreay. Figure 3 shows a cross section through the plant. It can be seen, particularly in the case of the low-active cell, that horizontal mixer settlers, i.e., infinite cylinders, are laid out on a lattice of approximately 2 ft 6 in. square pitch. Whilst individual pipes carrying solutions from one vessel to another are usually of too small a diameter significantly to affect the criticality problem, it is always possible that an assembly of pipes might do so, and the adoption of the lattice arrangement enables the designer to route his pipework approximately along the lattice points and thus avoid further criticality problems. Similar arrangements may be used for vertical systems such as columns where the pipe runs again tend to run parallel to the vessels.

The greatest problems in plant design are presented by those sections of the plant handling high-density materials, e.g. metal or oxide, and in par-

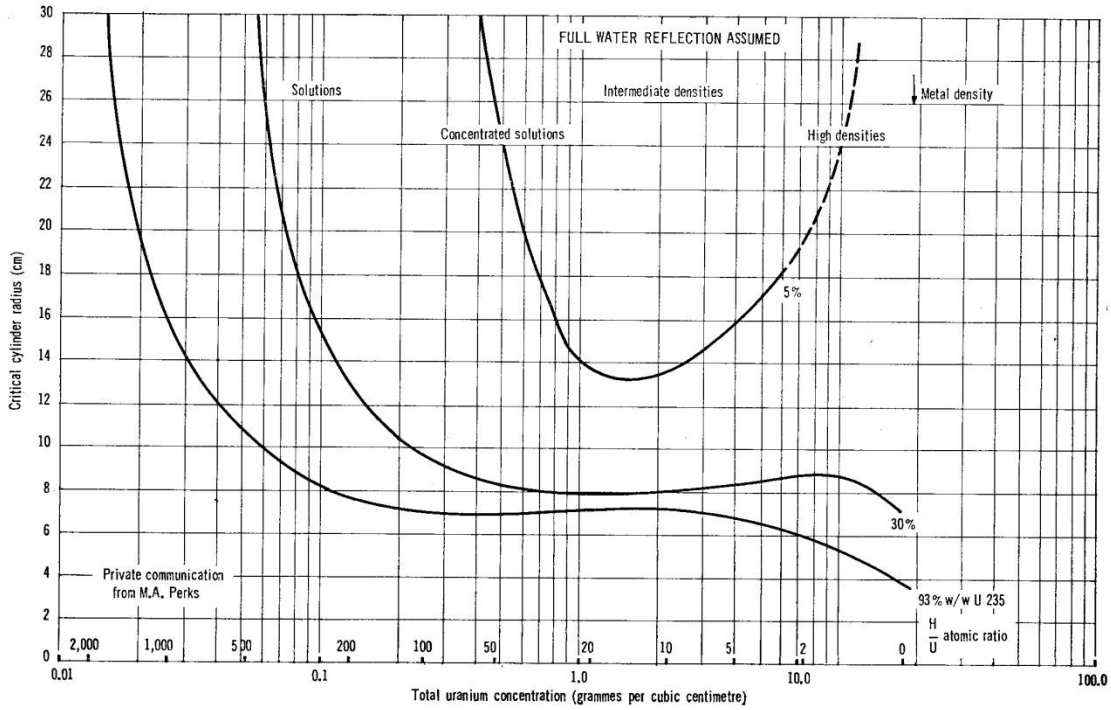

Figure 1. *Smallest critical infinite cylinder radius against uranium concentration for 5, 30, 50, 93 % w/w U 235.*

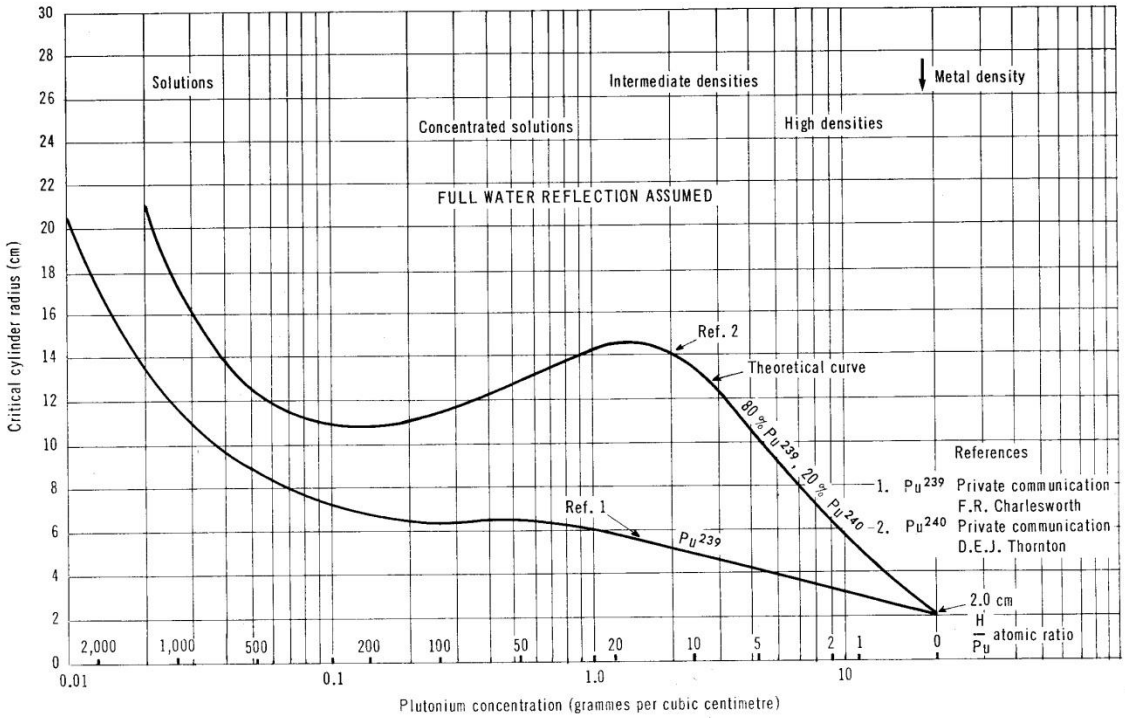

Figure 2. *Smallest critical radius of infinite cylinder against plutonium concentration.*

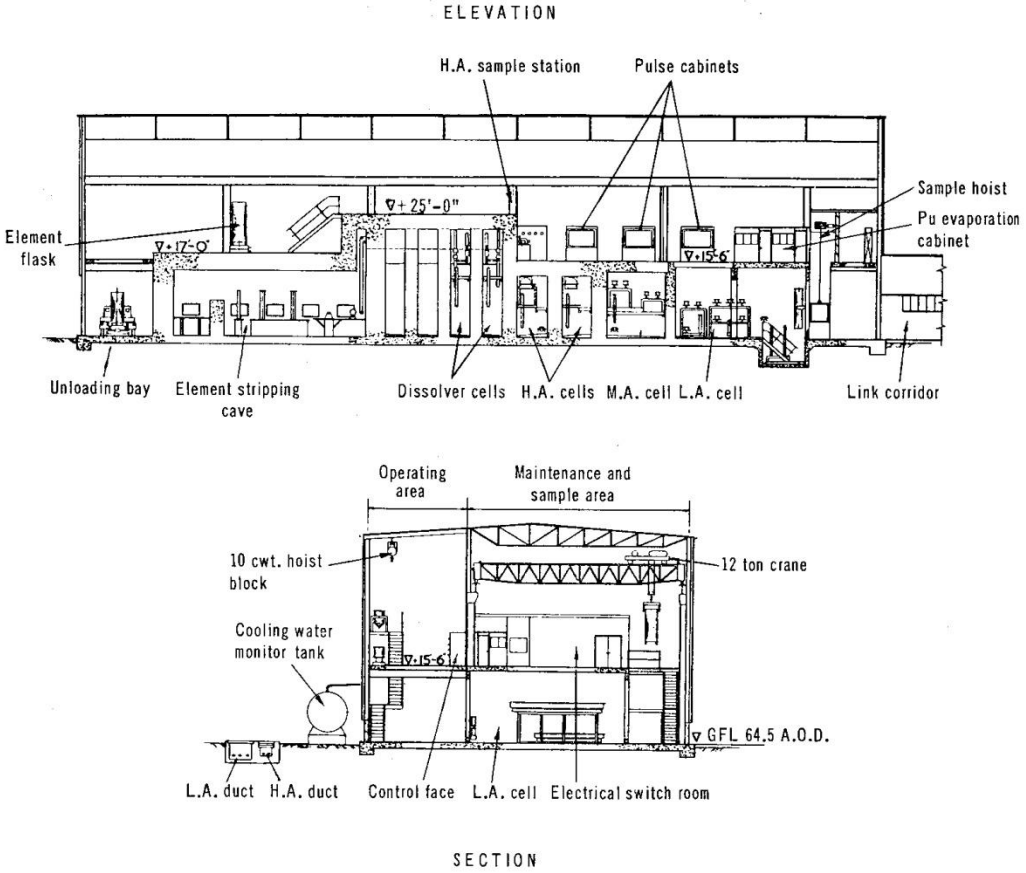

Figure 3. *Fast reactor chemical plant.*

ticular by dissolvers. In theory, by using suitable conditions, very high rates of dissolution of many compounds and the metals themselves are obtainable, but such rates of dissolution would present excessive entrainment and foaming problems. It therefore follows that any practical dissolver for metal or oxide is likely to have a very considerable hold-up of high-density material surrounded by the dissolving agent. The critical diameter for U 235 for such a system is about 2.8 in. and for Pu 239 about 1.5 in. and it follows that the capacity of such dissolvers is very small. Dissolvers made in slab geometry achieve a considerable increase in the hold-up capacity but are nevertheless still very restrictive. Figure 4 shows a batch dissolver built on a cylindrical basis capable of handling U 235: Figure 5 shows a slab dissolver for a similar duty. Both these dissolvers are based upon infinite geometries and have very restricted charging arrangements. If the charging arrangements or other process considerations require dimensions bigger than these infinite geometries, recourse can occasionally be made to increases in the critical masses and volumes available by utilizing cylinders or slabs of defined length-to-diameter ratio, although the advantages gained are very limited.

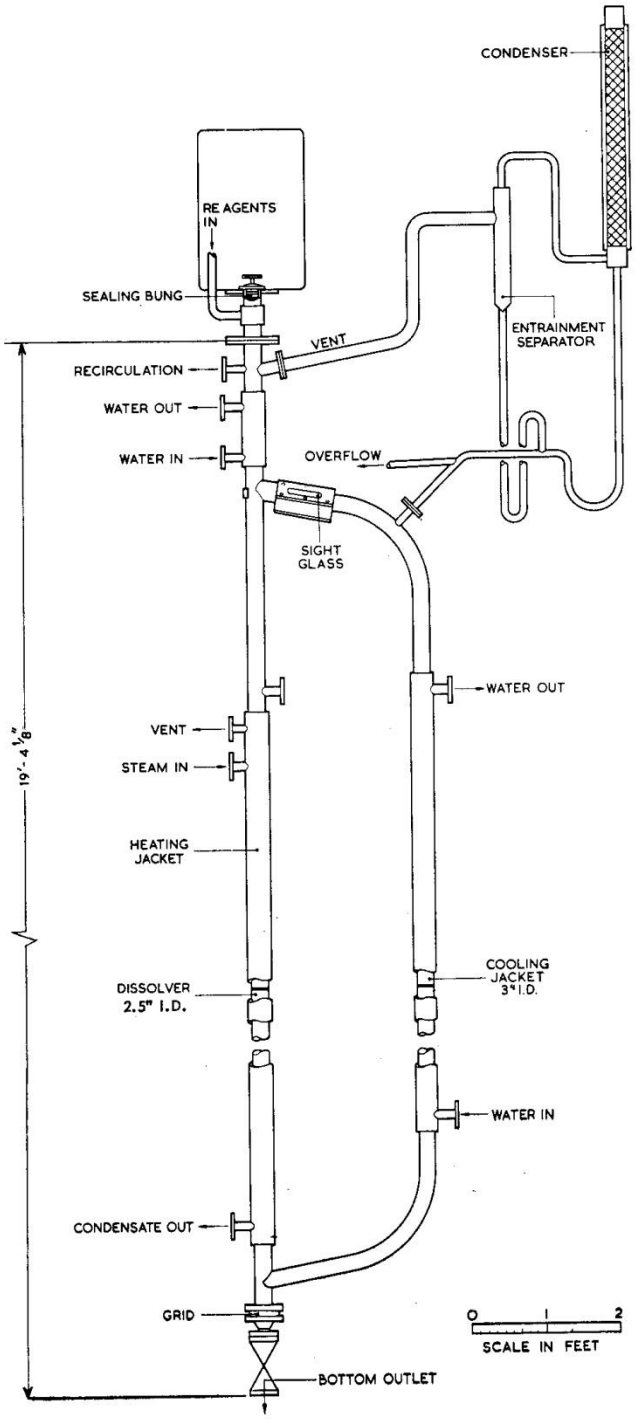

Figure 4. *Batch dissolver for U 235.*

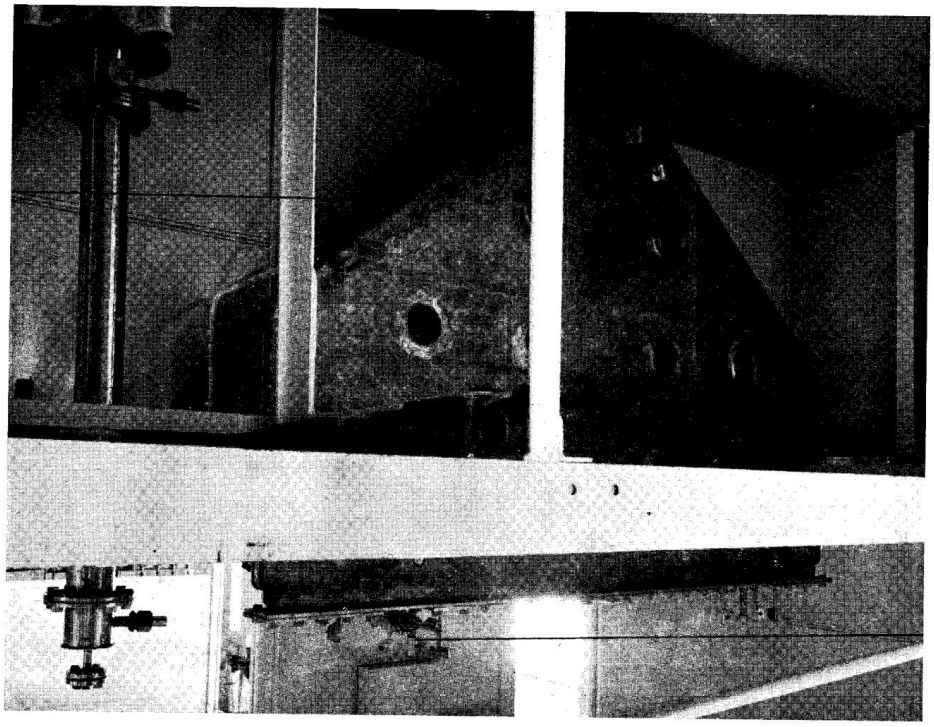

Figure 5. *Slab batch dissolver for U 235.*

If the use of these methods fails to produce a vessel of the required capacity, a combined control of mass, concentration and geometry must be resorted to. Figure 6 shows a continuous metal dissolver based on these principles. Hold-up in the dissolver is limited for safaty to 12 kg whereas the normal operating hold-up is 6 kg, these conditions being defined for the worst concentration conditions in the dissolver and the limited geometry of the dissolver. In operation the input and output are checked and operation can continue until the out-of-balance reaches 6 kg, a figure which in practice allows the dissolver to operate for considerable periods.

It is common in conventional chemical plant to change the cross section of the plant when the process requires it. The obvious limitation in these directions for safe-by-shape plants tended to lead to chemical engineering solutions which are compromises and retain the same geometry through all

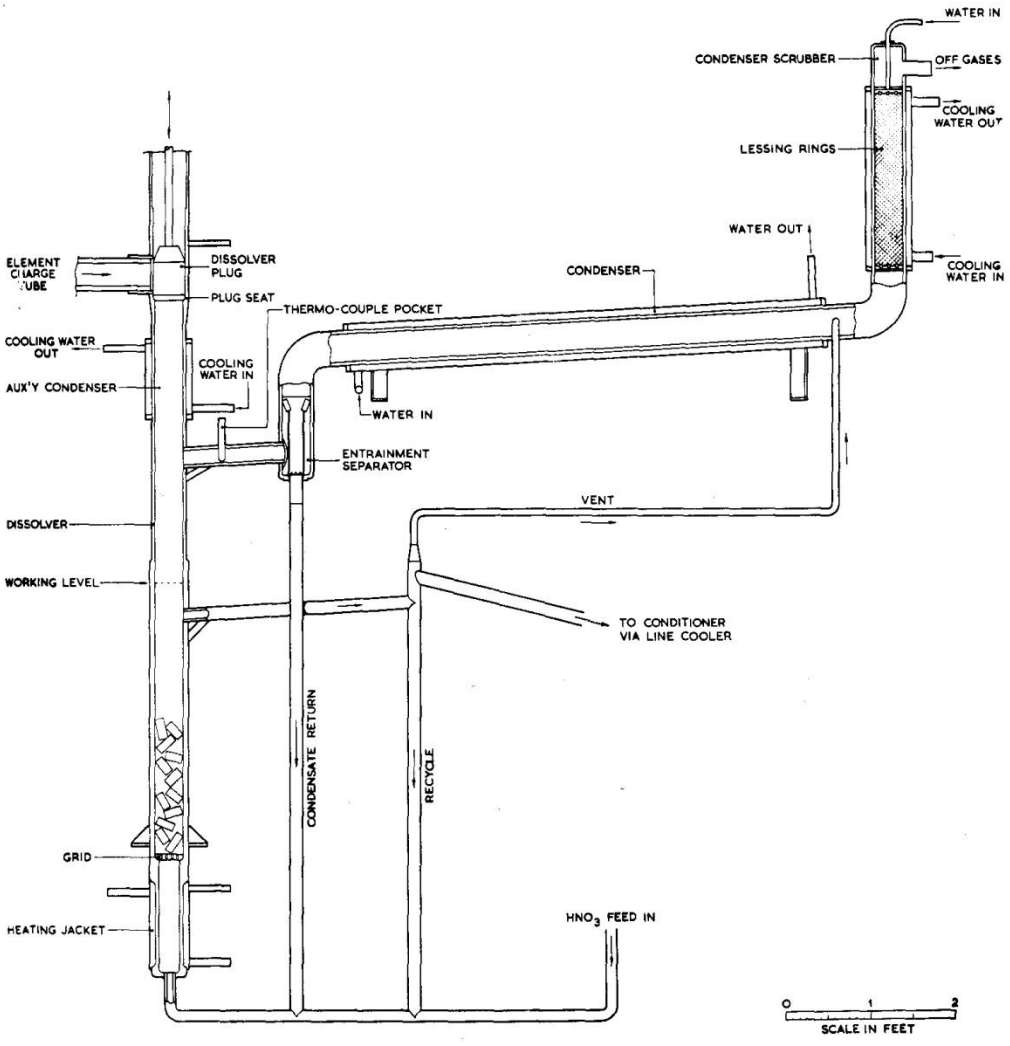

Figure 6. *Continuous metal dissolver for U 235.*

sections. This, however, is not necessary. As an example, a slab mixer-settler for use with Pu 249 solutions may be considered. In safe-by-shape plant the slab thickness will be limited to 0.83 in. for solutions, a figure which is not really adequate for satisfactory control of settling phenomena; but by reducing the slab thickness to 0.75 in., it is possible to have 3 in. dia. vertical cylinders 18 in. long intersecting the slab. These could be used for interface control and collection of clear settled liquids and additionally for the provision of agitator drives, etc. (see Figure 7). This example is not quoted as a necessarily satisfactory answer to the particular problem but merely to demonstrate the various ways in which criticality data may be used by the engineer.

A difficulty which often arises from the use of these approaches is that the engineer evolves a scheme which he thinks will satisfy the process and then consults the physicist as to the criticality safety of design. He then finds that the advantages which he thought to have gained by the particular design are lost because of the uncertainties in the interpretation of data which lead to higher safety factors being required on the unproved geometry. This problem may be overcome where a criticality facility with adequate resources of material is available to test the design directly. For those not so fortunately endowed the position is steadily improving, as more and more data on different assemblies accumulate. The concept of direct testing may also be usefully extended to many specific items of equipment such as pumps, special designs of evaporator, etc. In many cases it would be cheaper to test commercially available items of equipment which would appear to have a chance of avoiding critical dimensions than to develop special items.

Process Problems

In all safe-by-shape plants, sooner or later the process material that has been in the plant has to leave the safe geometry, and a key point in design is the decision on where this shall be. It is significant that most of the criticality accidents that have occurred in nuclear chemical plants have taken place at such changeover points.

The handling of the major proportion of the fissile material into and out of the plant is normally covered by transport considerations. Material arriving at the plant will come in transport containers designed to avoid criticality problems, and similar considerations apply to material leaving the plant. However, the plant has to be fed with non-nuclear materials in order to carry out the process, and effluent will accumulate from all purification stages; in addition, various ventilation systems, wash-down points, etc., all accumulate amounts of fissile materials to some degree and these must be disposed of. The decision has to be made on whether these should be of safe geometry or not.

Apart from the direct question of safety it is one of the primary objects of safe-by-geometry plant to avoid the expensive and tedious business of accurate fissile material accountancy directed at safety requirements. This accountancy can only be avoided if absolute guarantees can be given that at no time will fissile material accumulate in unsafe geometry. It is therefore necessary to ensure that all effluents enter drainage arrangements such that precipitation cannot occur. This can present problems in the case of unirradiated fuel plants since these are likely to be sited on general-purpose sites handling many other processes at the same time. It is good practice in general to operate factory drains under neutral or slightly alkaline conditions, but

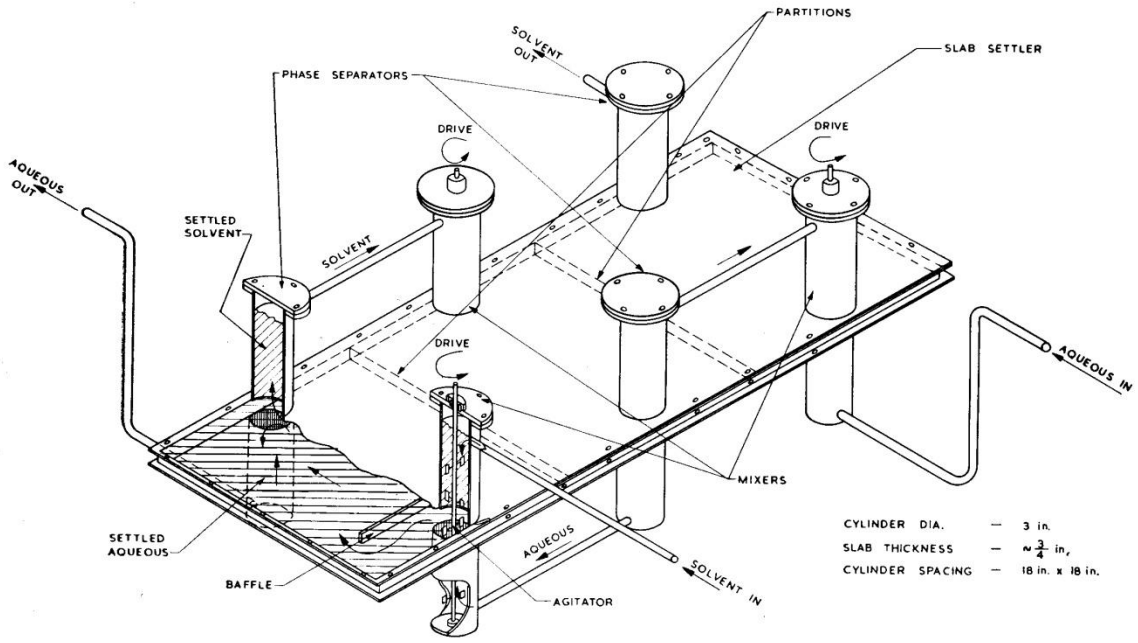

Figure 7. *Slab mixer settler for Pu 239.*

these conditions can produce embarrassing situations if fissile material is allowed to enter the drainage system. It is thus necessary to monitor reliably all material leaving the plant, a requirement leading to costly analyses since automated monitors cannot usually be relied upon to the degree of accuracy required. The alternative sometimes acceptable is to provide the plant with its own effluent-treatment system.

Irradiated fuel plants, on the other hand, will not have the problem of disposing of small quantities of fissile material by dilution in large quantities of the drainage. They have, however, the problem of the accumulation of fissile material over a long period of operation such that a criticality situation could occur in the fission product storage tanks. A recent development in fission product disposal makes it possible to avoid this situation. The fission product fixation process as developed at Harwell [1] proposes to incorporate fission products into glass which is manufactured in small-diameter, stainless steel cylinders, these cylinders being sent to permanent storage, thus preserving safe-by-shape conditions to the very end.

The principles of safe-by-shape design should be extended to the ventilation system of such plants, particularly where dry solids are being handled. In practice such dust as does settle out is likely to be spread throughout the system, but the possibility of an accumulation always remains, and under such circumstances prudence will dictate a return to accurate fissile material accounting unless the system is safe-by-shape. The use of fissile-material accountancy for controlling criticality involves as a principal element of its cost the shut-down of plant for washing out and general recovery of material. It is thus prudent to design safe-by-shape plants so that accountancy may be carried out readily if circumstances arise in which the original safe-geometry conception is not being fulfilled, and a prime requirement of this is the provision of adequate wash-down facilities. Plant leakages and spillages must be guarded against, usually by the provision of a suitably-tanked stainless steel floor, care being taken that this floor area is adequate to contain all material which can collect on it without exceeding the critical thickness of an infinite slab, and also that interaction with neighbouring pieces of equipment is avoided.

As mentioned previously, safe-by-shape plants are usually continuous processes; whilst this is in many cases dictated by problems of throughput it is also a very desirable feature to avoid the difficulties of handling material between batch processes. Figure 8 illustrates the problem in connection with batch calcination furnaces. It can be seen that a conveyor system has to be used in order to ensure that material passing to and from the furnaces follows a pre-determined path, and a system of interlocks is used to ensure that supercritical masses cannot accumulate at one point. On the other hand, a continuous fluid bed unit (see Figure 9) avoids this problem by using pneumatic conveying through safe-geometry pipes.

Although not strictly a chemical problem, many irradiated fuel plants have to de-can mechanically the fuel entering the plant. On the small scale this is best coped with by mass control. Future plants handling large quantities of fissile material from power programmes will have to reconsider the problem and it may well be that de-canning machines designed to preserve safe geometry during the de-canning operation and in the subsequent transport will be required.

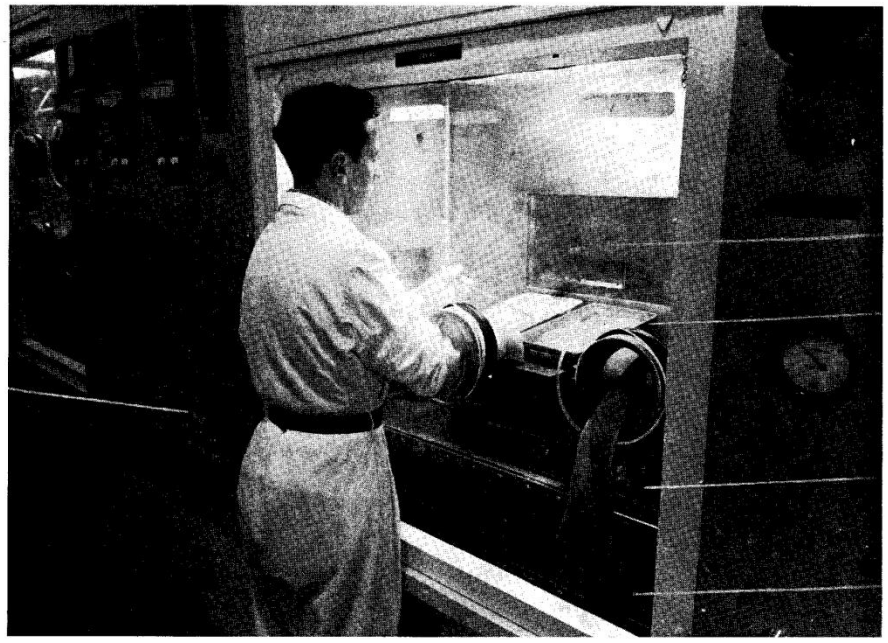

Figure 8. *Batch dryway furnace for U 235.*

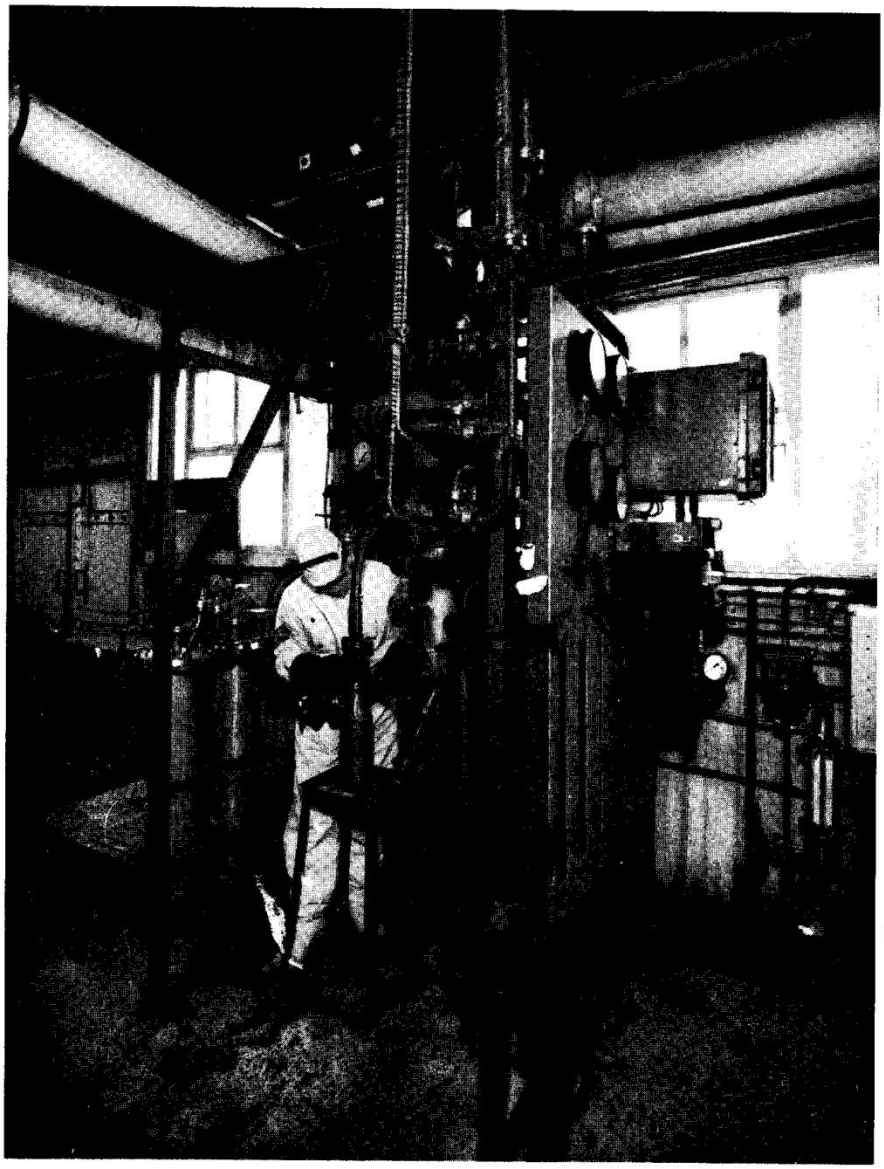

Figure 9. *Experimental 3 in. dia. fluid bed denitrator.*

The provision of adequate storage volumes for process purposes always presents a difficult problem for safe-by-shape plants. While storage in slab tanks tends to be more expensive, in terms of equipment, than storage in tubular tanks, the slab-tank system has many advantages with regard to the mixing and sampling of the contents. Both systems require an inordinate amount of space and many ingenious solutions have been suggested to enable the storage tanks to occupy volumes in the plant which would not otherwise

be utilized. The high cost of slab-tanks is largely due to the problem of building such tanks of adequate rigidity to ensure dimensional stability. There is considerable scope for improved design here, and this, coupled with the possibilities of much closer spacing than has previously been thought allowable, may well alter the position with regard to their economics.

The principles of design of plants for handling low-enrichment fissile materials are not essentially different from those of high fissile material content. In practice, plants at present handling low-enrichment materials tend to be mass- and concentration-limited, but this is largely by virtue of employing existing facilities designed for handling natural uranium. Plants specially designed for these materials would principally differ from plants handling material of high fissile content in that in many stages of the process the limiting dimensions would be infinite or so large as to cause no restrictions to normal plant design. In such plants the accumulation of critical masses of material involves the accumulation of such a large physical quantity that inspection can readily ascertain whether a dangerous situation is likely to exist or not. Nevertheless, it should not be thought that the problems of such plants are negligible, and the adoption of a large-scale nuclear power programme involving enrichments in the 2-3 % region would undoubtedly present many difficult criticality design problems.

Mechanical Design Problems

Safe-by-shape chemical plants present their own peculiar problems of detail design, particularly in the case of those handling irradiated fuel. The thickness of the vessel walls required for corrosion resistance is not less in safe-by-shape plants than in non-fissile plants; it follows therefore that the smaller safe-by-shape vessels require considerably smaller radius/thickness ratio bends, etc. An example of this problem is illustrated in Figures 10 and 11 which show cross sections of a mixer-settler as used in the experimental fast reactor chemical separation plant at Dounreay. This vessel is designed to the equivalent of a 4-in. dia. cylinder. To minimize the problems of weld corrosion it was desirable that the outer shell of this vessel be in one piece. The subsequent fabrication of these vessels up to 20 ft in length with many intermediate partitions, weirs, etc., presented many problems with regard to maintaining the necessary straightness and freedom from residual welding stresses. The finished vessels are shown in Figure 12 which also illustrates the arrangement of vessels and pipework upon a pitched lattice basis.

Many items such as pumps and agitators are not commercially available to industrial standards on this scale of operation and a great amount of design effort is required to produce them. The design costs of safe-by-shape chemical plants tend to be much higher, in relation to capital expenditure, than conventional plants; this is because the effect of scaling down shows up more in the capital costs of actual equipment than in the costs of designing units.

Chemical Engineering Design of Plant Units

Safe-geometry plant does not differ from conventional chemical plant in that it is governed by the same factors of chemical kinetics and the usual chemical engineering considerations of heat and mass transfer, etc.; more-

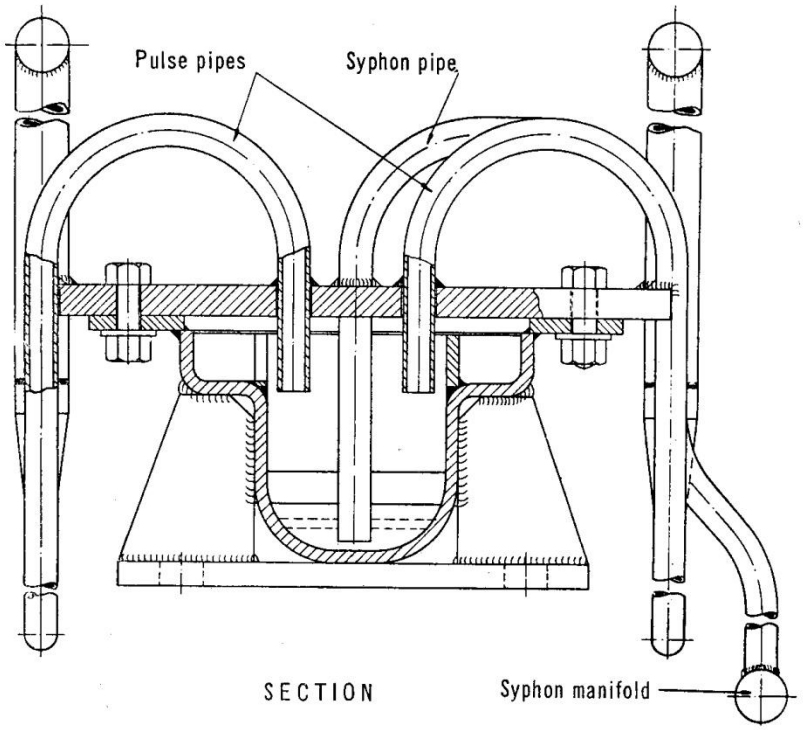

Figure 10. *Typical mixer settler.*

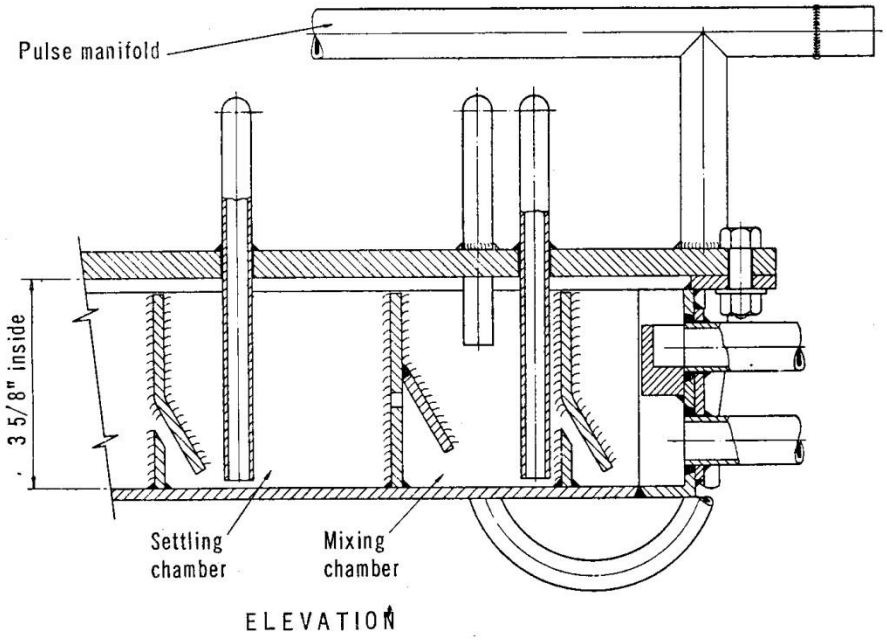

Figure 11. *Typical mixer settler.*

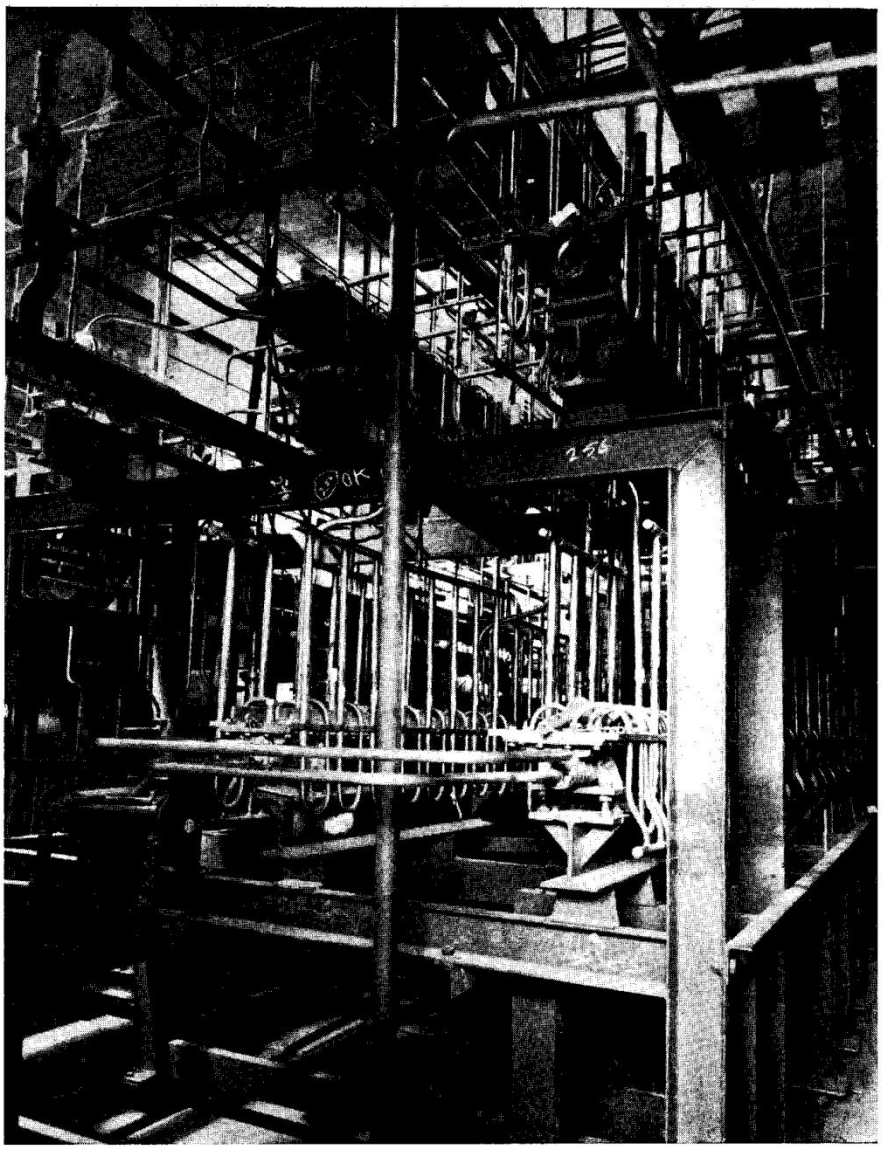

Figure 12. *Low-active cell, D.F.R. chemical plant.*

over, the throughput in terms of fissile material is largely a function of the flowsheet conditions. It would not, therefore, be relevant to this Symposium to discuss the detailed performance of particular items of equipment; the exception to this is those cases where the restricted geometry imposes special limitations to chemical engineering design.

The problems of foaming and entrainment in dissolvers and evaporators have already been touched upon. These problems are severe in the case

of safe-by-shape plant owing to the difficulty of obtaining adequate disengagement in narrow cross sections, tending to cause stabilization of foams. Similar problems arising from geometrical restrictions occur with fluid beds, where, to get adequate capacity and sufficient time for complete reaction, it is frequently necessary to consider beds of a length/diameter ratio which is excessive by normal standards. Problems of this type make it difficult to obtain unit cross section throughputs as high as those obtained in conventional plant, unless special precautions are taken.

Heat transfer from steam or cooling water presents a special problem in that, unless it can be guaranteed that no leakage will occur or that the pressure in a steam or water jacket will always be higher than that within the equipment proper, the design has to accommodate the heating or cooling jacket within the safe geometry. For example, in the slab dissolver illustrated previously (Figure 5), the heating jacket was arranged along the edges of the slab, thus not exceeding the slab dimensions. Similarly, the metal dissolver shown in Figure 6 incorporated its heating jacket in a narrowed section of the dissolver. It thus becomes difficult to incorporate large heat-transfer areas in such equipment and these can, in certain cases, become limiting factors to the design throughput.

The ultimate solution to the problem of high-capacity, safe geometry plant would appear to lie more in the field of radical changes of the process rather than in detailed chemical engineering considerations of particular designs. Thus the U.S. Atomic Energy Commission's design for a hex flame reactor [2], whilst not directed primarily at the problem of criticality control, shows admirably what can be achieved by departing to a radically new process (see Figure 13).

The Dounreay Fast Reactor Chemical Separation Plant

The process and general engineering design of the Dounreay fast reactor chemical separation plant has been described previously by Buck *et al* [3]. In the present paper it is proposed to describe some of the criticality thinking that went into the design of the individual items, but it should be realized that this is not necessarily the same as that which forms the basis of the independent criticality clearance approved by the Health and Safety Organization.

The plant was designed to handle 93 % U 235 although it will actually handle 46 % U 235 in its early life.

The irradiated fuel elements entering the plant are first of all de-canned in suitable equipment in an active cave. The portions of the fuel element are then fed by a conveyor to a dissolver cell. All vessels have been considered as infinite cylinders of a cross-sectional area not exceeding 13 in^2. These are laid out on a 2 ft 6 in. minimum-dimension approximately rectangular lattice. Such vessels are safe in plane array for U 235 concentrations not exceeding 2 g U 235/cm^3 [4] and the combined array is safe since vessels do not present more than one steradian of solid angle to any one vessel [5].

The dissolver itself is considered on a different basis, as previously described. On a volume basis the 4 in. cylinder of the dissolver becomes critical at an H/U ratio of 8. At this H/U ratio, however, the critical spherical mass is 15 kg [6]. It follows that 15 kg in a 4 in. cylinder is safe for all H/U. The right-angle bends in the 4 in. dia. cylinders forming the dissolver condenser require that some reduction be made in this amount, and a figure of 12 kg has been adopted. Because of the high fissile content, heterogeneity does

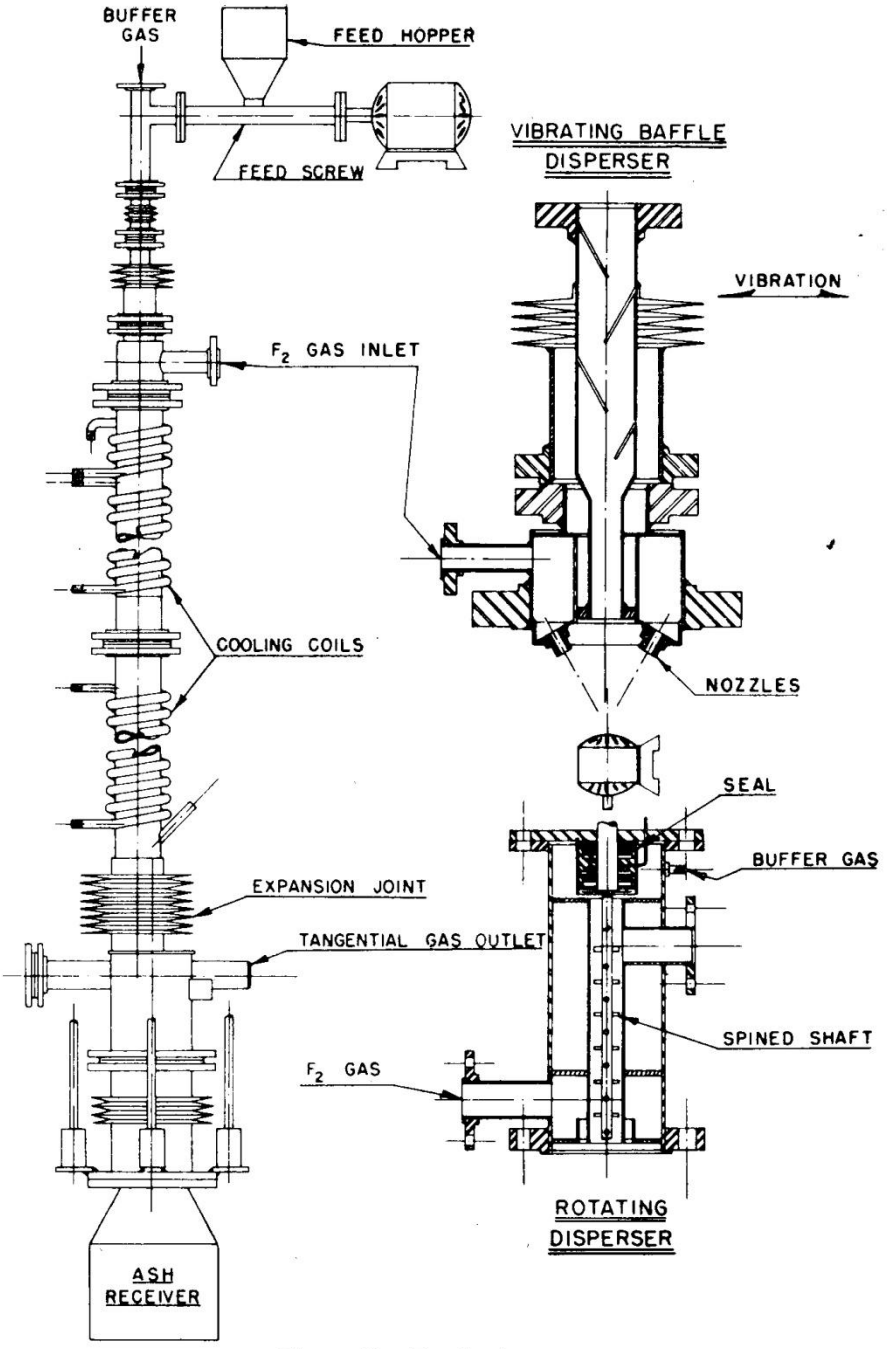

Figure 13. *Fluorination reactor.*

not adversely affect the result and the dissolver is therefore designed to be operated on a maximum out-of-balance, including the dissolver hold-up, of 12 kg. As previously pointed out, the normal operating hold-up is 6 kg, thus giving a considerable operational period between wash-outs.

The condensers and conditioners in the cell have water jackets of a diameter greater than 4 in. Corrosion data indicate for these vessels a life of ten years with a safety factor of 125.

The fuel coming into the plant will, of course, contain Pu 239 and this had to be considered in the design of the plant. It can be shown that 500 g Pu 239 in a 4 in. dia. vessel cannot exceed 150 g/litre without an increase in the safe mass to greater than 500 g owing to the decreased H/Pu ratio. It follows that the plant is safe for accumulation of Pu 239 up to 500 g, and operational control must be carried out to ensure that the out-of-balance does not exceed this amount. Figures 14, 15 and 12 show views of the highly-active, medium-active and low-active cells, respectively; the lay-out of the units follows the lattice arrangement already described for the dissolver cell. In all the cells some vertical cylindrical tanks are employed: Figure 14 shows how these are arranged to avoid interaction. In the low-active cell 1.5 in. thick slab tanks are utilized, these being placed in such a position as to avoid

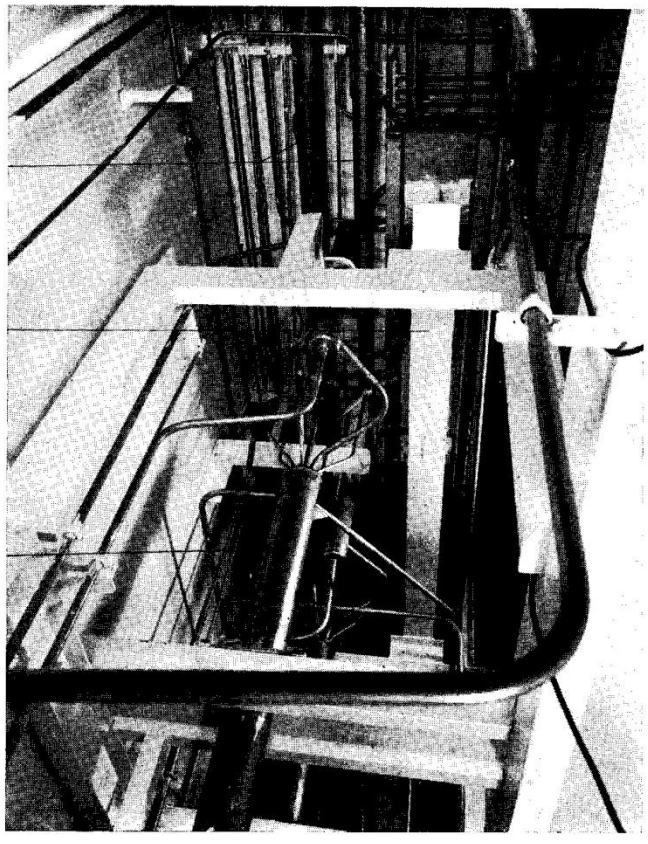

Figure 14. *Highly active cell, D.F.R. chemical plant.*

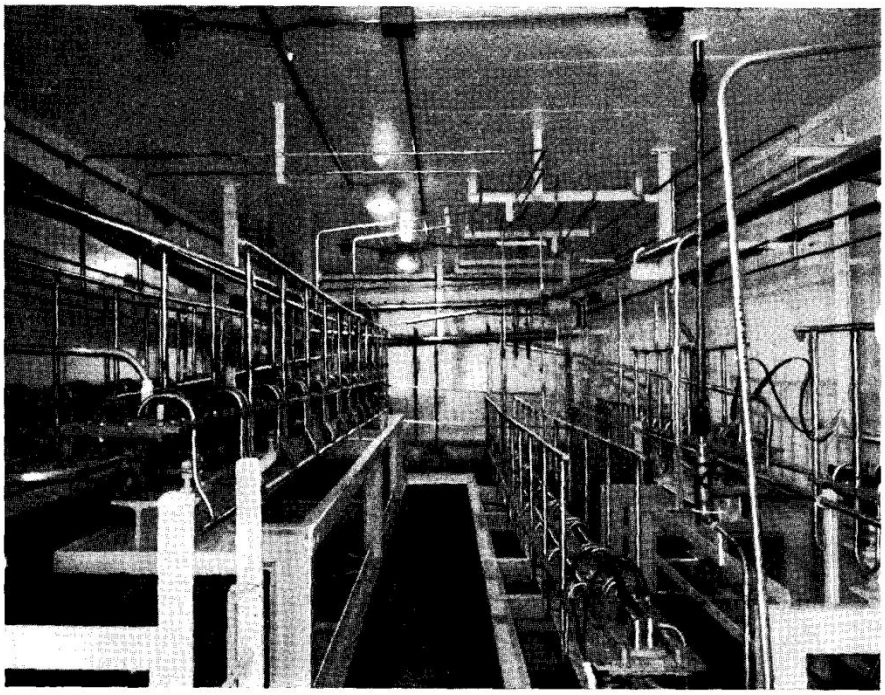

Figure 15. *Medium-active cell, D.F.R. chemical plant.*

interaction with the arrays of cylinders and where necessary incorporating nuclear shielding between the tanks.

At the time of design, few data were available on the settled density of compounds which could be precipitated from the solutions in the plant by admixture of any reagents used and at temperatures not exceeding the atmospheric boiling point of the free aqueous phases. A theoretical investigation showed no compounds of crystal density greater than 4, and since the plant does not handle uranium greater than 46 % U 235, it is clear that no precipitate could be produced of a density exceeding 2 g/cm^3. Data since accumulated on the actual density of settled sludges or uranium compounds show, of course, that this limitation could be eased considerably if required for handling 93 % U 235.

The plant does not incorporate any considerable volume of raffinate hold-up and has no facilities for recycle of this material. Since the active raffinates are sent to a highly-active evaporator of unsafe geometry and thence into large storage tanks, it was necessary to ensure that the uranium concentration of any raffinates leaving the plant was sufficiently low to allow for the subsequent evaporation and storage. Alpha monitors were therefore installed on the raffinate streams capable of detecting the plutonium associated with a breakthrough of 5 μg U/ml, this figure providing an adequate allowance for the subsequent evaporation and storage. It is significant that operations personnel have, from the viewpoint of economic operation, insisted upon being provided with more sensitive means of measurement. It can thus be

taken that the possibility of a significant quantity of U 235 leaving the plant via the active drains is so unlikely that it can be dismissed.

The plutonium content of the solutions passing through the plant is separated in the low-active cell, purified and subsequently concentrated. The plutonium stream is purified through mixer-settlers equivalent in cross section to a 3 1/8 in. dia. infinite cylinder, this dimension being safe for 2 g plutonium/cm^3. The plutonium stream then passes through a plutonium evaporator (Figure 16) consisting of 3 in. dia. cylinders disposed in plane

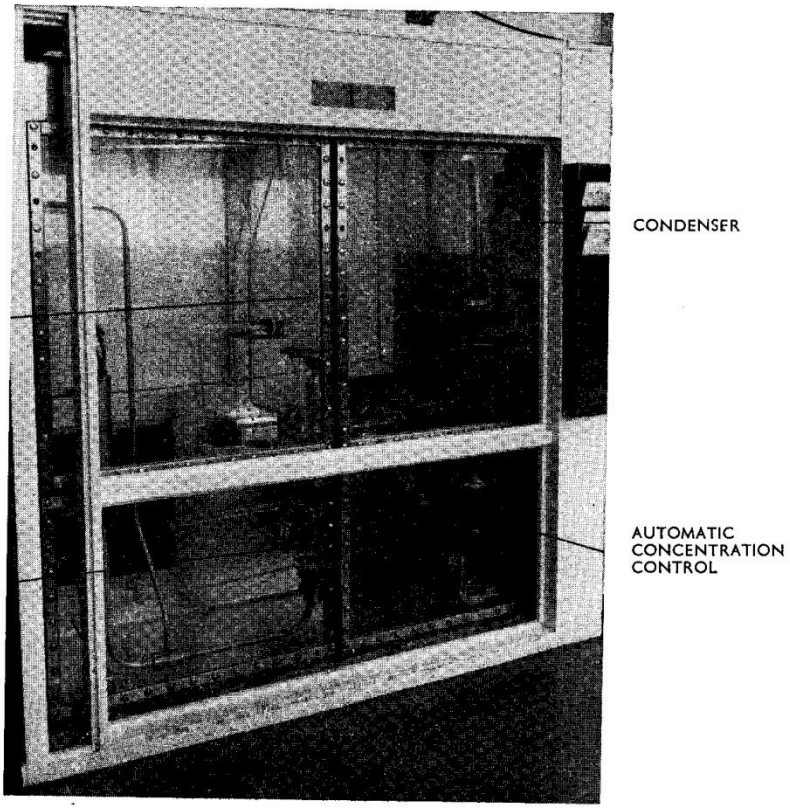

Figure 16. *Pu 239 continuous evaporator, D.F.R. chemical plant.*

array at 2 ft 6 in. centres, giving, therefore, a negligible interaction. Should a breakage occur in the equipment, the contents of the equipment would fill the bottom of the cabinet to a depth of about 0.25 in. It may be shown that this is safe since a 3 in. dia. cylinder may intersect an infinite slab of 0.25 in. thickness at a concentration of 2 kg plutonium/litre. There is, however, still the possibility that the incoming feed stream might fill the cabinet to a depth greater than this, and therefore, a small sump is arranged in the base of the cabinet with a detector which shuts off the feed and all water supplies in the event of a breakage.

Concluding Remarks

It is clear that adequate data exist for the design of fissile material plants to safe-by-shape standards and that it has become possible with experience to perceive what is good practice in the design of such plants.

The principal lesson of experience is that greatest caution and clearest thinking must be devoted to all those points where safe geometry equipment is connected to unlimited geometry and that the design must take account of operations not covered in the flowsheets or intentions of the designer.

Whilst much of this responsibility must fall on the operator, a truly safe-by-shape plant will always be designed to ' fail safe ' and to allow any failure to be corrected without requiring fissile material to enter unsafe geometry.

REFERENCES

1. M.N. Elliot *et al.*, " The Disposal of Fission Product Wastes by Incorporation into Glass ", *AERE-R 3610* (1961).
2. S.H. Smiley and D.C. Brater, " The Development of a High Capacity, Continuous Process for the Preparation of Uranium Hexafluoride from Uranium Oxides and Ore Concentrates " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 4*, 153.
3. C. Buck, G.R. Howells, T.A. Parry, B.F. Warner and J.A. Williams, " Chemical Processes at U. K. Atomic Energy Authority Works, Dounreay " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 17*, 25.
4. D. Callihan, " Nuclear Safety in Processing Reactor Fuel Solutions ", *Nucleonics 14*, No 7, 39 (July, 1956).
5. D. Callihan, W.J. Ozeroff, H.C. Paxton and C.L. Schuske, " Nuclear Safety Guide ", *TID-7016* (1958).
6. H.C. Paxton, " Basic Criticality Data for Homogeneous Systems ", *N-2-850.*

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DISCUSSION

A.J. Mallet, United States:

Although my paper is opposite to Mr. Hill's in the sense that his is geometric safety and mine is batch, I would like to point out that I must agree with him, that the psychology of the operator is immeasurably improved when his worries are eliminated.

CONTROLE DE LA CONCENTRATION ET APPLICATIONS CORRESPONDANTES

J. GUNTHER
Commissariat à l'Énergie Atomique, France

POSITION GÉNÉRALE DU PROBLÈME

Le contrôle par la composition

Pour être pleinement compris, le contrôle de la criticalité par la concentration doit être défini de la façon la plus générale possible, par opposition au contrôle par la masse et au contrôle par la géométrie. Nous dirons plutôt qu'on opère un contrôle par la composition, dont le contrôle par la concentration n'est qu'un aspect particulier, chaque fois qu'on se trouve en présence d'un système ne présentant pas une sécurité géométrique absolue et contenant ou pouvant contenir une masse de matière fissile supérieure à celle qui est considérée comme sûre dans les pires conditions imaginables.

On s'aperçoit aisément alors que, dans la plupart des cas pratiques, on exerce un certain contrôle par la composition. En particulier, les dimensions géométriques dites de sécurité ne le sont plus si l'on s'occupe de métal ou de sels à haute densité. De même, les masses considérées comme sûres pour du métal ou des sels anhydres, ne le sont pas pour des sels hydratés ou en solution, ou pour des copeaux de métal plongeant dans de l'huile.

Le contrôle des mélanges : Généralités

Il est toutefois de pratique courante de n'attacher une importance particulière au contrôle de la composition que lorsqu'on a affaire à des *mélanges* dont la nature exacte n'apparaît pas de façon évidente, tout en ayant une incidence notable sur les phénomènes de criticalité.

C'est pourquoi on définit des normes de sécurité portant sur la géométrie et sur la masse qu'on qualifie d'absolument sûres, même si ce n'est pas vraiment le cas, lorsque les phénomènes pouvant conduire hors du domaine de sécurité correspondent à des modifications de composition qu'il est très aisé de contrôler, ou qui ne peuvent survenir que dans des circonstances tout à fait invraisemblables. C'est le cas pour les deux exemples évoqués plus haut : apparition de composés à haute densité dans un récipient ne contenant que des solutions ou des sels à faible densité en métal fissile ; mélange de quantités notables d'eau lorsque l'on traite du métal ou des sels anhydres dans un local où rien ne peut en amener.

Par contre, le fait de traiter des mélanges, spécialement sous forme liquide, pour lesquels on ne désire pas s'imposer des règles fondées sur la masse et

la géométrie, pose des problèmes de nature spécifique sur lesquels sera spécialement axée la suite de cet exposé.

Le contrôle des mélanges : Aspects particuliers

Il existe trois cas où le contrôle de la criticalité par la composition d'un mélange présente une importance pratique essentielle. Ce sont :

— Le contrôle de l'enrichissement d'un composé contenant de l'uranium ;
— Le contrôle de la teneur en hydrogène, sous forme d'impuretés, dans des sels de métaux fissiles ;
— Le contrôle de la concentration d'un sel de métal fissile dans l'eau.

Le troisième cas est celui auquel on pense généralement lorsqu'il est question de contrôle de la concentration. C'est surtout pour nous le seul sur lequel nous ayons une expérience à la fois théorique et pratique.

Les deux premiers cas ne sont en effet étudiés au C.E.A., pour l'instant, que par application pure et simple des règles publiées par les États-Unis comme étant utilisées dans leurs usines [1]. Les variations des conditions gouvernant la sécurité sont données en fonction de l'enrichissement (pour la masse et la géométrie) et du rapport H/U 235 (pour la masse). Il est naturellement regrettable que l'on ne dispose pas de ces données pour la géométrie (en fonction du rapport H/U 235), et surtout qu'on ne puisse pas tenir compte de la variation simultanée de l'enrichissement et de la teneur en hydrogène.

Quoiqu'il en soit, nous ne désirons pas nous étendre sur ces points, encore que le contrôle en usine de l'enrichissement ou du rapport H/U présente manifestement des analogies avec le contrôle de la concentration en solution aqueuse. Mais cette dernière méthode ayant fait l'objet chez nous d'applications, nous nous y limiterons dans la suite.

Aspects physiques du controle de concentration en phase aqueuse

Délimitation du problème

Nous cherchons à définir les méthodes théoriques et expérimentales qui permettent de fixer des normes de sécurité basées sur la concentration. Les méthodes générales utilisées en matière de criticalité sont définies dans d'autres communications de la présente conférence, avec lesquelles nous ne désirons pas créer de double emploi.

Toutefois, un certain nombre de particularités surgissent dans l'application de ces méthodes générales à notre problème précis. Ces particularités découlent du fait que le contrôle par la concentration s'applique le plus souvent à des récipients de grandes dimensions. En effet, les récipients de taille moyenne sont justiciables du contrôle par la masse, du moins quand aucune de leurs dimensions n'est prépondérante. Les récipients encore plus petits dans l'une de leurs dimensions, sont bien entendu justiciables du contrôle par la géométrie.

Dans le cas du plutonium, et pour des récipients dont aucune dimension n'est prépondérante, on peut dire en gros que le domaine du contrôle de la concentration s'étend jusque vers 15 g/l, celui du contrôle de la masse de 15 à 50 g/l (car dans ce domaine, on est au voisinage de la masse critique

minimum), celui du contrôle de la géométrie au-dessus (car au-dessus de 50 g/l les dimensions critiques tendent vite vers une limite).

Lorsqu'une dimension est prépondérante (cylindre très long par exemple), on conçoit certes que le contrôle par la masse étant inapplicable, le contrôle par la concentration puisse être poursuivi plus loin. Mais on hésitera dans cette voie, car de tels récipients deviennent très sensibles à une inhomogénéité éventuelle.

En conséquence, les méthodes théoriques et expérimentales à utiliser seront caractérisées par le fait qu'on s'occupe de milieux à faible laplacien.

Méthodes théoriques

a) *Généralités*

Les cas qui nous intéressent sont parmi ceux où la théorie des réacteurs thermiques a le plus de chances d'être applicable, en raison de la bonne thermalisation des neutrons due à la grande dilution du corps fissile. Nous distinguerons ci-après le problème de l'étude microscopique du milieu qui doit nous fournir les constantes à utiliser, et le problème macroscopique où l'on emploie ces constantes à la détermination des tailles critiques, de récipients isolés ou non, réfléchis ou non. Nous donnerons ensuite des exemples d'application.

b) *Aspects microscopiques*

Nous nous proposons, à partir des sections efficaces connues, d'évaluer les constantes globales du milieu, à savoir essentiellement :

— k_∞ coefficient de multiplication infini
— L^2 aire de diffusion des neutrons thermiques
— τ aire de ralentissement des neutrons rapides (la notion d' « âge » n'a pas de sens pour les milieux hydrogénés).

De plus, il est nécessaire de connaître les libres parcours moyens de transport des deux groupes, si l'on désire tenir compte des effets d'extrapolation et de réflecteur.

Nous utilisons, pour déterminer ces grandeurs, des formules classiques ; nous exposons plus loin, à propos des aspects macroscopiques, les raisons pour lesquelles les constantes « ajustées » que l'on peut trouver dans certaines références [2], ne répondent pas au but recherché.

En ce qui concerne τ, nous le prendrons de 27 cm^2, limite inférieure des valeurs publiées, ce qui va dans le sens de la sécurité, car plus τ est petit, plus les dimensions critiques sont faibles. Par ailleurs, les coefficients de diffusion rapide et thermique (soit le tiers des libres parcours moyens de transport) sont pris égaux à ceux de l'eau pure, soit respectivement 1,135 cm et 0,16 cm.

Enfin, nous précisons que nous cherchons à faire le calcul sur des solutions contenant :

— de l'acide nitrique,
— du nitrate de plutonium,
— du nitrate d'uranyle dont l'uranium est plus ou moins appauvri par l'irradiation.

Le plutonium est susceptible de contenir un certain pourcentage de Pu 240 et de Pu 241.

Les sections efficaces classiques en fermis/atome et rendements en neutrons par fission des différents atomes en jeu sont :

Atome	Pu 239	Pu 240	Pu 241	O	H
σ_a	1.107 ± 16	510 ± 50	1.480 ± 150		0,33 ± 0,003
σ_f	770 ± 16		1.100 ± 100		
ν	2,91 ± 0,06		2,91 ± 0,10		
Atome	N	U 235	U 235	U 238	Th
σ_a	1,88 ± 0,05	591 ± 8	680 ± 10	2,75 ± 0,04	7,0 ± 04
σ_f		530 ± 8	576 ± 15		
ν		2,55 ± 0,06	2,47 ± 0,05		

Il est à noter que nous avons multiplié les sections efficaces à 2.200 m/s par le facteur f destiné à tenir compte, pour certains atomes, de la variation non proportionnelle à $1/v$ de la constante considérée.

Par ailleurs, le coefficient de fission rapide est toujours très voisin de 1 dans de tels milieux. Quant au coefficient anti-trappe p, il est donné en fonction du rapport H/U 238 par des formules classiques résumées dans le tableau ci-dessous.

H/U 238	20	60	100	200	400	1.000	2.000
p	0,88	0,92	0,94	0,96	0,975	0,985	0,99

En définitive, si N_i est le nombre d'atomes par cm³ du i-ème corps présent, σ_{ai}, σ_{fi} et ν_i ses constantes, on a :

$$k_\infty = p \frac{\Sigma \nu_i \sigma_{fi} N_i}{\Sigma \sigma_{ai} N_i}$$

On aura une valeur « probable » de k_∞ en prenant pour constantes les valeurs « probables » ci-dessus. On aura une valeur sûre en prenant, au lieu des valeurs « probables », les limites inférieures pour σ_a et les limites supérieures pour σ_f et ν. Cela présente un degré élevé de sécurité, les erreurs sur les constantes étant plus ou moins indépendantes.

On évaluera de même l'aire de diffusion du milieu en multipliant celle de l'eau (8,123 cm³) par le rapport : $N_H \sigma_H / \Sigma N_i \sigma_{ai}$ où N_H est le nombre d'atomes d'hydrogène par cm³ dans l'eau. Cela est assez approximatif, mais l'incidence sur le résultat de la valeur exacte de L^2 est faible ; mais k intervient par $(k_\infty - 1)$ tout en étant, dans les cas qui nous intéressent, voisin de 1 ; d'où la nécessité de l'évaluer avec soin, d'une part en valeur probable, d'autre part de la façon la plus pessimiste possible.

c) *Aspects macroscopiques*

Nous nous proposons de chercher la concentration critique dans un récipient de dimensions finies. Nous avons exposé plus haut pourquoi il y avait lieu de se limiter à des récipients dont le laplacien est assez faible.

La méthode classique pour traiter ces problèmes est celle de la diffusion à deux groupes, exposés notamment par Glasstone [3]. Nous avons des raisons d'ordre théorique pour penser que cette méthode est basée sur des approximations non valables dans le cas de milieux aqueux. Du reste, les efforts faits pour « ajuster » les constantes de la méthode à deux groupes aux résultats expérimentaux [2] montrent qu'on est obligé de faire varier ces constantes avec la concentration, beaucoup plus que la théorie ne peut le laisser supposer.

En conséquence, nous adopterons la théorie à un groupe, c'est-à-dire que nous écrirons l'égalité du laplacien géométrique et du laplacien matière, ce dernier étant donné par :

$$B^2{}_m = (k_\infty - 1)/(L^2 + \tau)$$

Quant au laplacien géométrique, c'est la première valeur propre de l'équation :

$$\nabla^2\varphi = \quad B^2\varphi$$

avec $\varphi = 0$ à la limite extrapolée du récipient.

Cette limite extrapolée distante de la limite réelle d'une « distance d'extrapolation » λ, diffère selon que le réacteur est nu ou réfléchi. Pour un grand récipient nu, le résultat classique [5] est :

$$\lambda = 0{,}7104 \times 3(D_1\tau + D_2L^2)/(L^2 + \tau)$$

Avec les valeurs de D_1 et D_2 données ci-dessus, on a :

$$\lambda = (65{,}27 + 0{,}341\ L^2)/(27 + L^2)$$

En pratique, L^2 est compris entre 2 et 4 cm^2 dans les cas qui nous intéressent. λ est donc compris entre 2,15 et 2,27 cm. Les résultats expérimentaux [4] indiquent 2,1 à 2,4 cm, en excellent accord. Nous prendrons partout 2,4 cm par souci de sécurité.

Lorsque le récipient est réfléchi, la seule méthode simple applicable est celle de la diffusion à deux groupes. Nous avons dit qu'elle ne nous paraissait pas utilisable de façon sûre. Nous allons cependant chercher, à titre indicatif, la limite de λ qu'elle donne pour une lame infinie réfléchie par de l'eau quand k_∞ tend vers 1. Lorsque D_1 et D_2 sont les mêmes pour le « cœur » et le réflecteur, on montre qu'en posant :

$L_{2c}{}^2$ = aire de diffusion du cœur
$L_{2r}{}^2$ = aire de diffusion du réflecteur
τ = aire de ralentissement du cœur et du réflecteur

$$\mu^2 \text{ et } -\nu^2 \text{ racines de } X^4 + (1/L_{2c}{}^2 + 1/\tau)X^2 + (k_\infty - 1)/L_{2c}{}^2\tau = 0$$

$$s_1 = 1/(1/L_{2c}{}^2 - \mu^2) \quad s_2 = 1/(1/L_{2c}{}^2 + \nu^2) \quad s_3 = 1/(1/L_{2r}{}^2 - 1/\tau)$$

$$K_1 = 1/\sqrt{\tau} \quad K_2 = 1/L_{2r}$$

on a :

$$\operatorname{tg}\left(\frac{\pi}{2} - \lambda\mu\right) = \frac{K_1K_2(s_1 - s_2) + K_1\nu(s_3 - s_2) + K_2\nu(s_1 - s_3)}{K_1(s_1 - s_3) + K_2(s_3 - s_2) + \nu(s_1 - s_2)}$$

Cette valeur de λ est aussi valable pour une sphère, mais non rigoureusement pour un cylindre. Si la concentration tend vers celle où $k_\infty = 1$, $L^2{}_{2c}$ tend vers une limite $L_o{}^2$, ν^2 tend vers $1/L_o{}^2 + 1/\tau$, μ^2 tend vers 0 et est équivalent à $(k - 1)/(L_o{}^2 + \tau)$, s_1 tend vers $L_o{}^2$, s_2 vers $-\tau$, $\operatorname{tg}\left(\frac{\pi}{2} - \lambda\mu\right)$ vers l'infini, équivalent à $1/\lambda\mu$.

Le produit $\mu \operatorname{tg}(\pi/2 - \lambda\mu)$ tend donc vers $1/\lambda$; d'où :

$$\lambda = \frac{K_1(L_o{}^2 - s_3) + K_2(s_3 + \tau) + \sqrt{1/L_o{}^2 + 1/\tau}(L_o{}^2 + \tau)}{K_1K_2(L_o{}^2 + \tau) + K_1\sqrt{1/L_o{}^2 + 1/\tau}(s_3 + \tau) + K_2\sqrt{1/L_o{}^2 + 1/\tau}(L_o{}^2 - s_3)}$$

Avec les constantes utilisées ci-dessus, le résultat est $\lambda = 6{,}15$ cm.

L'expérience [4] donne pour λ des valeurs allant de 6 à 7 cm, ce qui est en assez bon accord, du moins lorsque k_∞ est proche de 1. On sera du côté de la sécurité en prenant $\lambda = 7$ cm, et ceci pour tous les réflecteurs courants ; certains d'entre eux se révèlent parfois meilleurs que l'eau, mais ce n'est que pour de petits récipients où leur forte densité les favorise ; dans le cas qui nous occupe, à savoir les grands récipients, l'eau est le meilleur des matériaux courants, en tant que réflecteur.

Une fois λ déterminé pour chaque face du récipient, le laplacien géométrique s'obtient en résolvant le problème de valeurs propres de l'équation de diffusion à un groupe où Φ s'annule à la limite extrapolée.

Les résultats en sont bien connus dans un certain nombre de cas simples, rappelés dans le tableau ci-contre.

Les notations sont :

$J_n(x)Y_n(x)$ fonctions de Bessel de 1ère et 2e espèce d'ordre n, entier ou non ; $T_n(x) = J_n(x)/Y_n(x)$.

Un cas particulier important du secteur de sphère est la demi-sphère, où $n = 1$ et $p = 3/2$, d'où $\mu_r = 4{,}5$ environ.

Les cas du cône ou du tronc de cône, du prisme à section triangulaire ou trapézoïdale peuvent se ramener plus ou moins approximativement à ceux du secteur de sphère pleine ou creuse, et du secteur de cylindre plein ou creux.

Dans les cas plus complexes, la résolution sur machine électronique numérique de l'équation aux dérivées partielles, par la méthode des différences, donnera assez aisément B^2. Un certain nombre de méthodes analogiques sont aussi concevables : refroidissement d'un corps en régime permanent, vibrations de cavités ou de membranes. Devant la commodité des procédés de calcul numérique actuels, on n'a pas cherché à les développer.

Les méthodes ci-dessus, suffisantes pour un récipient isolé, convexe, et contenant un seul milieu multiplicateur, sont en échec si une de ces trois conditions n'est pas remplie.

Forme du réacteur	Définition des dimensions géométriques	Laplacien
Sphère	rayon r	$(\pi/r)^2$
Parallélépipède	arêtes a, b, c	$(\pi/a)^2 + (\pi/b)^2 + (\pi/c)^2$
Cylindre de révolution	hauteur h, rayon r	$(\pi/h)^2 + (2{,}405/r)^2$
Cylindre de révolution creux, l'intérieur absorbant les neutrons	hauteur h rayon intérieur r_1 rayon extérieur r_2	$(\pi/h)^2 + \mu^2$ où μ est la plus petite solution de $T_0(\mu r_1) = T_0(\mu r_2)$
Secteur de cylindre de révolution	hauteur h angle du secteur α rayon r	$(\pi/h)^2 + \mu^2$ où μ est la plus petite solution de $J_n(\mu r) = 0$ avec $n = \pi/\alpha$
Secteur de cylindre de révolution creux	rayon r_1 et r_2 angle α hauteur h	$\mu^2 + (\pi/h)^2$ où μ est la plus petite solution de $T_n(\mu r_1) = T_n(\mu r_2)$ avec $n = \pi/\alpha$
Sphère creuse, l'intérieur absorbant	rayons r_1 et r_2	$(\pi/(r_2 - r_1))^2$
Secteur de sphère	1/2 angle du secteur α rayon r	μ^2 où μ est la plus petite solution de $J_p(\mu r) = 0$ avec $p = n + 1/2$, n'étant l'ordre de la fonction de Légendre, dont la première racine est pour $\theta = \alpha$
Secteur de sphère creuse	rayons r_1 et r_2 1/2 angle du secteur α	μ^2 où μ est la plus petite solution de $T_p(\mu r_1) = T_p(\mu r_2)$, p étant défini comme ci-dessus

Le cas de récipients en interaction peut être résolu par des méthodes d'approximation classiques. Il n'en est pas de même de celui des récipients non convexes, souvent embarrassants. L'un et l'autre cas sont justiciables de méthodes résolvant l'équation du transport à deux ou trois dimensions, qui sont encore d'emploi difficile même sur les calculateurs actuels.

Les problèmes à plusieurs milieux (une phase aqueuse et une phase solvant contenant du Pu à des concentrations différentes, formation d'un précipité concentrant une masse appréciable de Pu, etc.) peuvent au moins formellement se résoudre par la méthode de diffusion à deux groupes avec l'aide, dans les cas complexes, des machines électroniques. Mais, pour les raisons indiquées ci-dessus, on ne peut faire grande confiance à ces résultats, sauf par exemple lorsqu'on se borne à rechercher une « perturbation » par rapport à un cas simple (phase solvant peu chargée en Pu, surmontant une phase aqueuse plus concentrée par exemple). De gros progrès restent à faire dans ces voies, notamment grâce à l'utilisation de programmes faisant appel aux équations du transport, qui sont dans le domaine normal des machines actuelles lorsqu'une seule dimension est en jeu.

d) *Exemples d'application*

Nous allons chercher la concentration de sécurité (probable et sûre) :

— pour une solution de Pu 239 pur dans l'eau en milieu infini ;

— pour une solution de Pu très irradié contenant 17 % de Pu 240 et 5 % de Pu 241, sous forme de $(NO_3)_2$ Pu, avec 2 d'acidité nitrique et 50 g/l de $(NO_3)_2$ UO_2, l'uranium étant irradié et ne contenant plus que 0,35 % d'U 235, la solution étant dans un réservoir cylindrique réfléchi de 1 m de diamètre et 1 m de haut.

Nous admettrons, pour simplifier, que la densité en hydrogène des solutions est la même que celle de l'eau pure, bien que, surtout dans le deuxième cas, la variation de cette densité soit en fait à considérer.

Les σ étant évalués à un facteur près, lié au nombre d'Avogadro, on peut prendre les N en atomes-grammes par litre. C'est ainsi que $N_H = 111{,}1$.

Dans le premier cas, on a le calcul suivant, c étant la concentration en Pu :

σ_a ↗ ↘

valeur probable : $111{,}1 \times 0{,}33 + 1.107 \times \frac{c}{239} = 36{,}66 + 4{,}632\,c$

valeur sûre : $111{,}1 \times 0{,}327 + 1.091 \times \frac{c}{239} = 36{,}33 + 4{,}565\,c$

$\nu\sigma_f$ ↗ ↘

valeur probable : $2{,}91 \times 770 \times \frac{c}{239} = 9{,}375\,c$

valeur sûre : $2{,}97 \times 784 \times \frac{c}{239} = 9{,}743\,c$

d'où l'équation donnant c :

— en valeur probable $\dfrac{9{,}375\,c}{36{,}66 + 4{,}632\,c} = 1 \quad c = 7{,}73$ g/l

— en valeur sûre $\dfrac{9{,}743\,c}{36{,}33 + 4{,}565\,c} = 1 \quad c = 7{,}02$ g/l

Dans le deuxième cas, le nombre d'atomes par litre est :

H	N	U 238	U 235	Pu 239	Pu 240	Pu 241
111,1	$2 + 4\frac{c}{239}$	$\frac{50}{238}$	$\frac{50}{238} \times \frac{0{,}35}{100}$	$\frac{c}{239} \times 0{,}78$	$\frac{c}{239} \times 0{,}17$	$\frac{c}{239} \times 0{,}05$

On a :

σ_a ↗ ↘

valeur probable :

$$111{,}1 \times 0{,}33 + 1{,}88\left(2 + 4\frac{c}{239}\right) + 2{,}75\frac{50}{238} + \frac{6{,}8 \times 0{,}35 \times 50}{238} + \frac{c}{239}(0{,}78 \times 1.107 + 0{,}17 \times 510 + 0{,}05 \times 1.480) = 41{,}05 + 4{,}317\,c$$

valeur sûre :

$$111{,}1 \times 0{,}327 + 1{,}83\left(2 + 4\,\frac{c}{239}\right) + 2{,}71\,\frac{50}{238} + \frac{6{,}7 \times 0{,}35 \times 50}{238}$$

$$+ \frac{c}{239}(0{,}78 \times 1.091 + 0{,}17 \times 460 + 0{,}05 \times 1.350) = 41{,}05 + 4{,}197\,c$$

valeur probable :

$\nu\sigma_f$ ↗

$$\frac{2{,}47 \times 5{,}76 \times 0{,}35 \times 50}{238} + \frac{c}{239}(0{,}78 \times 2.241 + 0{,}05 \times 3.200) = 1{,}046 + 7{,}983\,c$$

$\nu\sigma_f$ ↘

valeur sûre :

$$\frac{2{,}52 \times 5{,}91 \times 0{,}35 \times 50}{238} + \frac{c}{239}(0{,}78 \times 2.329 + 0{,}05 \times 3.612) = 1{,}095 + 8{,}356\,c$$

Par ailleurs :

$$\text{H/U 238} = \frac{111{,}1 \times 238}{50} = 535$$

d'où : $p = 0{,}98$ (valeur forte).

Enfin :

$$B^2 = \left(\frac{2{,}405}{57}\right)^2 + \left(\frac{3{,}1416}{114}\right)^2 = 2{,}539 \times 10^{-3}$$

Comme : $$L^2 = 8{,}123 \times \frac{111{,}1 \times 0{,}33}{41{,}50 + 4{,}317\,c}$$

On a l'équation donnant c :

— en valeur probable :

$$0{,}98\,\frac{41{,}46 + 7{,}983\,c}{41{,}50 + 4{,}317\,c} - 1 = \left(27 + \frac{297{,}8}{41{,}50 + 4{,}317\,c}\right)0{,}002\,539$$

soit : $c = 13{,}75$ g/l

— en valeur sûre :

$$0{,}98\,\frac{1{,}095 + 8{,}356\,c}{41{,}05 + 4{,}197\,c} - 1 = \left(27 + \frac{297{,}8}{41{,}50 + 4{,}317\,c}\right)0{,}002\,539$$

soit : $c = 11{,}93$ g/l

Ces exemples ont été choisis à dessein pour montrer la différence qui sépare, en matière de concentration de sécurité, un cas d'intérêt industriel comme le second, d'une abstraction telle que le Pu 239 pur en milieu infini. On se borne parfois, bien à tort, à cette abstraction ; pourtant, le fait de tenir compte de tous les éléments du problème (milieu fini, composition isotopique du Pu, impuretés diverses) relève de façon appréciable la concentration de sécurité.

En pratique, on aura intérêt à établir un réseau de courbes donnant les concentrations de sécurité en fonction des divers paramètres que l'on peut rencontrer.

Ce réseau, mis entre les mains de l'exploitant, peut être d'un précieux secours partout où le contrôle par la concentration est en usage.

Méthodes expérimentales

Ce paragraphe sera malheureusement court, en raison de l'avancement très insuffisant de la question.

Les nombreuses expériences faites en vue d'améliorer la connaissance des phénomènes de criticalité, et dont nous renonçons à donner ici une bibliographie, même sommaire, se limitent, du moins dans ce qui a été publié jusqu'ici, à rechercher les conditions critiques dans des récipients de dimensions modestes et à des concentrations assez fortes. Elles permettent de résoudre très correctement le problème du contrôle par la masse, un peu moins bien celui du contrôle par la géométrie, mais pas du tout celui du contrôle par la concentration. En effet, les laplaciens sont trop élevés pour que l'on puisse séparer dans le bilan neutronique les effets microscopiques et macroscopiques. Une telle séparation est pourtant essentielle si l'on veut se faire une idée de la validité des méthodes de calcul exposées plus haut.

On conçoit évidemment que les expériences sur grands récipients soient délicates, et cela pour deux raisons : besoins élevés en matière fissile, risques particuliers liés à la possibilité de rassemblement inopiné d'une masse supercritique par précipitation ou mauvais mélange.

Aucune de ces raisons ne paraît rédhibitoire ; la seconde notamment ne joue plus beaucoup à partir du moment où l'on réalise les expériences dans un local bien protégé, sans exclure, bien sûr, les précautions habituelles.

Nous nous bornons donc à souhaiter une extension des études expérimentales de criticalité dans cette voie, dont l'intérêt physique intrinsèque est par ailleurs notable.

ASPECTS INDUSTRIELS

Généralités

Les aspects industriels du contrôle par la concentration sont aussi divers que ses aspects physiques. Pour les mêmes raisons que celles exposées plus haut, nous nous limiterons à ce qui correspond à notre expérience en la matière, c'est-à-dire au contrôle de la concentration de phases aqueuses contenant du Pu plus ou moins irradié avec des impuretés diverses.

Cadre industriel

Le traitement des combustibles formés d'uranium naturel peu ou pas allié, irradié dans des réacteurs au graphite et à l'eau lourde, comporte deux étapes très différentes :

Dans la première, on opère sur de grandes quantités de matériau d'où l'on extrait le plutonium encore impur qui n'est présent qu'à l'état de traces dans le mélange initial. Dans la seconde étape, ce plutonium est purifié dans un appareillage qui ne reçoit que des débits horaires bien plus faibles.

Du point de vue de la criticalité, la conséquence en est que la première étape est justiciable du contrôle par la concentration ou la masse, la seconde appelant surtout un contrôle par la géométrie.

Cette première étape comporte les opérations suivantes :

— Dissolution à chaud dans l'acide nitrique de l'uranium irradié ;
— Extraction par un solvant (à base de tributyl phosphate) de l'uranium et du plutonium (à la valence 4) ;

— Réextraction dans l'eau du plutonium par réduction de celui-ci à la valence 3 ;
— Réextraction et purification de l'uranium ;
— Précipitation de l'hydroxyde de plutonium impur par une base, et redissolution dans un faible volume d'acide pour obtenir une solution relativement concentrée allant à la purification.

L'opération « précipitation » se contrôle par la masse, les autres par la concentration.

Celle-ci est très faible (moins de 1 g/l) et bien connue à la dissolution. Le seul problème se pose à l'extraction et à la réextraction du plutonium. Nous allons voir pourquoi.

a) *Les extractions : Problème chimique*

Les batteries d'extraction et de réextraction sont constituées d'une série de mélangeurs-décanteurs. Chaque mélangeur absorbe le solvant qui surnage dans le décanteur situé à sa gauche, et la phase aqueuse qui a décanté dans le décanteur situé à sa droite, et renvoie le mélange qu'il en fait dans son propre décanteur.

La batterie d'extraction (batterie I) est alimentée :

— à l'extrême gauche, en solvant pur ;
— au centre, en solution venant de la dissolution (« charge ») ;
— à l'extrême droite, en acide nitrique dilué servant au lavage.

Il en sort :

— à l'extrême droite, du solvant contenant l'U et le Pu ;
— à l'extrême gauche, une phase aqueuse acide contenant les produits de fission.

La batterie de réextraction (batterie II) est alimentée :

— à l'extrême droite, par une solution aqueuse réductrice ;
— au centre par le solvant sortant de la batterie I ;
— à l'extrême gauche, par un solvant de relavage.

Il en sort :

— à l'extrême droite, du solvant contenant l'U ;
— à l'extrême gauche, une phase aqueuse contenant le Pu.

Nous signalons tout de suite qu'en raison de la nature chimique des réactions qui se produisent dans la batterie II, les concentrations en Pu n'atteignent pas de valeur anormale ; le problème se pose à la batterie I.

Pour expliquer en quoi consiste ce problème, nous rappelerons brièvement la théorie des batteries d'extraction. Nous l'exposerons sous la forme graphique qui est la plus simple, et en supposant les volumes des phases invariables. Pour un constituant déterminé, les concentrations des phases aqueuses et solvant *sortant* de ce pot sont liées par une relation d'origine chimique qui, sur un graphique où les concentrations phase aqueuse sont en abscisses et celles solvant en ordonnées, se traduit par une courbe de partage. Ceci est la conséquence de l'équilibre qui est sensé être réalisé entre les phases, après mélange et décantation.

Sur le graphique ci-joint (figure 1), on a indiqué une courbe de partage unique, bien qu'en fait il y en ait une par pot, dont l'allure dépend de la concentration des autres constituants, supposée connue.

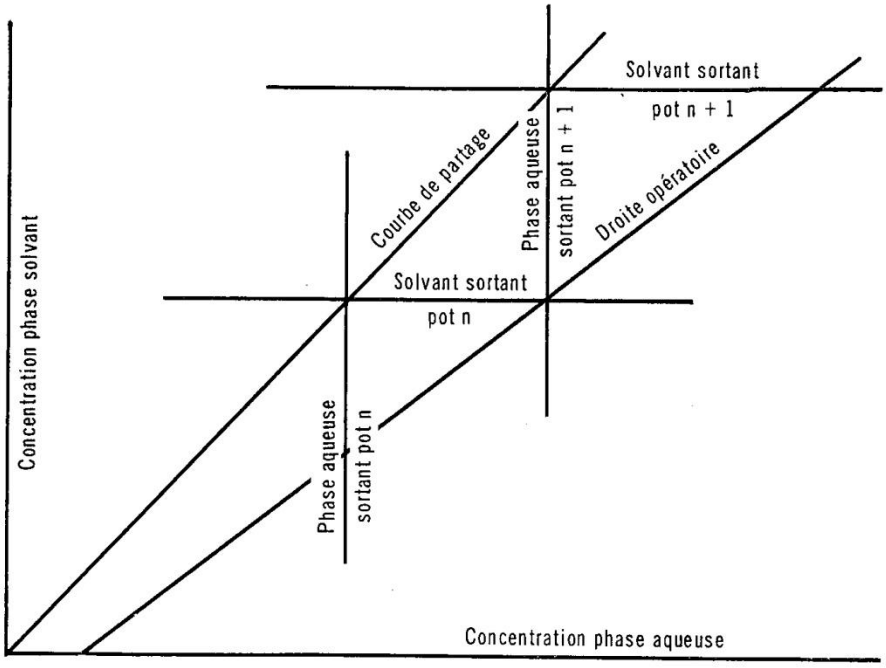

Figure 1. *Détermination graphique d'une batterie.*

Par ailleurs, on montre aisément, par un bilan matière, qu'il y a une relation linéaire, dont la pente est le rapport des débits, entre les concentrations du solvant sortant d'un pot et de la phase aqueuse qui y rentre. Cette relation est matérialisée par la droite opératoire.

Le graphique montre alors comment les caractéristiques d'un pot se déduisent de celles du précédent, par le procédé de la marche en escalier. Les données du premier pot se déduisent de la composition des charges, qui fixe également l'ordonnée à l'origine de la droite opératoire.

Bien entendu, au pot central où la charge est envoyée, on change de droite opératoire.

Pour les constituants « pondérables », à savoir l'uranium et l'acidité, les coefficients de partage de l'un dépendent des concentrations de l'autre et le résultat s'obtient par tâtonnements.

On peut alors en déduire le comportement des constituants « impondérables », Pu et activité, dont à chaque pot la courbe de partage est une droite différente passant par l'origine, et dont la pente dépend en principe seulement des concentrations en uranium et acidité.

La figure 2 donne le diagramme complet pour le Pu d'une batterie, en supposant que la droite de partage est la même dans tous les pots.

Les débits ont été supposés avoir les ordres de grandeur : 1 pour la charge, 2 pour le solvant, 1 pour l'acide de lavage.

On voit se produire sur graphique le phénomène du « pincement », qui se traduit par le fait que, si l'on augmente indéfiniment le nombre de pots de l'une ou l'autre section, les concentrations ne changent plus. Dans

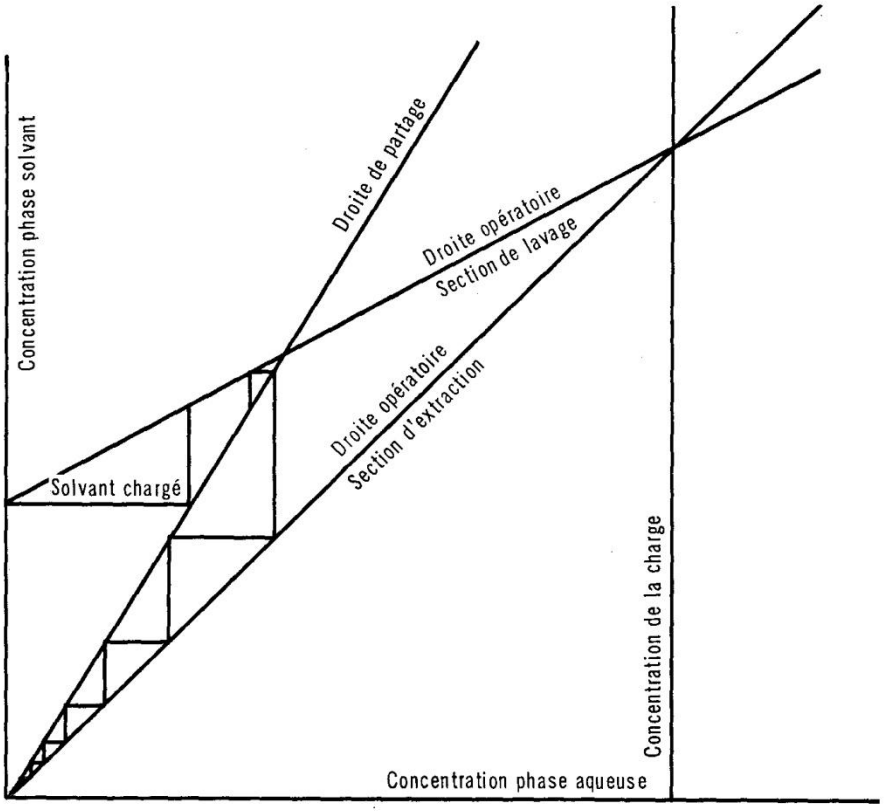

Figure 2. *Diagramme du Pu (cas idéal).*

la section d'extraction, les derniers pots à gauche sont tous à concentration nulle dans chaque phase. Dans la section de lavage, les pots de gauche sont tous à une concentration :

— $1/(2\lambda - 1)$ en phase aqueuse ;
— $\lambda/(2\lambda - 1)$ en phase solvant

si la concentration de la charge est 1, et la pente de la droite de partage λ.

On conçoit alors que la batterie soit très stable à toute cause de perturbation, tant que ce « pincement » se produit.

En pratique, les coefficients de partage sont plus faibles, pour diverses raisons, dans la section de lavage. Le graphique 3 montre ce qui se passe quand ils tombent en-dessous du rapport des débits (1/2 dans le cas présent). La figure se passe de commentaires ; on remarque qu'avec un coefficient de partage assez fort à l'extraction, on réduit très vite les concentrations, de sorte qu'on ne perd pas de Pu dans les pieds de colonnes. Mais, dans la phase aqueuse du pot de charge, se trouvent des concentrations plusieurs fois supérieures à celles de la charge. C'est là évidemment où se pose le problème de criticalité.

Bien entendu, en pratique, on essaie toujours de régler le rapport des débits pour avoir une situation genre figure 2 et non figure 3. Avec les coef-

ficients de partage déterminés en laboratoire, c'est ce qui devrait se produire. Mais, dans la pratique industrielle, des phénomènes perturbateurs apparaissent parfois : dégradation du solvant, mauvaise extraction due à des précipités d'interphase, introduction accidentelle de réducteur dans l'acide de lavage (qui abaisse le coefficient de partage, le Pu III étant peu soluble dans le solvant), etc.

L'expérience nous a en fait montré que des situations du type représenté par la figure 3 se produisent assez souvent. Il est donc nécessaire d'étudier de près les risques de criticalité qui en résultent.

b) *Les extractions : Problèmes de criticalité*

Le but du spécialiste de criticalité est de déterminer une concentration de sécurité dans le pot le plus dangereux, le pot de charge.

Pour cela, il dispose des méthodes esquissées plus haut. On conçoit qu'il faille tenir compte de tous les paramètres pour donner une limite réaliste, notamment :

— L'uranium toujours présent, qui constitue un facteur favorable ;
— La mauvaise composition isotopique du Pu qui s'observe justement pour des barreaux très irradiés où la concentration en Pu de la charge est déjà très forte ; il est important d'en tenir compte ;
— La dimension finie des pots, compte tenu surtout du fait que la concentration dangereuse ne se trouve que dans l'un d'entre eux.

Une fois en possession de cette concentration de sécurité, l'exploitant lui appliquera un coefficient de sécurité selon son appréciation personnelle, puis fera connaître à l'opérateur la concentration à ne pas dépasser. Compte tenu des débits, on en déduira la fréquence à donner aux analyses de Pu dans le pot de charge, de façon à ce qu'en aucun cas il ne rentre dans la batterie, entre deux analyses, plus de Pu qu'il n'en faut pour faire passer la concentration du pot de charge de la valeur précédente à la limite qu'on s'est fixée.

Pour terminer, signalons :

— qu'on admet toujours une répartition homogène du Pu dans chaque phase, rien ne permettant de mettre en doute cette hypothèse ; en particulier, on vérifie qu'il n'y a pas absorption de Pu sur le précipité d'interphase ;
— qu'un contrôle de la criticalité par la masse rendrait inexploitable une telle batterie pour des cartouches d'irradiation élevée, les concentrations résultantes étant bien plus faibles en raison du volume des pots ;
— qu'un contrôle par la géométrie condamnerait le principe de la batterie de mélangeurs-décanteurs au profit de la colonne pulsée, co qui présenterait de multiples inconvénients.

Conclusion

Nous avons essayé de délimiter au mieux les caractéristiques propres du contrôle de la concentration, tant dans ses aspects physiques que dans ses aspects industriels. Il nous reste à souligner à nouveau l'insuffisance des méthodes théoriques et des données expérimentales actuelles.

L'intérêt pratique élevé, du moins dans le cas développé plus haut, du contrôle de la concentration, justifie pleinement un effort dans ces directions.

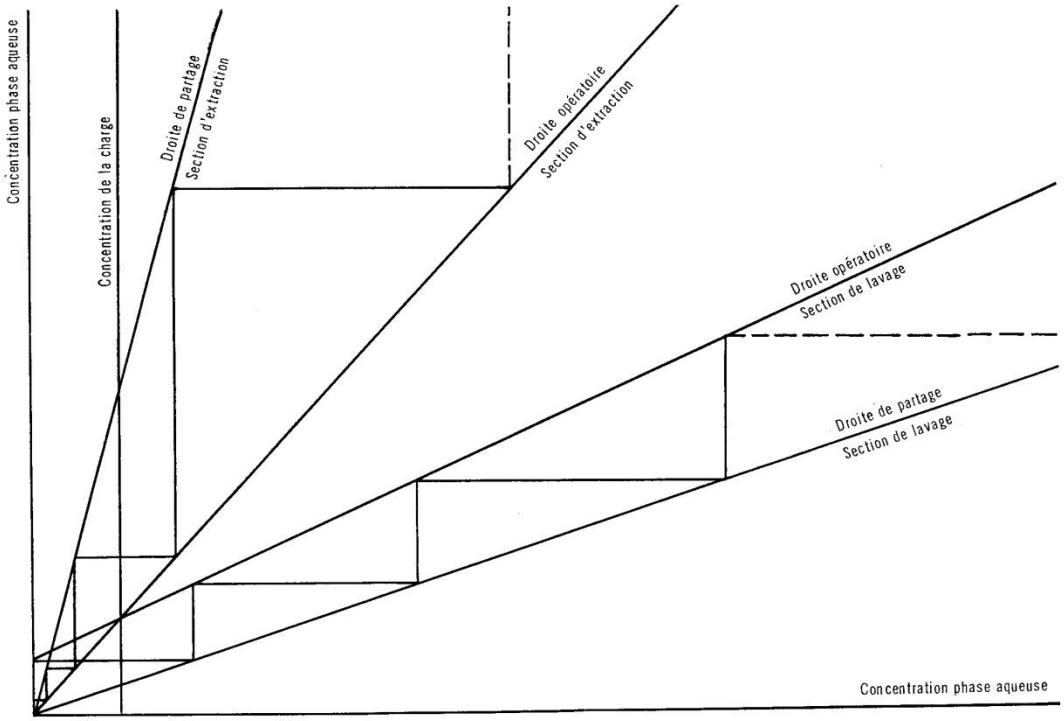

Figure 3. *Diagramme du Pu en cas de mauvais partage dans la section de lavage.*

REFERENCES

1. H.F. HENRY, A.J. MALLETT, C.E. NEWLON and W.A. PRYOR, " Criticality Data and Nuclear Safety Guide Applicable to the Oak Ridge Gaseous Diffusion Plant ", *K-1019 (5th Rev.)* (1953).
2. J.T. THOMAS, " Parameters for Two Group Analysis of Critical Experiments with Water Reflected Spheres of UO_2F_2 — Aqueous Solutions ", *ORNL-2081* (Nov, 1956),
3. S. GLASSTONE and M.C. EDLUND, *The Elements of Nuclear Reactor Theory*, Van Nostrand, New York, 1952.
4. H.C. PAXTON, " Critical Data for Nuclear Safety Guidance ", *LAMS-2415* (1960).
5. A.M. WEINBERG and E.P. WIGNER, *The Physical Theory of Neutron Chain Reactors*, University of Chicago Press, 1958.

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DISCUSSION

K.M. HILL, United Kingdom:

I would like to ask Mr. Gunther if he has considered the problem of the time scale of the build-up of plutonium concentrations at the feed - plate or any other particular part of the mixer-settler. Although there are a number of possible causes which could contribute to the build-up of concentration in one particular stage of a mixer-settler, this is usually caused by a failure of one of the feeds to the system. He did mention this question of having analysis carried out frequently, but when you are handling plutonium in any quantity through such a plant, the time-schedule is likely to be far shorter than the time that most analytical laboratories take to return a result. We have found some considerable problems, if not difficulties, in doing the chemical engineering calculations to assess the build-up of plutonium under these fault conditions. To specify the maximum allowable concentrations in a two-phase system where you do have different concentrations of plutonium in the two phases is not as easy as the more simple case that Mr. Gunther described.

J. GUNTHER, France :

Je répondrai à M. Hill qu'il n'y a pas eu de véritable problème chez nous en ce qui concerne le contrôle de l'accumulation, car nous disposons de récipients d'une assez grande dimension. Ceci comporte néanmoins des inconvénients ; en effet, comme le temps de contact entre le solvant et la solution est grand, on assiste à une radiolyse intense et à une dégradation rapide du solvant ; cependant, en ce qui concerne le contrôle en usine, tel qu'il se pratique chez nous, il n'y a pas eu, jusqu'à présent, de problème grave en ce qui concerne l'échelle de temps qui préside à l'accumulation de plutonium dans le pot le plus dangereux et dans les conditions les pires que l'on puisse imaginer. Il est possible que de tels problèmes se présentent à l'avenir ; l'expérience nous indiquera évidemment la conduite à tenir. Par ailleurs, M. Hill a également parlé du calcul de la concentration de sécurité quand on tient compte du fait que les deux phases ne contiennent pas la

même chose. Il est évident que ce cas est relativement complexe ; je souhaiterais pouvoir le traiter de la façon la plus exacte possible, mais je crains que les méthodes actuelles ne le permettent pas vraiment. Par contre, lorsqu'on admet que tout le pot est rempli de solution, les calculs sont assez faciles à effectuer et les résultats sont quand même beaucoup moins pessimistes que ceux relatifs au milieu infini, que certains ont tendance à considérer comme fournissant la seule concentration de sécurité acceptable dans de telles installations. Je reconnais qu'on peut améliorer les méthodes actuellement employées, et je souhaite qu'on les améliore.

J.T. DANIELS, United Kingdom:

Whilst Mr. Gunther has said that they had decided that the safe concentration would be from 3 to 4 g/litre, I would like to ask him at what concentration do they normally operate the mixer-settlers? In other words, do they operate to this safe concentration or is their operation level very much below this?

J. GUNTHER :

En général, on se tient nettement en-dessous de cette concentration de sécurité. D'ailleurs je dois vous signaler que j'ai indiqué 3 à 4 g/litre en tant qu'ordre de grandeur. En pratique, le problème ne s'est pas encore posé de façon cruciale et, le jour où il se posera, je pense qu'on pourra aboutir à un ordre de grandeur de ce genre. En réalité, les concentrations rencontrées dans nos usines sont nettement inférieures à ce chiffre dans les conditions habituelles, parce que normalement, en jouant sur les débits des phases aqueuses, les débits de solvants et les acidités, on essaye d'éviter ces accumulations de plutonium au pot de charge. Néanmoins, dans certains cas, on voit ces accumulations se produire et l'on peut arriver à des concentrations qui ne sont pas tellement inférieures à celles que j'ai indiquées. Toutefois, comme l'opérateur sait qu'il a encore une certaine marge avant d'arriver dans la zone véritablement dangereuse, il a le temps de prendre les mesures nécessaires pour faire disparaître cette accumulation, et revenir à des conditions de marche normales pour lesquelles les concentrations rencontrées sont d'un ordre de grandeur dix fois plus faible que celles que j'ai énoncées.

DISCUSSION ON PROGRAMMES FOR CRITICALITY RESEARCH AND ECONOMIC ASPECTS

After presentation of the papers of Dr. Paxton and that of Mr. Hill, Dr. Ackroyd initiated a vivid discussion on programmes for criticality research and the economic aspects of criticality.

R.T. ACKROYD, United Kingdom:

I have a question of a philosophical nature. This is: " Dr. Paxton has rightly drawn attention to the fact that there is a need to explain experimental points by advanced theoretical methods: but I think there is a danger in doing this, just as there is a danger in doing it by simple methods. And, to appreciate this danger, one must ask why we are interested in criticality? Criticality is a technology, and the only reason we are interested in both theoretical and experimental methods, in the end, is purely on the basis of economics. Can we save money in designing plants that are safe? Now, let us see where the two philosophies take you. Suppose you adopt the simple approach, where you use physical reasoning most of the time: you are asked to design a fairly complicated chemical plant; you are in a position to give ready answers upon which the designer can act; but what you do not know, is the accuracy of those answers. So your designer may find, subsequently, that his plant needs re-designing, re-building, to cope with its design throughput. This costs money. If you adopt the advanced method only, you will find that the designer puts a question to you, and you cannot answer this question rapidly enough. The designer will go ahead with his scheme, probably on a conservative basis. And you will have lost money again. Now, I think there is a real need to study, not only correlations from an advanced standpoint, but, having seen that the basic theoretical picture does fit in with experiments, there is a need to say, what are the important factors and what are the ones that can be forgotten? If you can delineate these factors, you can simplify the methods of calculation. Now, experimental assemblies are usually of simple geometry, plants are not; thus when you come to calculate the performance of a plant, either you have to say, " Well, I can extrapolate my advanced calculation to a more complicated geometry with some certainty " (and this is improbable at the moment), or you can say, " I know from an advanced analysis what are the important factors; I will perform a calculation on a complex geometry, taking only into account these important factors ". Now, we are still in the very earliest stages of our advanced calculations. There has been recently a Symposium in the United States organized by the American Mathematical Society, where some concern was expressed that certain of the mathematical tricks, which we had thought respectable, are no longer so. For example, that you can have a complete eigenfunction expansion for fluxes is certainly not true any longer. I think, if we are to get

the best plants for the least expenditure of money, we must get together with the experimentalists and the mathematicians in a sort of threefold attack, to pursue correlations on the basis of advanced methods. We shall then try to find out what are the dominant physical factors. A third step is that we ought to look at some of the bases of our advanced calculations and see if we can do something about reducing the cost thereof. In the paper by Mr. Pendlebury and myself, I have suggested an approach. The basic idea is that in the present state of advancement of calculation, we just go plodding on till convergency is achieved, and we hope that we have converged to a physical answer. Most of us know we have, but of course, there are one or two doubts in some cases, particularly where you have rapidly varying fluxes, say through a chain of slabs. Another approach is to adopt the method that Professor Birkhoff at Harvard and myself have been plugging, that one ought to use the methods of function spaces, because when they apply to numerical methods, you can tell the accuracy of your first iteration. If this is good enough from the point of view of plant design, stop and save yourself money. I do emphasize this point of cost, because not only do I have people doing criticality calculations, but I also have people designing reactors. I don't mind spending money on reactors — of course, they usually cost something of the order of say millions of pounds, and the thing I am making calculations on is going to go into the reactor core and to look like it — but I sometimes worry about the money we spend on advanced criticality calculations because they form a sizable part — say, 10 to 15 % — of the cost of the plant. I know the plant is not going to operate under conditions under which I have done my calculations: things are going to be found out. So I hope people here, with a more scientific outlook than myself, will excuse my interest in the economic aspect of the problem.

H.C. PAXTON, United States:

I agree that there should be much more said about money, and perhaps I did not explain in my paper the aims I had in setting up these correlations, in addition to the fact that this was an assignment. I believe I did mention that in order to accomplish anything within the allotted time, I had to limit myself essentially to what had been done — what I knew of what had been done — and had to avoid any treatment of the simpler computational methods. The purpose I had in mind was two-fold. One, to attempt to answer the question whether computations such as those of Mills provided a useful general supplement to experimental data. This was without any specific plant in mind; just a question of adding to the general body of information something that might be useful to anybody at any time in the future. I will confess that I also had several specific problems in mind. One was the problem of handling graphite-water-enriched uranium mixtures, which we have had to face at Los Alamos; another was the general question of transportation of low enrichment uranium which I believe I mentioned in my paper. So, in the first place, my outlook was very limited and in the second place, I had by and large no specific plant in mind. I might mention the conclusions I have drawn from this study. Actually, I like experimental data and I prefer this to any calculation. But, for some of these three — or four — component systems, life is simply too short to cover everything experimentally and experiments become very costly. It is possible to accumulate computational

values over a period of time, and my conclusion concerning the particular type that I reviewed — DSN, as it is now applied, with the specific cross section sets that were used — is that a computational survey of the three-component system can be of some value if experimental check points are judiciously spotted. As an example, I would hesitate to use the asymptotes on the under-moderated side of the relationship critical mass against H/U 235 for the U 235-U 238-water system without some experimental verification. I hope that what I have done will stimulate much better things, both experimentally and computationally.

E.R. WOODCOCK, United Kingdom:

I would like first of all to deplore the suggestion that economic factors are the only important criterion. Surely, money isn't everything in the scientific field? Criticality is an interesting mathematical study for its own sake, regardless of any economic factors. Another point I would like to make, is one which I think Dr. Paxton was also trying to make, namely that there is a place for both experiments and advanced calculations in this subject. For the practical implementation of criticality work, one needs a set of data, such as Dr. Callihan has presented, and these data can be obtained by a combination of the experimental and the calculational side. There are some basic points which can only be done by experiment; there is another series of points which can be done equally by experiments or calculation; and there are some factors which one can only do by calculation and which one cannot sort out by experiment.

F.R. CHARLESWORTH, United Kingdom:

I would like to ask a rather general question. Has anyone attempted to estimate what fraction of the cost of design or operation of a plant one should apply to acquiring criticality data?

H.C. PAXTON:

I have heard it stated that in connection with the fabrication of oxide fuel elements with low enrichment uranium, more than one-half the total overall cost is in some cases transportation cost. And I am sure that the bulk of this is because of special criticality requirements. This has no direct bearing on the question, but perhaps an indirect one, and there are bitter complaints in the United States about such as factor. In the plants that I have seen, it is very difficult to judge what criticality control has cost; in some cases what seemed to be very expensive geometric requirements have turned into rather efficient continuous operations, replacing earlier less safe batch operations.

K.M. HILL, United Kingdom:

The point raised by Mr. Charlesworth cannot be given a single answer. In any plant that one proposes to build, one has to develop a flow-sheet process for the whole bag of tricks. The criticality experimental and calculation work can play a bigger or lesser part and this depends upon the particular plant. The overall answer shows whether the proposed process will show a saving. If by putting a lot of money into criticality and very little

into plant development, you can make a saving, do it that way. Frequently, as Mr. Paxton has just mentioned, one thing leads to another: going to the safe geometry plant tends to lead to continuous processes and you may make savings both ways.

C. L. SCHUSKE, United States:

I would say that in some cases the initial criticality costs would be less than 1 % of the total capital cost and about 0.25 % as far as personnel requirements are concerned. The 0.25 % figure is based on the total personnel requirements for the production facility.

J. W. WACHTER, United States:

This sounds reasonable to me, since you are referring specifically to the criticality group costs. It seems to me that the cost depends in good part on the nature of the programme. One which is fairly long-continued — over five years or so — would usually be relatively inexpensive when compared to throughput. I think that criticality costs are not too high, either, in short-run production projects for which rather more strict criticality limitations can be imposed. But there is an intermediate type of project — something which would run three to six months say — where one could not allow administrative controls with quite the confidence that one could in a very short run, so that highly conservative answers are not feasible. In these intermediate ranges, I would think the cost would run quite high but I don't know exactly what the percentage might be. Although I have never pursued this, I don't think it could exceed 10 to 20 % — and then only if no alternative presented itself.

K. M. HILL:

If one is considering using an existing plant or existing facility for doing a specific job, and if one is talking about the cost of criticality control in putting through a batch of material through this plant, then the criticality cost could be quite a considerable fraction of the operational cost. I think that is what Mr. Wachter was referring to. In the other case, you have the problem of building a new plant, and investing money in criticality experiment and calculation in order to produce an economic unit. The latter case more or less stands on its own feet, since you are able to get a statement of all the savings and the costs fairly readily. On an operational basis, the costs are not likely to be so large in actual magnitude, although they may be quite considerable in terms of percentage of operational cost.

C. L. SCHUSKE:

I have one additional comment to make in regard to the criticality cost. For example, if one is to built a ten-million dollar plant — this is not a very large plant in the United States — I think it would be unusual indeed to spend as much as 1 % of that cost for direct criticality charges for both consultational and experimental work.

It must be remembered that one competent man with pencil and paper can handle all but a few unusual problems with the criticality data already available.

J.W. WACHTER:

Yes, I quite agree. In fact that would be a high percentage for the cost of criticality group and associated calculations. If one were to process the material as though it were an ordinary chemical and compare this cost with the cost for producing the same material as uranium, and consequently subject to criticality requirements, the result would be a far different number.

J.G. WALFORD, United Kingdom:

Is not the essential point here that the 1 % of process costs spoken of by Mr. Schuske as a top limit, is the extra money spent in applying the basic criticality data to day-to-day problems rather than the cost of obtaining the basic data in the first place? Surely it is this basic data, already available for application to plant problems, that is included in Dr. Callihan's and Dr. Paxton's papers? The question posed by Dr. Ackroyd seemed to me to be " How much money should be spent in obtaining basic data? " and I believe the 1 % referred to by Dr. Schuske is an additional expenditure that comes later.

A.J. MALLETT, United States:

One of the comments usually made to criticality people is that we increase the costs of operation by nuclear safety requirements. I would like to point out, however, that much money has been saved through safety of operations and without sacrifice of production throughout. The use of pilot plants in the development of operation processes is really no different from the criticality experimental work that must be done in order to lay a foundation to determine what is safe.

L.A. MEIERKORD, United States:

Most companies that are in business to make a profit have definite fiscal policy regarding the expenditure of funds for reduced cost and increased efficiency. This oftentimes takes the form of having the moneys expended for improvement returned to the company in profits, in something like two to five years; or in other words that the return on the invested money must be on an annual basis of twenty to fifty per cent. I see no reason why improvements in criticality, and cost reductions that would result from such improvements, should not be put on the same fiscal basis as any other process improvement or plant improvement.

F.R. CHARLESWORTH:

The reason for my original question on the cost of obtaining data was because the speakers have given the impression of a continuing study of criticality, perhaps over a number of years. It seems to me that there is a limited amount of information that we require. It is rather like putting lighthouses around the coast; if you have enough, you can navigate from one to the other. Therefore, one ought to forsee an end to criticality experimentation.

A.J. MALLETT:

The types of equipment involved and the types of nuclear safety questions raised as a result of processing operations leave one a little uncertain that we will have, experimentally, all the answers necessary relating to criticality.

E.D. CLAYTON, United States:

I would like to comment on how soon we might actually terminate the experiments on criticality. It depends on the kind of experiments which are undertaken. I believe the emphasis should be placed on the basic types of experiments in plain geometry, which will be amenable to theory, so that a correlation can be made between theory and experiment. If we do these kind of experiments, we will then be able to terminate the programme. If we continue to do so-called *in situ* type experiments, purely engineering experiments, such as odd geometries for specific plant application, then we will be in a position of having to continue experiments for an indefinite and long period of time.

K.M. HILL:

I would like to ask Dr. Clayton if he thinks, with the completion of these clean and nice experiments, which enable theory to be improved, that the safety factors which are applied to most criticality determinations could be reduced; because it seems to me that one of the big advantages of dunking a piece of equipment in U 235 and determining whether it is going to be critical or not, is that your safety factors are much more related to the true situation, whereas if you are going to hum and haw over interpretation, put a safety factor on for this, and a safety factor on for that, I often think that it is hardly worth carrying out the experiments in the first place.

E.D. CLAYTON:

I think the kind of safety factor one has to apply really depends on how well the operating conditions in the plant can be defined. For example, if one knows exactly what concentration is required for criticality, this is one thing, but if the people who are operating the plant cannot maintain the concentration to within $\pm 20\,\%$, then it is necessary to apply some sort of safety factor which really does not have anything to do with our knowledge as to what criticality is, but rather depends on the ability of the operating people to run the plant.

K.M. HILL:

I think there is a point here. In my paper I stressed this point about designing for fault conditions, and I think that if you are going to design safe-by-shape plant, this is very important because of the complete freedom it gives the operator. These fault conditions can be defined in many situations, fairly closely, in terms of the maximum conceivable density one is likely to arrive at. For example, in the type of plant I described, the data is on a fairly flat portion of its curve at those densities, and it is the safety factors which you apply after having determined the chemical conditions, i.e. the maximum density, to which I was referring.

R.T. ACKROYD:

I would like to perhaps correct some impressions which may have arisen from my original comment. I would agree with the need for experiments in clean geometry, and with the need for correlation with calculation, but what I think has been lost sight of, is a point which has been put very ably by E.P. Wigner and A.M. Weinberg, namely, that we must have physical understanding,

that a code is no substitute for the physical understanding. We have moved away from the era of the early days, when we used physical understanding because we could not compute to any degree of accuracy by advanced methods. Now we seem to have fallen into the snare of just being content with getting a correlation between a prediction from a mathematical equation and experiment. We still have been missing the most important thing in this, that is physical understanding, i.e. what terms are operative in equations and what terms are not. Now, if we don't reform, I think our experimentations and our calculations are going to move at a pedestrian pace. Sooner or later, people will be asking when is this all going to end. We have to decide not to cover the range slowly, step by step, but to delineate those areas where crucial decisive experiments can be made. Now, Mr. Hill did show a graph of the possible effect of Pu 240, and it may seem that in a certain range the Pu 240 curve came out rather high, but this was done deliberately; I may say, in fact, it was done some time ago, to delineate an area where experiments with Pu 240 might be the most profitable. When, for example, the people who supply our funds see positive results from our being bold in choosing where to experiment, I see the possibility of the life-span of criticality experiments being increased. But if we continue to do one experiment next to the one previously, and proceed in a rather pedestrian fashion, I would fear that if one were not very careful, in some places the axe might fall.

H.C. PAXTON:

I agree heartily with everything Dr. Ackroyd has said.

J.G. WALFORD, United Kingdom:

This discussion on the cost of criticality studies started with a question concerning the expenditure considered reasonable for theoretical or computational work and has since moved rather towards the experimental side. I would like to ask the American speakers if they can suggest what the relative expenditure on these two types of study should be, assuming it is possible to draw a line between them; do they consider that more money should be spent on experiments rather than on computation at the present time?

E.D. CLAYTON:

I think we should spend more money on both experiments and calculations. Up to now, I feel we probably have not spent quite enough money on calculations to go along with the experiments. I am not sure that I speak for any of the other American delegates, but this is my own personal feeling concerning the work that we have done at Hanford. I feel that we have to expand our computational programme with the experimental work.

H.C. PAXTON:

I agree with Dr. Clayton's statement. I would say that in some cases there may be an opportunity to gain information parasitically. We happen to be in the particularly fortunate situation of having a critical assembly laboratory and rather abundant computational facilities, and we try to keep in mind the possibility of gradually accumulating information just as a matter of principle.

J.G. WALFORD:

I am really trying to enquire how much, relatively, the two types of study do cost, rather than how much we ought to spend on each of them. I have encountered the impression that a programme of criticality experiments is expensive and this is, I think, fairly obvious to anybody. But the impression is also sometimes given that computations are much less expensive. Having heard during the first two Sessions of some rather sophisticated types of computation, I wonder whether this really is so, and whether there are not occasions when an experimental programme to obtain some required data can be pursued for a sum of money comparable to the cost of the appropriate calculations ?

E.D. CLAYTON:

If we assume a typical critical mass programme, costing in the neighbourhood of perhaps $ 500,000 per year, then a reasonable computational effort in support of the experiments would be about $ 75,000, which would include the cost of machine computations, and the personnel working on computer codes; in other words, for a budget of the above size, about 15 % of the total funds might be used to advantage in theoretical support.

J.G. WALFORD Kingdom:

I would like to ask Dr. Clayton whether the $ 500,000 annual operational cost includes anything for the preparation of fuel materials in the form in which the critical mass experiments require them.

E.D. CLAYTON:

Yes, the cost quoted would include the preparation of special fuels. I might mention that perhaps the estimate is low. We are just now getting our plutonium critical mass laboratory into operation, and we may find that our costs somewhat exceed the figures quoted. But these are estimates only and they do include the cost of preparing fuels, say plutonium oxide-polystyrene mixtures and reprocessing, and so-on.

E.R. WOODCOCK:

I would like to comment on Mr. Walford's question on the relative costs of experiment and calculation. It is one which has not got an overall answer, for it depends on what particular system is considered. I think it would be true to say that for some experiments, such as Mr. Walford was describing at the beginning of the discussion, with 1.4% enriched uranium, there is very little to choose between the cost of the experiment and the calculation; in fact, it may well be that to do an advanced calculation on that system would be rather more expensive than to do an experiment. Whereas, at the other extreme, if one, say, has to take a solid sphere of U 235 metal, then there is no doubt at all that an advanced calculation is extremly cheap compared with the experiment.

A.N. DICKSON, O.E.E.C. High Temperature Reactor Project (Dragon):

Much of this discussion about the economics of criticality strikes me as being not dissimilar from many other engineering problems. I would like

to take as an analogy the question of the strength analysis of an engineering structure. Here we have an elegant body of mathematical theory done by people who have a bent in this direction, and most advanced communities feel they can support such people although their economic worth is perhaps difficult to measure. Their theories are generally based on some idealization of the real behaviour of materials.

Consider now the practical problem of designing a pressure vessel; at some points, say at a nozzle entry, the geometry may be extremely difficult from the mathematical point of view. If the penalty for increasing the thickness of material in such a region is just the cost of the extra material, then there is clearly little justification for employing a lot of clever mathematicians on stress analysis.

On the other hand, in an aircraft structure the penalty for needlessly increasing the weight far exceeds the cost of the material involved and in this case analytical methods may be justified. If the calculations are very complicated, however, an ad hoc test would be adopted and most engineers feel this to be more reliable than calculations.

If we turn now to our chemical processing plant, if the use of criticality data for simple geometries such as spheres and cylinders leads to a satisfactory design in which you are not paying big penalties, the matter would rest there. If the problem were very esoteric and complicated, the designer would probably require an ad hoc test. Thus the problem of criticality does not seem to be different from other problems of engineering design.

J.T. DANIELS, United Kingdom:

Whilst I agree on many points with the sentiments that Mr. Dickson has expressed, I feel that he has omitted one point, that, in dealing with safety in the atomic energy field, we are not merely dealing with the physical consequences of an accident. There is a large measure of cost, which it is quite impossible for anybody to estimate, tied up in the political and public relations ramifications of an accident. I am speaking principally of the countries where the scale of operations has not been such that they have had the misfortune to have an accident. Our communities are quite used to people being killed on the roads, I apologize for pointing out this terribly obvious fact, but the day we have a serious radiation accident, be it from the mishandling of radioactive material or from a criticality accident, the cost of this accident in the political sense will be almost without measure. Therefore, in weighing the money and the effort to be spent in ensuring safety, it is not sufficient merely to consider the capital investment in the project.

W. SCHÜLLER, Eurochemic:

I should like to raise another point, associated with the discussion of the relative merits of experimental and theoretical work. In comparing these, there is one question which is not associated with purely financial terms, and this is the availability of the large quantities of fissile materials required for experimental work. The scientists from the United States and the United Kingdom, and also from France, are in the happy position of being able to say, " Well, we want to make criticality experiments, we need 150 kg U 235 or so-much plutonium solution ". This, not only for financial but also for political and other obvious reasons, is certainly not possible for quite a num-

ber of countries who are, of course, also interested in the subject. I want to raise the question as to whether there is any possible way of working experimentally in the field without having large quantities of fissile material at one's disposal.

D. CALLIHAN, United States:

There are, of course, ways of experimentation using substantially less than critical quantities, such as replacement experiments in critical assemblies and low-power reactors, and exponential experiments as examples. On the other hand — I may perhaps have misunderstood Dr. Schüllers's question — if it is a question of experiments in support of designs for processing operations in which, in turn, the inventory is limited, I wonder whether one does in fact need information on such large quantities ?

W. SCHÜLLER:

It is perfectly true that even with limited amounts of fissile material one can make experiments, such as replacement experiments, but these also require a reactor facility and essentially the problem boils down to the same question: what type of work other than calculation and theoretical work can be done by those groups or those countries which cannot afford a large-scale experimental programme? Is there anything?

With regard to Dr. Callihan's question, I would say that you might be in a position to deal with a large amount of fissile material, without having it available for critical experiments alone.

D. CALLIHAN:

As I said, there are indirect methods for measuring the nuclear properties of materials which, after all, are the things we are speaking of. And these do give and have given in the past years acceptable results which can be used at least to substantiate and verify analytical procedures. And this, from the standpoint of experiment, seems to be the method which might be applied in the case Dr. Schüller described.

Session IV

APPLICATION OF CRITICALITY CONTROL PRINCIPLES TO PLANT DESIGN *(Continued)*

APPLICATION DES PRINCIPES DE CONTROLE DE LA CRITICALITÉ DANS LA CONCEPTION DES INSTALLATIONS (Suite)

Chairman - Président :

Prof. A. F. ORLICEK

(Austria)

MASS (BATCH) CONTROL AND ITS APPLICATIONS

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INTRODUCTION

The purpose of this paper is to discuss some of the problems of nuclear safety control associated with various operations in which the quantity of uranium materials handled is specifically referred to as a safe mass or safe batch. Although the topics covered are generally applicable to solutions, solid compounds, metal, chips in solution, etc., the discussion will be limited principally to solutions and salts of uranium. Much of the material mentioned may be found in the Criticality Data and Nuclear Guide Applicable to the Oak Ridge Gaseous Diffusion Plant [1] or in the Nuclear Safety Guide (Revision I) [2] which also includes considerable information concerning metals.

The mass or batch size method of control for nuclear safety is actually quite analogous to other industrial processes wherein control of the amount of the individual ingredients added to the mix is necessary for good operation, where ' good ' also implies safe processing. The baker must know the type and quantity of yeast or the pan overflows; the powder-maker the purity and composition of his chemicals as well as pressure and temperature limitations; and the steel-maker the right ore to add and the percent required in the manufacture of certain steels. Other important factors related to safe operation include a knowledge of the compatibility or incompatibility of the various chemicals involved and of the possible individual or resultant explosive, corrosive or toxic effects.

In the handling of materials such as uranium or plutonium, there is one important and differing characteristic, other than those which are chemical in nature, which must be taken into account, namely, the ability to undergo fission. This is a condition in which a neutron enters the nucleus of an atom causing it to ' split ' or separate into two identical units with the release, on the average, of 2.46 additional neutrons per fission. If these neutrons are either captured in, or escape from, a given system so that fewer neutrons are available in succeeding fissioning generations, the system will be subcritical. If conditions are such that neutron production is an increasing process, then a criticality condition can occur for a specific mass, volume, or geometry parameter. Thus, since all that is needed to cause a ' chain ' reaction is to have a sufficient mass available, batch operation is simply a type of processing operation wherein various controls are established and main-

tained to prevent the accumulation of more than a specific quantity of uranium material which has been designated as a nuclearly safe mass.

Factors Affecting Nuclearly Safe Masses

To the uninitiated in nuclear safety, batch operation is where you dump material into a tank of solution, stir, filter and use; a simple operation essentially devoid of problems. It is actually much more complex, especially where the material is of such a nature that the end result, if adequate planning and attention to detail are not consistently followed, may be a criticality incident.

Several factors are involved in determining these safe conditions, of which the amount of uranium material considered to constitute a safe mass or batch is one. These include:

1. *Enrichment.* This is the percent of U 235 isotope in the uranium content of the material and is determined analytically by means of a spectrograph or a spectrometer, the latter giving more precise measurements. The U 235 content in uranium materials normally found in nature is about 0.71 % and this value is frequently referred to as normal enrichment; conversely, the U 235 content of UF_6 where isotopic separation of the U 235 and U 238 has been effected, may be over 90 %.
2. *Moderation.* Where an associated material other than the U 238 causes a slowing-down of fast neutrons to thermal velocities, thus resulting in a higher probability of neutron fissioning. Hydrogen, as found in ordinary water, is considered among the most significant moderators, and the degree of moderation is usually expressed as a ratio of the number of hydrogen atoms to U 235 atoms (H/U 235) or to the number of uranium atoms (H/U).
3. *Density.* This refers to the uranium density and not to the material density. It varies not only with the type of uranium material but with the conditions under which the material is made or processed. For example, the density of uranium metal is about 18.7 g/cm^3 but the maximum uranium density of UF_6 is 3.2 g/cm^3 (198 lb/ft^3). As an illustration of the relationship of density and moderation, Figure 1 indicates the variation in the uranium density of a UO_2F_2 water solution as the hydrogen/uranium ratio varies, the density decreasing as the solution becomes more dilute until a value is reached at which any volume of the solution will be subcritical. Conversely, density increases will result in decreased nuclear safe mass or volume.
4. *Reflection.* Certain materials have the property of reflecting escaping neutrons back into the mass of uranium material; this increase in the number of neutrons available for fission can result in a critical mass which is smaller than that for the unreflected condition. The hydrogen atom which was previously noted as an excellent moderator is also considered a significant reflector.
5. *Neutron poisons.* Materials which can absorb neutrons and thus remove them from the system, particularly in the neutron energy ranges where fission is most probable, are referred to as neutron poisons. The size of the capture cross section of the nuclei of the material used determines whether it is a mild poison, such as nitrogen or hydrogen, or a strong poison such as cadmium or boron.

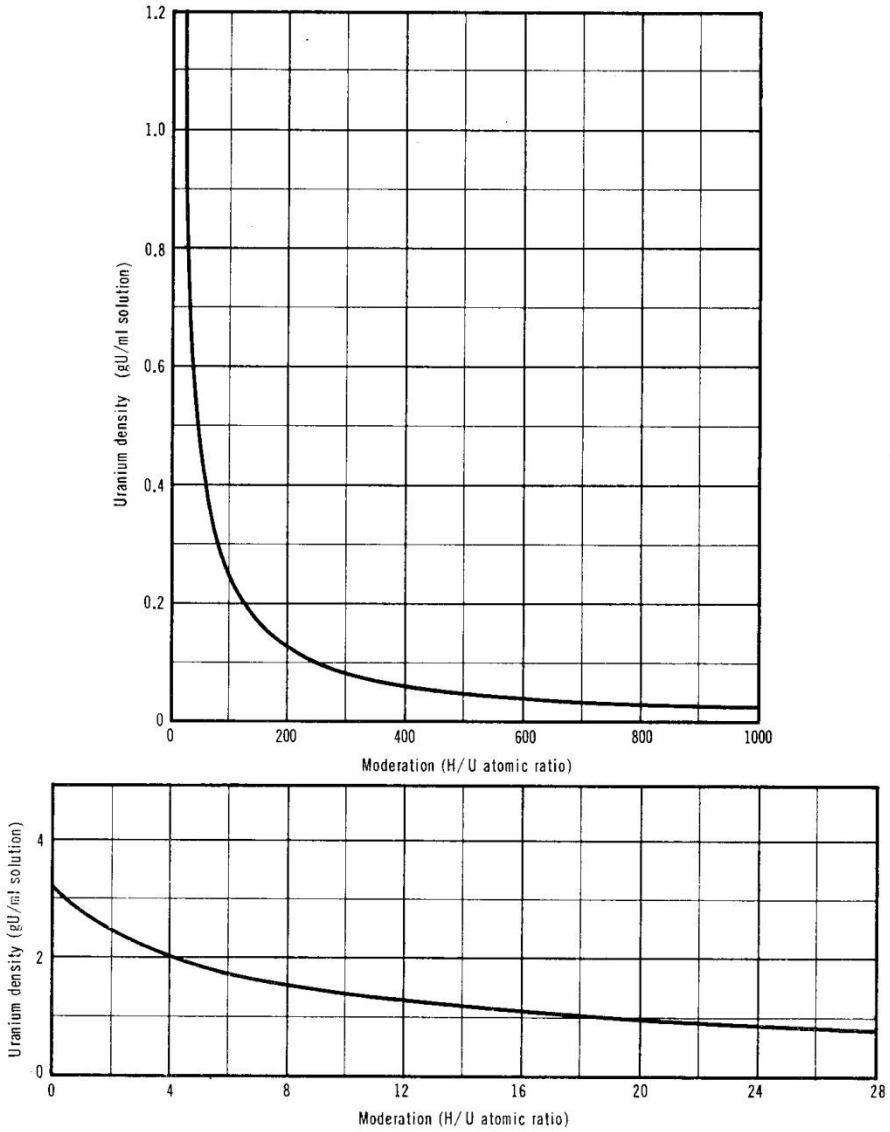

Figure 1. *Uranium densities for specified moderation (UO_2F_2-water solution).*

The use of neutron poisons as a method of criticality control can enable larger batch quantity operations, or provide greater assurance of safety against the contingency of multi-batch operation. The principal problem, once the effectiveness of the neutron absorber has been determined, is that of assurance that the poison will not be removed from the system or so changed as to lose its effectiveness.

In addition, other factors which are standard operative and administrative functions also influence the control of safe batch operation, since

they introduce the possibility of undetected error with its subsequent increased effect on the potential hazard of the operation. Included in this group are a wide variety of functions such as sampling, laboratory analysis, weighing, identification, record keeping, operator checks, transfer and storage problems, equipment isolation, the ever-present problem of attention to detail where routine has been established, and, one of the most important, possible changes in state: for example, precipitation where uranium concentration in solution is the method of control. These aspects of nuclear safety control and their application to specific plant problems will be discussed in more detail shortly.

Safe Masses

Over the years, a continuing programme of criticality experiments with uranium solutions, and with UF_4 intermixed with plastic materials to form a pseudo UF_6 and compressed in block form to an approximate uranium density of 3.2, has provided a considerable amount of data. These data, which include conditions of moderation have, in conjunction with theoretical studies, resulted in the establishment of appropriate mass values for the various enrichments which are considered to be nuclearly safe if fully-moderated and reflected or nuclearly safe if under-moderated and reflected.

Figure 2 depicts nuclearly safe masses with respect to enrichment. It may be noted that, as the enrichment approaches 0.95 %, the mass approaches infinity. However, there are some experimental data at present which indicate that 1.0 % enrichment may be the lower limit at which criticality will not occur. The limiting enrichment value for uranium metal where no re-entrant surfaces are available is considered to be 5.0 %. The limiting safe mass values as the enrichment increases have been established for uranium compounds at 350 g U 235 for uranium salts and solution; for metals, it is about 10 kg.

A plot of the experimental critical masses of U 235 *versus* the hydrogen to U 235 atomic ratio indicates the mass curve to exhibit a rather broad and flat minimum of about 800 g U 235 over a H/U 235 range of 400 to 600; the solution volume for this H/U 235 range varies significantly, increasing from about 10 to about 20 litres. On the other hand, the minimum critical volume of 6.4 litres occurs at an H/U 235 atomic ratio of about 50 and its corresponding critical mass is approximately four times that of the minimum critical value. The minimum critical mass of about 22 kg for uranium metal occurs when the pure metal is in a ball form, without re-entrant surfaces. Since the density in this case does not vary, the minimum critical volume is concomitantly determined. It should be noted that all the above values are obtained with highly enriched uranium and water-reflected systems. The values given would be considerably lower were reflectors such as carbon or beryllium used.

Safety Factors on Mass

The safety factor by which critical mass values are reduced for safe mass operation is one involving the individual organization for nuclear safety control and the extent of effective process control by the operating groups concerned. Thus, using the same materials, one facility may choose to operate on a safe mass basis of 350 g U 235 to allow for the possibility of double

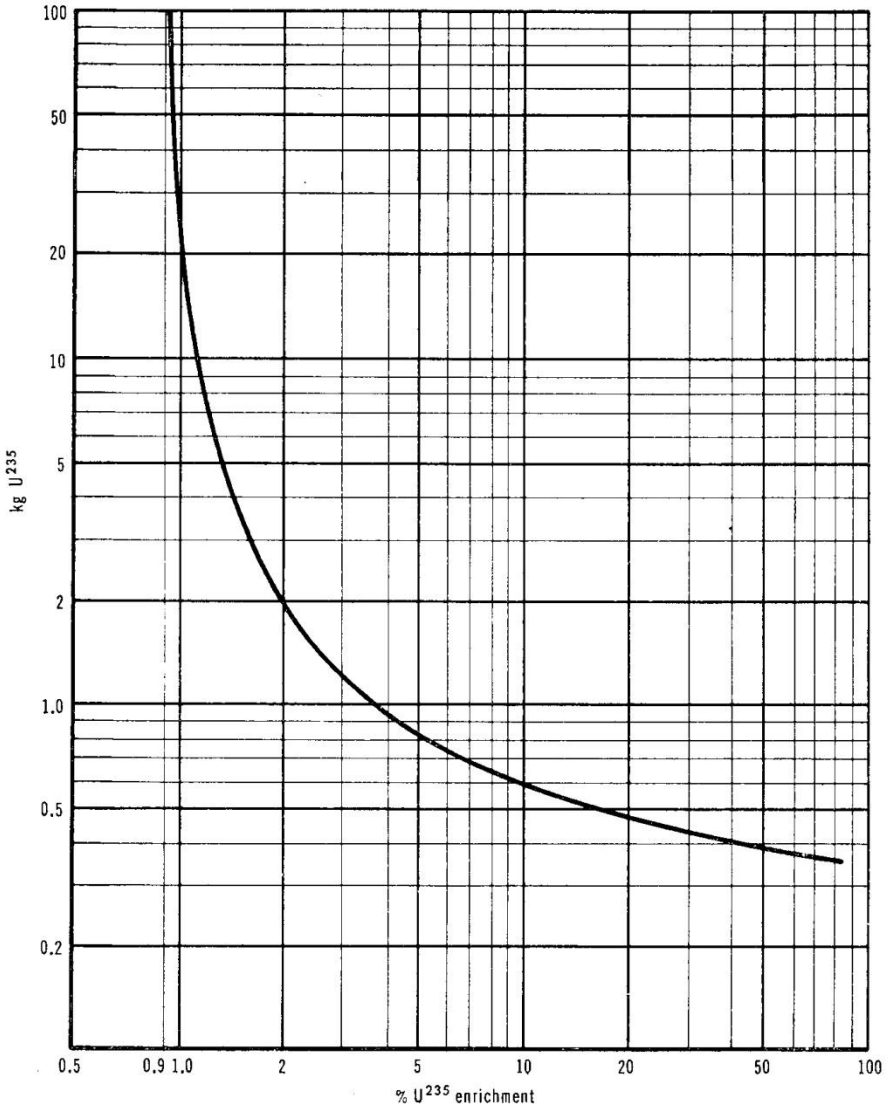

Figure 2. *Nuclearly safe masses for specified U 235 enrichment.*

batching while another facility may decide on a 400 g or even a 500 g limit, dependent on their consideration of the adequacy of control of the processing operations. The safe masses for solution or salts, shown in Figure 2, represent a decrease from measured or calculated critical values by a factor of 2.3, a value which provides a margin of safety were the contingency of double batching to occur. Usually, the 350 g U 235 limit is taken as a standard safe mass value where the handling of highly enriched uranium materials is of concern. There are conditions, however, depending on the type of material processed, the difficulty in securing a representative sample, and the

accuracy and reliability of laboratory analyses, where further reductions in the allowable mass limits may be required for nuclear safety. In some instances, operations supervision have, from their own evaluation of the control problems associated with certain processes, additionally limited the batch quantity processed regardless of higher allowable nuclearly safe values. Generally, the specified nuclearly safe mass for an operation also includes allowance for analytical error.

Hazards of Batch Operation

Once the nuclearly safe criteria have been established in terms of the batch size — and this includes consideration of the above items for a particular operation — it is the prime responsibility of the processing group concerned to ensure that the safeguards for mass control remain effective as far as possible. Generally, the controls which govern batch processing are such that multiple deviations from the normal must occur before a critical condition could exist. Thus, while operation with batch quantities is not necessarily unsafe — *per se* — it is obvious that it does lack some of the highly desirable features inherent in the use of safe geometry, in that careful and continuous evaluation of all phases is necessary to avoid a criticality condition, and additional mechanical safeguards and warning devices are usually required for preventive action.

Nuclear safety is in fact the mutually interdependent responsibility of all the groups concerned: nuclear safety, design engineering, laboratory and processing, etc. In this respect, I would like to discuss in more detail some of the problems of batch operation and some specific design and operational controls which have proved effective.

Operation and Equipment Controls

Sampling

One of the greatest hazards to safe batch operation is the use of poor techniques in sampling. No matter how excellent the analyses may be, they are meaningless unless good representative samples have been secured. From the simple possibility of an operator including solution hold-up in sample lines as part of the sample, to the more complex problems of sampling mixed solids and solutions such as aluminium-water vapour blast mixtures or other undissolved particles in solution where settling out occurs, emulsions or entrained solvents where separation is possible, or mixtures of solutions or solids of varying concentrations, the importance of an adequately representative sample in true determination of batch size cannot be over-emphasized. In such cases, we have found it advisable to take two separate samples, with a time interval between samples, and to request individual analysis for each sample.

The following case histories will illustrate types of sampling problems which may be encountered, and some of the techniques employed in securing representative samples.

Case 1. Contaminated combustible materials such as paper, rags, wood or oils which contain uranium materials over a wide enrichment range are burned in a furnace and the ash is collected in geometrically safe con-

tainers. Large quantities have been collected and it is considered expedient to process this material through screening, blending and dissolving operations in batch quantities based on the highest enrichment for various specified ranges. Since the material per container could vary considerably in isotopic content, the sampling procedure considered adequate requires that a full-length tube sample be taken of the contents of each container, and this material is then blended, sampled and analyzed for total uranium and U 235 content. Enrichment ranges are then established from the data obtained and containers constituting the specified mass for a particular range are grouped together. As a further precaution, additional samples for comparison purposes are also taken after each batching operation so that any errors which may occur will be detected prior to the leaching operation. Although material hold-up in the large equipment used for screening and blending is possible, this is controlled by weighing each batch prior to and after each operation and either reducing the size of the next batch or cleaning out the equipment.

Case 2. Equipment from locations of differing but known enrichments was spray-decontaminated and the resultant solutions, which contained small particles of undissolved solids, were collected and stored in a series of interconnected safe-geometry pipes. Due to conditions at the time which prevented processing this material in safe equipment, it was proposed to withdraw the solution into drums on a batch basis and then to process the material in large tanks.

Since the solution contained undissolved solids, the principal problem was that of assuring representative sampling. This was solved by recycling the solution in the interconnected pipe storage array until the analysis results of two samples, taken with a time interval between, agreed within 5 %; the minimum recycling time found adequate for this condition was twelve minutes. With the maximum enrichment known for each series of equipment decontamination, and the uranium concentration of the homogeneous solution readily ascertained, the volume of solution containing a safe batch quantity was easily determined and controlled. However, there have been instances where the material was of such a nature that analytical results indicated sampling errors by as much as a factor of ten, thus necessitating a corresponding reduction in the allowable safe mass from 350 g U 235 to 35 g.

Analysis

Other examples of problems where the technique of sampling is an important factor could be cited, but one can undoubtedly recall instances from one's own operations. It is of interest that, regardless of the sampling method, the results of laboratory analysis are generally accepted for use without question since the analytical procedures are known to be carefully determined and the reliability and accuracy of the method used well established. However, the operator should be trained to view all results with a certain degree of scepticism until he has made comparison checks, or reviewed aspects of the problem which tend to support the laboratory results. Conversely, while the chemist seldom has the opportunity to learn the various process operations, he has frequently proved instrumental in detecting errors of misoperation, or of poor sampling, by noting when the analysis results were not in line with samples for verification. In these cases, it is desirable that two independent samples be requested for the check test.

Administrative Controls

In the electronic age which is now unfolding, the functions of the operator are being superseded largely by automatic instrumentation controls. However, such changes primarily affect only the situations concerned with continuous processing, those pertaining to batch operation remaining very much operator-dependent. Hence, written procedures covering each phase of an operation, including the necessary nuclear safety precautions to be followed, should be available and all operators, even temporary, made familiar with those which concern them. Instructions should be clearly given, preferably in writing, and it is often desirable to have either the operator or the foreman, or both, sign a check-off sheet on which have been listed the special controls necessary to assure nuclear safety. This is of particular importance when the operation involves a change from that conducted in nuclearly safe geometry to that of batch operation in unsafe equipment. The use of transfer sheets for incoming or outgoing material, which list container numbers, weights, type of material, U 235 enrichment range and source location, and are checked against the data on each container tag, serves as an excellent means for detecting possible errors. The use of these transfer records should be emphasized and frequent supervisory checks made to ascertain that their purpose is not disregarded. Frequently, it is also important that verification be made concerning material received from other plants, and that reliance be not placed completely on analyses as given in the transmitted records. This may also be applicable to certain within-plant operation.

Although the laboratory is usually considered as a unit apart from other plant operations, its functions are very closely allied and similar problems of administrative control exist. While it is important to the process operation, for example, that the analytical results be obtained as quickly and as accurately as possible, it is equally as important to the laboratory that such results do not involve potential hazards due to a misplaced decimal, calculation error, a reversed number, the misreading of data, and others which could seriously affect those operations which are based on safe mass or safe volume. Thus, not only are tested procedures in effect, but careful records are kept of data concerning incoming materials, and special attention is given to the analytical results, with cross-checks used to note mechanical errors of calculation, or possible process errors, as determined from the sample.

Controls in Batch Processing

One advantage of batch operation is that it enables the expedient use of equipment of varied geometry for processing large volumes of dilute solution at any enrichment, for processing large quantities of solid materials at specified low-range enrichments, or for handling materials which are not only of low enrichment but are also moderation-controlled. Although over the years many of the processes used have evolved from those of batch operation to those of safe geometry, as indicated in Figure 3 which depicts some of the equipment in a geometrically safe uranium recovery facility, there can remain a number of instances where it is not feasible to go to safe geometry. Hence, batch processing continues as an important function in plant operations. Although some processes involving such operations appear

Figure 3. *Recovery facility, main floor.*

relatively simple and the possibilities of difficulties which could lead to a nuclear incident also appear quite remote, the hazards of such an occurrence and the associated ramifications of the problem are considered of such significance as to warrant the employment of precautions in even the most doubtful case.

The following examples of problems encountered in batch operation and some of the design controls used in assuring the nuclear safety of the operation may be of interest. They are not distinctively different from similar problems elsewhere but serve to highlight the importance of batch control from the preliminary sample to the end result. You will note that operator dependence, while still important, can be considerably reduced by means of equipment controls as indicated in the first example which illustrates the use of multiple controls, these including warning devices, which are considered necessary from a nuclear safety viewpoint but which may be considered by operations personnel as somewhat excessive control features. However, in this case, the operator could not be immediately available at all times.

Case 1: feed operation. Several cylinders, each containing a moderation-limited safe mass of UF_6 are to be fed to the process operation. The cylinders are to be mounted in tanks and steam heated so that the UF_6 is fed in the gas phase. The hazard evaluated was the possibility of an unsafe, moderated accumulation in the geometrically unsafe tanks in the event of a line rupture or valve leakage, other conditions of operation being such that rupture of a cylinder was considered essentially impossible.

Conductivity cells connected to a recorder were used to monitor the steam condensate at each unit and, in the event of a UF_6 release, these could actuate audible and visible alarms in several operator locations, actuate to the closed position solenoid valves in both the steam and UF_6 feed lines, and actuate a water spray system for ' freeze-down ' of the UF_6 at the point of leakage. Other precautions included additional valving in the UF_6 feed lines which could be closed by remote control, or manually at the feed station; emergency power to the alarm circuits; and provisions for safe geometry control of any uranium solution formed in the system. It is perhaps advisable at this point again to direct attention to the advisability of not overlooking any facet of a problem where nuclear safety is concerned. As an illustration and in reference to the above problem, is the discarding of low uranium concentration solutions to storm sewers. Since this is a possibility, knowledge is required of the possible hold-up locations where uranium, over a period of time, could slowly accumulate due to other materials which could cause precipitation, as unsoluble solid particles settling out, and — perhaps most serious — as the result of a sudden release of large quantities of material through equipment failure or misoperation.

Case 2: large equipment usage. A further word of warning may not be amiss concerning operations which involve dissolution of batch quantities in unsafe equipment containing large volumes of solution, or the precipitation of material from such solution. Regardless of the effect of mechanical stirring, residue build-up on the walls may occur and not be detected due to problems of sampling and analysis, or the small cumulative processing losses which may be noted may be considered acceptable to the operation; moreover, it may be that the operator on visual inspection considered the deposition as only a small and negligible layer. However, over a period

of time, it is possible for a multi-batch accumulation to occur and the steam cleaning or washing down of this material could result in an unsafe accumulation on the base of the tank. Again, there is some question concerning the advisability of adding the undissolved batch to the agitated solution rather than pouring solution on to that which, through error, might be a multi-batch quantity.

Case 3: volume-mass control. Where the handling of large quantities of low-enrichment, solid materials is necessary, other than in geometries considered safe for the assays involved, a combination of mass and volume control rather than mass alone is often effective. Thus, if the volume of a container is such that it cannot hold more than 80 % of the minimum critical mass for the enrichment in question, which corresponds to 1.84 times the safe mass, the unit is considered safe. Density is a principal consideration in this operation, and the material density for uranium determination is based on the maximum tap density encountered in the particular operation, although the free flow material densities after pulverizing or processing operations are, of course, considerably less. However, it is possible that unexpectedly higher densities could occur through caking or fusing if units were electrically heated and without adequate warning devices and controls in the event of a short in the electrical circuit.

Generally, the equipment to be used in the mass-volume control operation noted above is of such dimensions as to require some type of modification to meet the volume condition. Several methods can be used, varying from an actual volume reduction by the insertion of fixed spacers or the remodelling of equipment to size, to volume control through use of the inlet feed line. Many of us can probably remember childhood days when we filled a pail with sand and discovered that, if a piece of cardboard or some other wedge were placed in a partially-filled pail, one could then fill the pail with sand on one side of the cardboard without raising the level of the sand on the other. This method would be a simple solution to volume control, rather than inserting a volume displacement unit, provided it were in a fixed position. Similarly, the inlet pipe could be so positioned that flow would be stopped by the rising bed of material. In such cases, the volume of the inlet pipe for several feet is considered as part of the volume occupied by that of the specified mass, thus controlling not only the required element but also avoiding a problem of interaction. In addition, the screw feed mechanisms used to fill such units can be equipped with overload alarms, and with overload devices for shut-down; shear pins can also be used in the feed screw mechanism which will withstand pressures only slightly above those for normal loads.

In other cases where large containers are used in processing operations involving low enrichment, solid materials and the safe mass in each unit may total several thousand pounds; they are mounted on special scales which are connected to micromaxes for continuous recording of weight changes. Load cells, for example, which operate on a strain-gauge principle, are frequently used. Other controls consist of load limit audible and visible warning devices, usually in two locations, and frequent inspection checks, which involve the use of check-off sheets, are also made by operating personnel. As in other operations, instrument maintenance checks of the control equipment are made frequently, since considerable reliance is placed on instrument dependability and accuracy.

Case 4: moderation control. In the preceding illustrations on the handling of large masses of uranium materials, the material is assumed to be moderated and reflected, as previously defined. There are other operations, particularly those involving UF_6 withdrawals, in which large masses may be safely handled in unsafe equipment as a batch operation provided the amount of intermixed moderator, such as hydrogen, is held to very low levels and adequate means of control are effected to prevent the specified limit from being exceeded. As an example, a maximum of 5,000 lb UF_6 at 2 % U 235 enrichment may be placed in a 30 in. dia., 6 ft long cylinder provided the amount of intermixed hydrogen is maintained below an H/U 235 moderation ratio of 3.7. Further, four of these cylinders could be safely stored in any configuration, while any number of such cylinders could be stored in a single row side-by-side with their axes lying in the same plane. Separation from other uranium materials, obviously, is a pertinent requirement, and no reflector worse than water should be placed near such units. Since both enrichment and moderation are essential controls, U 235 enrichment analyses are made prior to and during the withdrawal operations, the purity of the material must meet highly restricted standards, weight checks are continuously made for mass control, and temperature and pressure ranges of condenser operation are maintained by frequent operator checks; audible and visual alarm signals are provided to warn if the allowable limits of variation are exceeded. Further, transfer pumps are equipped with automatic cut-off switches which are interconnected with the alarm instrumentation and actuate when the set limit conditions are exceeded; they may also be actuated by pump overload conditions.

There are also other instances in which specific quantities of UF_6 at high U 235 enrichment, and in excess of amounts normally considered nuclearly safe, may be contained in non-geometrically safe equipment. Each container holding these quantities is also considered safe and specified spacing must be maintained. Figure 4 indicates those safe masses for specific moderations.

Case 5: density control. Whether the uranium materials are fully-moderated or under-moderated, one of the factors of concern in determining the safe mass is density. Normally, most of the uranium materials processed have uranium densities which are not greater than 3.2 g/cm^3 although crystal densities greater than this are indicated for a number of compounds. When it is necessary to process those materials which have densities greater than 3.2 g/U/cm^3, safe parameters are established for these materials by simply reducing the safe parameters normally used. This is done by multiplying the safe parameters by the ratio of the density (ρ_0) of a UO_2F_2 water solution, at a moderation corresponding to that for the material of higher density concerned, to that of the density (ρ) of this material. As a general criterion, the uranium densities used may be based on unmoderated materials, in which case ρ_0 is 3.2 g/U/ml. The following formulae give conservative values; however, it is necessary that the densities be rather carefully measured.

$$\text{Linear } (\rho_0/\rho), \qquad \text{Mass } (\rho_0/\rho)^2, \qquad \text{Volume } (\rho_0/\rho)^3$$

Case 6: other problems. Occasionally, there are certain proposed short-term or temporary operations which can present unexpected nuclear safety control problems, or which appear to present no immediate nuclear safety problem, but there do exist one or more doubtful possibilities which have to be taken into account. Thus, where evaporation, precipitation, or extrac-

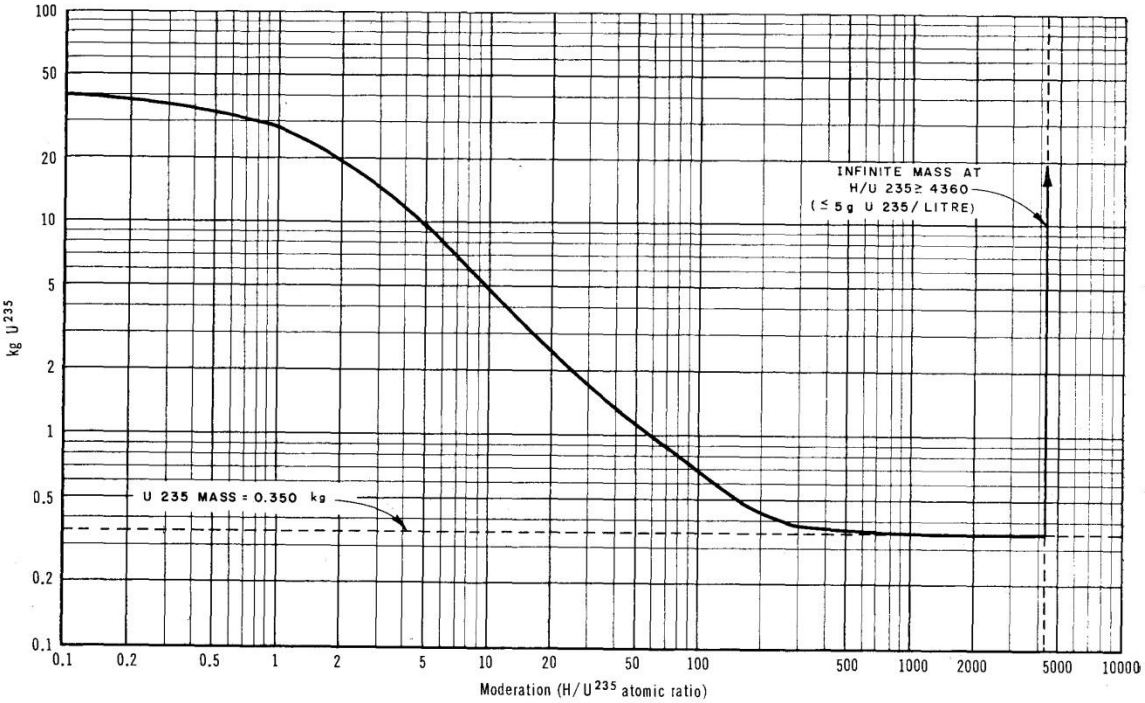

Figure 4. *Nucleary safe masses for specified moderation, any U 235 enrichment.*

tion phases are required for large volumes of solutions containing an estimated safe mass, based on laboratory analyses, one must be certain that there have been no errors in the sampling procedures used, or in the analytical results obtained. Normally, the use of double samples tends to preclude this possibility. However, it is frequently desirable as an augmentive safeguard that additional samples be taken and analyzed either prior to or during succeeding steps of the operation, and that arrangements be made with laboratory personnel for prompt handling and reporting which will be within a time interval such that continuation of the processing could not create a hazard. Although this may occasionally result in small delays, these can be considered negligible compared to the difficulties which could ensue were an indicated low result to be in error by a factor of ten, due to a misplaced decimal.

In the opposite direction, for example, water-abrasive mixtures have been used in the vapour blasting of equipment which has been through decontamination processes but which retains some residual quantities. The batch limitation is established by the maximum enrichment at which the equipment was used, but is reduced by the errors associated with the problem of representative sampling where material settles rapidly from solution. Since the quantity of uranium removed at each operation is very small, a considerable time lapse will be required to approach the specified mass limitation. Thus, chemical analyses for uranium content are sufficient, and the tanks for the vapour blast mixture and the water rinse can be considered as a single unit for batch control, although the capacities and the limitation on grammes of uranium per litre vary for each unit. A potential error here is administrative in that other than specified equipment could be cleaned in the unit.

Interaction

Although mass, or geometry, limitations are the basic criticality control methods for individual containers, consideration of the problems of interaction or neutron exchange and of spacing requirements is necessary where more than one container is involved. Several approaches have been made to this problem of neutron exchange, particularly between batch quantities, and to the determination of adequate spacing. Spacing at the Oak Ridge Gaseous Diffusion Plant is based on an interaction solid angle, and this in turn is related to several factors included in a determination of the k_{eff} or nearness to critical, assuming $k = 1$, of an individual critical unit, and considering the U 235 enrichment to be the maximum possible. The variation in multiplication factor, k_{eff}, for spherical vessels, each of which contains a constant safe mass of 350 g U 235, is shown in Figure 5. The k_{eff} value varies with the volume and reaches a maximum of about 0.65 at 12.5 litres. It decreases as the container volume becomes larger, primarily due to non-fission capture in the dilute uranium solution, and decreases for smaller volumes due to the increased neutron leakage probability. Hence, a value of $k = 0.65$ is used for computing batch separations at high enrichments. It is also considered applicable for low U 235-enriched materials where the mass is dependent on moderation control.

If the multiplication factor, k, cannot be readily calculated but, from experiment, the unit is known to be as safe as the parameters used for highly enriched material, then a k value of 0.80 may be used; this includes safe geometries and volumes at low U 235 enrichments and those volumes which contain no more than 80 % of the minimum critical mass. As an illustration,

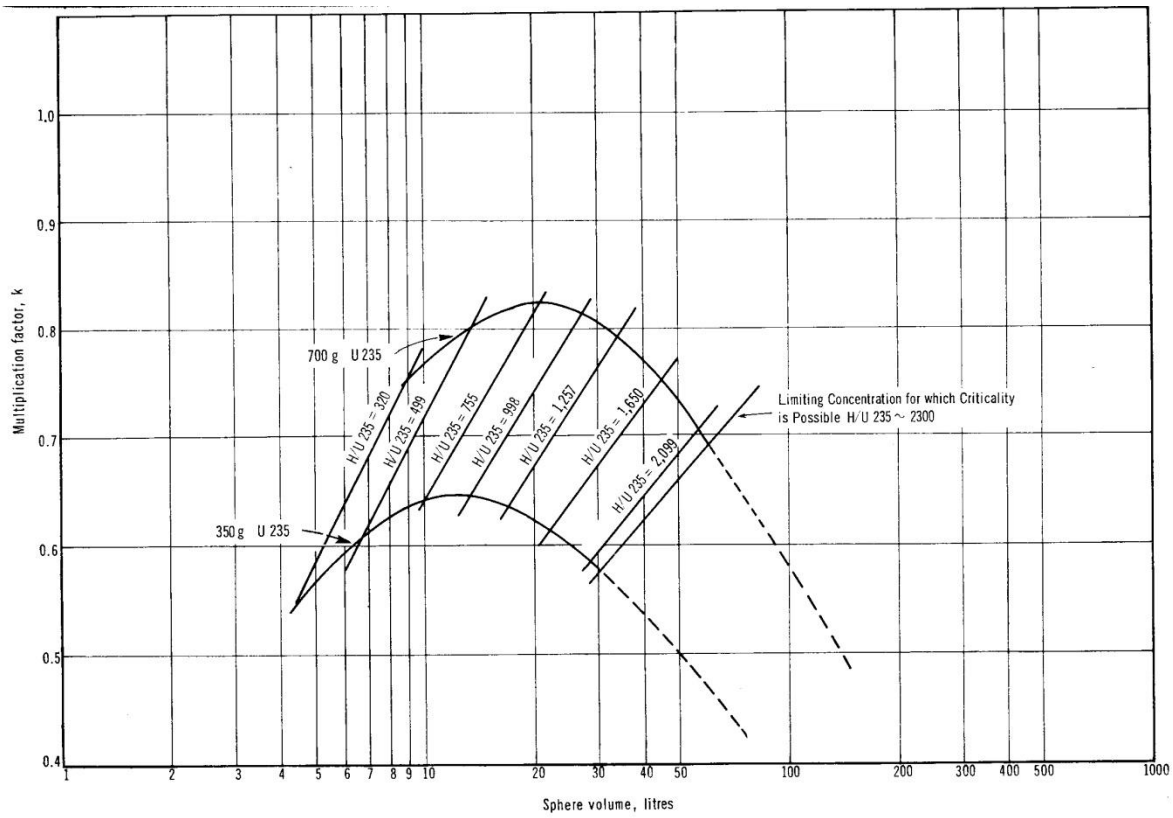

Figure 5. *Multiplication factor vs. sphere volume.*

the interaction fractional solid angle, Ω_f, for a k value of 0.65 is 0.20 (2.5 steradians); the fractional solid angle for a k value of 0.80 is 0.08 (1.0 steradian).

Another method frequently used in determining the required separation between units where the safe mass is contained in a large volume of dilute solution and the concentration could be altered by freezing or precipitation, is to consider the uranium in solution as being concentrated to 5 g U 235/litre; the dimensions of the occupied volume are then used in the pertinent solid angle formula. This, of course, is not applicable for those instances where concentration control rather than mass control is involved, a value of 5 g U 235/litre being generally considered as the upper limit in concentration at which an infinite volume of solution is considered safe.

Reflection

It was noted earlier that an individual unit, whether of geometry or mass, is considered safe only if it is safe under the two restrictive conditions of full moderation and reflection. Further, that water is considered as both a moderator and a reflector of neutrons and, in the latter rôle, provides opportunity for additional fissioning to occur, thus reducing the mass or volume required for criticality. One can also visualize that this reflective effect would not be very much for a thin layer of mist or frost on a container, but that it would be considerable were the unit to be embedded in a large cake of ice or immersed in water; in fact, it has been experimentally proved that approximately 12 in. of water will essentially attenuate all the neutron exchange between containers. Conversely, if one visualizes a number of containers placed close together and then flooded, there is not only the effect of reflection but the water in the interstices between the containers acts as a moderator, increasing the reactivity of the system.

Thus, it would appear that the use of large safe masses based on moderation control also presents problems in storage and shipping if the system could be flooded. A moderated individually safe mass could be placed in a single container, or in several containers grouped together, and immersed in water without presenting a nuclear safety problem. Similarly, a safe 'dry' or undermoderated mass in a single cylinder would also be safe immersed in water, but if placed in a number of containers positioned close together, could be made critical when immersed in water since it is then not only reflected but moderated. Hence, the safety factor in handling a safe mass of such material lies in having the units spaced with at least a minimum separation of 1 ft.

Nuclear Poisons

In line with an earlier comment that safe geometry operation had superseded many batch operations, I would like to present, briefly, the solution devised for a special problem, based on neutron poison data obtained in a series of criticality experiments with Pyrex rods, Raschig rings, and a special experiment with copper rods. Normally, nuclear poisons such as cadmium or boron, when used internally, are considered an additional safety feature only, and the poisoning effect of the nitrogen in a nitrate solution as a means of control for mass or concentration increase is also considered in this category. However, where standard nuclear safety controls are not feasible, internal nuclear poisons are used, provided the adequacy of the poison has been rigorously determined.

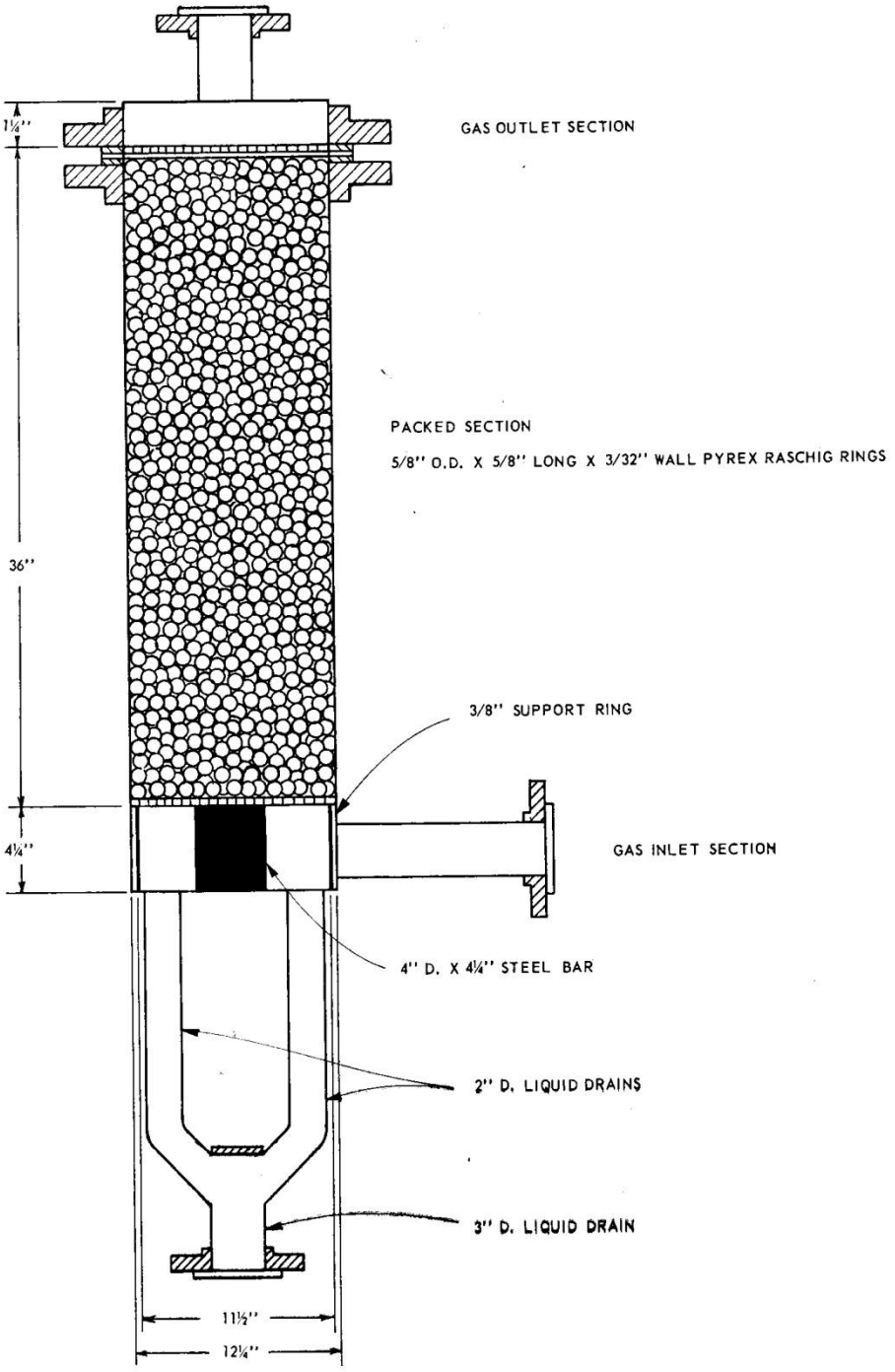

Figure 6. *Nucleary safe entrainment separator.*

Figure 6 shows the design of an evaporator entrainment separator which is considered nuclearly safe if filled with uranium solution at any enrichment provided the packed section is filled with 5/8 in. O.D. Pyrex Raschig rings which contain a minimum of 3.7 % boron by weight and occupy at least 27 % of the free volume available. Administrative controls include quality control specifications to ensure that the specified boron content for purchased rings is met, as well as annual inspection of the unit for detection of possible channelling, breakage, etc., and to check that the position and dimensions of the 4 in. steel bar in the inlet section remains unchanged. The Pyrex Raschig rings selected are smaller in diameter than similar materials used in the criticality experiments — thus higher in poison efficiency — and the specified volume of the rings is also much greater than that of an experimental lattice which could not be made critical in a larger cylinder. Since the evaluation was based upon the results of experimental and theoretical considerations of various neutron absorbers in well-moderated thermal systems, it should be noted that it is not applicable for unmoderated or slightly moderated systems.

Although one can readily visualize other types of equipment and operations where the use of fixed nuclear poisons may be quite useful, the conditions for which the poison is to be used should be carefully investigated, its effectiveness substantiated experimentally, and adequate administrative controls governing its use and retention put into effect. I am reminded of a pharmacist I knew a number of years ago who never had what you requested but always had something " just as good ": one should not be too trusting of such statements.

Concluding Remarks

In conclusion, it is perhaps desirable to restate the various points presented in the discussion: they are considered as part of a basic operation philosophy and may be summarized briefly as follows:

1. An operation will be considered safe if it requires that a double contingency must occur before a uranium configuration can become unsafe.
2. Only specifically designated groups are authorized to make up safe amounts of uranium and to process these safe amounts through recovery operations.
3. No uranium-containing equipment other than that specifically approved for an operation may be brought into an operating area.
4. In general, uniformity of sampling is controlled through the use of two separate samples, taken at different times during the blending operations and individually analyzed. The results of the analyses should agree within 5 %.
5. Where the U 235 enrichment is unknown and it is impractical to obtain isotopic analysis, chemical analyses are obtained and uranium material is handled in equipment which is not geometrically safe based on a total of 350 g uranium.

Finally, and of particular importance where batch operation is involved, there is the policy of not making assumptions as to what others have done, may do, or are supposed to do. The key to safe operation is frequent checks of every phase of the operation.

REFERENCES

1. H.F. HENRY, A.J. MALLETT, C.E. NEWLON and W.A. PRYOR, " Criticality Data and Nuclear Safety Guide Applicable to the Oak Ridge Gaseous Diffusion Plant ", *K-1019 (5th Rev.)* (1959).
2. A.D. CALLIHAN, W.J. OZEROFF, H.C. PAXTON and C.L. SCHUSKE, " Nuclear Safety Guide ", *TID-7016 (Rev. 1)* (1961).
3. H.F. HENRY, " Studies in Nuclear Safety ", *K-1380* (June 1957).
4. W. SCHULLER and M. LUNG, " Criticality Study ", *Eurochemic Technical Report No. 68* (Sept. 1959).
5. *Handbook of Criticality Data* (for Plant Designers and Operators), Risley (1960).
6. H.F. HENRY, " Guide to Shipment of U-235-Enriched Uranium Materials ", *TID-7019* (June 1959).

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DISCUSSION

H. HOWELLS, United Kingdom:

Mr. Mallett, you referred to the fact that you have a large number of alarm systems in your plants and you mentioned for example the use of strain-gauges for this purpose in some instances. Do you get any false alarms on these units?

A.J. MALLETT, United States:

No, we do not. Tests are made quite frequently by operating personnel and instrument maintenance to assure that the alarm systems are in operating condition. It is a very rare case when the alarms are inoperable or do not work and the correction is made immediately. It may be of interest that the nuclear safety department also makes fairly frequent field audits of all equipment and operations. The fewer people we get, of course, the fewer checks we make, and so we try to designate them in order of importance. If we should find anything that was inoperable at the time we made the check, and it was a nuclear safety requirement, it must be reported in writing; the inspection sheet being signed by the supervisor of the operation concerned and by the nuclear safety engineer The fault is then corrected as soon as possible.

C. ERGINSOY, I.A.E.A.:

I would like to ask Mr. Mallett what the policy is regarding scraps, filings, borings and any materials of which the composition, enrichment, density, moderation and other parameters are not precisely known.

A.J. MALLETT:

Normally, such material is sampled and the samples blended and re-sampled. Isotopic analysis is made and the amounts are handled in safe

mass quantities for the U 235 enrichment of concern less an allowance for analytical error for the type of material. The U 235 enrichment may also be based on accountability records of equipment usage or the maximum enrichment for a specific area; in either case, a uranium check (ppm) is made and the safe mass is that for the enrichment considered. In some cases, no analysis is made and the total weight is considered to be all uranium. Again, if the uranium content is known, the mass is often based on an assumed enrichment of approximately 90 %. We usually consider materials to be moderated except in the case of UF_6 withdrawals. Also material densities are of the order of 3.2 g U/cm^3 or less.

H. HOWELLS:

In the case of batch control, all depends on the operator carrying out the instructions which have been laid down. Do you make any special provisions for the training of these operators?

A.J. MALLETT:

Yes, that is the responsibility of the supervisor of the area concerned. We do have training sessions where we review the nuclear safety problems of the operation, both in meetings with the supervisors and often with the operators. Firstly, we train supervisors in nuclear safety and they, in turn, train their operators. We check frequently by inspections and often discuss problems with the operators and supervision. The hazard here is principally where a man is absent on vacation, personal leave, or sick, and the supervisor borrows a replacement from another location but does not tell him anything about the nuclear safety aspects of his job. He tells the operator only what he needs to do such as, " Open this valve, close this one, use this bucket, drain this." This is a bad situation, and the principal cure is repeated training of your supervisors and close follow-up by them of the job requirements.

E.R. WOODCOCK, United Kingdom:

Figure 2 of Mr. Mallett's paper shows nuclearly safe masses *versus* specified U 235 enrichment. This graph is apparently based upon the curve for homogenous mixtures. In his paper Dr. Callihan showed us a graph for lattice arrangements, which gave for the low enrichments a considerably lower critical mass. Should not Figure 2 be based on the lattice arrangement, rather than on the homogeneous mixtures?

A.J. MALLETT:

If we had types of materials by which we got into that sort of an arrangement, I would say yes. Ordinarily the materials we handle are either in solution or they are in a solid state with uranium densities below 3.2 g/cm^3. Even if that type of material in solid form is ground up and mixed with water throughout, it would not be considered as a heterogeneous system and we check it as a homogeneous system.

E.R. WOODCOCK:

You said that the materials you used were in solutions, but when we get to these low enrichments — and we are approaching the minimum critical mass in a homogeneous system — you have a wet solid and not a solution. It seems to be asking rather a lot, that you should expect these wet solids to be a homogeneous system, particularly if there is some other water around too.

A.J. MALLETT:

But the uranium density we use is still relatively low and the average density would be even lower.

F.R. CHARLESWORTH, United Kingdom:

I would first of all like to make an observation on the general topic of batch limitation and then ask a question. First of all, the safe masses that we have seen in the Figures were all for unlimited geometry conditions. If one is prepared to limit the geometry of the container, one can get great relaxation on the mass of material that one can put in the system. In fact, if one takes the specific example of a slab system for 93 % enriched material, where the density will not exceed 3.2 g/cm^3 of uranium, we know that the minimum critical slab of infinite extent is 1.8 in. This obviously requires an infinite mass of U 235 to make it critical. If one goes to a slab of 2 in. thickness, one still requires a very large mass of U 235, of the order of several hundred kg, to make it critical, and one could therefore, in a slab of that size, store or process a very large amount of material, so that the tank is virtually over-safe for the process in question. I would like to ask now, " Does anyone use such a geometry plus mass combination? "

A.J. MALLETT:

By the use of buckling equations one can go from infinite to finite dimensions. For instance, a 5 in. dia. cylinder which is safe for our material if infinitely long; and a shorter and larger diameter cylinder may be equally safe with the same material content, or the infinite, 1.5 in. thick slab become finite and thicker; but this is more a question of geometry control, not batch control. We tend to view everything as if it were infinitely long, and reflected, except for special occasions.

F.R. CHARLESWORTH:

My point was, that if one could design a tank which would hold perhaps several hundred kg of U 235, one could be relatively certain that such an amount could never be out of balance or exceeded during any condition of maloperation in the process in that particular vessel, and to all intents and purposes that tank is safe for any maloperation in the process.

K.M. HILL, United Kingdom:

When Mr. Charlesworth said, does anybody use this, I think he really meant anybody outside the United Kingdom. In the paper I gave during Session III, the dissolver was a case in point, using this principle in part.

And also, in the same plant, the product storage tanks are safe on a rather similar principle. The thickness of these tanks is greater than you would use for safety under all conditions and there is this element of there being an impossibility of getting a sufficiently large mass in the tank to render it critical.

A.J. MALLETT:

I might mention, without getting too far over into other nuclear safety problems, since this topic is batch control operation, one or perhaps two items of unexpected problems.

Over a period of time, while using a 1,500-gallon tank for batch operation, a coating formed on the sides of the tank. It was considered negligible by operations and was eventually removed by steaming and washing but was collected on the bottom of the tank. Analysis indicated a rather large amount of uranium in the solution although far from critical due to low enrichment. Hence, one cannot overlook all phases of an operation, and it indicates that safe geometry is a desirable method of operation. Where you have batch operation there remains a continuous worry although it can be done safely provided all operations connected with it are closely checked.

K.M. HILL:

I would like to ask Mr. Mallett how he writes off quantities of fissionable material which do get out of balance on these mass operations.

A.J. MALLETT:

There is at our plant a uranium accountability control group whose responsibility is to know where almost all quantities are, minute or large, or where they have gone, i.e., accumulated in equipment or discarded, and what criteria can be set up to discard material not worth recovery. Thus, some material has been buried; other, as solution, discarded. Burial would be with respect to safe mass or less distributed such that an unsafe accumulation in any manner is not foreseeable.

K.M. HILL:

In any accountancy operation there are errors in analysis, which inadvertantly accumulate as an apparent out-of-balance on any process: although you can treat these by statistical processes it is in the nature of things that such errors will be erased when plant supervisors wish to make sure they have not been responsible for some loss of yield. In any mass control plant, where there are places where uranium can conceivably accumulate — in my paper, for instance, I mentioned ventilation systems — there will always be this little bit of doubt as to where material has gone to. The problem I was raising was, how far one should go in determining just where these out-of-balances have disappeared to.

A.J. MALLETT:

There will be a slight continuing loss over-all. For example, UF_6-metal reactions, losses due to spills, etc., non-worthwhile recovery operations. The problem is not only where material can accumulate but also which area or

operation is charged for the gain or loss. Equipment which requires change-out is contaminated and must be cleaned. Records must be maintained to show the quantities recovered and the U 235 enrichment of the materials. Special accountability is often made for particular operations. Occasionally, the recovered values may be credited directly to an operation or pro-rated. It may be noted that plant inventory checks are made at specified intervals.

K.M. HILL:

If we take the figure of 350 g of U 235 being a minimum safe mass, when a section of the plant over which an accountancy check has been made has, over a period of months, years even maybe, accumulated an out-of-balance of 350 g, it is conceivable that these 350 g are lodged in one place that you haven't thought of, and that a critical accident might occur. This surely is the fundamental principle of accountancy control. There is, I think, a serious problem in the operation of any real plant where you are going to get out-of-balances which do not necessarily refer to the actual presence of physical material. They may be due to errors in analysis or errors in arithmetic; the material which appears to be out-of-balance may not actually be in the plant at all. There comes a stage when you have to stop your operations, or to write this material off. Because a surplus is very, very rare, the fissile material is always missing. There is a serious problem here, about what one does with a plant under these conditions.

A.J. MALLETT:

We try not to emulate the fellow who withdrew the solution from a tower, in which there was suspended material which settled out. He took a sample at the top and sent it to one laboratory for analysis, and a sample of solids at the bottom which he sent to another laboratory, for batch control. Fortunately it was discovered. There is a fair amount of material which, over a period of time, will disappear, one knows not where in all cases. As an accumulation in equipment it can be detected with gamma monitoring instruments; in other cases, by material analysis. In some, it is irretrievably lost. Nuclear safety people have the responsibility of periodically asking themselves concerning equipment and operations, " Where have I not been, what have I not seen or checked, where could material possibly be? " An illustration of this is in the laundering of contaminated coveralls and gloves which, over a period of years, resulted in a large sludge accumulation in the holdup tanks for the wash water. Recently, samples were obtained of this sludge and a quantity of uranium was found therein, although it was considerably less than the safe mass for the U 235 enrichment indicated by analysis. However, it was where one would not ordinarily expect an accumulation to be. Thus, your checks must be made at periodic intervals of filters, sumps, piping, and all types of equipment and locations imaginable to prevent conditions which could lead to a critical situation.

H.C. PAXTON, United States:

I would like to say that at Los Alamos, there are numerous situations in which we use geometry that we consider safe for the specific process. This depends upon the fact that we are confident that we will be given an opportunity to re-evaluate things, if the process is changed. One specific example

of this is a crucible dissolver, where the dimensions that would be normally considered safe, have been relaxed considerably because of limitations of charge.

A.J. MALLETT:

I foresee the time when geometrically-sized equipment, sized for different enrichments within given ranges, may perhaps be expanded through the use of fixed poisons; this will, of course, lead almost to the elimination of batch operation. Batch operation is a necessary thing under certain conditions, but the conditions and their tenure should be carefully evaluated.

E.D. CLAYTON, United States:

I would like to mention another example, which would fit the category to which Mr. Charlesworth has referred, and that is, in the Hanford Plutonium Critical Mass Laboratory, we have some slab tanks for plutonium solutions which are 2.5 in. thick, and they are what I refer to as limited safe. They are safe for the concentrations which we normally expect to run. If we exceeded these concentrations, the critical mass limits would be quite large, larger than the amount of plutonium we would normally have in the facility.

W. SCHÜLLER, Eurochemic:

In the right lower corner of Figure 4 of Mr. Mallett's paper the curve, " Safe mass *versus* moderation ", has been simplified in a conservative sense. Does this simplification reflect Mr. Mallett's belief that in the well-moderated region one should not make use of the relaxation of safe masses at low concentration, but should rather use either an always-safe concentration, or an always-safe mass?

A.J. MALLETT:

In terms of safe concentration, we would operate with solutions up to 5 g/litre U 235 for an infinite volume. We would very much hesitate to do that, however, except under very special conditions, supervised and followed much more than in routine operations. On a safe mass basis, you really don't care too much about the different processes it goes through, and the different equipment it is in: it is safe by itself.

W. SCHÜLLER:

I am raising this question because the same thing is true for geometry, and these are just the cases which were quoted in the earlier discussion. In many cases one does not take the always-safe geometry for any solution concentration, but a safe geometry for a specified maximum solution concentration, which is reasonable in a specific plant or operation. This practice would contradict the part of the diagram I am referring to, with the only difference that here, we are dealing with masses. So I am asking, do you consider it unsafe if one takes a solution of, let us say, 20 g/litre of fully-enriched uranium, and would allow a higher mass than 350 g?

A.J. MALLETT:

Obviously, one can have much more than 350 g U 235 on a concentration control basis as indicated previously, but, so far, we have not used a combined geometry and concentration control.

H.C. PAXTON:

The question refers to the artefact on the limit curve of Figure 4. On the low concentration side you have levelled the curve off, instead of giving it a normal critical mass form. Does this reflect some belief that you have about it being undesirable to make use of the full allowance in that region?

A.J. MALLETT:

This curve reflects an upper limit on the safe mass of a few years ago, and has not been changed since that time. I believe the safe mass could possibly be higher in this region.

J. GUNTHER, France:

M. Schüller a demandé s'il paraissait opportun d'augmenter la masse critique, dite de sécurité, lorsque les concentrations étaient de l'ordre de 20 g/litre. Lorsqu'on opère à des concentrations intermédiaires, situées entre la concentration de sécurité absolue, c'est-à-dire 5 g/litre avec le facteur de sécurité, et la concentration qui donne la masse critique réellement minimum, c'est-à-dire environ 35 g/litre pour l'uranium, je crois personnellement qu'il ne faut pas considérer la masse comme le facteur susceptible d'assurer la sécurité, mais qu'il convient de jouer sur la concentration elle-même, compte tenu de toutes les réserves que j'ai énoncées en ce qui concerne l'utilisation de la concentration comme variable de sécurité. A partir du moment où l'on opère sur des solutions aqueuses, je crois qu'il est peu opportun d'avoir des masses de sécurité plus élevées sous prétexte que la concentration est faible et qu'il vaudrait mieux s'efforcer de déterminer la concentration de sécurité pour le récipient utilisé.

A.J. MALLETT:

We find, when we talk about these 350 g U 235 for high enrichments, that there are times when we have reduced it to as low as 35 g, because of the difficulty of analysing accurately the type of material involved. Operationally, where we have specified limits, people reduce them even more because they are a little uncertain as to the reliability of their operation control.

HOMOGENEOUS AND HETEROGENEOUS POISONING AS POTENTIAL METHODS FOR CONTROLLING CRITICALITY

J. W. WACHTER
Union Carbide Nuclear Company, Y-12 Plant, Oak Ridge, United States

INTRODUCTION

General

All fissionable materials contain at least trace quantities of contaminants having neutron-absorbing properties. In reactors such materials compete for neutrons with the fissionable elements, thus decreasing the reactivity of the fuel, and tending to ' poison ' the reactor. At certain stages of reactor fuel manufacture, particularly in the salvage cycles, these poisons may be present to a sufficient degree as to affect criticality. General nuclear safety practice, however, ignores the existence of such poisons in defining the safety requirements of equipment because their presence is incidental to the processing itself and cannot be guaranteed. In general, only two cases of inherent poisoning are recognized in nuclear safety rules: the U 238 poisoning at the lower isotopic enrichments and the hydrogen poisoning of dilute aqueous solutions.

On the other hand, the planned addition of poisons to render specific operations safe has always been attractive to the nuclear safety specialist. Even in 1945, drums of dilute fissionable solutions were packed with thin wall Pyrex tubing to safeguard against criticality by making use of the boron content of the borosilicate glass. In general, such usage of a nuclear poison has been restricted to applications in which it provides an additional safeguard beyond those normally considered adequate. Relatively recently, however, under the impetus of economic pressures and with the routine processing of increasingly large amounts of material, the use of poisons to achieve nuclear safety is being re-examined. New experimental and calculational information has been developed, and its effect may be a re-definition of the philosophical bases for the use of poisons and lead to their greatly expanded use.

Definitions

Although any material which is a strong neutron absorber can properly be considered a nuclear poison, we shall consider as poisons only those materials that are specifically added for nuclear safety purposes. *Soluble poisons* are those added to process solutions under direct administrative control to

render safe such equipemnt as dissolvers, salvage solutions and pickling or cleaning solutions. *Fixed poisons* are those built into processing equipment so that the equipment becomes an individually safe vessel or so that its safety in a group of other containers is assured. By common usage, certain materials which are not particularly good neutron absorbers are sometimes referred to as poisons. This includes certain *poor moderators* which may be substituted for water or other moderating material to provide nuclear safety.

Homogeneous poisoning implies that the poison is uniformly distributed throughout the fissionable material and can be effected by the proper use of soluble poisons. Fixed poisons can be used to approximate homogeneous poisoning if at least one dimension is small, and if the pitch is small.* Otherwise, fixed poisons lead to *heterogeneous poisoning*, in which the relatively large size of the poison materials leads to a less efficient neutron absorption.

Nuclear Characteristics of Poisons

Effects due to Resonance Absorption

The effectiveness of a particular element as a poison depends strongly on the reactive system to which it is to be applied. For most elements, neutron absorption takes place because of resonances in their neutron absorption cross sections so that only neutrons of certain energies are strongly absorbed. Neutrons whose energies do not lie in these resonances are only weakly absorbed. The practical importance of this fact can be seen by Table I in which the relative effectiveness of several of the more common poisons is compared.**

Table I. Relative Effectiveness of Various Poisons

Material	$\frac{\Sigma_a}{\rho}$, cm²/g			
	Maxwell-Boltzmann Distribution	Thermal Reactor	Intermediate Reactor	Fast Reactor
Cadmium	11.67	7.33	3.16	0.00
Boron	37.40	21.19	8.44	0.01
Nitrogen	0.07	0.04	0.02	0.00
Hydrogen	0.18	0.10	0.04	0.00
Stainless Steel	0.02	0.01	0.01	0.00

The first column lists the effective poisoning ability of each of the elements on the assumption that the only neutrons present are thermal neutrons having a Maxwell-Boltzmann energy distribution. Even in a thermal reactor, however,

* The dimension and pitch are considered ' small ' if they are of the order of the neutron mean free path for the poison or fissionable material, respectively. For thermal neutrons this is about 0.010 cm for cadmium and boron sheets, and about 10 cm for an aqueous solution.

** Relative effectiveness is here measured by calculating the flux-averaged macroscopic absorption cross section per unit density of poison. Thus, these numbers are effective barns/g for the flux being considered, and multiplication by the poison density in g/cm³ yields the one-group macroscopic cross section for the poison material.

neutrons are born in the fission energies and spend appreciable time in the epithermal range before slowing down to thermal energy. The second column assumes a flux appropriate to a well-moderated solution reactor. At 2% isotopic enrichment, a reactor at optimum moderation does not involve pourable solutions and may have a neutron energy spectrum considerably faster than that at top enrichment. The third column compares the relative effectiveness of these poisons in this range. The last column assumes a neutron energy spectrum which is only slightly degraded from the fission distribution, corresponding to a dry metal reactor.

Comparing these values it can be seen that poisons such as cadmium which are black to thermal neutrons are not necessarily the best nuclear poisons. Unlike cadmium, boron is not a resonance absorber* and is therefore effective in the epithermal range of energies. It is seen to be more effective in any practical application, including the pure thermal reactor, than is cadmium. Poisons which are highly effective when used as an additive to process solutions have greatly decreased efficacy when applied to dry or metallic systems and, in some cases, are quite worthless. The choice of a poison must therefore be guided by knowledge of the nature of the critical system which might occur in the process material.

Reflection by Poison Sheets

The effectiveness of the nuclear poison depends also upon the specific manner in which it is utilized. Although cadmium will poison a process solution, it is effective only if it is dispersed throughout the solution. Wrapping a sheet of cadmium around an isolated process vessel is not only worthless as a means of poisoning but actually makes the vessel more nearly critical. This is because neutrons which are absorbed by the cadmium sheet would not normally have returned to the vessel in any case, and some of the neutrons which strike the cadmium are reflected back into the vessel, thereby increasing its reactivity. Of course, cadmium wrapping of vessels is effective both in preventing interaction among groups of subcritical vessels (since the cadmium is now interspersed within the scattered fissionable material) and also in reducing effects of reflection, since neutrons thermalized in the reflector are absorbed by the cadmium while returning to the vessel.

Effects of Heterogeneity

Poisons are most effective when placed in the region of highest neutron flux, as at the axial centre of cylinders. If the poison is introduced in the form of rods (or plates) and is uniformly distributed, it is less effective than an equivalent weight of poison distributed homogeneously. This is because, in the neutron energy region of strong neutron absorption by the poison, only the outer layer of the rod is fully effective. Relatively few neutrons can penetrate the skin of the rod. The mass of poison in the interior of the rod is thus shielded from most of the neutrons and contributes little to decreasing the reactivity of the vessel. This situation parallels that which applies to reactor fuel latticing, in which the interior of the fuel rod contributes little to sustaining

* The neutron absorption cross section for boron exhibits no resonance and falls off the neutron energy as $E^{-1/2}$; i.e. inversely to the neutron velocity, and is therefore called a ' pure 1/v absorber '.

the chain reaction. The magnitude of both effects can be expressed in terms of a ' disadvantage factor', calculated in a similar fashion.

Undesirable Side Effects

In assessing the value of poisons for a particular application, attention must be given to possible side effects. The density of the original solution will be reduced by use of a poison due to the displacement by the poison material, and, if the poison is in a soluble form, the moderation may be increased. If the fissionable material was originally dry, the increase in moderation might be sufficient to override the effect of neutron absorption in which case it might be hazardous to use such a poison. In general, the nuclear parameters of the unpoisoned system are not merely changed in value by the poison but are also changed in their relative relationship. Thus the moderation at which minimum mass occurs is generally decreased through the addition of poisons.

It is clear that the effective use of poisons depends upon a judicious choice of both the poisons to be employed and the nature of the material to be rendered safe. We shall now examine certain specific applications of both soluble and fixed poisons together with the calculational or experimental information supporting their use.

HOMOGENEOUS POISONS

Unfortunately, many practical difficulties, such as those of effective administrative control, complicate the nuclear safety situation in considering the addition of a soluble poison to a fissionable process solution. From the purely nuclear point of view, however, such poisoning is highly effective in that homogeneity can usually be assured and the least possible amount of poison need be added to ensure safety. Such fixed poisons as foils, thin walled tubing, and the like may approach soluble poisons in their uniformity of distribution and effectiveness, and the information given below could be extended to include them.

Infinite Aqueous Systems

Figure 1 indicates the minimum amount of poisoning required to render an infinite system of aqueous uranium solution critical. Two curves are shown for both cadmium and boron, one giving the required density of the poison and the other giving the mass ratio of the poison to U 235. The four-factor formula was used for this calculation and it was assumed that $\varepsilon p = 1$ and that $\eta = 2.12$.

It is seen that higher densities of cadmium are necessary to poison the reactor as the moderation is decreased. The mass ratio curves show that this density increase occurs primarily because of the increased amount of uranium in the more concentrated solution. In the region of non-pourable solutions, the neutron energies at which the nuclear reaction is propagated are much higher, and one would expect the levelling of the weight ratio curve to be replaced by a sharp rise, corresponding to the decreased efficiency of cadmium for fast systems. The mass ratio curve decreases in the region of the more moderated system, corresponding to the increased effect of the hydrogen absorption. The corresponding curves for boron show that the required

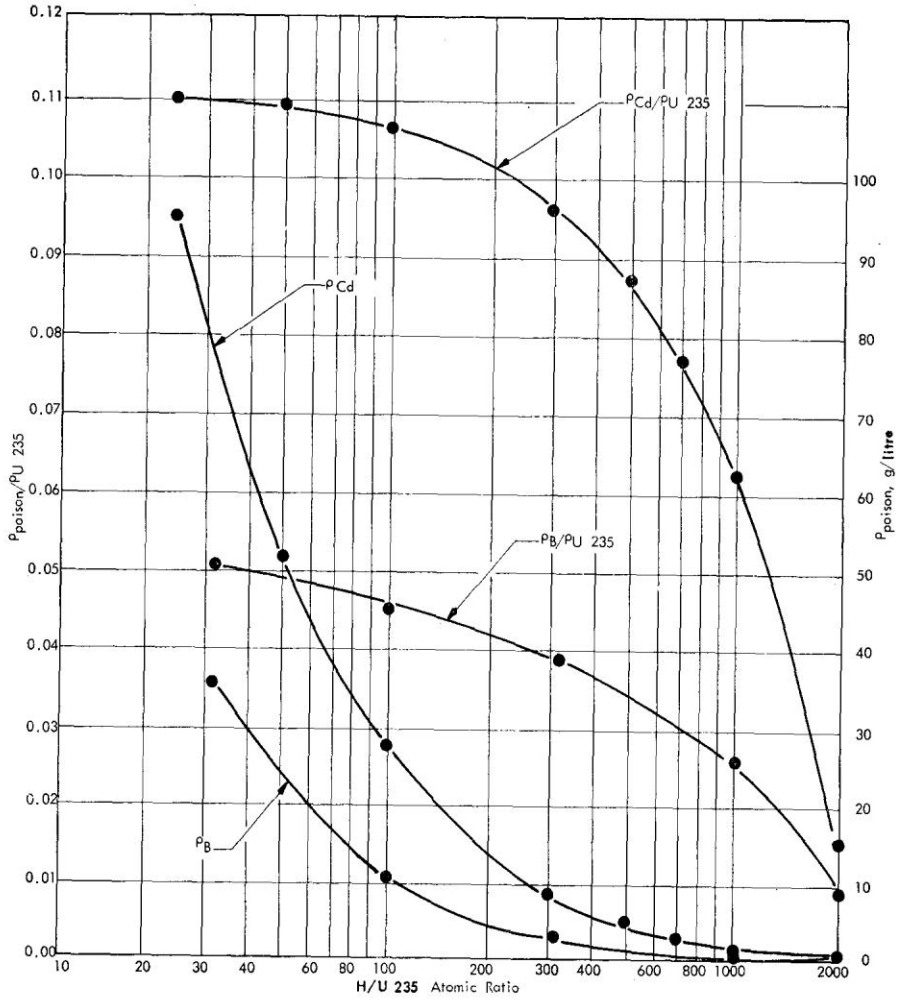

Figure 1. *Minimum amount of poison required to render an infinite system of aqueous uranium solution critical.*

density of boron is less than that required for cadmium, due primarily to the fact that these tables relate to the mass, rather than the atom, requirements.

The above discussion dealt with solutions containing uranium at approximately 90 % isotopic enrichment. Less poisoning is required at the lower enrichments due to the presence of the U 238. Ketzlach [1] has calculated that approximately 1 g/litre boron will render a 3 % U 235-enriched uranium solution in water just critical at an H/U 235 ratio of 320, and a g boron/g U 235 ratio of 0.015. This is approximately one-half the amount that would be required at the same degree of moderation for a fully-enriched aqueous solution.

The use of sufficient poison for an infinite system has practical advantages. Firstly, no account need be taken of the container volume into which the

solution might, in the future, be put. Secondly, the problem of interaction of containers is greatly reduced, since control need only be extended to cover unpoisoned solutions and any containers for which the reflection offered by poisoned solutions is a problem. The curves showing the required weight of poison for a given weight of fissionable material are of particular benefit in practical applications, since the amount of fissionable material in a container is usually well established by accountability measures. If soluble poisons are at all usable, the quantity of poison required is sufficiently small for differences between poisoning a finite system and an infinite system to be of little practical importance. For this reason it is frequently convenient to choose the maximum mass ratio value from these curves and poison all solutions to this extent, without regard for their actual uranium concentration. If the solution is actually quite dilute, the margin of safety becomes correspondingly greater.

Finite Aqueous Systems

Figure 2, due to Goodwin and Bidinger [2], shows calculated values of the bare spherical mass of U 235 for various mixtures of uranyl nitrate and cadmium nitrate aqueous solutions. The lower curve represents the unpoisoned reactor with a critical mass of approximately 1.5 kg U 235 at a density of about 0.5 g U/cm^3. As increasing amounts of poison are added, it is seen that the value of the minimum mass rises sharply and occurs at increasing values of uranium density.

These curves indicate that relatively small amounts of cadmium poison are sufficient to gain large margins of safety in any practical situation. Although it requires approximately 1.5 g cadmium for 10 g U 235 to render the solution safe in an infinite volume, no more than one-half this amount is required, under unreflected conditions, to raise the minimum critical mass of any solution by a factor of ten. In each of these cases, the minimum mass occurs within a relatively small range of volume. For these curves, the volume at which minimum mass occurs rises from about 30 litres to only about 70 litres. It is clear, however, that for a solution of low concentration, small amounts of poison quickly render it safe in large volumes.

Figure 3 shows the corresponding curves for plutonium solutions.

Special Cases

Soluble poisons can also be used in situations where metallic uranium is placed into a moderating material to which the soluble poison is added. Two special cases have been considered, the first involving the addition of soluble poisons to a dissolver and the second appropriate to a pickling operation with large metal pieces.

Ketzlach [1] has considered the dissolving of 1 % enriched U 235 fuel elements which, in the absence of poison, requires a limitation of the equipment to a safe diameter of 55.88 cm. Both thorium and boron were considered as poisons to be added to the dissolver solution. He concludes that approximately 1 g boron/litre would make the dissolving operation safe in an infinite system. The use of thorium as a poison was less effective, but 230 g thorium/litre was sufficient to increase the safe diameter of such a system to approximately 68.58 cm as compared to 55.88 cm for the unpoisoned system. This corresponds to a fourfold increase in the minimum critical mass.

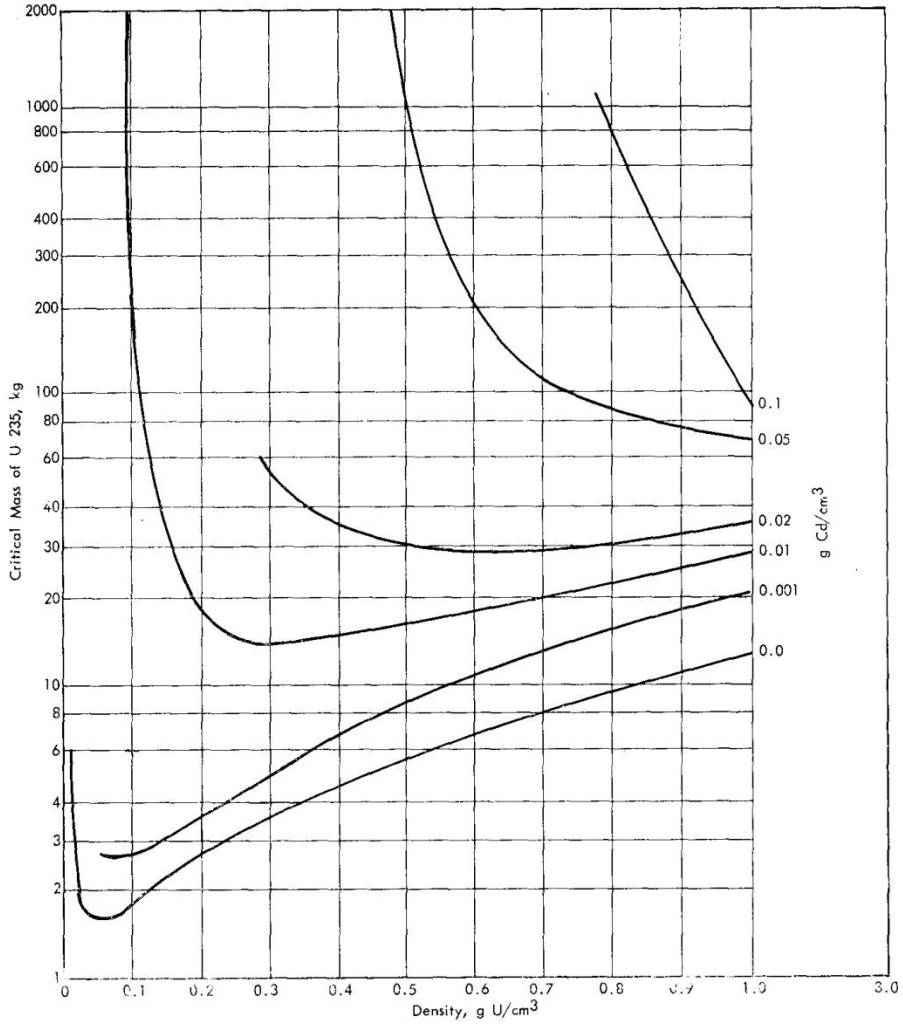

Figure 2. *Bare spherical mass of U 235 in $UO_2(NO_3)_2$ and $Cd(NO_3)_2$ mixtures.*

Schuske *et al.* [3] have experimentally determined the poisoning effect of a cadmium nitrate solution as a reflector of a 25 × 41 cm Oralloy slab. They find the critical slab thickness to be 3.3 cm with an unpoisoned water reflector and to increase to approximately 5.7 cm as the cadmium concentration in the reflector is raised to approximately 6 g/litre. When the slab is reflected by a uranium solution, increased amounts of cadmium must be added to counteract the reactivity presented by the uranium solution itself. In this situation the results are not universally applicable, since the overall size of the reflector contributes to the reactivity. It may be noted, however, that in this instance the ratio of weights of cadmium and U 235 in the solution was of the order of 0.14 g cadmium/g U 235.

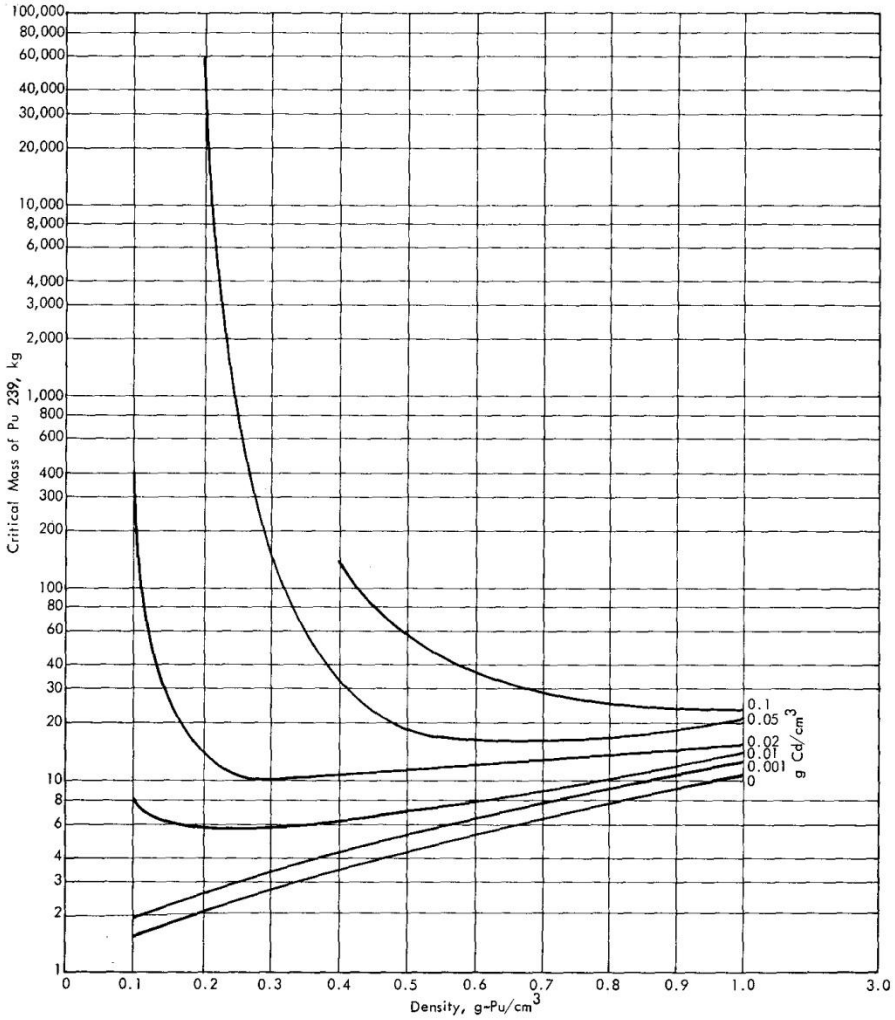

Figure 3. *Bare spherical critical mass of Pu 239 in $Pu(NO_3)_7$ and $Cd(NO_3)_2$ mixtures.*

In a practical pickling operation, uranium is continuously dissolved from the metal slab, which is then reflected by a uranium-acid mixture. Figure 4 illustrates the effect of poisoning in such a situation. The poison acts both to poison the solution and to reduce the effectiveness of the water reflector.

In certain operations involving dry materials, the use of a poor moderator as a poison is often highly effective in enhancing nuclear safety. Perchloroethylene has found wide acceptance as an antipyrophoric liquid to be used in place of oil for the storage of metallic uranium chips and fines. McLendon and Morfitt [4] have shown the effectiveness of this substitution in increasing the critical masses. True poison solutions have been used with otherwise dry materials for cleaning purposes; but, as expressed earlier (see page 292), this should only be done after careful consideration of the moderating effects of the solution.

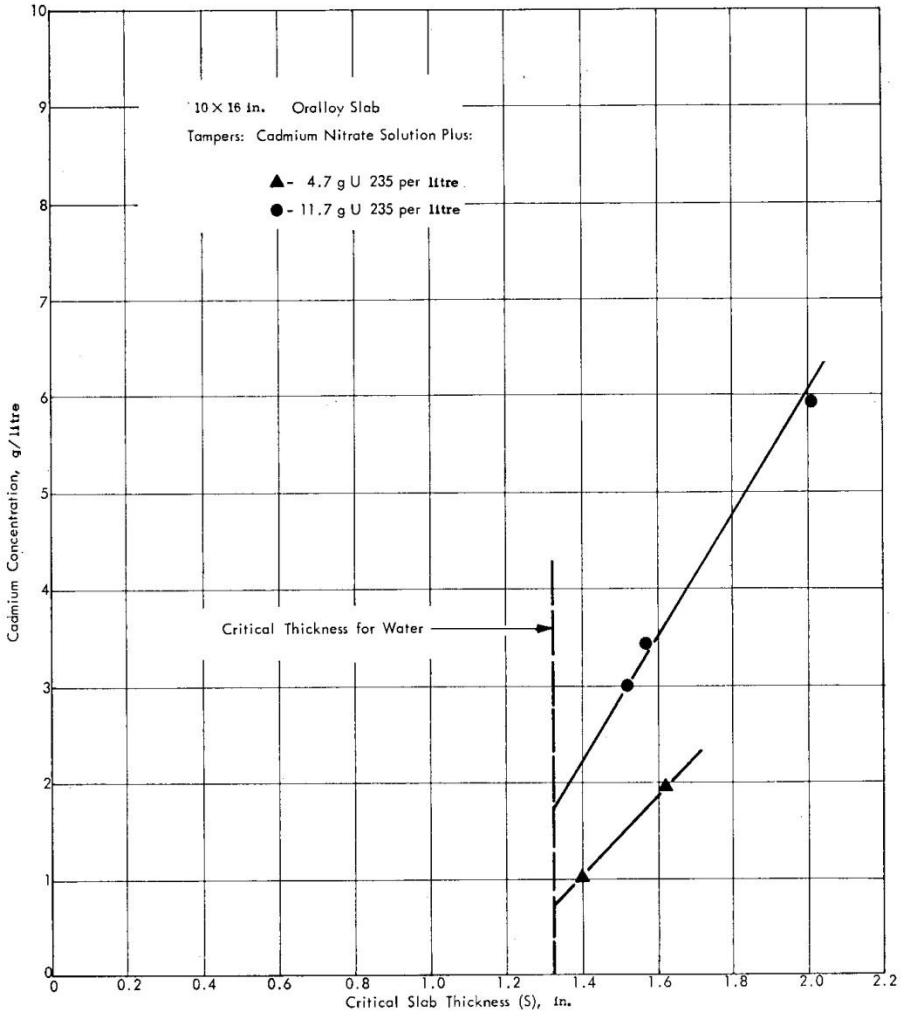

Figure 4. *Poisoning effect of cadmium.*

Special situations such as those described above can only be approached on an individual basis. These results are not readily applicable to any other situation and are included here only as a means of indicating the sort of special benefits that could be derived from the use of soluble poisons.

Heterogeneous Poisons

The use of heterogeneous, fixed poisons in solid form is intended to convert a specific piece of equipment into a nuclearly safe vessel. Fixed poisons are particularly advantageous when used in vessels which form a portion of a continuous train of processing equipment. In such cases, the require-

ments on the poisons are less stringent and the confidence with which the nature of the process material is known is considerably higher. Economically such poisons are desirable, not merely because they allow equipment of larger volumes to be constructed but also because poisoning can frequently be accomplished so as to use commercially-available process equipment. Unlike the use of soluble poisons, which can be mixed intimately with the fissionable material, it is usually necessary to conduct specific experimentation or calculations in order to establish the nuclear safety of a specific piece of equipment. Generally speaking, most of the data so far acquired on poisoned systems are related directly to specific equipment and are not suitable for wide application. They indicate, however, the variety of applications in which poisoning may advantageously be employed.

Boral Plates

Gilley *et al.* [5] have investigated the effect of boral plates uniformly spaced within a vessel of uranium solution having a 1.5 m $\times$ 0.75 m cross section. The critical height was determined as a function of the centre-to-centre spacing of the poison slabs. The thickness of the boron was 0.64 cm or about 300 mg B/cm^2. A consequence of the thickness of these poison plates, as discussed previously, is a depression of the neutron flux in the interior of the slab. This flux depression causes a reduction in the efficiency of the boron poisoning, although this depression would not be so severe at higher neutron energies.

Figure 5 shows the experimental data. It is seen that for the half-reflected case, a maximum spacing of 4.9 cm completely poisons the solution.

Borosilicate Glass Pipes and Rings

Additional experiments [6] have been conducted using Pyrex glass pipes as poison rods. These glass pipes contained boron oxide to approximately 12 wt % or about 4 % elemental boron. The pipes were of various diameters and about 1 m in length and were mounted in several hexagonal lattices to achieve the various glass-to-solution volume ratios indicated. The results are given in Figure 6. The unpoisoned critical heights for both bare and reflected cases are indicated on the ordinate and are the values for an aluminimum cylinder 51 cm in diameter and reflected to solution height. The critical height rises at first slowly and then quickly approaches an asymptote at a glass-to-solution volume ratio of approximately 0.17 for the reflected case and 0.13 for the bare case.

Raschig rings are segments of glass pipe whose height is approximately the same as the diameter. As a poisoning medium they have the great advantage that they can be poured into a vessel of any size or shape without the need for special supports and with relatively little concern for the placement of the access ports. In use, they drain well and provide a quite uniform distribution of poison. Experiments using such rings have given results comparable with the glass pipes. Figure 8 presents data points from experiments by Fox and Gilley [6]. Figures 7 and 8 give calculated values of the critical masses for uranium and plutonium due to Goodwin and Bidinger [2] and include a measured point reported by Bidinger *et al.* [7].

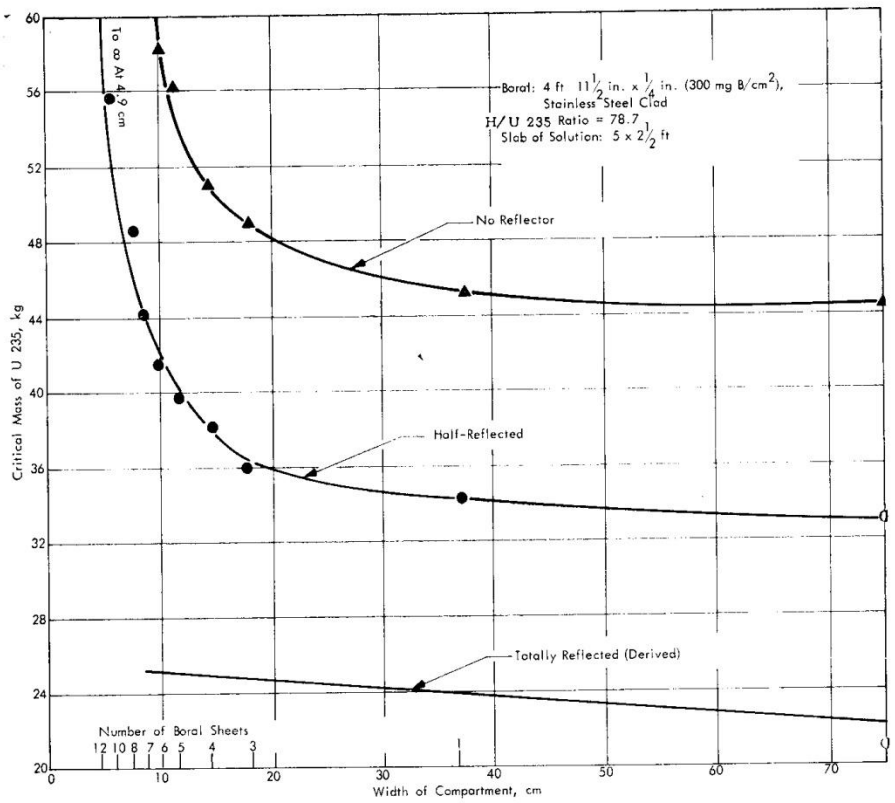

Figure 5. *Effect of boron poisoning on critical height of a slab of uranium solution. Half-reflected points were taken with the level of the reflector water at the height of the centre of mass of the fuel solution. Uranyl fluoride enriched to 93.3 % U 235 was used. (Gilley, Cronin and Harness, " Boron Poisoning in Critical Slabs ", ORNL-1715).*

Poison Sheathing of Solutions

Cadmium sheathing is highly effective in reducing reactivity when placed between an individual container and its reflector. In plant applications, reflection is frequently due to nearby walls, personnel, and the like, and the wrapping of cadmium around the vessel serves to isolate it from its environment. In this case, the fact that this increases the reactivity of the container itself is of small importance compared with the desirability of reducing criticality restrictions which otherwise would be necessary. Where the normal plant situation involves equipment that is unshielded, except when personnel are performing maintenance or operating functions in its vicinity, the use of sheathing may permit a relaxation from those safe dimensions which are based upon full reflection.

Cadmium wrapping of vessels of well-moderated aqueous solutions is also of direct benefit in reducing interaction. As seen in Figure 9, due to Fox *et al.*[8], the cadmium sheathing increases the critical heights of arrays of cylinders. Cadmium sheathing may be of questionable value in connection with poorly moderated and metallic units, but it appears to be a positive

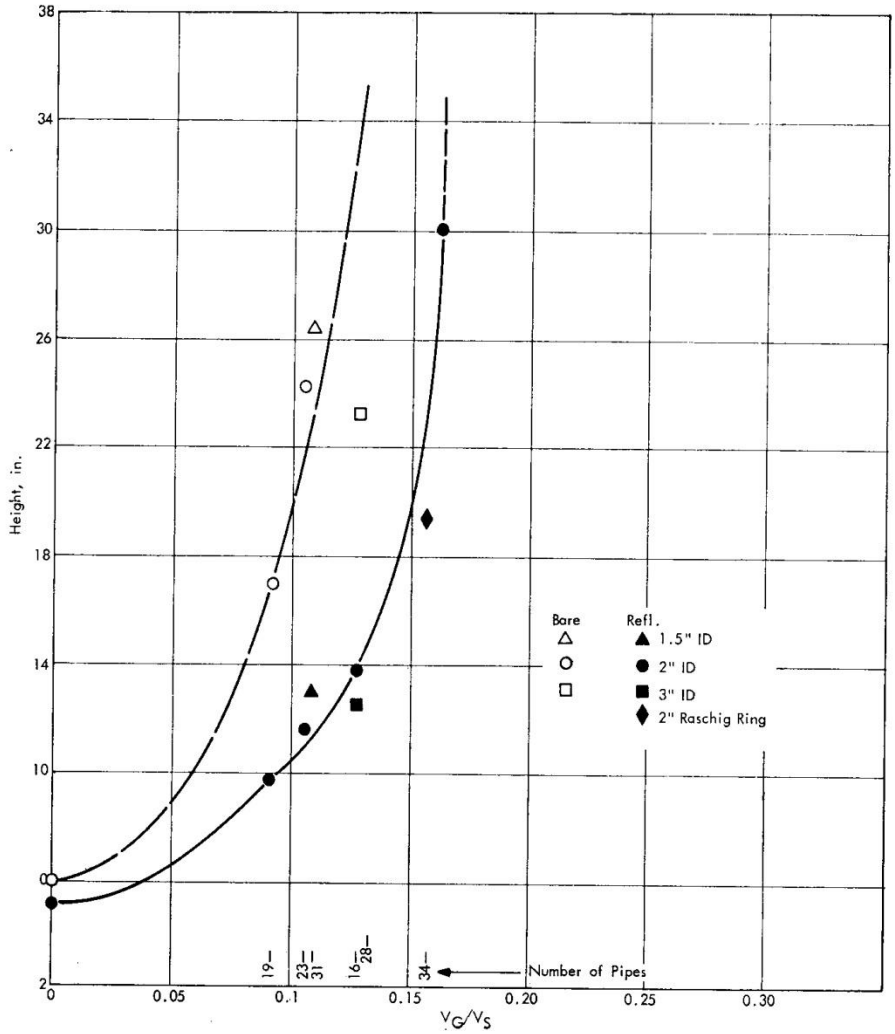

Figure 6. *Critical height of uranyl nitrate solution in a 20-in. I.D. SS cylinder filled with pyrex glass pipe versus the glass-to-solution volume ratio. The uranium was enriched to 87 % U 235, and the H/U 235 atom ratio was 85.*

benefit in any solution situation. Advantage has been taken of this fact in numerous applications, particularly in the field of reactor fuel element shipments. Since the fuel elements have been irradiated, the presence of cooling water is essential, and the resultant well-moderated system can be effectively reduced in reactivity by grouping the fuel rods into compartments made up of poisoned barriers. At lower enrichments where less poisoning is required, use has been made of pipes surrounding each fuel element. In each of these cases, it is essential to provide means of ascertaining that the poison sheets are in place and remain effective. Where the poison sheets are not available for visual inspection, multiplication methods have been employed.

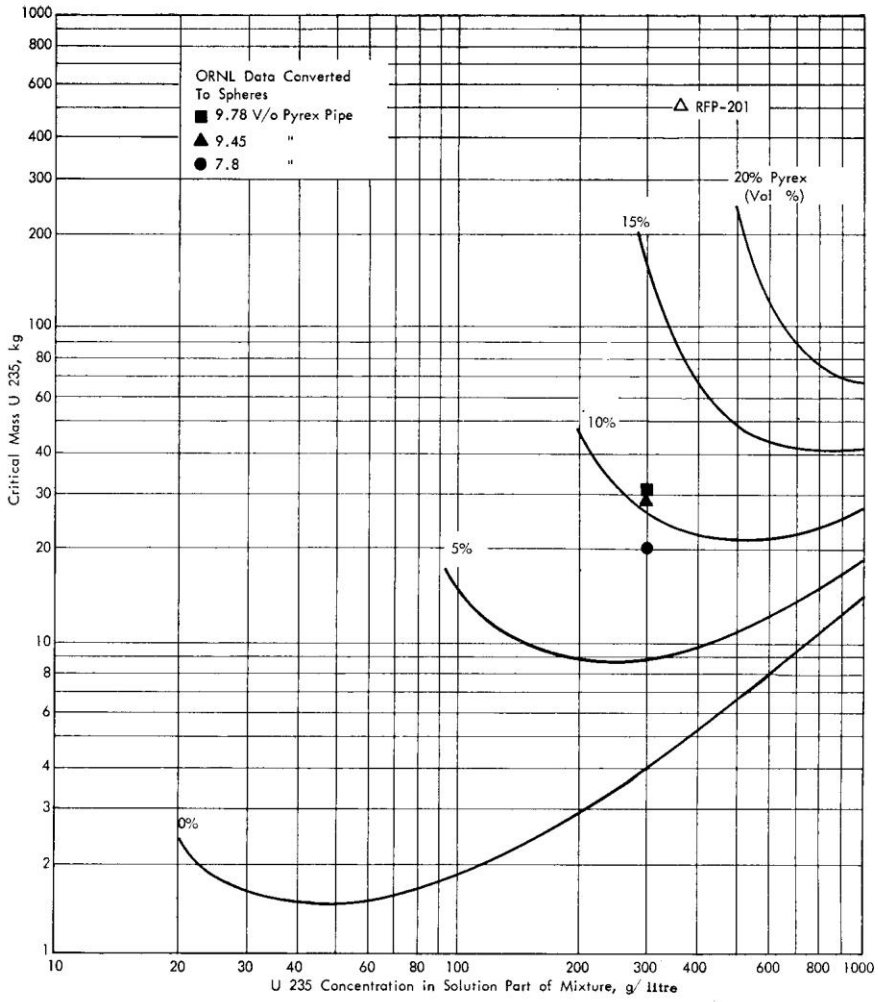

Figure 7. *Spherical critical pyrex poisoned U 235 solution, pyrex 12.5 per cent* B_2O_3.

Poison Sheathing of Metals

Metallic poison sheets can be applied as sheathing only with caution lest the degree of reflection given to the individual member of the array prove more hazardous than the original configuration where poison was not used. This caution applies most strongly to interacting arrays of metallic units which may form a critical system propagated by fast neutrons. It appears that cadmium sheathing on the walls of a vault containing metal units has virtually no effect. While it might appear that the use of a plastic containing large amounts of boron would be effective in such a case, this is not always so, and the use of boronated plastic sheets may decrease the number of critical units in an array.

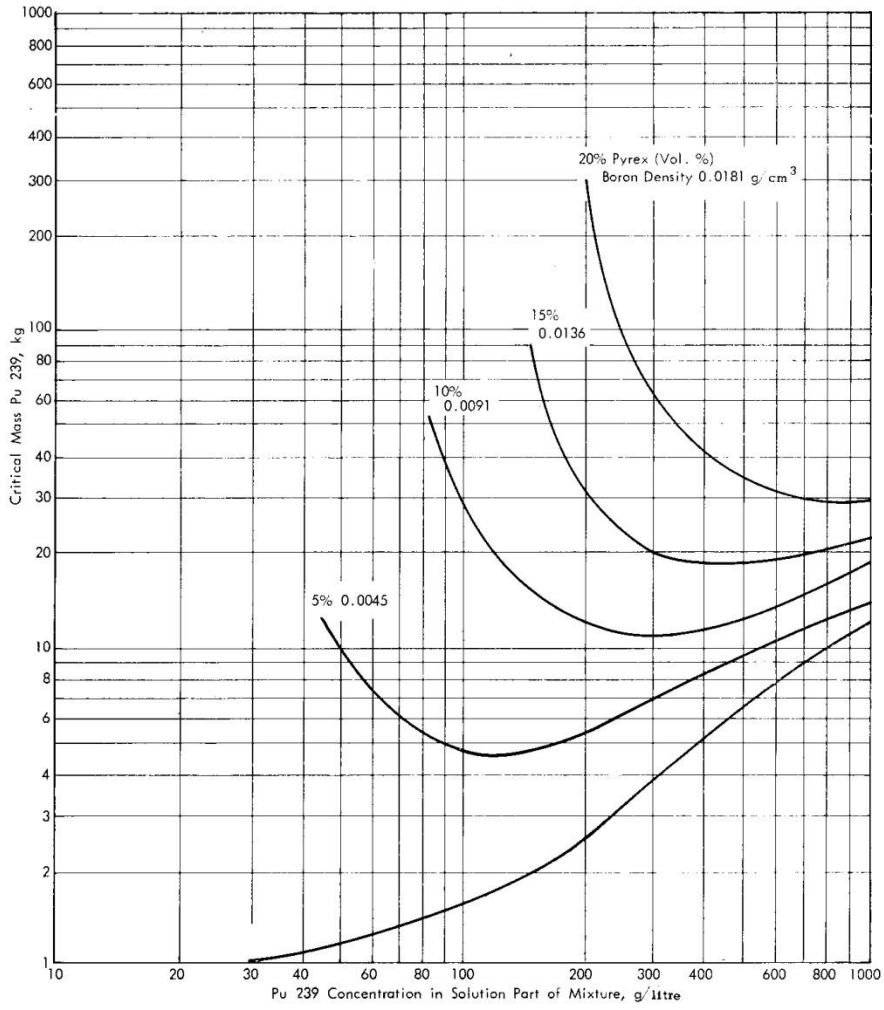

Figure 8. *Spherical critical pyrex poisoned Pu 239 solution, pyrex 12.5 per cent* B_2O_3.

In special cases, sheathing of metals may be effective, however. The most hazardous situation involved in the storage and transport of metallic uranium might not be that of complete water inundation (which would merely isolate units separated by 21 cm) but rather the formation of a thin layer of moderating material between each pair of units, as by ice encrustation. Mihalczo and Lynn [9] have conducted experiments involving plates of uranium weighing approximately 25 kg and measuring 2.54 × 20.3 × 25.4 cm. In Figure 10, the lowest curve is the multiplication of an array of such slabs on 30.48 cm centre-to-centre separations versus the number of fissile units. The individual slabs are not significantly reflected by their supporting members and are separated only by air. When sheets of plastic 2.54 cm in thickness were placed within the array and spaced equidistant from adjacent uranium slabs,

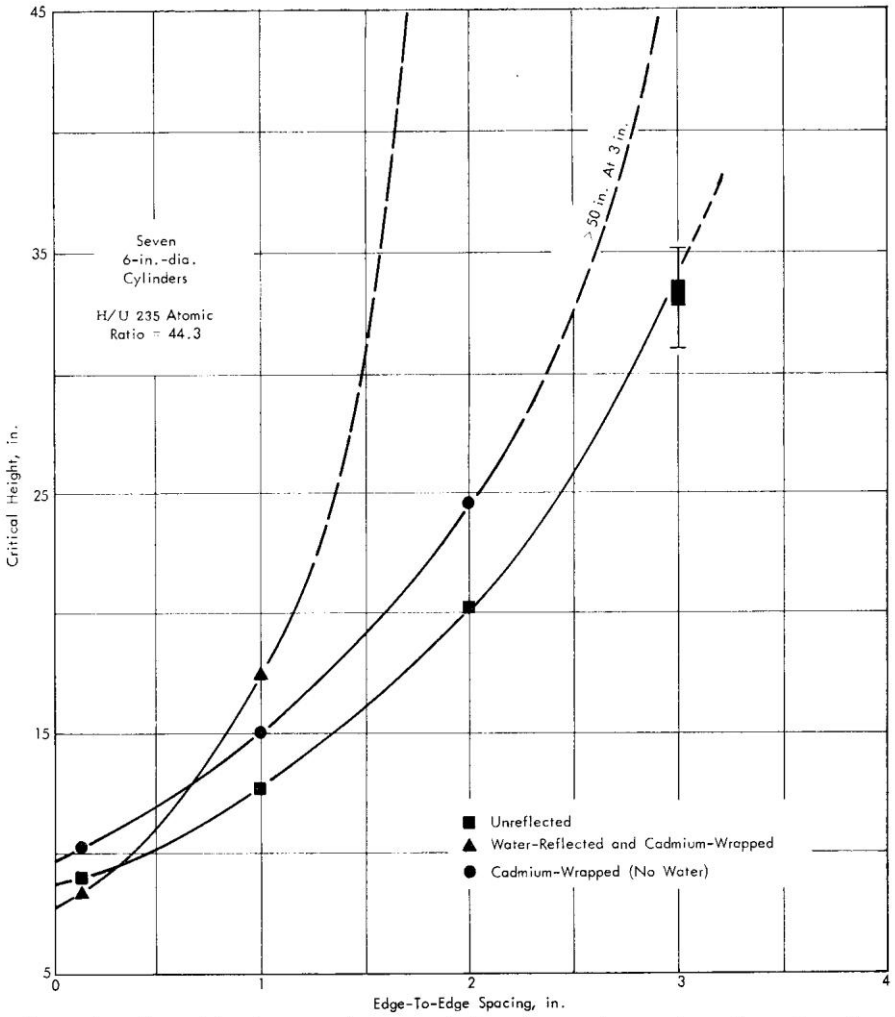

Figure 9. *Critical height as a function of the edge-to-edge spacing of unreflected and water-reflected 6-in. diameter aluminium cylinders containing an enriched U 235 solution: hexagonal arrays, with and without cadmium wrapping.*

the reactivity of the array was greatly increased, as seen by the third curve. The addition of Styrofoam, an expanded plastic, to fill all remaining voids between slabs, resulted in the uppermost curve. When the Styrofoam was replaced by Foamglas, a low density, foamed, borosilicate glass containing approximately 2 wt % elemental boron, the cross-multiplication of the array was reduced almost to that experienced by the slabs alone. The Foamglas thus counteracted the thermalizing effect of the interspersed Plexiglas to a degree greater than its effect in providing reflection to the individual elements of the array. One might infer from the slab geometry of each unit that the change introduced by crushing the foam glass into a higher density glass sheet surrounding each unit would not be large. On the other hand, had the

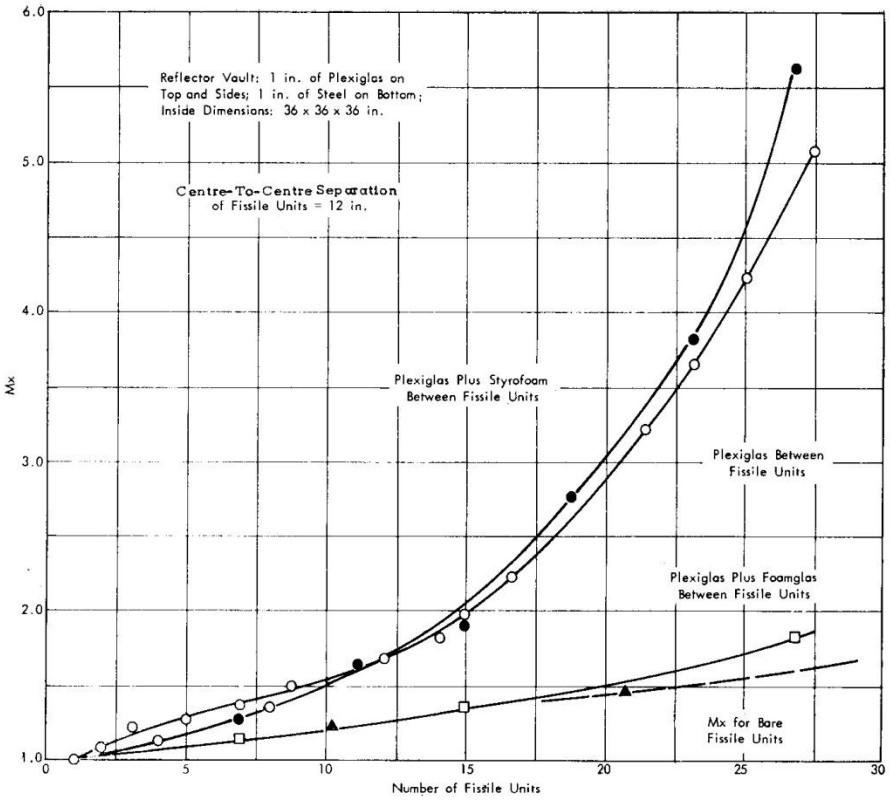

Figure 10. *Cross-multiplication of arrays of fissile units separated by Plexiglas alone, Plexiglas and Styrofoam, or Plexiglas and Foamglas.*

individual units been spherical in shape, the same operation probably would have led to a much higher reflection of the individual unit and a corresponding decrease in the effectiveness of the poison.

Such considerations are indicative of the difficulty of providing adequate poison without recourse to a critical experiment closely duplicating the intended application.

Practical Considerations

The experimental and calculational information given above suggests the wide variety of applications for which nuclear poisons have been considered. But quite apart from the nuclear problem in assessing the value of poisons in a particular application, there are certain practical considerations to be weighed. Clearly, a poisoned vessel must not be employed unless it meets certain requirements with respect to:

a) Prevention of product contamination;
b) Guarantees of poison integrity;
c) Suitability for administrative control;
d) Favourable overall cost picture.

These requirements are determined almost entirely by the nature of the process material and by the situation at an individual plant. The comments thereon given below have been related fairly specifically to the use of Raschig rings. This is not merely to give definiteness to such a discussion, but also because Raschig rings, whether made of glass, boron steel or other poison material, have a very wide application throughout the chemical processing field. In a sense, they combine the flexibility in application of the soluble poisons with the freedom from administrative control of the fixed poisons.

Product Contamination

Clearly the advantages to be derived from the use of poisons are largely erased if it becomes necessary to introduce a purifying process step in order to remove the poisons from the final product. Fixed poisons can usually be encased in a non-reactive metal such as stainless steel to obviate the problem. Where unclad fixed poisons are considered for use, attention must be given to their corrosion characteristics under the conditions of acidity or alkalinity which may exist. Soluble poisons are usually feasible only where the process itself incorporates a purification step. Even in this instance, the nature of the soluble poison may greatly affect costs, either by the necessity of incorporating additional equipment because the purification system is ineffective against the particular poison employed, or by the need for refluxing to bring the product down to necessary specifications.

The use of soluble poisons in a purification plant is not without a worthwhile by-product: the waste products and solvent streams, which are normally of such low uranium concentration as to invite the use of large volumes, become loaded with the poison which has been stripped from the product stream. The result is a large safety factor against the possibility of any malfunction in the purifying system which would divert large amounts of uranium into the salvage and solvent streams.

Borosilicate glass presents a high order of resistance to chemical attack, amounting to less than 0.2 wt % boron/year. It should be noted that such glass has a maximum concentration for chemical attack (corresponding to approximately 0.5 normal for most acids) so that tests conducted using very high concentrations of acid are not necessarily the most meaningful. In addition, the degree of corrosion depends upon the temperature and upon the degree of boron enrichment.

Nature of Product Material

Quite apart from the possibility of contaminating the product material, the loss of poison through corrosion or other processes cannot be tolerated. Clearly, the chemical nature of a soluble poison must match that of the product to avoid the pitfalls of precipitation, phase separation or the like.

The physical form of the fissionable material may also be of importance. If it is gummy or contains tars or is highly viscous in nature, the use of fixed poisons may be complicated by plugging of the process stream or by difficulties in cleaning equipment. Soluble poisons may not be effective in salvage mixtures containing large chunks of foreign matter. The poisoning of slurries and materials below the saturation limit poses difficulties. At the lower enrichments, below 2 %, the optimum moderations for homogeneous systems occur as wet salts and the difficult problems of plugging and clogging appear.

For some dry materials, the introduction of a soluble poison may cause, rather than prevent, a hazard as a result of the moderation introduced in the poison itself.

Nature of Process and Environment

The confident use of poisons requires the assurance that such poisons will be in their proper location and fully effective for a period of time longer than that normally required for review and inspection of the equipment. Although the corrosion resistance of the poison may be sufficiently high that problems of product contamination do not appear, it may well be that the loss of poison is great enough to affect the nuclear safety. High temperatures accelerate the corrosion process and sudden changes in temperatures, as by hot product materials dripping onto the poisons, may introduce thermal stresses tending to fracture the poison or open fissures in its cladding. This is particularly true of glass poisons since the effects of corrosive attack result in a mechanical weakening of the material. Under chemical attack, borosilicate glass suffers both a loss of boron from the surface layer and, in addition, a migration of soda from the glass. The soda diffuses through the glass, resulting in low soda content in patches. This, in turn, sets up stresses which render the glass easily broken. Chemical resistance of the glass can be significantly increased by use of a titanium oxide coating.

Mechanical motion may also affect the integrity of the poison, and fixed poisons built into the movable parts of production equipment must be designed with corresponding care. The use of glass poison in movable equipment, such as drums, is suspect, particularly when the glass is so supported that cavities may occur through breakage without being visually obvious. Mechanical vibration occurs in most plant environments and may be due to rotating equipment located some distance from the poisoned vessels. In practical applications of Raschig rings, it is found that the rings settle to a higher packing density after a few weeks of use. Moreover, if a layer of Raschig rings is placed in a large vessel, vibration may cause piling up of the rings against a side of the vessel. Both effects lead to unpoisoned volumes within the vessel, and it is therefore necessary to inspect and to add more rings after a few weeks of use.

Administrative Controls

Fixed poisons have the enormous advantage that their presence can be guaranteed by visual inspection. As indicated above, such inspection must be carried out at predetermined intervals and the equipment torn down or disassembled as necessary to establish the presence of the poison. Particularly where aqueous solutions are involved, instrumental methods of assuring the effectiveness of the poison by neutron absorption can be set up. Such a case has been reported in connection with a cadmium-poisoned cask for transport of fuel elements. Normally, however, the necessity for visual inspection should be taken into consideration during design of the equipment. Moreover, continuing attention must be given to the equipment to guard against its possible employment in an operation for which the poisons are not adequate. That is, the tendency of persons to consider equipment, once approved, as ' nuclearly safe ' rather than ' nuclearly safe in a particular application ' should be counteracted by firm administrative controls.

The greatest disadvantage of soluble poisons is the extreme difficulty in establishing such administrative controls. The presence of the poison can be determined only by nuclear or chemical analysis. Chemical analysis normally involves an undesirable delay. Only in exceptional circumstances or with special equipment can the presence of the poison by means of its neutron absorption be determined easily. On occasion, the Y-12 Plant has made use of a subcritical reactor to determine the reactivity of liquid samples, but, however effective, this method hardly commends itself for routine production or universal application.

In general, instrumental methods to ascertain the presence of the poisons are both cumbersome and expensive in a production milieu. Instead, the ubiquitous batch card (more nearly the symbol of fissionable material processing than the usual nucleus with orbiting electrons) is used as a means of keeping track of poison additions. Further, multiple additions of poisons may be employed, any one of which is sufficient to poison the solution to the desired degree. Each person making such a poison addition enters upon the batch card the quantity of the poison liquid added and the equivalent amount of elemental poison, and attests to this addition by signature.

The methods by which it is ascertained that poisons are present and that the correct procedures are being carried out will differ from plant to plant. The usual practice seems to support inspections both by members of the production organization directly and also by members of the criticality group who, through the organizational structure, are relatively insulated from the pressures of production commitments, equipment efficiency and the like. To aid such inspections, liberal use is made of limit signs, distinctively coloured stickers for batch cards, and floor markings. The administrative controls over poisoned equipment also have their place in the usual complex of nuclear safety educational measures, criticality approvals and the like, which support a programme of nuclear safety and serve to emphasize its importance.

Costs

The savings derived from the use of poisoned equipment can be very large. Although mention has been made of the larger amounts of poison required when introduced in the form of thick plates or rods, this consideration is actually quite minor on a cost basis. The cost savings do not always result from the direct construction of equipment. Because of criticality, the required size of unpoisoned equipment is frequently not commercially obtainable since it lies intermediately between batch-type laboratory equipment and the much larger, continuous, commercial equipment. By suitable poisoning, however, it may be possible to make use of commercial equipment with relatively minor modifications.

In selecting a poison for specific application, attention must be given to its neutron absorption properties relative to its cost. Table II, derived from a table given earlier, indicates the relative cost for the use of various poisons in various applications. It is seen that, for all applications, boron is more effective on a barn/dollar basis than is cadmium. Some of the rare earths, notably samarium and gadolinium, may be most effective poisons on account of their very large neutron absorption cross section. It appears, however, that their cost would be ten to forty times greater on a dollar/barn basis than is boron.

TABLE II. RELATIVE COSTS OF VARIOUS POISONS

Material	barns/dollar			
	Maxwell-Boltzmann Distribution $\times 10^{24}$	Thermal Reactor $\times 10^{24}$	Intermediate Reactor $\times 10^{24}$	Fast Reactor $\times 10^{24}$
Cadmium	2,917	1,833	790	0
Boron	37,400	21,190	8,440	10
Stainless Steel	6.66	3.33	3.33	0

Table based on following prices:
$Cd(NO_3)_2.4H_2O$ in 300-400 lb lots is \$ 0.67/lb. Cd is \$ 1.83/lb or \$ 0.004/g.
H_3BO_3 in 100 lb lots is \$ 0.081/lb. B is \$ 0.46/lb or \$ 0.001/g.
Stainless steel (304 L) rods, 1.27 cm dia. are \$ 0.016/linear cm. Stainless steel is \$ 1.36/lb or \$ 0.003/g.

The costs of using poisons are not entirely associated with the initial purchase of the equipment. Even though the poisons are of a fixed kind, the user commits himself to a continuing programme of inspections and other administrative controls. Although the poison may not constitute the primary control over criticality, these continuing controls remain necessary. Largely, this is because poison may be lost from a piece of equipment in a relatively subtle, and usually invisible, way; whereas geometrically safe equipment loses its effectiveness by the messy but obvious method of leaking on the floor or on neighbouring equipment.

Nuclear Safety Considerations

Experimental and calculational evidence and practical considerations are but two of the three ingredients which go into the formulation of nuclear safety rules. The third may be termed nuclear safety philosophy. It seeks to define the nature and extent of the risks of a critical incident which plant management is willing to undertake in the fulfilment of its production goals. To a certain extent, but at present to a very limited one, these have been defined by law. To a far greater extent, it is set by the general body of opinion established by criticality experts as good nuclear safety practice. As yet, no legal statement or universal opinion concerning the employment of poisons has appeared.

Consequently, it is incumbent upon individual plant management to establish its own nuclear safety philosophy regarding poisons. This may take the form of acceptance of existing limits without modification but using poisons as an added safeguard. Where the plant philosophy is defined in terms of the allowable degree of risk, as with the double- or triple-contingency criteria, the philosophy regarding poisons does become thoroughly defined. Basically, there appear to be three separate approaches to the use of poisons, depending on the degree of reliance placed upon their presence. Poisons can be employed to provide added control, primary control, or contingent control over criticality. For the purposes of this discussion, the amount of poison contained in a piece of equipment can be considered to be identical in each case. This discussion does not involve the design features of the placement of the poison, but only the reliance placed on its presence.

Poisons as Additional Safeguards

Poisons are used as additional safeguards if the nuclear safety restrictions on the equipment are not relaxed from those normally considered adequate in the absence of the poison. This does not imply that poison is added without specific reason. Rather, the hazard against which the poison is a safeguard is so remote as to fall into the category of an acceptable risk.

Good nuclear safety practice allows solutions whose uranium concentration is 5 g U 235/litre or less to be handled without imposition of criticality limits. Experiments show that they must be evaporated to one-half their volume before any significant nuclear hazard exists, and it is clear that double-batching or multiple mixing produces no hazard. Unfortunately, this does not mean that the contents of two 100-litre vessels, whose batch cards each read 500 g U 235, may be combined into a 200-litre vessel without hazard. Indeed, the degree of hazard in any batching operation will depend on such questions as: " Is this a product solution or a salvage solution? Are the batch card estimates based on analysis of one or two aliquots? How many samples were taken? Have the mass entries been checked for decimal errors? For figure inversion? Could this batch card have been switched with another? How long has this material been stored? Inside or outside? Could precipitation have occurred? Could organics be present? Has phase separation occurred? What is the pH? Does it favour the organic or aqueous phase? Has evaporation occurred?... "

Obviously, no such inquisition will accompany a production batching operation. The chances that the history of the material, its analysis or its accountability measures have led to any hazard whatever are extremely remote. But they exist, and the use of fixed or soluble poisons to remove them might well be considered. If product material is involved, poisons would probably not be employed because it is a uniform material whose characteristics are carefully specified. Salvage solutions, on the other hand, may advantageously be handled in poisoned equipment as an additional safeguard. In specialized situations, soluble poisons can be added in ten times the amount given in Figure 1 (i.e. 1 g Cd/g U 235) to safeguard against the tenfold mass batch estimates which result from decimal and figure inversion errors.

As a second example, consider a battery of geometrically safe solution evaporators having common steam and condensate manifolds. Condensate from the steam jackets of evaporators should contain no uranium and can usually be sewered directly. The steam jackets may leak to the interior of the evaporator, but such a leak would result in the high pressure steam going into the evaporator rather than the solution going into the condensate. Only during infrequent shut-downs is the situation reversed so that concentrated solution finds its way into the steam jacket. Shut-down and start-up procedures are therefore specified which should detect such a leak and prevent solution from getting into the unsafe hold-up tank where the condensate is stored awaiting analysis prior to discard. One cannot really expect that more solution than the contents of one evaporator could ever find its way into the hold-up tank without being highly diluted — except, of course, for the further unlikely event that the condensate tank has just been dumped. All in all, the possibility of nuclear hazard depends upon an exceedingly long and improbable chain of events and lies well within the range of acceptable risks. Fixed poisons in the hold-up tank, however, provide a firm, inexpensive and desirable safeguard.

The use of poisoning materials in lieu of conventional materials to reinforce the existing control measures is another method of providing added safeguards. Such a case might be an oil bath of considerable size in which uranium plate is heated and cleaned. Oxide cleaned from the surface sifts down to the bottom of the tank. The active criticality control, in this case, is the mass of uranium oxide accumulation in the tank estimated from experimental knowledge of the weight of oxide stripped off the uranium per square foot per minute of submersion in the tank. Baffles along the floor of the tank inhibit stirring up of the oxide. By making these baffles of stainless clad boral, the tank is rendered safe against errors in the estimate of contained uranium; for, if the uranium is concentrated at the base of the tank, it is rendered ineffective by the boral and if it is distributed throughout the tank, it is safe by dilution.

Poisons as Primary Controls

Poisons are used as primary controls if loss of the poison could result in criticality under normal operating conditions. The nuclear safety limit of the equipment is relaxed to the point where the vessel would be comfortably subcritical in the presence of the poison; i.e. a sufficient margin of safety is allowed so that any reasonable loss of poison that might occur between inspection periods would not be sufficient to allow criticality. Table III gives examples of practical limits in use at the Rocky Flats Plant. Restrictions may also be imposed on the closeness to critical to be allowed in the poi-

TABLE III. CRITICALITY LIMITS FOR USE OF RASCHIG RINGS

Volume of Vessel (*litre*)	Simultaneous Requirements	
	Mass Limit (*kg U 235*)	Concentration Limit (*g U 235/litre*)
< 200	10	50
> 200	20	25

Source: C. L. Schuske, private communication, January 23, 1961 [2].

soned state: in a specialized case, an effective multiplication of 0.90, and a mass limit of 75 % of the critical mass, have been suggested as such a restriction [10]. Equipment which is poisoned in this fashion is assumed to be safe within the meaning of the word ' safe ' as applied, say, to a 12-in. (30.48 cm) diameter pipe used for 3.5 % U 235 enrichment. Controls which must be established to guarantee the presence of the poison have their counterparts in those necessary to prevent cross-over of materials of different enrichments. Significantly, the safety of the 30.48 cm pipe is considered absolute only in plants where no higher enrichment is handled, and the use of poison-safe equipment has found greatest application in the shipment of fuel elements whose unique geometries safeguard against cross-over.

The necessity for controls to guarantee the presence of the poison cannot be overemphasized, but the basic requirement of poison-safe equipment is that no reasonable accident can cause loss of the poison. Placement of the poison into a process stream of high uranium concentration opens questions of corrosive ability, but wrapping a sheet of poison around a vessel and suitably protecting it from environmental effects is obviously quite another matter. The situation is also changed if the poison, even though placed in a process stream, is subject to continuous monitoring. Such monitoring is possible through neutron absorption measurements, or by direct multiplication measurements; but such methods can only be applied where personnel shielding is provided, as in irradiated fissile material processing. Whether monitored or not, shielding provides the special circumstances required for the use of poisons as primary controls.

The use of poisons as a primary control is clearly indicated only where fixed poisons are employed, where the poisons are so placed as never to come into contact with corrosive chemicals, and where the possibilities of mechanical dislodgment are negligible. Primary dependence of poisons must be viewed as an individual problem requiring very special circumstances, and the equipment it poisons must be viewed as a power reactor with control rods in place. It is essential that adequate experimental data confirm the mechanical and/or chemical stability of the poisons and that the nuclear safety of equipment incorporating them shall have been experimentally established by a directly applicable criticality test.

Poisons as Contingent Controls

We have now discussed the two extremes in the use of poisons where either no reliance or full reliance is placed upon their presence. In industrial criticality control, the most effective use of poisons may occur in the intermediate range between the two extremes.

For any application in which the use of poisons is contemplated, there currently exists an alternate method not employing the use of poisons. Depending entirely upon the nature of the material and the process, this alternate method may involve safe geometries and/or safe batching. In every case it must be firmly established that the poisoning will result in a system that is at least as safe as the alternate approach. The use of poisoning as a primary control, therefore, requires a prejudgment as to its permanency. Where less reliance need be placed on the poisons and where the plant nuclear safety philosophy is defined in terms of the extent of risk to be undertaken, the judgment as to the relative safety of the poisoned and unpoisoned alternatives can be determined. Such a situation exists in the so-called double-and triple-contingency statements of nuclear safety philosophy. The discussion given below is framed in terms of the double-contingency criteria; its extension to other criteria is obvious.

Poisons are used as contingent controls if, in the absence of the poison, the equipment would not be critical under normal operating conditions but would be critical if one of several other possible but unlikely accidents were to occur. The accidents referred to here are essentially defined by the equipment itself and the nature of the process operation and include such occurrences as double batching, water flooding, change in fissionable material concentration, failure of overflows, water inleakage, analysis errors and the like. It is further assumed that administrative controls are set up to render unlikely

any of the above accidents and that similar controls guarantee the presence of the poisons, as by requiring that soluble poisons be considered as a contingent control only if they are added on two separate and independent occasions.

The use of soluble poisons in this fashion might be illustrated by considering a fuel element dissolving operation conducted on a batch basis in an unsafe vessel. Standard practice allows a 350-g U 235 batch limit in this dissolver. Double batching is a distinct possibility in this situation and batch card errors may also occur although they are much less likely. Consequently, poison might be added in gross amounts, say 1g cadmium to 1g U 235, to safeguard against these events. Because it is an added safeguard, the batch limit for the process remains at 350 g. Suppose, however, that poison is pre-mixed with the dissolving acid in a separate and independent step. The requisite safeguards for the consideration of poisons as a contingent control have now been established. Consequently, the batch limit may now be increased to 700 g since even if the poison, through human error, were omitted in both instances, the batch limit for 700 g would still be safe. The equipment would only be critical if the second mistake of double batching had also been made. The use of the poison therefore guarantees safety in the event that double batching at the new and higher batch limit were to occur. This example is given only because it illustrates the contingent use of poisons in terms of concepts and limits that are generally understood. In point of fact, the use of soluble poisons as contingent controls should only be approached with great caution. The most important usage of poisons should be the replacement of such geometrically unsafe equipment as the dissolver cited above, by equipment rendered safe by fixed poisons.

A similar example involving the use of fixed poisons might occur at the feed point of a purification system. Normally the feed is at low concentrations, so that the extraction columns are set to be most efficient in this range. However, for purposes of pH adjustment, feed solutions may be blended with highly concentrated, but impure, solutions. The possibility therefore exists that, through human error, the dilution and mixing does not occur and solutions of optimum concentration enter the purification system. Safeguards in the form of administrative controls or instrumentation would be set up to render this possibility highly unlikely. But for the possibility of such valving errors, a 30-gallon vessel would be quite safe for the concentration range normally encountered, even considering equipment malfunctioning or major variations in the process operating variables. The addition of fixed poisons to this vessel, in the proper amount to render the vessel safe at optimum concentration, results in a system considered safe by this criteria.

In each of the above cases, the omission of the poison has been considered as a possible but unlikely event which, if no other malfunction occurs, would still not result in criticality. There is, however, a foreseeable event not outside the bounds of possibility which would render the equipment critical were it not for the poison, namely a net relaxation of the nuclear safety requirements, either through an increased mass batch limit or an increased dimension of the equipment. Except where local conditions alter the requirements, this usage of poisons essentially allows a just-subcritical equipment dimension in the absence of poison, with the poison providing the safety factor against any and all conditions not usually encountered. It might be noted that this application of poisons need not assume, as above, that all the poison is omitted. The assumption of a specific degree of partial loss of the poison is, however,

one to be approached with great caution. In most cases of fixed poisons, a mechanism seems to exist by which poison can be lost at a precipitous rate following a long period of low, or no, loss.

Concluding Remarks

Poisons and methods of poisoning have certain nuclear characteristics which must be taken into account in their application. It is clear that much experimental and calculational effort has been directed toward the problem of firmly establishing the effectiveness of poisons under a great variety of circumstances encountered in chemical and metallurgical plants. Practical difficulties may exist, depending upon the application, but none of these appear to be insurmountable and most have more to do with the selection of the poison and poisoning method than the efficacy of poisoning *per se*. Generally speaking, it appears that poisons must be applied on a purely individual basis. A criticality group must consider each possible use of poisons, bearing in mind the problems involved in the administrative supervision of such equipment, as well as the more obvious technical problems concerned with both the poison and the equipment.

At the present time, it appears that Raschig rings are the most reliable and flexible method of providing a poison medium. The data so far obtained have testified to its general usefulness, and further experimentation will doubtless provide a basis for a greatly expanded use of these rings.

The use of soluble poisons in any application other than that of an added safeguard poses difficult problems of administrative control. It seems essential never to rely upon a single addition of poison, but rather to insist on separate and independent additions, each sufficient to do the job. It further seems that an essential factor in weighing the possible use of soluble poisons is the determination of its chemical stability in the intended application. Basically, it must be concluded that the use of liquid nuclear poisons in a process is an administrative form of nuclear safety control and should not be undertaken until the reliability of this control has been thoroughly established for the intended use.

The use of fixed poisons as primary controls depends upon the individual application. It can only be concluded that, where adequate experimental data confirm the mechanical and/or chemical stability of fixed neutron absorbers and the nuclear safety of process equipment incorporating them has been experimentally established, such equipment should be regarded as being as safe as its geometrically restricted counterpart. Firm administrative controls to ensure the presence of the absorbers is essential.

The best nuclear safety practice insists that the physical design and construction of all process equipment incorporate its guarantees of nuclear safety. Normally this is done by geometric restriction. The use of fixed nuclear poisons appears to provide a means by which such practices can be extended into areas in which geometric safeguards either have not been feasible, have hampered production, or have been economically unjustifiable. The use of fixed poisons does not eliminate the need for administrative control any more than does the use of geometrically restricted equipment. Poisons can, however, greatly reduce such administrative controls and, in those applications where they provide contingent controls, their use makes it highly improbable that an outright unsafe condition would result from a temporary loss of administrative controls.

REFERENCES

1. N. KETZLACH, " Soluble Poisons for Slightly Enriched Uranium Systems ", *HW-50155* (1957).
2. C.L. SCHUSKE, Private communication, January 23, 1961. A report of these calculations is at present in preparation by A. Goodwin Jr. and G.H. Bidinger, Rocky Flats Plant, Dow Chemical Company.
3. C.L. SCHUSKE, M.G. ARTHUR and D.F. SMITH, " Neutron Multiplication Measurements on Oralloy Slabs Immersed in Solutions ", *RFP-66* (1956).
4. J.D. MCLENDON and J.W. MORFITT, " Critical Mass Tests on U-235 Machine Turnings ", Y-12 Plant, *Y-A2-71* (1952).
5. L.W. GILLEY, D.F. CRONIN and V.G. HARNESS, " Boron Poisoning in Critical Slabs ", Physics Division Semi-Annual Progress Report for Period Ending March 10, 1954, *ORNL-1715*, p. 12 (1954).
6. J.K. FOX and L.W. GILLEY, " Critical Parameters for 20-inch Diameter Stainless Steel Cylinders Containing Aqueous Solutions of U 235 Poisoned with Pyrex Glass " Neutron Physics Division Annual Report for Period Ending September 1, 1959, *ORNL-2842*, p. 78 (1959).
7. G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Nuclear Safety Experiments on Plutonium and Enriched Uranium Hydrogen Moderated Assemblies Containing Boron ", *RFP-201* (1960).
8. J.K. FOX, L.W. GILLEY and D. CALLIHAN, " Critical Mass Studies, Part IX, Aqueous U 235 Solutions ", *ORNL-2367* (1958).
9. J.T. MIHALCZO and J.J. LYNN, " Multiplication Measurements with Highly Enriched Uranium Metal Slabs ", *CF-59-7-87* (1959).
10. D. CALLIHAN, W.J. OZEROFF, H.C. PAXTON and C.L. SCHUSKE, " Nuclear Safety Guide ", *TID-7016 (Rev. 1)* (1961).

ÉTUDE D'UNE MÉTHODE D'EMPOISONNEMENT HÉTÉROGÈNE ET DE SON APPLICATION PRATIQUE

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INTRODUCTION

La sécurité des usines traitant les solutions de matière fissile pose de gros problèmes. De nombreuses publications américaines et anglaises permettent d'établir, dans un certain nombre de cas simples, des règles de sécurité.

Les normes de sécurité absolue s'avèrent souvent trop rigoureuses, sinon incompatibles avec les nécessités de production des éléments fissiles. Il faut donc adapter les règles de sécurité à chaque type de récipient. Nous nous placerons ici dans le cas des récipients cylindriques contenant des solutions aqueuses de matière fissile (Pu ou U 235), entourés d'un réflecteur latéral infini d'eau.

COEFFICIENT DE SÉCURITÉ

Pour chaque récipient contenant une solution de matière fissile, on peut définir un coefficient de sécurité. Nous distinguerons deux cas :

a) *La solution reste toujours homogène*

Le coefficient de sécurité peut être défini comme le rapport de la masse critique minimum que l'on peut atteindre dans le récipient entouré d'un réflecteur d'eau d'épaisseur infinie, à la masse maximum de matière fissile contenue dans le récipient.

Cette définition du coefficient de sécurité est sans doute la meilleure lorsque la concentration critique en Pu ou U 235 correspondant à la masse critique minimum est mal connue, ce qui arrive souvent dans la pratique.

b) *La solution est amenée à précipiter*

On peut toujours envisager le rassemblement de la matière fissile sous une forme favorable à la divergence, tout au moins lorsque la forme et les dimensions du récipient le permettent.

Nous distinguerons plusieurs cas, suivant que le rayon R du récipient est plus grand ou plus faible que celui de la sphère de même solution réfléchie par un réflecteur infini d'eau, et correspondant à la masse critique minimum.

Nous savons en effet que la courbe de la masse critique, en fonction du rayon, passe par un minimum pour R = 16 cm. Elle atteint une valeur de 5,7 kg pour un rayon de 11 cm. Pour un rayon inférieur, il faut des $\frac{H}{U\,235}$ ou des $\frac{H}{Pu}$ tels que la divergence est impossible à partir d'une solution homogène.

$$R < 11{,}5 \text{ cm}$$

La sphère ayant un rayon inférieur à 10 cm ne peut diverger. Si 10 cm $< R <$ 11,5 cm, la masse critique de la sphère de solution est supérieure à la masse critique minimum du cylindre. On prendra donc les règles de sécurité relatives aux cylindres de solution homogène.

$$11{,}5 \text{ cm} < R < 15 \text{ cm}$$

Nous prendrons comme coefficient de sécurité pour chaque rayon, le rapport de la masse critique de la sphère inscrite dans ce cylindre à la masse maximum de matière fissile contenue dans le récipient.

$$R > 15 \text{ cm}$$

Nous sommes amenés à prendre pour coefficient de sécurité le rapport de la masse critique minimum en géométrie sphérique à la masse maximum de matière fissile présente dans le récipient.

Remarque. Le rassemblement de matière fissile en une géométrie favorable à la divergence, durant la précipitation, est évidemment très improbable. Mais il est difficile de chiffrer la probabilité de réalisation de ce phénomène.

Accroissement du coefficient de sécurité par un apport d'absorbeur de neutrons

C'est un problème complexe, puisque chaque cas est un cas particulier. On peut augmenter le coefficient de sécurité en mettant dans la solution de matière fissile un absorbeur de neutrons, tel que B 10 ou Cd.

On peut envisager deux types d'empoisonnement : l'empoisonnement homogène et l'empoisonnement hétérogène.

a) *L'empoisonnement homogène*

Il est très efficace. Cependant, l'absorbeur de neutrons peut être difficile à éliminer. Il n'est pas évident, d'autre part, que l'absorbeur suive la matière fissile dans toutes ses transformations.

b) *L'empoisonnement hétérogène*

Il est beaucoup moins efficace, puisque l'absorption est plus localisée. Le coefficient de sécurité qu'il apporte sera donc plus faible, mais il a l'avantage d'être un apport d'antiréactivité sûr, si la matière absorbant les neutrons (Cd ou B_4C) est suffisamment bien protégée de la corrosion ou de l'entraînement dans la solution.

L'étude que nous ferons dans ce rapport est celle d'un cas particulier d'empoisonnement hétérogène. Elle a une application pratique : les réci-

pients cylindriques dans lesquels on précipite une solution de matière fissile (précipitation du Pu sous forme d'oxalate, au cours de la purification de ce métal).

Etude de la criticalité d'un cylindre contenant un croisillon fait d'un absorbeur de neutrons thermiques

Les pots cylindriques où la solution de matière fissile (Pu 239) est amenée à précipiter (précipitation à l'état d'oxalate de Pu, lors de la purification de ce métal) contiennent des agitateurs pouvant porter des palettes radiales. Ces palettes ont un rayon inférieur à celui du cylindre. Pour améliorer l'agitation, on fixe sur les parois intérieures du cylindre des plaques radiales (figure 1). Ces différentes plaques contiennent un absorbeur de neutrons (B 10 ou Cd).

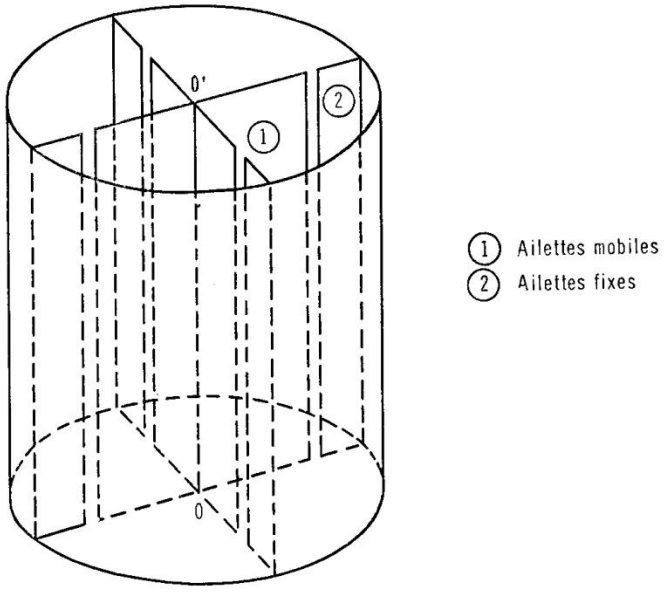


Figure 1. *Cylindre avec agitateur à palettes.*

On schématisera l'agitateur par un croisillon ayant un nombre de branches égal à celui de l'agitateur.

Les plaques fixes forment un croisillon dont les branches ne se rejoignent pas.

Nous nous proposons d'étudier l'influence de ces absorbeurs hétérogènes sur la masse critique dans le cylindre.

Nous nous bornerons, pour simplifier, au croisillon à quatre branches. Nous prendrons les géométries suivantes :

Cylindre avec croisillon complet, absorbant les neutrons thermiques

La longueur de chaque branche est égale au rayon du cylindre.

La hauteur de chaque branche est égale à la hauteur de la solution.
Le cylindre est réfléchi par un réflecteur latéral infini d'eau.

Cylindre avec demi-croisillon, absorbant les neutrons thermiques

La longueur de chaque branche est égale à la moitié du rayon du cylindre.
La hauteur de chaque branche est égale à la hauteur de la solution.
Le cylindre est réfléchi par un réflecteur latéral infini d'eau.

Ces deux problèmes ne sont pas susceptibles d'une solution analytique rigoureuse. Le premier, cependant, peut être résolu de manière approchée.

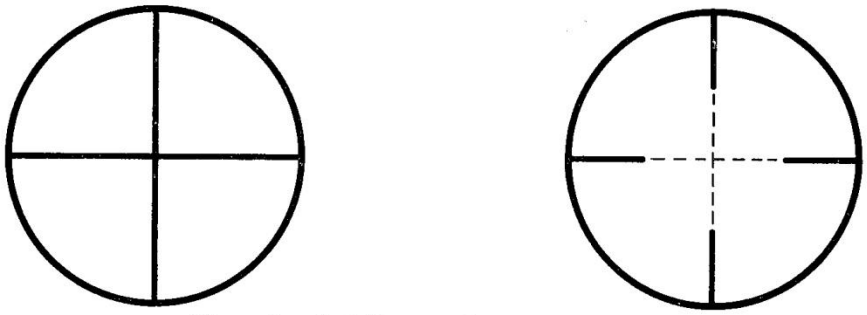


Figure 2. *Croisillon complet et demi-croisillon.*

Cylindre avec croisillon complet, réfléchi par un réflecteur latéral infini d'eau

a) *Théorie utilisée*

Nous avons utilisé, pour la résolution de ce problème, la théorie à deux groupes d'énergie : un groupe de neutrons rapides et un groupe de neutrons thermiques.

Le seul problème que l'on peut résoudre par l'analyse est le suivant : calcul de la masse critique ou de la concentration critique d'un secteur cylindrique nu, de rayon R et de hauteur H déterminés.

On sait que le laplacien matière B^2 est solution de l'équation :

$$(1 + L^2B^2)(1 + \tau B^2) - k = 0$$

avec :

L^2 = longueur de diffusion des neutrons thermiques dans la solution
τ = âge des neutrons thermiques dans la solution
k = coefficient de multiplication en milieu infini.

Le laplacien géométrique est donné par :

$$B^2 = \left(\frac{\alpha_{\frac{n}{2}}}{R + \delta_1}\right)^2 + \left(\frac{\pi}{H + 2\delta}\right)^2$$

avec :

$\alpha_{\frac{n}{2}}$ = première racine de la fonction de Bessel d'ordre $\frac{n}{2}$: $J_{\frac{n}{2}}(x)$

n = nombre de branches du croisillon

$\delta_1 = \delta + \dfrac{\delta}{\sin\dfrac{\theta}{2}}$ (la ligne en pointillé de la figure 3 montre les points où les flux s'annulent)

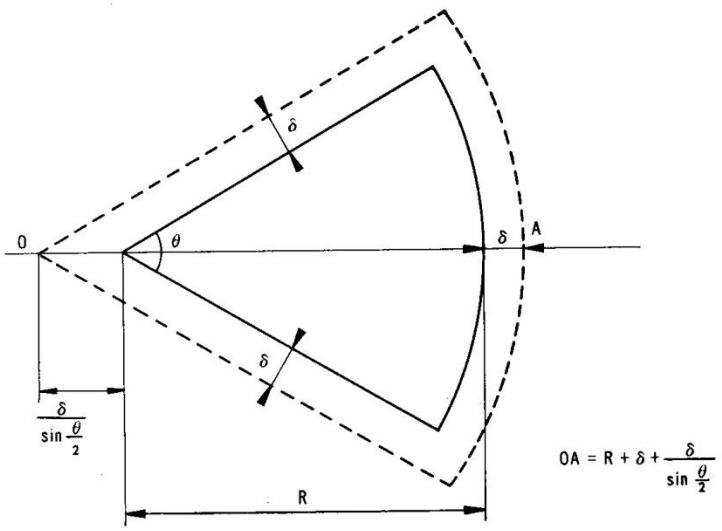


Figure 3. *Secteur cylindrique nu.*

b) *Choix des constantes neutroniques*

i) *Constantes du modérateur*

Nous avons pris les valeurs données dans le document ORNL-2081.

L_0^2 = longueur de diffusion des neutrons thermiques dans l'eau : 8,123 cm^2

τ = âge des neutrons dans l'eau : 27 cm^2

D_1 = coefficient de diffusion des neutrons du groupe rapide : 1,135 cm :

D_2 = coefficient de diffusion des neutrons du groupe thermique 0,160 cm.

La solution considérée est une solution aqueuse de Pu 239.

ii) *Sections efficaces utilisées*

H_2O = section efficace macroscopique d'absorption : 0,0197 cm^{-1}

Pu { section efficace microscopique d'absorption : $\sigma_a = 982$ barns
Pu { section efficace microscopique de fission : $\sigma_f = 682{,}5$ barns
Pu { nombre moyen de neutrons émis par fission : $\nu = 2{,}91$.

Les constantes utilisées sont évidemment sujettes à discussion, mais il s'agit ici d'un problème où l'on regarde surtout la variation de masse critique apportée par la présence de l'absorbeur.

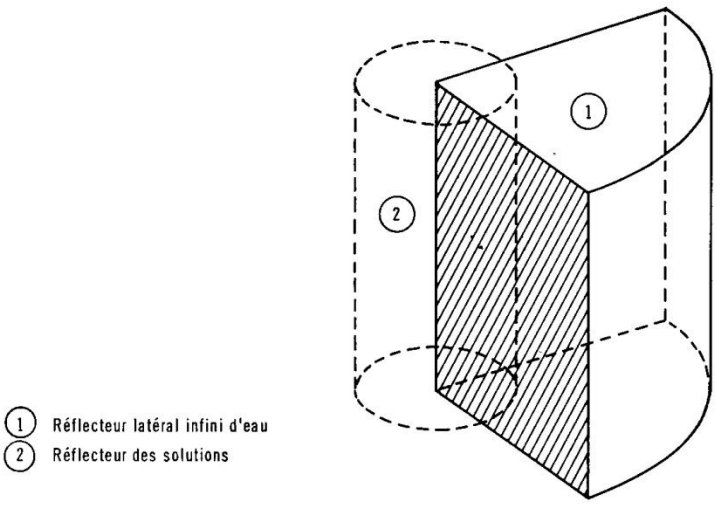


Figure 4. *Secteur cylindrique réfléchi.*

c) *Gains de rayon dus aux réflecteurs*

La présence d'un réflecteur peut s'interpréter, dans le cas d'une sphère ou d'un cylindre réfléchis par un réflecteur d'eau ayant les mêmes éléments de symétrie, comme un gain de rayon ou de hauteur. Nous admettrons que ceci est encore vrai dans le cas du secteur cylindrique réfléchi sur ses trois faces latérales.

Nous considérerons deux gains dus au réflecteur : gain sur la face cylindrique réfléchie par l'eau et gain sur les faces planes recouvertes de cadmium et réfléchies par un réflecteur de solution de même concentration que dans le secteur et d'épaisseur variable.

i) *Gain de rayon dû au réflecteur d'eau*

Ce gain de rayon calculé dans le cas d'un cylindre réfléchi par un réflecteur latéral infini d'eau, varie peu avec le rayon de celui-ci. Lorsque le rayon varie de 10 à 20 cm, il oscille entre 5,55 et 5,85 cm. On le désigne par $R_0 - R$.

ii) *Gain de rayon dû aux parois latérales*

Le calcul du gain du réflecteur est plus compliqué. Nous l'avons réalisé dans les hypothèses suivantes :

On prend un cylindre ayant une surface de section droite égale à celle du secteur cylindrique et même hauteur, entouré latéralement d'une solution identique à celle du cylindre, et d'épaisseur égale à l'épaisseur moyenne du réflecteur entourant le secteur cylindrique (figure 5).

Le cylindre est entouré de cadmium sur sa surface latérale.

L'épaisseur du réflecteur latéral est ramenée à une valeur constante égale à $\frac{\pi}{4} R$.

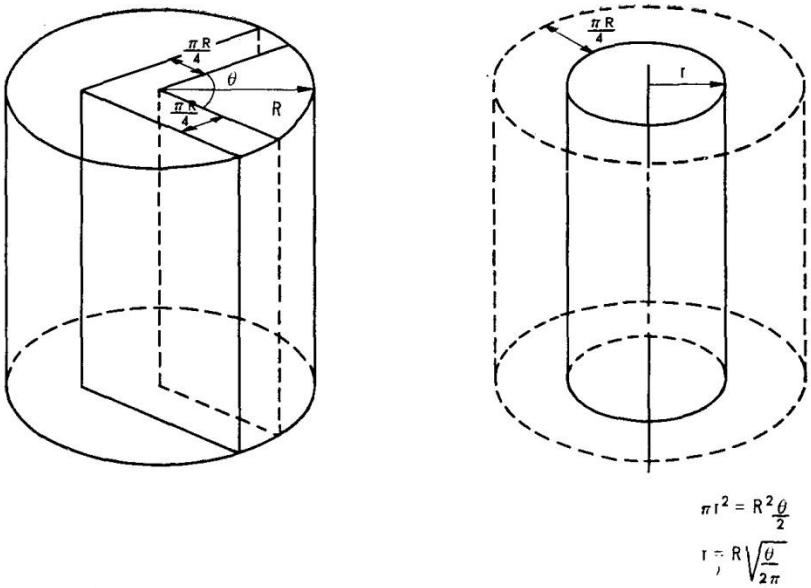


Figure 5. *Équivalence secteur cylindrique-secteur réfléchi par un réflecteur latéral de solution de matière fissile.*

d) *Validité des hypothèses*

Nous voulons obtenir une valeur minimum du coefficient de sécurité apportée par la présence des plaques absorbantes.

Le remplacement du secteur cylindrique de rayon R et de hauteur H par un cylindre de rayon r et de hauteur H, tel que :

$$\pi r^2 = \frac{R^2 \times \pi}{4} \quad r = \frac{R}{2}$$

diminue le rapport $\frac{\text{surface de fuite}}{\text{volume}}$, donc la masse critique.

Le fait de supposer le cylindre entouré d'un réflecteur latéral homogène d'épaisseur donnée, donne une valeur maximum du gain de réflecteur pour cette épaisseur.

L'hypothèse consistant à remplacer le réflecteur d'épaisseur maximum R par un réflecteur homogène d'épaisseur $\frac{\pi}{4}$ R n'est pas justifiable théoriquement. Mais l'influence de cette hypothèse sur la masse critique est certainement plus faible que les deux premières.

e) *Résultats obtenus*

Le calcul du gain de rayon dû au réflecteur infini d'eau nous donne, pour des rayons de cylindre variant entre 10 et 20 cm, une valeur moyenne de 5,7 cm.

Le gain de rayon dû au réflecteur solution varie avec le rayon r et la hauteur H du cylindre équivalent.

Les courbes donnant la variation du gain de rayon en fonction de r et de H sont données dans l'annexe à ce rapport. On appelera ce gain de rayon $R_1 - r$.

f) *Assimilation du cylindre avec croisillon réfléchi par un réflecteur latéral d'eau d'épaisseur infinie à un secteur cylindrique nu*

La figure 6 montre que le secteur cylindrique nu équivalent peut être pris comme le secteur de rayon égal à :

$$R_2 = \frac{R_1 - r}{\sin \frac{\theta}{2}} + R + R_0 - R$$

avec :

θ = angle dièdre du secteur $\left(\frac{\pi}{2}\right.$ dans le cas d'un croisillon à quatre branches).

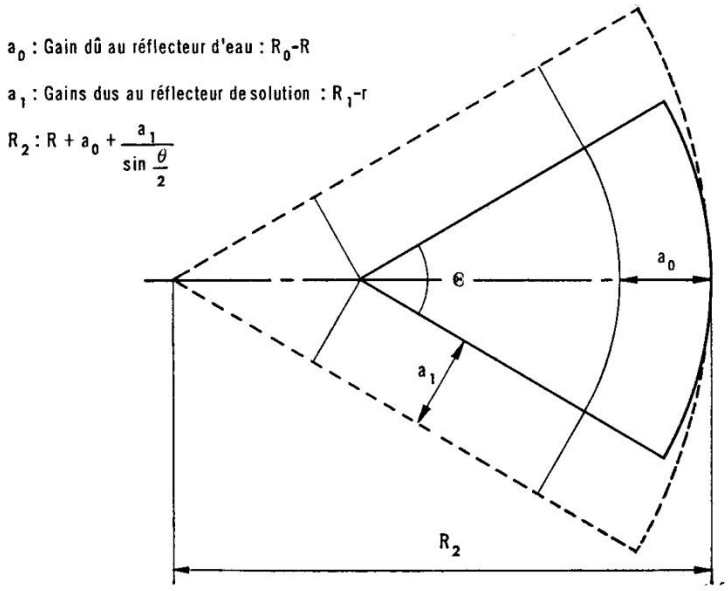


Figure 6. *Secteur cylindrique nu équivalent.*

Le rayon ainsi défini est le rayon extrapolé. Le flux s'annulera aux limites de ce nouveau secteur cylindrique.

L'équation critique, dans le cas d'un croisillon à quatre branches, s'écrit :

$$\left(\frac{\alpha_2}{R_2}\right)^2 + \left(\frac{\pi}{H + 2\delta}\right)^2 = B^2$$

avec :

$\alpha_2 = 5{,}13562$;

B^2 = racine positive de l'équation bicarrée

$$(1 + L^2B^2)(1 + \tau B^2) - k = 0.$$

Résultats obtenus. Ils sont condensés dans les courbes de la figure 10. On voit que la masse critique minimum, dans le cas d'un croisillon à quatre branches, est égale à :

$$M_c = 1.400 \text{ g}$$

Il apporte donc un coefficient de sécurité égal à 1,8.

Cylindre avec croisillon incomplet

La mise en place d'un agitateur à palettes au centre d'un cylindre où précipite la solution de Pu, pose souvent des problèmes d'ordre pratique.

Nous avons étudié l'influence d'un croisillon incomplet, formé de quatre branches radiales, fixées à la périphérie du cylindre (figure 7).

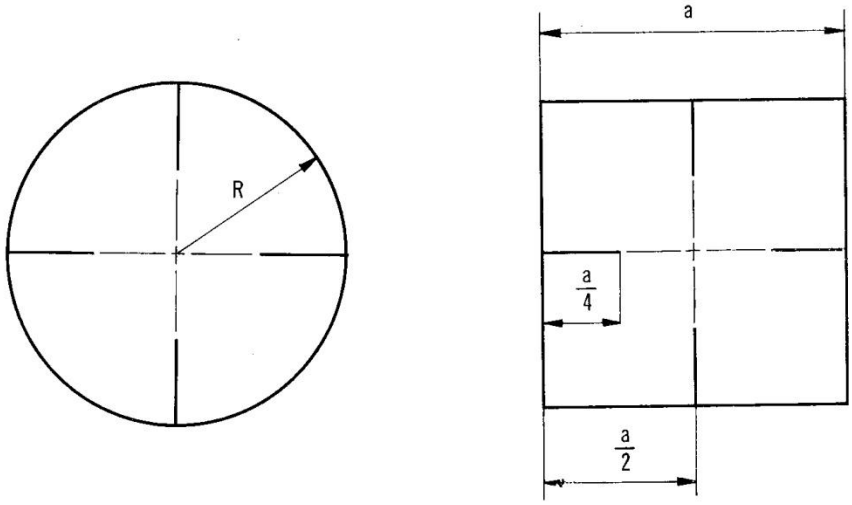


Figure 7. *Cylindre avec croisillon incomplet.*

Le calcul n'est pas susceptible de solution analytique. Nous avons donc modifié la géométrie de l'appareil étudié, de façon à l'adapter à un programme de calcul existant.

Le programme utilisé est le programme PDQ sur IBM-704 et IBM-7090.

Le cylindre étudié est assimilé au parallélépipède de même section droite et de même hauteur. Nous étudions les cas suivants :

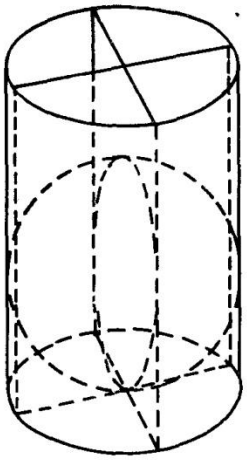
— Parallélépipède nu avec croisillon complet ;
— Parallélépipède nu avec croisillon incomplet ;
— Parallélépipède réfléchi par un réflecteur infini d'eau avec croisillon complet ;
— Parallélépipède réfléchi par un réflecteur infini d'eau avec croisillon incomplet.

Les calculs sont faits pour un ensemble de longueurs d'arête et de hauteurs.

Les résultats obtenus sont publiés dans une annexe à ce rapport.

En prenant pour masse critique minimum du cylindre réfléchi par un réflecteur latéral infini d'eau (calculée dans les mêmes conditions), $M_c = 780$ g, les coefficients de sécurité sont :

— croisillon 2
— demi-croisillon..... (entre 1,2 et 1,4)



$10 < R < 15$ cm

Figure 8.

a) *Étude et discussion des résultats obtenus*

Nous avons fait ici l'étude de l'influence d'un croisillon à quatre branches. Il est certain qu'un croisillon ayant un nombre de branches plus élevé aurait une influence plus grande.

Dans le début de ce rapport, nous avons distingué plusieurs catégories de cylindres, suivant leur rayon. L'introduction d'un croisillon va nous permettre d'établir de nouveaux coefficients de sécurité.

Cylindres de rayon $R < 10$ cm

La présence d'un croisillon à quatre branches augmente la masse critique dans un rapport supérieur à 2.

Cylindres de rayons tels que $10 \text{ cm} < R < 15$ cm

Tout se passe comme si la sphère qui peut se former par précipitation à l'intérieur du cylindre était partagée en quatre branches par le croisillon. Celui-ci apporte donc un coefficient de sécurité voisin de 2.

Cylindres de rayon $R > 15$ cm

Nous devons distinguer plusieurs cas :

$15 \text{ cm} < R < 30$ cm

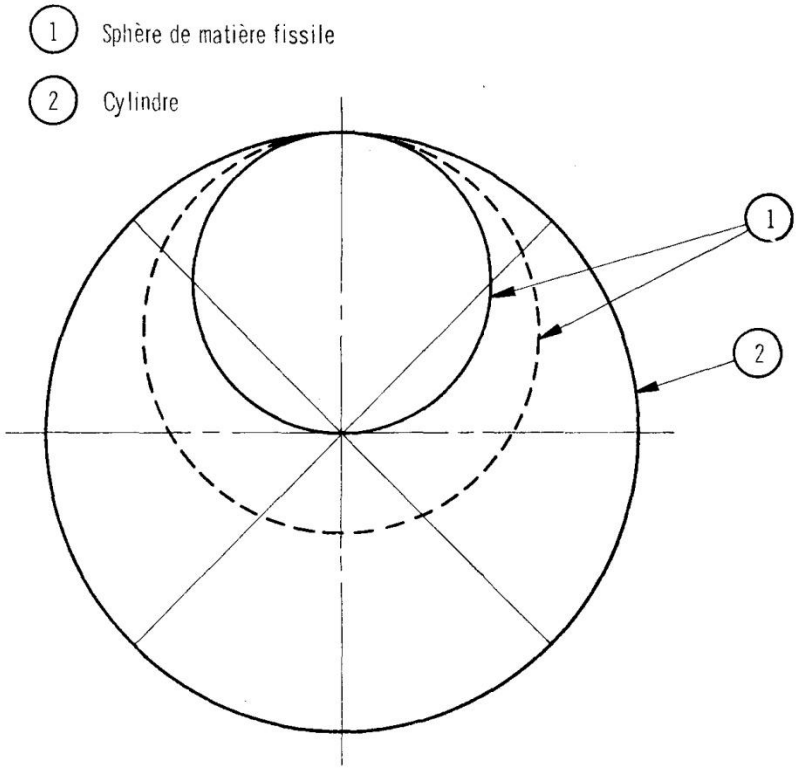


Figure 9.

La sécurité ne pourra être assurée que grâce à un croisillon à huit branches. Le coefficient de sécurité est alors au moins égal à 1,5.

$$R > 30 \text{ cm}$$

L'augmentation du coefficient de sécurité grâce à un croisillon, ne donne pas à ce moment là toutes garanties.

b) *Les risques de rassemblement d'une quantité importante de matière fissile peuvent etre considérés comme nuls*

Dans ce cas, les normes relatives aux masses critiques dans les cylindres sont applicables.

Les courbes établies à partir de la méthode de calcul à la machine seront présentées en annexe.

Conclusion

Les calculs et les résultats présentés nous montrent, sur un cas particulier, la difficulté d'établir des règles générales de sécurité.

L'empoisonnement hétérogène peut être malgré tout avantageux dans

les cas où il permet d'augmenter le facteur de sécurité d'un facteur 2, et même d'un facteur 1,5.

Nous avons cherché à obtenir dans les calculs une valeur minimum du coefficient de sécurité.

Les résultats obtenus, basés sur la théorie de la diffusion et sur des constantes ajustées d'après des résultats expérimentaux obtenus à partir de manipulations sur les solutions de UO_2F_2, sont évidemment discutables.

Le réacteur ALECTO, destiné à tester la sécurité des récipients cylindriques contenant des solutions de plutonium, nous permettra également de faire une étude sur la sécurité apportée par les absorbeurs hétérogènes.

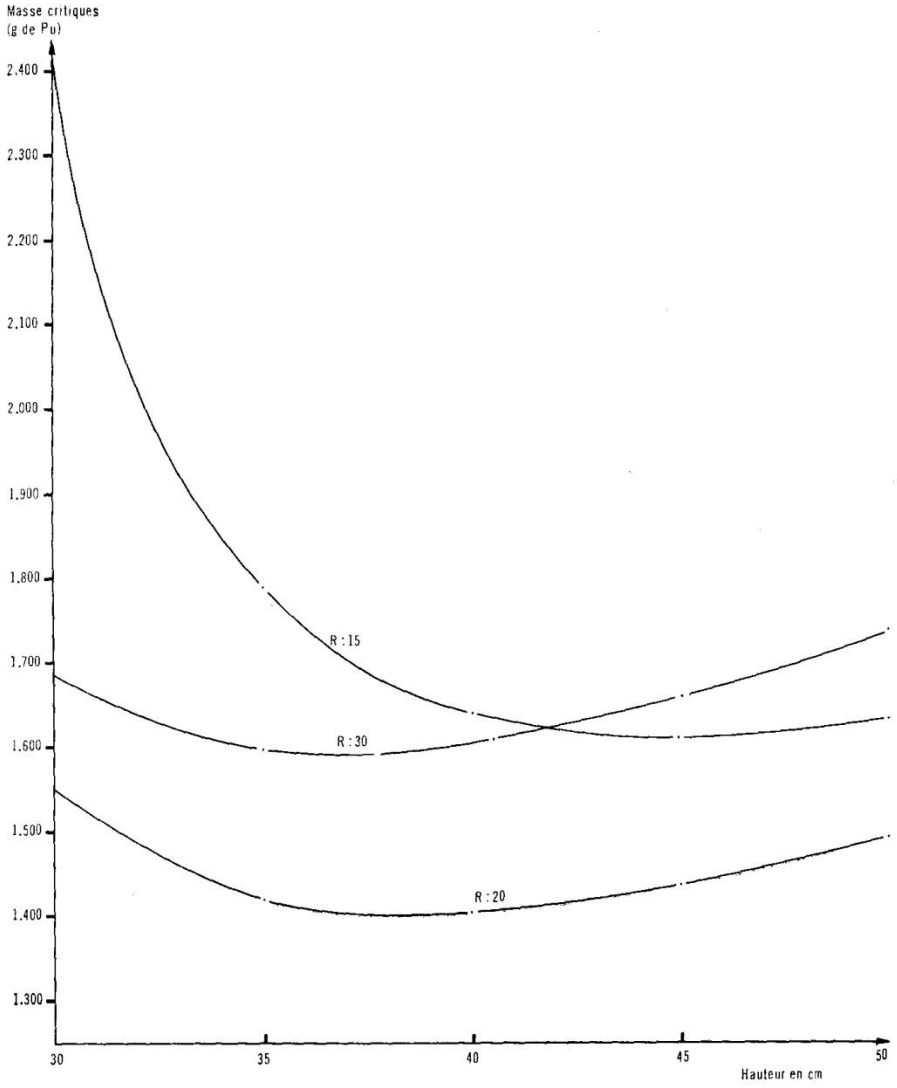


Figure 10. *Masses critiques des cylindres avec croisillon complet, réflecteur latéral infini d'eau (Rayon R constant).*

ANNEXE

INTRODUCTION

Nous nous proposons, dans cette annexe, de donner la méthode utilisée pour le calcul de l'influence du croisillon sur la concentration critique d'un cylindre de solution de matière fissile, de diamètre et de hauteur donnés, ainsi que les résultats détaillés obtenus par les différentes méthodes de calcul.

INFLUENCE D'UN CROISILLON A *n* BRANCHES

Nous supposons évidemment les n branches réparties de façon régulière. Deux branches consécutives font entre elles l'angle $\theta_1 = \frac{2\pi}{n}$ (figure 1).

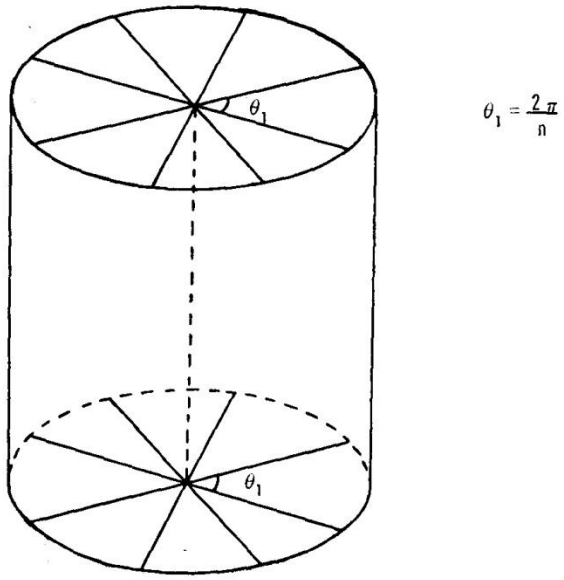


Figure 1. *Croisillon à* n *branches.*

Criticalité d'un secteur de cylindre nu d'angle dièdre

a) *Equations utilisées*

On a, avec les notations classiques, les équations de diffusion en théorie deux groupes :

$$D_1\nabla^2\Phi_1 - \Sigma_1\Phi_1 + k\Sigma_a\Phi_2 = 0$$
$$D_2\nabla^2\Phi_2 - \Sigma_a\Phi_2 + \Sigma_1\Phi_1 = 0$$

avec :

Φ_1 = flux de neutrons rapides

Φ_2 = flux de neutrons thermiques

D_1 = coefficient de diffusion des neutrons rapides

Σ_1 = section efficace de transfert du groupe rapide au groupe thermique

D_2 = coefficient de diffusion des neutrons thermiques

Σ_a = section efficace d'absorption par les neutrons thermiques

k = coefficient de multiplication en milieu infini.

On sait que B^2, laplacien matière, satisfait à l'équation :

$$(1 + L^2B^2)(1 + \tau B^2) - k = 0$$

avec :

$$L^2 = \frac{D_2}{\Sigma_a} \qquad \tau = \frac{D_1}{\Sigma_1}$$

La solution du système d'équations différentielles est de la forme :

$\nabla^2\Phi = -B^2\Phi$ ($B^2 = \mu^2$ racine positive de l'équation en B^2)

L'équation solution s'écrit, en coordonnées cylindriques (figure 2) :

$$\frac{\partial^2\Phi}{\partial r^2} + \frac{1}{r}\frac{\partial\Phi}{\partial r} + \frac{1}{r^2}\frac{\partial^2\Phi}{\partial\theta^2} + \frac{\partial^2\Phi}{\partial z^2} + B^2\Phi = 0$$

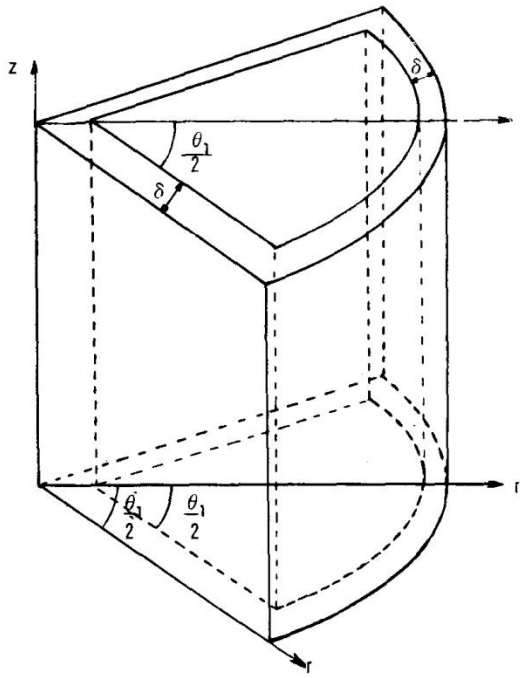


Figure 2. *Secteur cylindrique nu (limites extrapolées).*

On cherche une solution de la forme :

$$\Phi = R(r)\ \Theta(\theta)\ Z(z)$$

ce qui nous donne :

$$\frac{1}{R}\cdot\left(\frac{d^2R}{dr^2}+\frac{1}{r}\frac{dR}{dr}\right)+\frac{1}{r^2}\frac{1}{\Theta}\frac{d^2\Theta}{d\theta^2}+\frac{1}{Z}\frac{d^2Z}{dz^2}+B^2=0$$

Le flux s'annule aux limites extrapolées indiquées sur la figure 2. On admettra pour frontières où s'annule le flux :

$$r = 0$$

$$r = R + \delta + \frac{\delta}{\sin\left(\frac{\theta_1}{2}\right)}$$

(δ : distance d'extrapolation)

$$z = \pm\left(\frac{H}{2}+\delta\right)$$

b) *Conditions aux limites*

i) $$\frac{1}{\Theta}\frac{d^2\Theta}{d\theta^2} = -\alpha^2 = \text{constante}$$

d'où $\Theta = \cos(\alpha\theta)$.

Pour $\theta = \pm\frac{\theta_1}{2}$ $\quad \cos\left(\frac{\alpha\theta_1}{2}\right)=0 \quad \frac{\alpha\theta_1}{2}=\frac{\pi}{2}$

$$\alpha = \frac{\pi}{\theta_1} = \frac{\pi}{\frac{2\pi}{n}} = \frac{n}{2}$$

ii) $$\frac{1}{Z}\frac{d^2Z}{dz^2} = -\beta^2 = \text{constante}$$

$$Z = \cos(\beta . z)$$

Pour

$$z = \pm\left(\frac{H}{2}+\delta\right) \qquad \cos\left(\beta\frac{H+2\delta}{2}\right)=0$$

$$\beta = \frac{\pi}{H+2\delta}$$

En remplaçant dans l'équation en R, Θ, Z, $\frac{1}{\Theta}\frac{d^2\Theta}{d\theta^2}$ et $\frac{1}{Z}\frac{d^2Z}{dz^2}$ par leurs valeurs, on a :

$$\frac{1}{R}\left(\frac{d^2R}{dr^2}+\frac{1}{r}\frac{dR}{dr}\right)-\frac{\alpha^2}{r^2}+B^2-\beta^2=0$$

En posant : $B^2-\beta^2=B'^2$, on a :

$$r^2\frac{d^2R}{dr^2}+r\frac{dR}{dr}+((\beta' r)^2-\alpha^2)R=0$$

Appelons u la quantité βr.

On a :

$$u^2 \frac{d^2R}{du^2} + u \frac{dR}{du} + \left(u^2 - \frac{n^2}{4}\right) R = 0$$

La solution de cette équation qui satisfait à notre problème est la fonction de Bessel $J_{\frac{n}{2}}(u)$, qui s'annule pour $u = 0$, et qui a pour première racine non nulle $\mu_{\frac{n}{2}}$.

On doit donc avoir :

$$B' \left(R + \delta + \frac{\delta}{\sin \frac{\theta_1}{2}} \right) = \mu_{\frac{n}{2}}$$

$$B' = \frac{\mu_{\frac{n}{2}}}{R + \delta + \frac{\delta}{\sin \frac{\theta_1}{2}}}$$

On arrive à l'équation critique :

$$\frac{\mu^2_{\frac{n}{2}}}{\left(R + \delta + \frac{\delta}{\sin \frac{\theta_1}{2}} \right)^2} + \left(\frac{\pi}{H + 2\delta} \right)^2 = \mu^2$$

Calcul de l'influence des réflecteurs sur le secteur de cylindre

On a vu dans le rapport que l'on devait considérer deux gains de rayons dus au réflecteur (figure 3) : le gain de rayon dû au réflecteur latéral infini d'eau et le gain de rayon dû au réflecteur latéral de solution.

a) *Gain de rayon dû au réflecteur d'eau*

On a défini le gain de rayon dû au réflecteur d'eau dans le cas d'un cylindre de rayon de section droite R, possédant un réflecteur latéral infini d'eau, critique pour une hauteur H et une concentration c(g Pu/litre), comme la différence $R_0 - R$; R_0 est le rayon extrapolé du cylindre nu de hauteur H, contenant une solution de concentration c(g Pu/litre), tel qu'il soit lui aussi juste critique.

$R_0 - R$ varie avec R et H.

Le tableau suivant donne une idée de la variation de $R_0 - R$ en fonction de R et H (exprimés en cm) :

R	10	14,7	20	25
H	$R_0 - R$	$R_0 - R$	$R_0 - R$	$R_0 - R$
25	5,57	5,70	5,70	5,80
35	5,57	5,76	5,80	5,80
45	5,58	5,81	5,87	5,87
55	5,58	5,84	5,94	5,94
60	5,58	5,86	5,97	5,97

1. Solution de Pu
2. Reflecteur latéral infini d'eau
3. Plaques radiales absorbant les neutrons thermiques

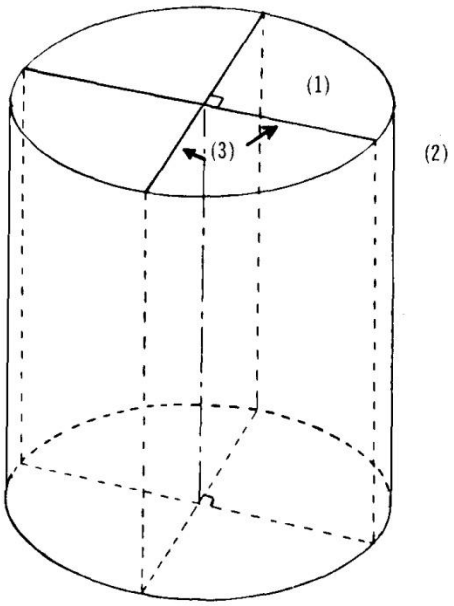


Figure 3. *Cylindre avec croisillon*

a_0 : gain R_0-R (réflecteur d'eau)

a_1 : gain R_1-r (réflecteur solution)

$$R_2 \simeq R + a_0 + \frac{a_1}{\sin\frac{\theta}{2}}$$

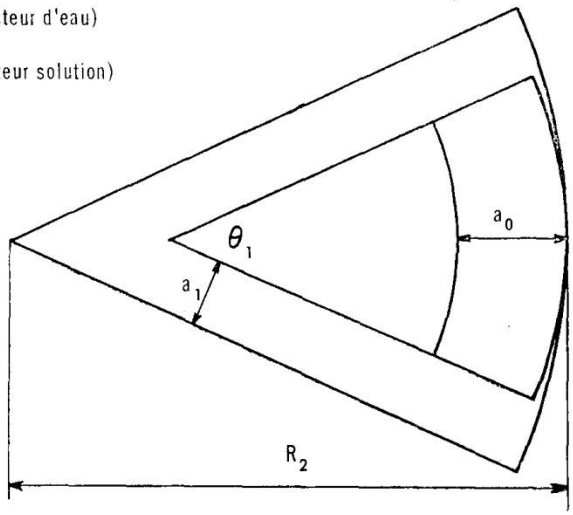


Figure 4. *Secteur cylindrique nu équivalent.*

On voit que $R_0 - R$ varie relativement peu (de 5,55 à 5,95 cm) lorsque R varie de 10 à 30 cm, et H de 20 à 55 cm.

b) *Gain de rayon dû au réflecteur de solution*

Nous avons considéré le cylindre de rayon r et de hauteur H ayant un volume égal à celui du secteur cylindrique de rayon R et de hauteur H ; r est tel que :

$$\pi r^2 = R^2 \frac{\theta_1}{2} = R^2 \frac{\pi}{n}$$

$$r = \frac{R}{\sqrt{n}}$$

Les surfaces latérales de fuite des deux solides sont respectivement égales à :

$$2\pi r H \qquad R.\frac{2\pi}{n}.H + 2RH$$

Il est facile de voir que la surface latérale du cylindre de rayon R est la plus faible. Le calcul fait sur le cylindre équivalent donnera donc une limite supérieure du gain de rayon dû au réflecteur.

Définition du gain de rayon

Nous sommes obligés, dans le cas du secteur de cylindre, de prendre une épaisseur équivalente de réflecteur. La figure 3 montre quelle est l'approximation faite. On prend une épaisseur de réflecteur e, telle que :

$$e \times R = \frac{\pi R^2}{4}$$

$$e = \pi \frac{R}{4}$$

Nous considérerons le cylindre de rayon r et de hauteur H, entouré latéralement d'une couche absorbant les neutrons thermiques, d'épaisseur négligeable. Ce cylindre est entouré d'un cylindre coaxial de même hauteur et de rayon r.

Les cylindres contiennent une solution à la concentration c (g Pu/litre).

Il est important de noter l'influence que peut apporter le réflecteur infini d'eau sur le gain de rayon. Nous donnerons dans le tableau du paragraphe suivant, les gains de rayon en supposant d'abord le cylindre extérieur non réfléchi, puis réfléchi par un réflecteur latéral infini d'eau.

On appellera gain de rayon, pour les valeurs R et H du cylindre avec croisillon, la valeur de $R_1 - r$, avec $r = \frac{R}{2}$, R_1 étant le rayon extrapolé du cylindre nu de même hauteur H, tel que le cylindre soit juste critique à la concentration c.

Calculs faits et résultats obtenus

Les calculs des hauteurs critiques H en fonction des concentrations c pour différentes valeurs de R, ont été faits sur machine Ferranti.

Les constantes utilisées ont été données dans le rapport. Elles sont redonnées dans la suite du texte.

Les deux tableaux suivants donnent la valeur de $R_1 - r$ pour $\left(n = 4,\ r = \frac{R}{2}\right)$, lorsque le cylindre de rayon $R' = \frac{R}{2} + \pi\frac{R}{4}$ est nu, et lorsqu'il est réfléchi par un réflecteur latéral infini d'eau.

TABLEAU I. CYLINDRE DE RAYON DE SECTION R′ NU

$r = \frac{R}{2}$	7,5	10	12,5	15
H	$R_1 - r$	$R_1 - r$	$R_1 - r$	$R_1 - r$
25	9,92	10,60	11,16	11,85
35	9,12	9,81	10,53	11,24
45	8,88	9,59	10,31	11,10
55	8,75	9,46	10,16	11,02
60	8,71	9,41	10,10	11,00

TABLEAU II. CYLINDRE DE RAYON DE SECTION DROITE R′ RÉFLÉCHI PAR UN RÉFLECTEUR LATÉRAL INFINI D'EAU

$r = \frac{R}{2}$	7,5	10	12,5	15
H	$R_1 - r$	$R_1 - r$	$R_1 - r$	$R_1 - r$
25	11,60	12,06	12,32	12,60
35	10,84	11,43	11,9	11,94
45	10,41	11,50	11,68	12,30

Secteur cylindrique nu équivalent

Nous voulons avoir une valeur minimum de la masse critique pour un rayon R et une hauteur H du cylindre avec croisillon. Nous remplacerons donc ce cylindre par le secteur cylindrique nu équivalent, de rayon R_2, tel que :

$$R_2 = \frac{R_1 - r}{\sin\frac{\theta_1}{2}} + R + R_0 - R$$

Equation critique

Elle s'écrit sous une forme simple :

$$R_2 = \left(\frac{5{,}13562}{R_2}\right)^2 + \left(\frac{\pi}{H + 2\delta}\right)^2 = \mu^2 \quad (n = 4)$$

μ^2 racine positive de l'équation en B^2

$$(1 + L^2B^2)(1 + \tau B^2) - k = 0$$

On a :

$$L^2 = \frac{L_0^2}{1 + z} \qquad k = 2{,}023\,\frac{z}{z + 1}$$

avec :

$$z = \frac{\Sigma_{a\ Pu}}{\Sigma_{a\ H_2O}}$$

Σ_{aH_2O} = 0,0197 cm^{-1}
$\Sigma_{a\ Pu}$ = section efficace macroscopique d'absorption du Pu dans la solution.

Les constantes utilisées ont été données dans le rapport.
L_0^2 = longueur de diffusion de l'eau : 8,123 cm^2
τ = âge des neutrons dans l'eau : 27 cm^2
D_1 = coefficient de diffusion des neutrons rapides : 1,135 cm
λ = distance d'extrapolation correspondant aux neutrons thermiques : 0,34 cm
D_2 = coefficient de diffusion des neutrons thermiques : 0,160 cm
$\sigma_{a\ Pu}$ = 982 barns
$\sigma_{f\ Pu}$ = 682,5 barns
ν = 2,91 neutrons émis par fission d'un atome de Pu.

Masse critique

La masse critique est donnée par :

$$M_c = \pi R^2 HC$$

Résultats obtenus

Les résultats obtenus par ce mode de calcul ont déjà été présentés dans ce rapport. Ils sont redonnés dans la figure 5. On voit que la valeur de la masse minimum est voisine de 1.400 g.

CROISILLON INCOMPLET

La résolution des équations de la diffusion, dans le cas d'un croisillon incomplet à quatre branches, ne peut se faire qu'à l'aide d'une ordinatrice de grande dimension (IBM-7090, par exemple).

Les calculs ont été faits sur IBM-704 et IBM-7090, en programme PDQ. Le cylindre a été assimilé à un parallélépipède à section carrée, de même hauteur et de même surface de section droite. Les résultats obtenus sont condensés dans les figures 6, 7, 8, et 9.

Les masses critiques ainsi obtenues sont légèrement supérieures à celles que l'on attendrait en milieu cylindrique, pour les mêmes surfaces de section droite et les mêmes concentrations, mais la différence est faible : de l'ordre de 5 %.

On peut établir les tableaux suivants :

MASSES CRITIQUES MINIMA EN GRAMMES,
EN FONCTION DES RAYONS DE SECTION DROITE R DES CYLINDRES

CYLINDRES NUS			
	Pas de croisillon	Demi-croisillon	Croisillon complet
R *(cm)*	M *(g)*	M *(g)*	M *(g)*
14,7	3.590	5.390	13.855
20	1.628	2.110	3.595
25	1.677	2.020	3.095
30	1.890	2.230	3.250
CYLINDRES REFLECHIS PAR UN REFLECTEUR INFINI D'EAU			
	Pas de croisillon	Demi-croisillon	Croisillon complet
R *(cm)*	M *(g)*	M *(g)*	M *(g)*
12,5	810	1.307	1.668
14,7	800	1.245	1.550
22,6	1.130	1.440	1.835
25	1.350	1.600	1.992

Nous en déduisons les coefficients de sécurité apportés par les demi-croisillons et les croisillons pour les différents rayons (le coefficient de sécurité pour un rayon de section droite R est défini comme le rapport entre la masse critique minimum avec absorbeur hétérogène, et la masse critique minimum sans absorbeur).

On a :

CYLINDRE NU COEFFICIENT DE SECURITE		
R *(cm)*	Demi-croisillon	Croisillon complet
14,7	1,50	3,86
20	1,30	2,21
25	1,20	1,84
30	1,18	1,72
CYLINDRE REFLECHI PAR UN REFLECTEUR INFINI D'EAU COEFFICIENT DE SECURITE		
R *(cm)*	Demi-croisillon	Croisillon complet
12,5	1,61	2,06
14,7	1,55	1,94
22,6	1,27	1,62
25	1,18	1,47

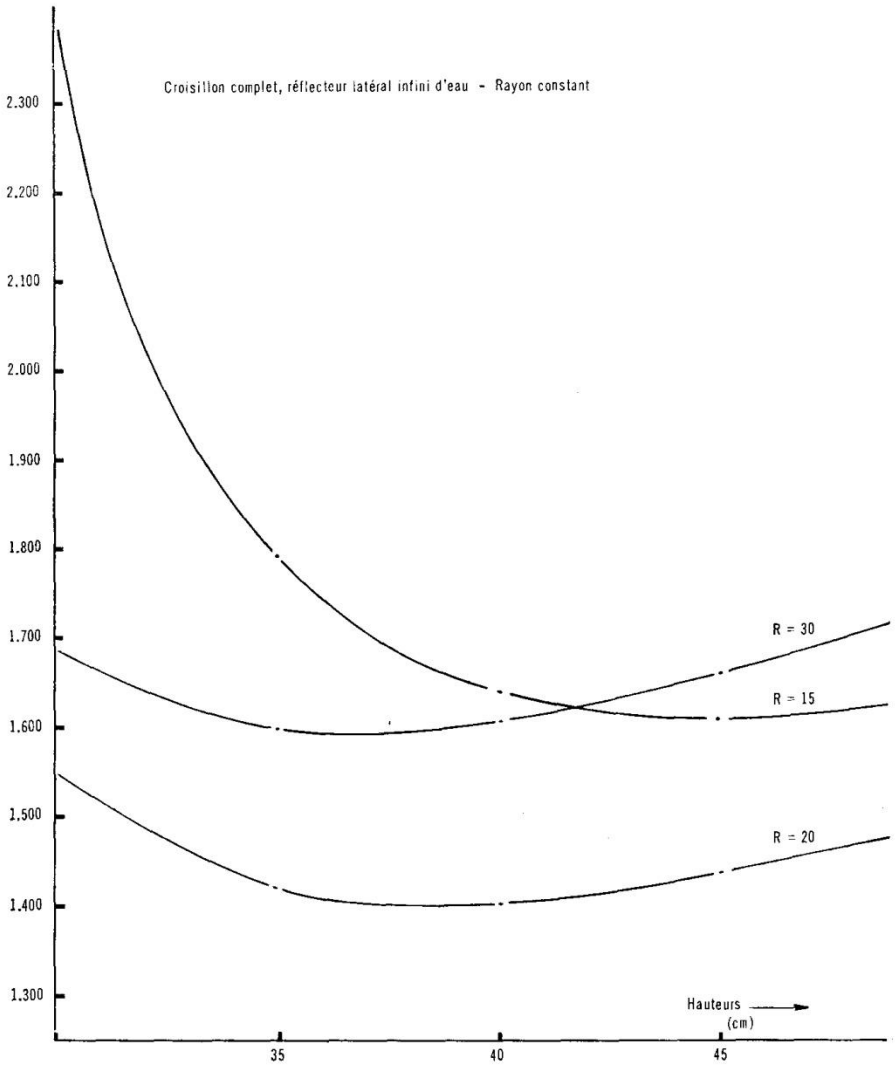


Figure 5. *Masses critiques des cylindres.*

On voit que, dans le cas du réacteur réfléchi, le coefficient de sécurité varie de 1,6 à 1,18 pour le demi-croisillon, et de 2 à 1,47 pour le croisillon complet, lorsque le rayon R varie de 12,5 à 25 cm.

Ces résultats ne sont valables que pour les cylindres de solution homogène. Nous avons exposé, dans le rapport, les réserves qu'il convient de faire lorsque la solution est amenée à précipiter.

Le calcul montre que le croisillon complet, et même le demi-croisillon, apportent, par des rayons inférieurs à 20 cm, un coefficient de sécurité non négligeable.

Les valeurs plus faibles trouvées pour les masses critiques dans le calcul au moyen des gains de réflecteur s'expliquent très bien puisque l'on a pris dans ce cas les hypothèses destinées à donner une valeur minimum des masses critiques.

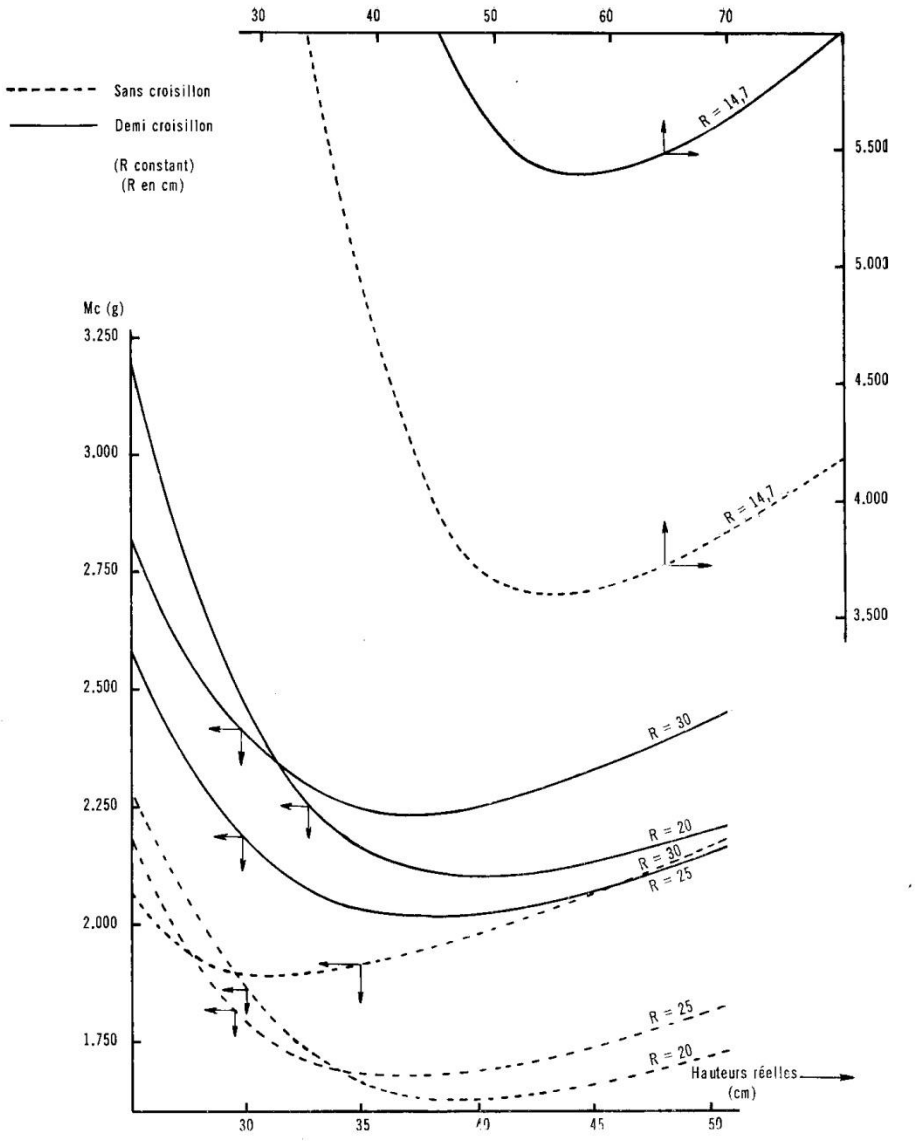


Figure 6. *Cylindres nus (masses critiques en fonction de la hauteur).*

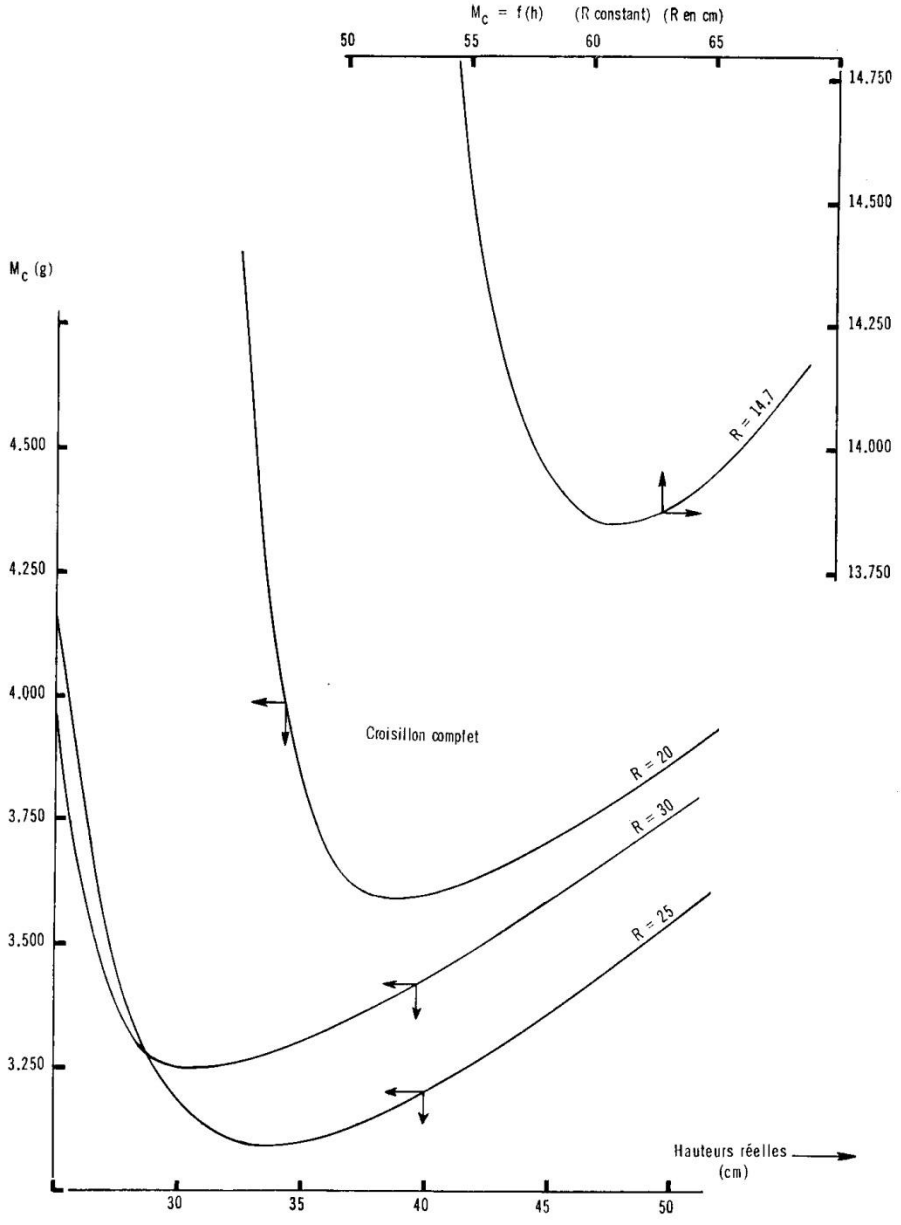


Figure 7. *Cylindres nus.*

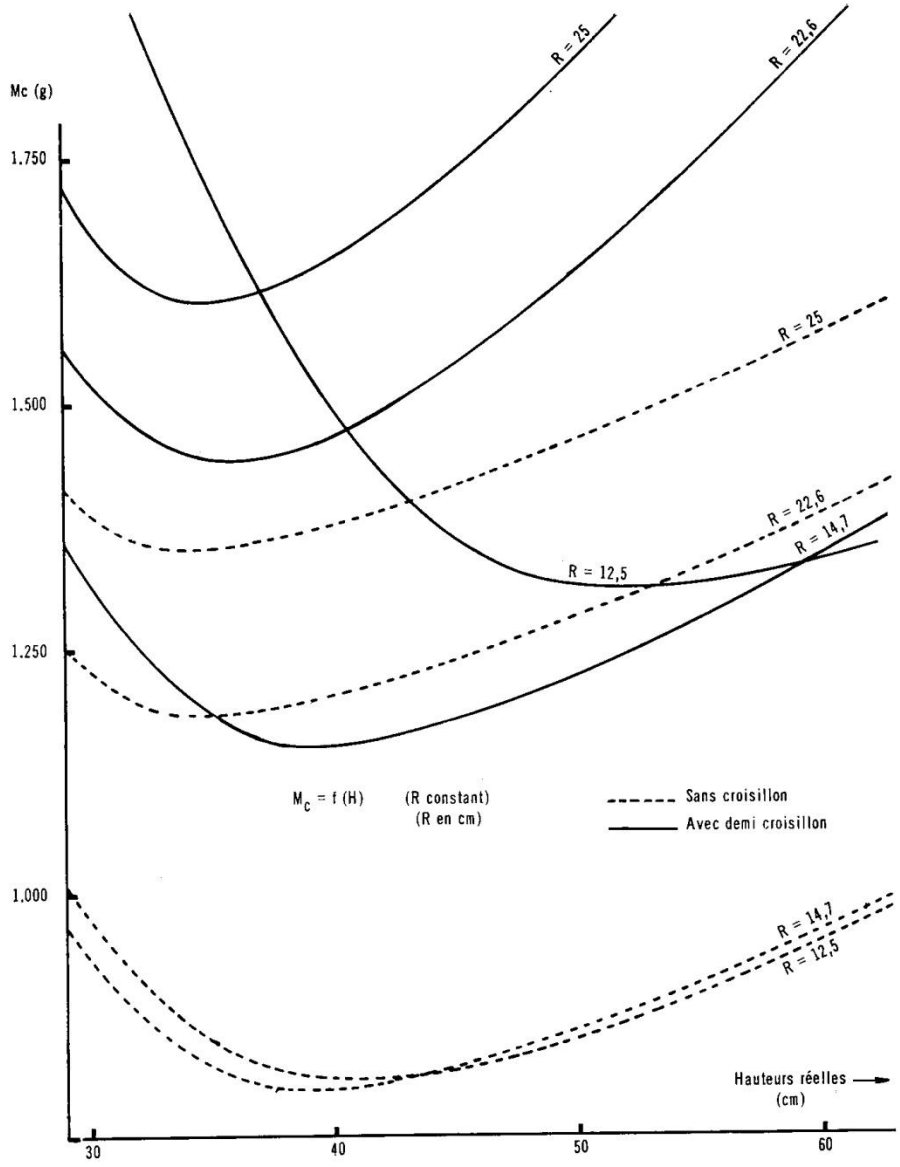


Figure 8. *Cylindres avec réflecteur latéral infini d'eau.*

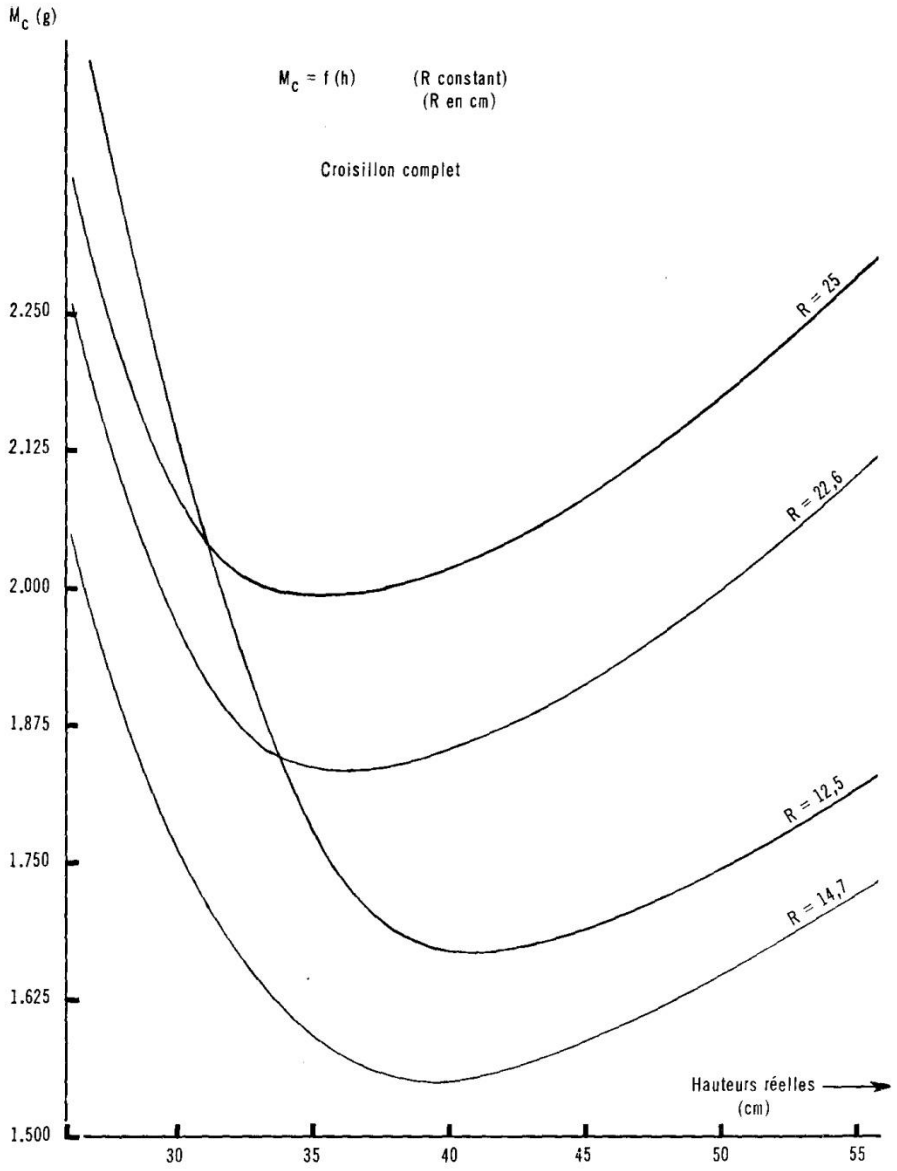


Figure 9. *Cylindres avec réflecteur latéral infini d'eau.*

DISCUSSION

E.R. WOODCOCK, United Kingdom:

I have a question for Mr. Wachter on his paper. Figure 1 therein was, he said, only an approximate indication of the amount of poisoning, which was calculated without allowing for any spectrum changes. Table 1 showed that the effect of spectrum changes is quite important. I presume that the later figures did not ignore the spectrum changes, and perhaps I could ask if he could give an indication of how accurate Figures 2, 3, 4, 7 and 8 are. I think you said these were the Rocky Flats calculations.

C.L. SCHUSKE, United States:

The Figure with the metal slabs in solutions and cadmium added to the solution is an experimental figure. The accuracy of those particular experiments is in the neighbourhood of $\pm$ 10 %. The other figures are calculations for which there are very few experimental checkpoints. For those calculations, I would estimate the accuracy to about $\pm$ 25 %.

G. BLÄSSER, Euratom:

Il apparaît à la page 2 du rapport de M. Caizergues que, pour un rayon inférieur à 11,5 cm, le cylindre a une masse critique plus faible que la sphère correspondante. Je crois que le cylindre a un rayon critique plus faible que la sphère corespondante, mais je crois également que, dans tous les cas, un cylindre critique doit avoir une masse critique supérieure. Conformément au rapport de M. Woodcock, nous avons trouvé que les masses critiques d'une sphère et d'un cylindre peuvent au maximum être égales, mais que, d'ordinaire, la masse critique du cylindre est supérieure.

R. CAIZERGUES, France:

Il est certain qu'en général la masse critique minimum du cylindre est supérieure à celle de la sphère. Mais, si l'on diminue suffisamment le rayon R du cylindre et celui de la sphère qui a même rayon R, la sphère obtenue à partir d'une solution aqueuse homogène de matière fissile ne peut plus diverger lorsque le rayon est de l'ordre de 10 cm.

R.J. SLOAT, United States:

Does Mr. Wachter have any information on the radiation degradation of these boron glasses and what would he consider a reasonable radiation level for application of these boron glasses?

J.W. WACHTER, United States:

I regret I have no information regarding the radiation level for these glasses but I am reasonably certain that such number has been established. My only interest in this has been that the glass manufacturers do make a high-boron glass. Since radiation problems do not appear in my criticality work I have not pursued such enquiries.

Session V

CRITICALITY CONTROL IN THE FABRICATION OF FUEL ELEMENTS

CONTROLE DE LA CRITICALITÉ DANS LA FABRICATION DES ÉLÉMENTS COMBUSTIBLES

Chairman - Président:

Mr. H. HOWELLS
(United Kingdom)

A TYPICAL MANUFACTURING PLANT FOR ENRICHED UO_2 FUEL ELEMENTS

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INTRODUCTION

The manufacturing plant designed to process enriched, unirradiated uranium usually assumes unique characteristics because of the requirements of nuclear safety. These requirements impose a variety of restrictions on the design engineer, who must abandon, or severely modify, the vessels and equipment that are conventional to his art.

Although the fundamentals of nuclear safety are generally known, the practice of nuclear safety soon resolves itself into an applied art. A balance must be established between adequate safety and minimum operating costs; between restricted equipment geometries and administratively controlled batching processes; between dependence on mechanical safeguards and reliance on human psychology. Chemical conversion of the fissile material involves a variety of special considerations. The safe recovery of uranium from salvage and scrap materials presents a very real problem. The distribution of uranium over a wide area makes accountability a major choice. Increasing inventories magnify the situation.

With a complicated overall picture and with high stakes involved, nuclear safety must be given serious and thoughtful consideration by everyone involved in the design, construction and operation of the plant. Those associated with the project must have both a general knowledge of the nature of nuclear hazards and an accurate definition of the risks specific to the production programme.

To carry out such objectives, the criticality specialist must be intimately associated with such a project from its inception. As the programme moves forward, he will be concerned with the nuclear characteristics associated with the material under the special conditions that may attend either the process or material, with the chemical features of the operation and the design of the equipment, and, finally, with the administrative and operational procedures which must be invoked in order to fulfil the basic processing objectives. The process by which these considerations are applied in practice is unique to each facility, but the nature of the problems is common to all. In the following discussion, a brief review of some of the general problems encountered is given. Attention is then focussed on problems associated with UO_2 production and, finally, detailed consideration is given to a particular fuel element manufacturing plant.

General Considerations

The effort to achieve criticality control in the processing facility is a continuing one; and the nuclear safety programme itself parallels that of the overall programme, becoming increasingly more specific and more firmly intermeshed with it. The general considerations given below illustrate the manner in which a nuclear safety programme evolves, and are indicative of the general sequence in which criticality problems must be resolved.

Feasibility and Cost Analyses

Nuclear safety considerations become necessary upon formulation of the most elementary material flow diagram, since the control of criticality hazards often influences the choice among possible chemical or physical operating techniques. It may also affect the utilization of existing facilities. For example, if various materials of different fission properties (enrichment, say) are to be processed simultaneously or in campaigns in a single facility, the entire facility should be designed for the material requiring the most stringent nuclear safety measures.

The requirements of nuclear safety are best served by a closed, continuous-processing train of equipment which is safe by reason of its design. Such a chain of safe equipment is much to be preferred to a process flow involving unit transfers of material, whether or not in safe equipment. A continuing effort in criticality control will result in a reduction of the need for administrative control, and this is best effected at the outset by use of automated equipment. Costs favour such expensive equipment only where a long-term or high-production programme is involved. As either the term or rate of production is decreased, it becomes increasingly necessary to depend on such manual operations as hand transport of materials and mass batching in order to meet cost and production specifications. The overall nuclear safety becomes correspondingly more dependent on pure administrative control and is weakened by this dependence on the human factor. This is not to imply that effective controls cannot be established, but rather that they require a continuing, positive effort on the part of both production and criticality personnel to maintain their effectiveness.

Where the nature of the process requires the exercise of administrative controls, each operation must, unless exceptional circumstances prevail, require the occurrence of at least two independent contingencies* in order for a critical incident to be possible. This double contingency requirement should apply not only to manual operations for which administrative controls are the only ones possible, but should include the ' safe ' equipment where safety is dependent upon the degree of enrichment of the uranium, the absence of reflection, the presence of poisoning materials and the like.

Equipment Design and Engineering Layout

During the design phase, the selection of the materials of construction and the equipment sizes and shapes form the nucleus of the nuclear safety effort. Nuclear safety should be incorporated into the design and construc-

* A contingency is a possible but unlikely and uncontrollable change in one or more of the conditions originally specified as essential to the nuclear safety of a specific operation or activity such that its nuclear safety is decreased.

tion of all process equipment involving fissionable materials. Such designs should take full advantage, through administrative control, of the characteristics of the material and process; but every effort should be made to place minimal dependence on such administrative control. Difficulties may be encountered at early stages of planning if specific information regarding possible equipment is not available. This problem occurs most frequently in connection with commercial equipment; the readily-available information concerning throughput and quality specifications rarely permits a full evaluation of the possible hazards. The ' normal ' hold-up of material in the equipment is of less concern than the maximum possible hold-up. The nominal size may differ widely from the true size. In general, a firm approval of a piece of equipment requires a complete assembly blueprint, for which manufacturer's pamphlets do not suffice.

The relatively large masses of material included in the main line of production require careful choice of equipment dimensions and operating procedures to ensure nuclear safety. Even so, the resulting process may not differ greatly from its counterpart in a plant processing non-fissionable material. But the handling of scrap and salvage materials is invariably a radical departure from standard practices. At virtually every point of salvage generation, there exists the potential for diversion of large amounts of uranium into a stream which is otherwise merely contaminated with uranium. Nor is this true only of salvage materials; steam and condensate lines, cooling water, ventilation systems, vacuum lines, and all such auxiliary service systems must be carefully scanned to ascertain that no potential hazards exist. The utilization of existing equipment or plant areas, whether previously used for non-fissile or fissile materials, may magnify these problems, particularly since a change in isotopic composition may be involved. The possibility of such conversions for future programmes may also be a factor in the purchase of new equipment or the establishment of administrative controls.

Construction and Start-Up

Every phase of construction of the facility should be inspected with the requirements of nuclear safety specifically in mind. It is essential that the construction be followed up to certify that nuclear safety requirements incorporated in the approved plans are accomplished in the construction of actual equipment. Only after appropriate consideration should in-the-field changes be made, since their effects on nuclear safety control may be subtle but important.

Start-up activities often generate unexpected problems. Safety controls may be inadvertently invalidated in the pressures to make deadlines and in the unevenness with which various parts of the production line may come on stream. For such reasons it is desirable that specific nuclear safety approval be required of all equipment just prior to its use for fissionable material. Although the equipment will already have been approved for purchase or construction, a review of this approval should be made which takes into consideration the special circumstances of start-up as well as the future conditions in the vicinity of the equipment.

Routine Operations

Every operation with fissionable material should be made pursuant to written standard operating procedures. These procedures should not merely

detail the process steps, but should also identify the nuclear safety requirements imposed on equipment and personnel. Mere statement of the requirements for nuclear safety is not sufficient; effective means of communication, which have been established with attention to the nuclear hazards involved in the process, should be used among responsible personnel. Such communications are of particular importance when a change in responsibility takes place. In general, these communications should be written; and attention should be given to the use of forms, check-lists, log books and the like, which are tailored to the specific purpose of enforcing nuclear safety through accountability methods and the delineation of responsibility.

Changes in process operations and equipment are inevitable both as an immediate and unforeseen necessity and as long-term effort to improve the process and its safety. In such cases, a criticality approval should be required. Such an approval is basically an agreement between the operations and the nuclear safety groups incorporating both the conditions of the operation and their consequent nuclear safety considerations and cautions. In order to provide a firm basis for such mutual approval, all requests for approvals should be in writing and sufficiently detailed to allow criticality analysis. Likewise, all approvals should be in writing and sufficiently detailed that those responsible for supervision are fully cognizant of the hazards involved in the process and are thus equipped to evaluate, for example, courses of immediate action to be taken in case of breakdown, fire or other emergency.

During routine operations, audits or inspections by qualified nuclear safety personnel should be made, to assist in keeping the operation free of non-standard operating conditions or techniques. Such inspections may be supplemented by an occasional (say, biennial) audit by a team of criticality consultants who are otherwise not associated with the plant. Non-standard conditions which are discovered in the course of audits or inspections are termed nuclear safety violations and are studied with great care by the criticality specialist as an index of the degree of effectiveness of the nuclear safety programme.

It is important that all persons associated with the processing of fissile material have a basic understanding of the ' do's ' and ' don'ts ' of nuclear safety, particularly where dependence is placed on administrative controls. In general, the supervisory services will be informed, in detail, of the requirements of criticality control through the nuclear safety approvals, clearances, comments or other formal channels. In addition, a training programme must be instituted to maintain a continuing awareness of the requirements of nuclear safety. Such a programme will normally include safety meetings, training films, pamphlets and the like, and may include individual discussions between supervisor and worker with a prepared check-list of subjects to be covered.

Special Considerations

A plant for the preparation and processing of UO_2 has process steps involving all forms of material: solutions, slurries and wet cakes, as well as oxide as dry powder, pellets or fuel elements. Not only do different nuclear safety considerations apply to each of these material forms, but also they change for each material as different isotopic enrichments are considered. The discussion below is intended to examine such considerations and thereby

indicate more exactly the extent to which the manufacturing plant to be described is unique or typical.

The effect of decreased isotopic enrichment in increasing both the minimum size and mass of critical homogeneous systems is well known. Figures 1—4 [1] show the trend of the data and Table I [2] lists generally-accepted values of the safe parameters for a variety of isotopic enrichments.

TABLE I. SAFE LIMITS FOR INDIVIDUAL SYSTEMS OF HOMOGENEOUS U 235 SOLUTIONS FOR VARIOUS ISOTOPIC COMPOSITIONS

(Tabulated Values Give the Maximum Allowable Inside Dimensions of the Containing Vessel or the Maximum Allowable Mass or Internal Volume)

U 235 Assay *(wt % (U 235)*	Volume		Infinite Cylinder Diameter		Infinite Slab Thickness		Mass *(kg U235)*
	(litres)	*(gallons)*	*(in.)*	*(cm)*	*(in.)*	*(cm)*	
100.0	4.4	1.16	5.0	12.70	1.5	3.81	0.350
75.0	5.0	1.32	5.2	13.21	1.6	4.06	0.360
50.0	6.0	1.58	5.7	14.48	1.9	4.83	0.390
40.0	6.7	1.77	6.0	15.24	2.0	5.08	0.410
30.0	7.7	2.03	6.3	16.00	2.2	5.59	0.440
20.0	9.5	2.50	6.9	17.53	2.7	6.86	0.480
15.0	11.0	2.90	7.4	18.80	3.1	7.87	0.520
12.0	12.5	3.30	7.8	19.81	3.4	8.64	0.583
10.0	14.0	3.69	8.2	20.83	3.6	9.14	0.600
8.0	16.0	4.22	8.7	22.10	3.9	9.91	0.650
6.0	20.5	5.41	9.6	24.38	4.5	11.43	0.710
5.0	27.0	7.13	10.25	26.04	5.0	12.70	0.800
4.0	33.8	8.92	11.2	28.45	5.5	13.97	0.930
3.5	40.0	10.60	12.0	30.48	6.0	15.24	1.04
3.0	49.2	13.00	12.8	32.51	6.5	16.51	1.20
2.5	64.6	17.10	14.0	35.56	7.1	18.03	1.47
2.0	95.1	25.10	16.0	40.64	8.7	22.10	2.00
1.75	126.0	33.30	17.6	44.70	9.6	24.38	2.56
1.5	186.0	49.10	20.0	50.80	11.0	27.94	3.60
1.25	308.0	81.30	24.0	60.96	13.5	34.29	6.67
1.0	731.0	193.00	33.0	83.82	19.5	49.53	22.70
0.8	3,917.0	1,040.00	58.0	147.32	36.0	91.44	36.00
0.72	∞	∞	∞	∞	∞	∞	∞

Source: Reference 2: interpolations were made from the source data as plotted in Figure 1 (reciprocal mass vs. log U 235 isotopic concentration).

Generally speaking, the optimum hydrogenous moderation for minimum mass is constant for all enrichments, so that the atom ratio of hydrogen to U 235 remains about 500. This means that there is a wide variation in the corresponding hydrogen-to-uranium atomic ratio, which, over the range from 2 to 100 % U 235, must change by a factor of 50. Since the chemical form of the uranium salt solution depends only on the uranium content, independent of its enrichment, it is clear that the molarity of the water-salt solutions must also change by the same factor of 50. This change in molarity represents the full range from a true salt solution at 100 % enrichment to a wet, unsaturated, undissolved salt at 1 to 2 % enrichment.

Figure 5 indicates how the critical masses change with H/U atomic ratio. It is seen that each minimum mass occurs at a lower value of H/U

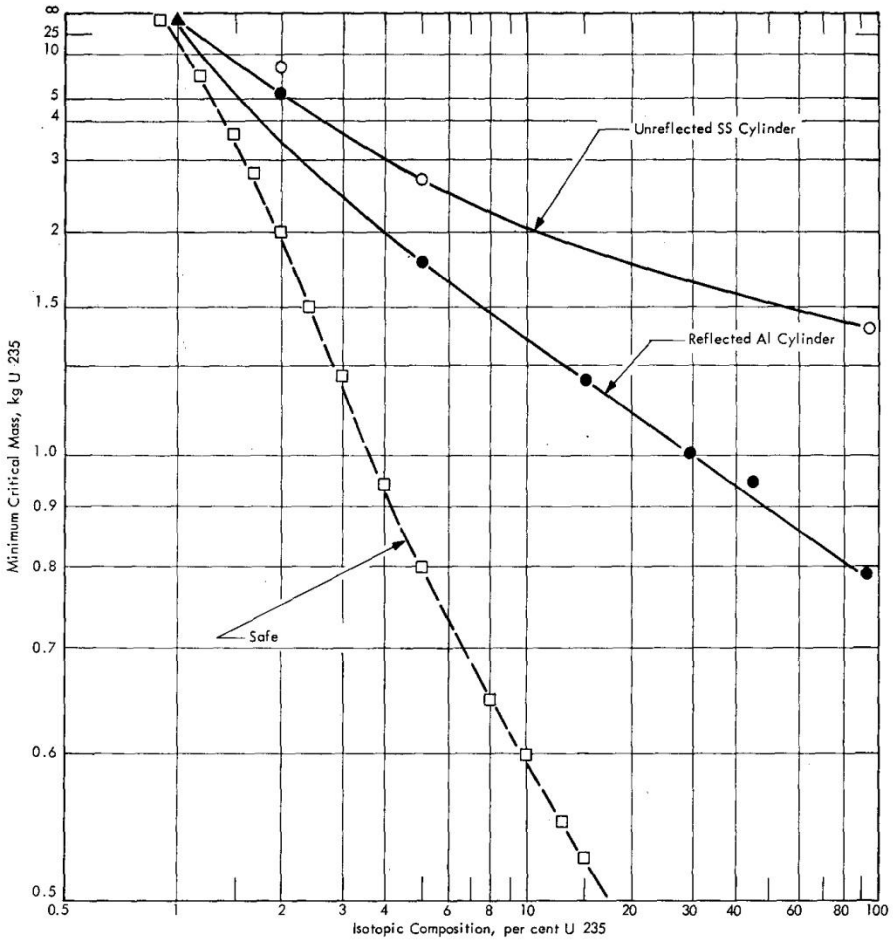

Figure 1. *Minimum critical mass versus U 235 enrichment.*
Variation of estimated minimum critical mass of aqueous uranium solutions (reciprocal scale) with U 235 isotopic composition (log scale). Critical data have been reduced to equivalent spheres by equibuckling transformations.

as successively smaller isotopic enrichments are considered, even though the H/U 235 ratio is about the same in each case. The physical form of the material, which is entirely a function of H/U, is indicated along the bottom of the graph for UO_2F_2 material. It is clear from these curves that any evaluation of the hazards associated with handling a particular form of uranium material must carry with it a specification of the enrichment. All solutions of 50 g U/litre (H/U $\sim$ 500) are identical in appearance; but the one at 100 % enrichment is of optimum concentration for criticality, while another at 2 % enrichment is far too dilute for critically. Since it is the uranium content that determines the chemical processing, identical chemical processes may or may not involve hazardous materials, depending on the enrichment involved.

The handling of dry materials is a case in point. Water-saturated uranium salt has a H/U of about 9, so that at 100 % enrichment the moderation

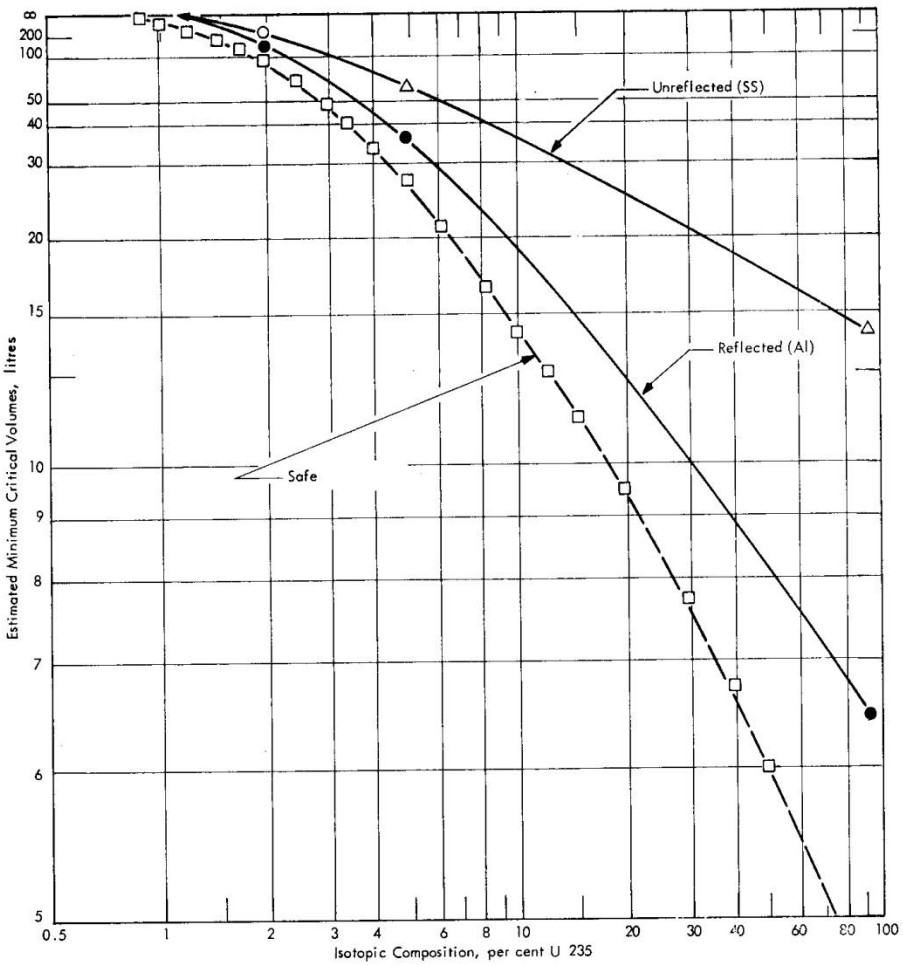

Figure 2. *Estimated minimum critical volume versus U 235 enrichment.* Volume is plotted on a reciprocal scale. Solid and broken lines merely connect estimated points and have no other signifiance.

is the same; i.e. H/U 235 = 9. At 2 %, on the other hand, the H/U 235 is about 450, so that the process of wetting the oxide has brought the moderation through its optimum value for criticality and thence to a system too dilute for criticality. In fact, the pick-up of moisture from the air can reach as high as an H/U of 0.5, which for 2 % is an H/U 235 of 25. In effect, therefore, moisture pick-up from the air produces greater moderation for 2 % enrichment than complete water soaking at 100 % enrichment.

The change in the nuclear characteristics of the various processes in a UO_2 production plant is of importance to the chemical processing supervisor who seeks to evaluate the relative hazard associated with the material involved in each processing step. Such an evaluation is desirable only because of the possibility of abnormal operation of the equipment or of an outright emergency such as fire. Under normal operating conditions, the safety of

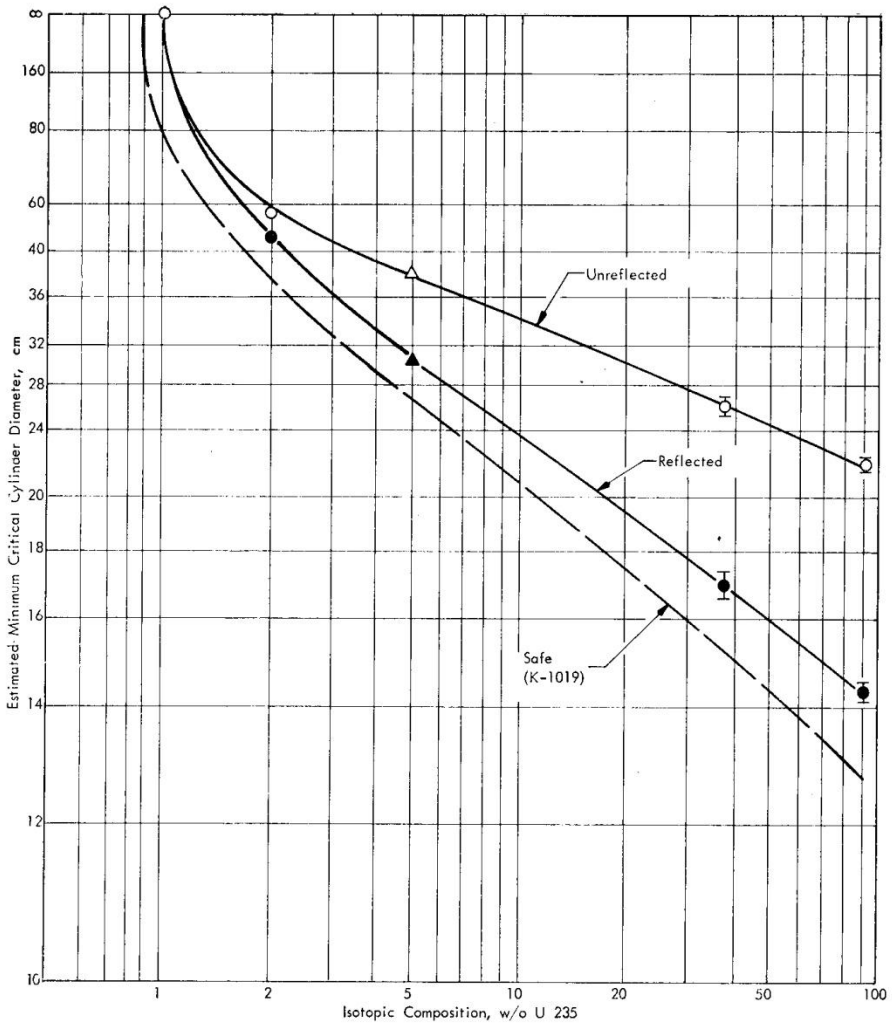

Figure 3. *Infinite cylinder diameter versus U 235 enrichment.* The estimated minimum critical cylinder diameter of aqueous uranium solutions (reciprocal scale) is plotted *versus* the U 235 isotopic composition (log scale). Circles and triangles denote aluminium and stainless steel containers, respectively. Limit strikes indicate the extreme range of possible critical values, as inferred from related critical and subcritical experiments. The dashed curve connects the safe values of Reference 2.

the equipment is not affected by its contained material. The safe geometric and mass values of Table I are based on the most reactive form of the material for each enrichment considered.

Description of a Plant

There are several methods that can be employed for producing UO_2 acceptable for reactor fuel. The method used in any given facility will depend on the type of raw material available to the manufacturer. The design engi-

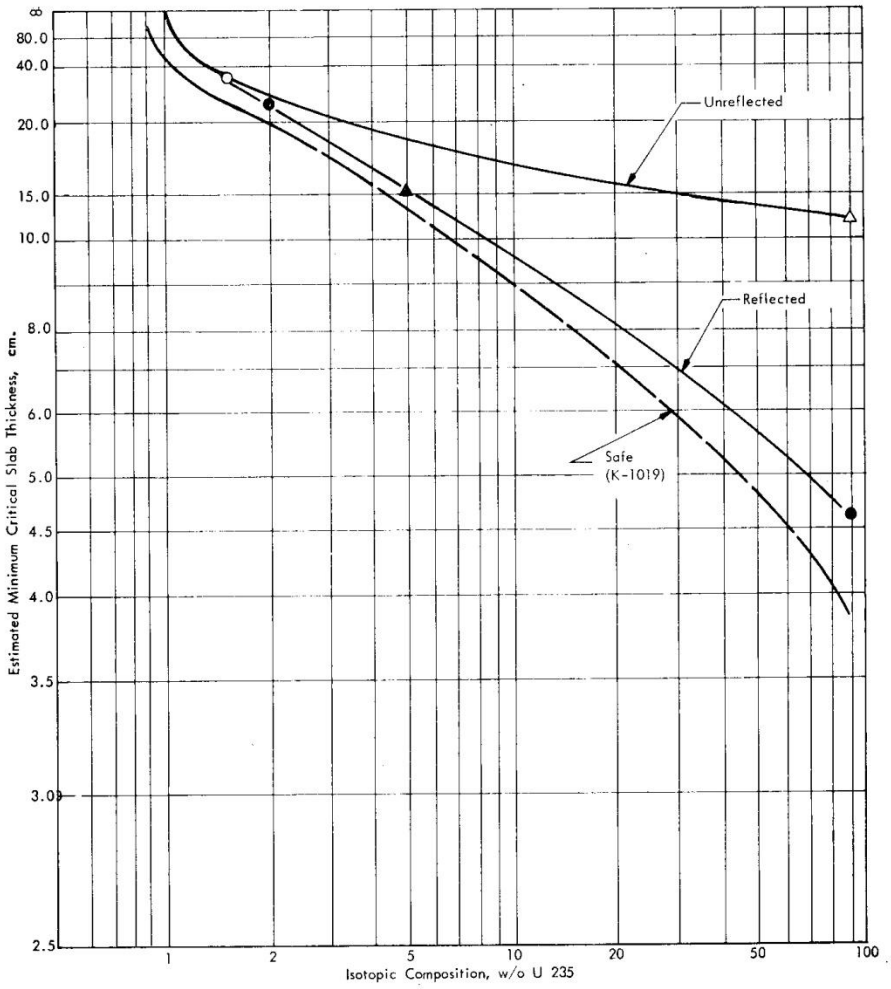

Figure 4. *Minimum critical slab thickness versus U 235 enrichment.* Estimated minimum critical slab thickness of aqueous uranium solutions (reciprocal scale) *versus* per cent U 235 isotopic composition (log scale). The dashed line connects the safe values given in Reference 2. Triangles denote stainless steel containers; circles denote aluminium containers.

neer will be guided by the most economical method that can be adapted to his raw materials and which will meet his product specifications.

Figure 6 illustrates one possible process line including, somewhat incidentally, the production of the enriched UF_6. The UO_2 facility is actually presumed to commence with receipt of the UF_6; other raw materials might include uranium metal, U_3O_8 or uranyl nitrate solution. The processing of the UF_6 involves most of the feed preparation steps necessary with these materials, and comment is made concerning them at the first step of the processing line which is common to both.

The comments given for each operation are intended to indicate the nuclear safety problems involved rather than specify an exact situation.

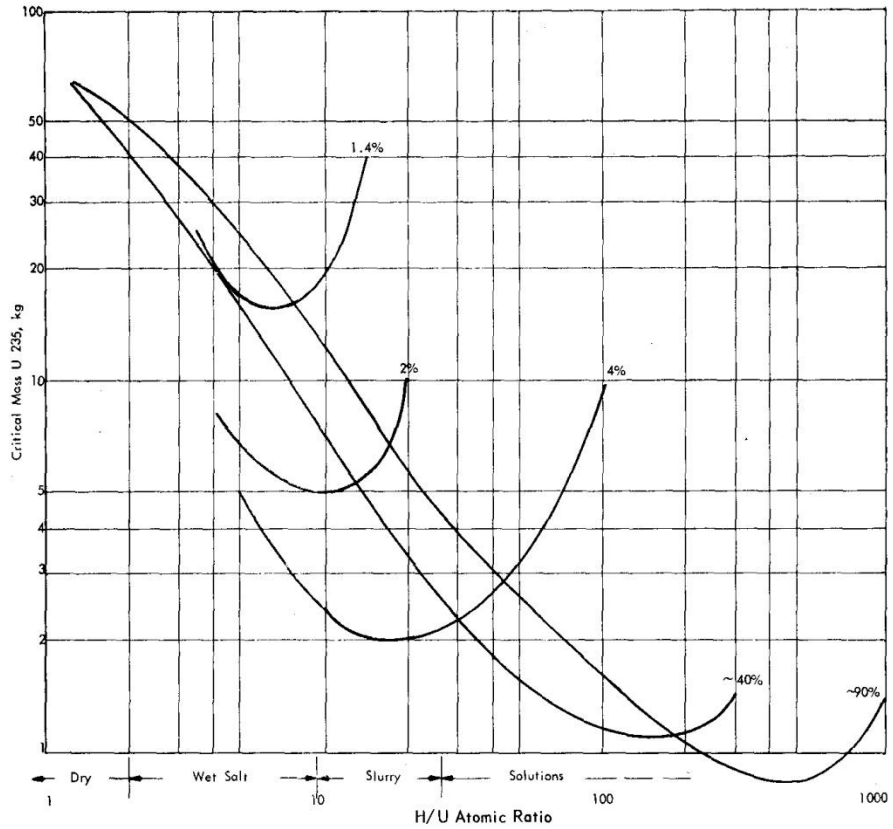

Figure 5. *Minimum critical mass versus H/U atomic ratio.*

Complete details, in the form of a compilation of the nuclear safety analyses of each step of such a plant, are available elsewhere [3]. Comments are made regarding alternative methods for certain operations: the continuous operation equipment depicted in Figure 6, and a possible manual operation. In every case, the continuous line of equipment is much to be preferred from the nuclear safety point of view because it eliminates hand operations requiring administrative controls. The following description of the plant processes and their appropriate nuclear safety considerations pertains very specifically to the assumed isotopic enrichment of 5 %. It will, however, indicate the nuclear safety problems encountered in UO_2 fuel element fabrication at any enrichment.

Feed Preparation

Receipt of Enriched UF_6. The solid UF_6 is transferred from the gaseous diffusion plant in pressurized cylinders. The transfer dolly is of a size appropriate to the U 235 enrichment both as to the inner container, which must be of a safe diameter, and the outer frame, which determines the number of such transfer dollies that can be stored or transferred at a single time. In

Figure 6

Typical manufacturing plant for enriched UO_2 fuel elements.

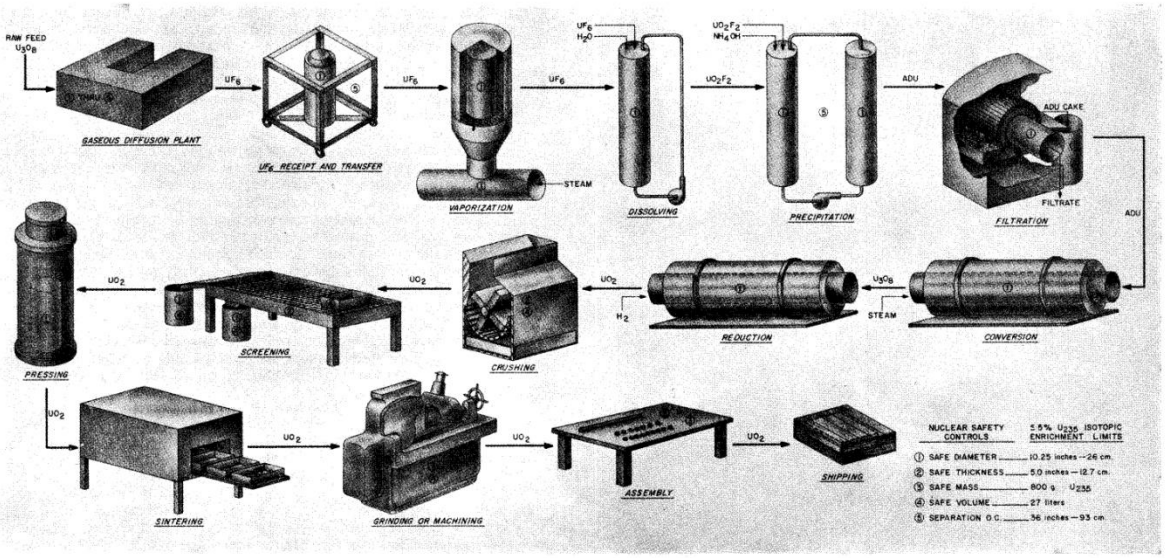

Figure 6. *Typical manufacturing plant for enriched UO_2 fuel elements.*

the 5 % isotopic enrichment range, this transfer of UF_6 is made by a truck in which the cylinders are placed horizontally on wooden structures which provide adequate separations. The separations are maintained by notching 20.32 cm × 20.32 cm wooden timbers lying on the floor of the truck to form saddles for the cylinders at the specified spacing. Chains secure the cylinders in the saddles. Storage of this material is accomplished in a similar fashion, the storage area being set off from those used for non-fissionable materials and marked with criticality limit signs.

Maintenance of the separations between cylinders and prevention of inleakage of water to the UF_6 are the primary bases for nuclear safety control. For UF_6 the required separations are usually determined by use of the solid angle method. The most practical procedure for smaller plants is often that of retaining the shipping containers for use in storage areas. Transfers of cylinders are accomplished by means of a saddle-type dolly, only one cylinder being transferred at a time. Handling equipment must be such as to render unlikely the possibility of one cylinder falling against another. Handling equipment other than that used for fissionable material must also be considered when storage areas are laid out. Hoists and cranes should not be allowed to transport heavy objects over arrays, and suitable bumpers around arrays may be necessary to prevent hazards from fork lifts and other mobile equipment. Route designations for each cylinder transfer must be specified so as to minimize the effects of interaction with other arrays.

Vaporization. A cylinder of UF_6 is placed in a pressurized vessel and super-heated steam is allowed to surround it. When the proper temperature is reached, connecting valves are opened to allow the gaseous UF_6 to pass from the cylinder to the dissolving step of the operation. The temperature is maintained above that necessary to form gaseous UF_6 so that condensation does not occur in the transfer lines. The steam is allowed to condense in the pressure vessel and a safe reservoir is supplied at the bottom.

The pressurized vessel, and the steam, normally present only nominal reflection to the inner cylinder, although the system is made safe against the possibility of full reflection and optimum concentration in the inner cylinder. The most important nuclear safety requirement in this operation is the prevention of an appreciable depth of liquid in the steam jacket. Either the condensate reservoirs must be made of safe diameter piping, as here, or multiple overflows must be provided to guard against the possibility of UF_6 leakage into the condensate. The arrangement shown guards against rupture of the cylinder by maintenance of the water level well below the cylinder and by suitable condensate overflows to prevent filling of the outer cylinder with water.

Dissolving or Hydrolysis. The gaseous UF_6 is passed to a vertical cylinder through a jet-type nozzle where it unites with de-ionized water. Proper ratios of each are essential to proper dissolution. Non-corrosive materials such as plastics must be used because of the hydrogen fluoride present in the solution. The components are continuously agitated to assure complete dissolution of the UF_6. The resultant uranyl fluoride (UO_2F_2) solution may be stored in safe geometry equipment or immediately transferred to the precipitation operation.

Safe geometry is used throughout the dissolving operation since the material being handled is in the most reactive form for this isotopic composi-

tion. Attention must therefore be paid to the elimination from the area of such hazards as open-topped, unsafe vessels and clusters of safe containers into which the solutions may leak. A further hazard is the well-intentioned placement of such vessels under a leak by janitors, maintenance men or others who are unaware of the hazard.

Precipitation. The uranyl fluoride solution is transferred from the dissolver to one or more vertical cylinders where the solution can be continually circulated. It is then reacted, in fixed molar ratios, with ammonium hydroxide and the solution is continually agitated until thoroughly mixed. The proportions of ammonium hydroxide and uranyl fluoride must be carefully controlled in order to achieve a specific particle size through precipitation. The precipitate from the resultant operation is ammonium diuranate (ADU). Rather than allow the ADU to precipitate, it is maintained in slurry form until transferred to filtration.

Safe geometry equipment is essential to a continouus operation of this nature. Any equipment malfunction during this operation may bring about a wide variation in uranium concentration. Dilution of the uranyl fluoride may occur if the precipitating agent is too dilute. Failure to circulate the material results in a precipitation of the ADU, in the course of which virtually any moderation may occur and the possibility is that the material will pass through the optimum moderation for minimum mass or volume.

This operation is important in defining the particle size of the resultant UO_2. For this reason, other feed materials may be especially processed so as to incorporate it. Uranium metal, for example, could be burned in oxygen, resulting in U_3O_8, and then converted directly to UO_2. Specifications on the grain structure may not allow such an abbreviated feed preparation, however, and the metal must then be dissolved in nitric or sulphuric acid. The resulting solution is then precipitated with ammonium hydroxide, as described above*. When U_3O_8 is the feed material, it may be dissolved and precipitated in a similar fashion. Solution feed materials are, of course, precipitated directly.

Filtration. Many types of filters may be used to collect the ADU. The rotating drum filter and the plate-and-frame filter press are considered here. The first is a continuous process. The ADU slurry is pumped into a sump of safe slab dimensions, and a perforated drum covered with a filter cloth rotates through the sump. A vacuum is pulled on the drum causing the ADU cake to adhere to the filter and to dry as the drum rotates through about three-quarters of a revolution, where it is scraped off into the discharge chute. The filtered solution passes through the drum and is piped to storage for further processing or for analysis prior to discard. The discharged cake is slurried with water in a safe tank and pumped to a wash filter identical with the primary filter. An alternate filtration process might use a plate-and-frame filter press in which a vacuum draws the filtrate through the filtering medium so as to deposit the ADU cake on a filter cloth supported by frames. This process must be interrupted periodically and the filtered cake manually removed from the press and transferred to a tank for slurrying with water. The slurry is pumped to an identical filter press for washing.

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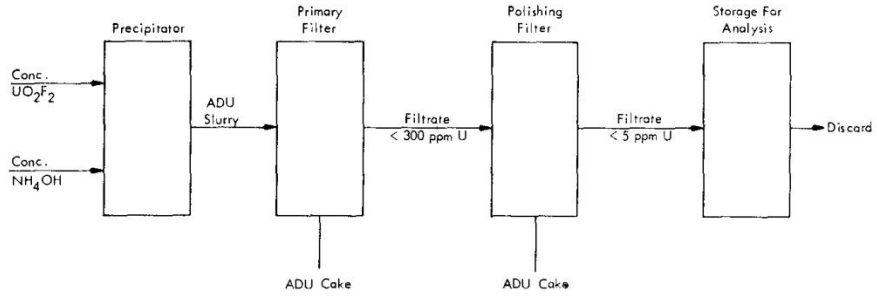
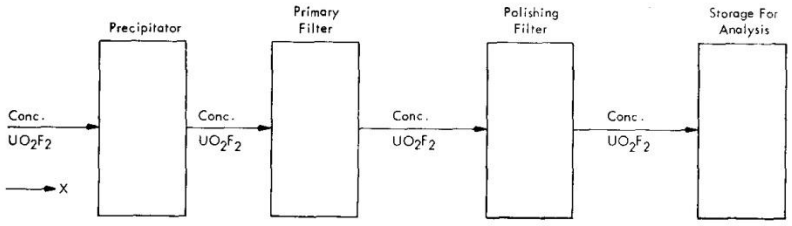
* The dissolving operation must be performed in equipment of which the size is limited by the safe dimensions for the metal or solution (whichever is the smaller).

The use of the plate-and-frame filter press for this operation is controlled by limiting the number of frames to those necessary to achieve a safe volume or slab thickness. The rotary drum filter, on the other hand, poses a number of difficult nuclear safety problems. It seems wise not merely to limit the depth of the sump to a safe dimension by means of weirs, but also to restrict the overall size of the equipment to a safe volume, even though, under normal circumstances, the amount of uranium contained both in the sump and on the drum and on the ADU cake discharge trough is quite small relative to the applicable critical mass.

Filtrate System. Filtrate from the primary filter is pumped to a polishing filter to remove traces of ADU, and then to a hold-up tank for analysis before discarding. The nuclear safety considerations for the filtrate system are indicative of the alertness to human and equipment maloperation that must be maintained if nuclear safety is to be effected. Figure 7*a* shows the normal situation for this system, in which the primary filtrate contains only minimum quantities of uranium. The polishing filter removes most of these, and the resultant material, having only trace quantities of uranium, can normally be sewered after analysis. Despite this low concentration, safe equipment is essential even for the final storage tanks, to guard against the possible, but unlikely, occurrence of an interruption in ammonium hydroxide feed to the precipitation apparatus three steps before. Figure 7*b* shows the result of



(a) Normal flow for filtrate system.



(b) Filtrate system flow in the event of an interruption in the ammonium hydroxide supply to the precipitator.

Figure 7. *Effect on filtrate system of interruption in flow of precepitation agent.*

such an omission. Since no precipitation occurs, the slurry does not develop and uranyl fluoride goes directly to the primary filter, passing through it as a filtrate. Unless caught by alert supervision, it could easily pass through the polishing filter also and be held up in the final storage tanks. In the continuous train of processing equipment depicted in Figure 6, the occurrence might not be discovered at all except by the uranium analysis routinely made before discard.

The use of safe equipment for the filtrate system can be relaxed by employing fixed poisons. Indeed, this filtrate system provides a perfect example of the use of poisons as a contingent control [4] since the hold-up tanks can be of large volume and filled with Raschig rings. Corrosion is no problem. The filtrate is normally little more than contaminated water, but the tanks must be safe, under normal conditions, in the absence of the rings. In the event that precipitation does not occur, and the tanks become filled with UO_2F_2 feed solutions, the rings must be sufficient to prevent criticality.

Drying and Conversion. Drying and conversion may be accomplished by a continouus operation in which the wet ADU cake is screw-fed to one end of an indirectly heated rotary calciner. By regulating the internal temperature and introducing steam countercurrent to the flow of the ADU cake, it is converted to a dry uranium oxide (U_3O_8). A second means of drying and converting the ADU cake is a manually-operated furnace. The wet ADU cake is manually placed into individual pans and the pans inserted into an oven. The material is allowed to stay in the furnace under controlled temperatures and steam atmospheres until converted to uranium oxide. The uranium oxide powder must then be removed manually and transferred to storage or to the next step of the operation. Both temperature and steam atmosphere must be rigorously controlled if the product of the conversion is to be of ceramic grade.

The batch processing of the ADU cake can be rendered safe by the use of safe geometry cans and by the imposition of the necessary administrative controls to ensure separation of the individual batches. Although numerical limitations on the number of batches or cans that may be placed in the furnace, or that may be used at each work station, provide conventional controls, these may be supplemented by the requirement that pans always be handled in the same plane. The pans may be shaped in such a way that they do not invite stacking. In the oven and in the storage racks, relatively simple fixtures can be designed so as to permit only one layer of pans to be used.

The rotary calciner may represent a case where the desirable features for good chemical engineering and the requirements of nuclear safety do not agree. It may be necessary for the design engineer to suppress his desires for a calciner of a large diameter, and consequent large internal surface area, in favour of a smaller but longer calciner having a safe internal diameter for the enrichment involved. Plugging of the calciner is a distinct possibility through poor operation of the equipment or by an interruption in the heat. Either occurrence allows insufficient drying of the ADU, which may then stick and plug the equipment. This may be particularly important at the lower isotopic enrichments since the ADU cake, in drying, is very likely to pass through its optimum moderation.

Reduction. The nuclear safety considerations in this operation are identical with those in the drying and conversion step, largely because the

equipment is automatic and no visual check is made on the dryness of the calciner product.

Processing to Granular Characteristics

The specifications, and particularly the density requirements, on the final fuel element frequently require an extensive degree of working of the uranium dioxide to produce the desired granular structure. To accomplish this requires preliminary processing of the dioxide aimed primarily at obtaining a specific particle size and particle size distribution.

The nuclear safety considerations in the following steps are the same for each and are concerned almost entirely with the prevention of hazard in the event of water leakage into the dry material. For this reason, safe geometries are used throughout. While in the dry state, the oxide is relatively non-reactive since little neutron moderation is provided by the oxygen. Moreover, this reactivity is further reduced for most of these steps because of the low density of the oxide particles. To maintain dryness, attention is given to all sources of water in the production area so as to remove, as far as practicable, any possible moderation of the product material. However, at the lower enrichments, very little moisture is required to bring the moderation into the range of minimum masses; and safe geometries and safe spacing must be maintained.

Pre-pressing. Pre-pressing frequently is used in a typical manufacturing plant when the final product is to be in a pellet or rod form. Pre-pressing utilizes a single-acting die which presses the raw UO_2 into a solid cylinder to give better granular structure to the final product.

Crushing. The UO_2 can be crushed for better distribution of particle sizes. The crusher can be any standard model that is designed to accomplish the desired particle size distribution.

Screening. A screening operation is usually desirable in order to segregate the oxides into proper particle sizes. The use of screens on a power-vibrating mechanism is a standard means to accomplish this operation. The material is collected from the screen at two locations: one for oxide of the desired sizes, the other for oxide particles which are oversize and must be returned to the mill.

Fabrication of Fuel Rods or Pellets

The operations given below assume that the oxide is processed into rod or pellet form for use as reactor fuel. It may be, however, that the screened oxide is tightly packed into tubing and used directly. Alternatively, it may be dispersed in such matrix materials as iron, aluminium or zirconium, and cast and/or extruded into the finished fuel rod. The use of fertile materials as the matrix, such as depleted uranium or thorium, might also occur.

Pressing. If the UO_2 powder is to be in the form of a pellet or rod, it is pressed to an oversize dimension in a suitable press. The operation might result in a single rod or pellet or, as in many instances, may involve multiple pressings.

Sintering. The sintering process is one in which the pressed rod or pellets are loaded in boats and the boats loaded in a furnace. Indirect heat is applied to the furnace and the temperature increased slowly until the sintering temperature is reached. After the sintering temperature is maintained for a designated time, the furnace is allowed to cool slowly. A cracked ammonia atmosphere is maintained throughout the sintering process.

Grinding or Machining. Either grinding or machining can be used for obtaining the diameters of pellets or rods containing UO_2. If dimensional tolerances are acute, a centreless grinder will be used in preference to a machine such as a lathe. A lathe cannot easily be used for machining UO_2 pressed pellets or rods for standard machine surfaces, but close dimensional tolerances can be achieved as efficiently as with the grinding process. Nuclear safety in these processes is enhanced by the fact that they involve dry materials. The use of plasticizers in the pressing operation with a subsequent heating process to drive off the plasticizer would require separate consideration, since the oxide is thereby moderated. The comments made earlier regarding batch conversion of the ADU cake would then apply. Apart from such deviations from the above programme, the maintenance of mass batch limits and the designation of limit areas provide the nuclear safety controls over main line operations.

If a coolant is used, special attention must be given to the salvage materials from the grinding or machining operations. At the higher isotopic enrichments particularly, modifications to the lathe or grinder may be advantageous in restricting the volumes for safety purposes. The normal coolant sumps may be very large and must be restricted to a minimum depth at the normal overflow and further safeguarded by multiple overflows at the safe depth (safe thickness or volume for the enrichment concerned). When this is not practicable, as for high enrichment, the normal sump may be eliminated and a smaller pan placed directly under the work. Firm administrative controls must be set up around the handling of the coolant materials which should be filtered as soon as possible, preferably before re-use, and certainly before being handled in unsafe geometries. Even filtered coolant is preferably handled in drums filled with Raschig rings as a poison.

In general, however, most such operations would be accomplished under dry conditions. Attention must be directed to the vacuum system and its associated filters. The possibilities of moderation of the oxide in the filters, including the moderation posed by the filter medium itself, should be examined. It is frequently desirable to handle the filters in plastic bags, since this prevents water inleakage and, further, reduces the health physics problems.

It should be noted that the magnitude of the nuclear safety problems of handling salvage depends strongly on the enrichment and the production rate. A highly-enriched programme, or a 5% enriched programme of some magnitude, requires close control over salvage. On the other hand, the whole problem may evaporate for a limited manufacturing run at a low enrichment for which all the salvage developed may not comprise a safe batch.

Assembly of Fuel Elements. The 'typical' fuel element is made up by inserting the uranium dioxide pellets or rods, with or without additional materials, into a pre-formed tube of a structural material such as alumi-

nium, Hastalloy or stainless steel, possibly by means of a loading device. The loaded tube may then be placed in a vacuum chamber which is evacuated and back-filled with a noble gas such as helium. Then the end cap is welded in place within the chamber. The resultant fuel rods are handled in specially designed trays which serve the double purpose of preventing rod breakage and maintaining numerical batching for nuclear safety purposes. These trays may then be handled so as to maintain a safe slab for the heterogeneous system that would result from water flooding. A cluster of such rods may be placed in a welding fixture and secured together by means of end plates, spiders or the like. This may complete the fuel element assembly, or it may represent a sub-assembly of a still more complicated fuel element.

These operations may form an assembly line whose complexity and area requirements match those of all preceding operations. The nuclear safety controls are entirely those established by administrative measures. For this reason, it is essential that a detailed standard operating procedure delineate the operations to be performed at each stage of the assembly. It is usually easy to ensure that the rods, sub-assemblies and fuel elements move in a common plane. Numerical limit restrictions can be established around the various process and storage facilities and their effect reinforced by use of signs stating the criticality limits, and by firm accounting methods.

Shipping. Containers for the shipment of completed fuel elements and/or assembled moduli of a reactor are fabricated of metal or wood. They are constructed to be waterproof and to withstand a certain degree of crushing in the event of a transfer vehicle wreck. The containers are normally fabricated to carry a specific type of element and are not necessarily interchangeable.

The nuclear safety criterion for shipment is the establishment of a safe individual system by making use of safe geometry, mass or volume. Determination of the proper spacing that should be incorporated in the construction of the container must be made on the basis of the arrangement of the array desired (plane or cubical), the reactivity of each container, and the mode of transportation to be used. Such determinations may be made by a solid angle, a lattice density or other method, and the safety factors applied in consideration of whether or not the shipment has exclusive use of the transport vehicle.

Concluding Remarks

The essential characteristics of a nuclear safety programme for a uranium manufacturing plant have been examined. Criticality considerations become necessary in the very early stages of planning for such a plant and may enter importantly into the cost picture. Where possible, the facility should be designed as a closed, continuous processing train of equipment which is safe by reason of its design; such a process line is to be preferred to one involving unit transfers of material, whether or not in safe equipment. Where manual handling of material is unavoidable, a continuing positive effort on the part of both production and criticality personnel becomes necessary to maintain effective administrative control.

The requirements for routine maintenance of a nuclear safety programme have been discussed and are found to include detailed standard operating procedures, effective communications among responsible personnel, written

nuclear safety approvals, audits and inspections, and a continuing programme of education.

A typical process line for the production of UO_2 reactor fuel elements from a hexafluoride feed material has been described and the nuclear safety considerations applicable to each unit operation briefly discussed. The specific example chosen presumed an isotopic composition of approximately 5 %, but the approaches to the nuclear safety problems remain the same for any isotopic enrichment.

Nuclear safety is a fundamental consideration in processing enriched uranium, regardless of its isotopic content, and it must not be compromised for mere expediency in accomplishing production commitments. Criticality is a hazard which must be prevented or avoided rather than corrected.

REFERENCES

1. J. W. WACHTER *et al.*, " Nuclear Safety Handbook ", *Y-1272*, Y-12 Plant, Union Carbide Nuclear Company (to be published).
2. H.F. HENRY, A.J. MALLETT, C.E. NEWLON and W.A. PRYOR, " Criticality Data and Nuclear Safety Guide Applicable to the Oak Ridge Gaseous Diffusion Plant ", *K-1019 (5th Rev.)* (1959).
3. W.T. MEE, T.J. CAGLE and J.W. WACHTER, " Nuclear Safety Anlysis of a Typical Production Line for High-Fired UO_2 at $<$ 4.5 % U 235 Enrichment ", *Y-1273*, Union Carbide Nuclear Company (1959).
4. J.W. WACHTER, " Homogeneous and Heterogeneous Poisoning as Potential Methods for Controlling Criticality ", This Conference, p. 289.

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DISCUSSION

H.J. HEIN, Germany:

Why do you heat your UF_6 bombs in a pressurized vessel with super-heated steam instead of more easily and cheaply under normal pressure in an open pot? Did you ever hear of ruptures of UF_6 cylinders?

J.W. WACHTER, United States:

Under normal circumstances one could drive UF_6 from the cylinders either way. This happens to be a convenient method of doing it. So far as I know, there is no advantage to be gained by using super-heated steam in this step. With regard to the second question, I do not know that there have been such breaks, but I think that some have occurred due to faulty valves. I understand that experiments have been made at the Oak Ridge Gaseous Diffusion Plant with respect to the rupture of UF_6 cylinders, and perhaps Mr. Mallett would care to comment on this matter.

A.J. MALLETT, United States:

Cylinders which are to be used must pass through a special group, whose responsibility is to pressure-test them at very high pressure; the standards required are at least 100 % above any pressure that they will actually get, and the same cylinders are generally re-tested every five years. We have had only one known case of rupture, where temperatures were too high, but the cylinder was overfilled beyond capacity. Normally the quantity of material placed in such cylinders is limited to an amount which will fill the cylinder and take liquid form at a temperature considerably above any that will normally be used, unless you have electric heating without adequate controls to stop it in event of shortage.

R. GALLEY, France:

Nous avons beaucoup apprécié l'exemple choisi par M. Wachter sur la précipitation de l'ammoniaque. Dans un tel cas, l'ingénieur de génie chimique est tenté de choisir l'une des deux méthodes suivantes : la première méthode consiste à faire confiance au personnel d'exploitation, afin de s'assurer que, par le moyen de check-lists, l'ammoniaque a été ajouté à la solution d'UO_2F_2 pour la précipitation ; la deuxième méthode consiste à mettre un appareil automatique, par exemple un pH-mètre, pour interrompre automatiquement l'opération si l'ammoniaque n'a pas été ajouté. Dans le premier cas, on fait appel à la conscience professionnelle du personnel et, dans le second cas, à un appareillage automatique. M. Wachter pourrait-il me préciser les règles de génie chimique qu'il applique dans de pareilles circonstances et la méthode à laquelle il fait généralement confiance.

J.W. WACHTER:

As to the two methods, the administrative control and the instrumental control, I think the chemical engineer seems rather to depend upon the administrative control with the nuclear safeguards which I have discussed. So far as nuclear safety is concerned, one would assume that in both cases there would be some failure involved. Both these controls, administrative and instrumental, are what we refer to as safeguards. One would assume that there should be at least two safeguards — one might prefer two instruments, for example, rather than only one. And, generally speaking, one would make the assumption for nuclear safety purposes, that there is a possibility of both failing. Therefore, I don't think that primary dependance would ever be put on either one or the other. In point of fact, we consider instruments as a means of administrative control.

J. GUNTHER, France:

Il ressort de l'exposé de M. Wachter que le gâteau de diuranate d'ammonium est filtré sur un filtre rotatif et que les dimensions globales de l'appareil sont telles que la sécurité est atteinte, même si tout l'intérieur du filtre rotatif est en fait rempli de précipité. Nous avons dû faire face à un problème analogue et nos spécialistes en matière de filtration ont estimé qu'il était très difficile de fabriquer un tel type de filtre, qui réponde aux conditions de sécurité nucléaire. Il s'agissait alors de savoir si on ne pouvait pas donner à ce filtre des dimensions supérieures, en utilisant le volume intérieur pour le remplir d'un poison quelconque destiné, d'une part, à empêcher toute

interaction du point de vue nucléaire entre les deux extrémités du filtre et, d'autre part, à éviter l'introduction physique de matières à l'intérieur du filtre, ce qui permettrait d'obtenir des dimensions bien supérieures. Il faut remarquer que nous avons renoncé à nous engager dans cette voie, mais nous serions heureux de savoir si la question s'est également posée pour M. Wachter.

J.W. WACHTER:

Yes, indeed, we had a problem with the one which you saw. It ended up with a relatively safe and quite small continuous filter. It seems to me that your suggestion of poisoning the interior is a very good one — an excellent application in that one does not really expect to find corrosive materials in this plant stream, since it is essentially water. I would be interested in your progress in this respect.

A.J. MALLETT:

I am rather interested in the type of filter, in the sense that one could use the internal volume merely as the steam heating chamber — a solid surface container, let us say — pass steam through the centre and use the exterior as you are drying, instead of a paper-filter. I wonder why you didn't do that?

J.W. WACHTER:

I imagine because this is a standard, off-the-shelf item, so dear to the chemical engineer. That is, the continuous filtration equipment which I described is commercially available.

G.W.K. FORD, United Kingdom:

I would like to ask whether you have any criticality alarm arrangements in this plant? Also, what are your rules for fire-fighting, and your precautions against fire?

J.W. WACHTER:

We have criticality alarms in any portion of the plant which handles more than a critical mass. They are distributed on 60 or 80 ft centres, and are a standard gamma radiation alarm. With regard to fire precautions, I presume you are referring to the fact that there are certain areas in the plant where the firemen may not use water. Wherever possible, in handling oxide, we seek to keep it covered, and consequently to expand those portions of the dry areas, where one can use water, if required, in place of the various dry compounds. I am afraid I cannot give you much of an answer on what specific precautions are taken to prevent fires. But I do know that, in connection with the handling of chips and fines and so on, one does use perchloroethylene — if you were referring to the pyrophoricity problem.

J. GÉRARD, Belgique:

L'utilisation d'hydrogène pour le processus de réduction d'U_3O_8-UO_2 et le danger d'explosion susceptible d'en résulter augmentent-ils le danger de criticalité ? Comment pourrait-on éventuellement se protéger contre ce danger ?

J.W. WACHTER:

The step in which the hydrogen is used is actually a dry step. The only case in which one has highly radioactive material would be in the event that the preceding step, involving the steam, were to fail so that the material came directly down. Actually, that is most unlikely, and consequently, I think that an explosion, assuming even that it was quite a bad one, would cause a dispersal of the dry material. I would therefore think that no very great additional hazards are involved.

NUCLEAR SAFETY IN CHEMICAL AND METALLURGICAL PROCESSING OF PLUTONIUM

E.D. CLAYTON
Hanford Laboratories Operation, United States

INTRODUCTION

When fissile materials are handled in significant quantities, such as in chemical separation plants for reprocessing spent reactor fuels and concomitant metallurgical processes, a potential criticality hazard exists. The prevention of a criticality incident, or inadvertent uncontrolled critical nuclear reaction, is of primordial importance because of the potentially serious consequences which can result.

The nuclear safety problems which are encountered in the handling and processing of fissile materials outside of reactors are especially complex; not only must criticality be considered for the standard operating conditions of the plant, but also for those off-standard conditions which are physically possible and, at the same time, perhaps most favourable for the chain reaction. In determining the nuclear safety of a chemical and metallurgical processing plant, all aspects of reactor theory may be involved, including both moderated and unmoderated systems. The problems are further enhanced because of the lack of certain experimental data and the uncertainty which exists in the theoretic values.

This paper is concerned primarily with nuclear safety considerations in the handling and processing of plutonium. Among the questions discussed are: What types of criticality problems are encountered in a typical plutonium processing and metal fabrication plant? What criticality data are needed, and what are some of the methods and procedures used to achieve nuclear safety in operation?

NUCLEAR SAFETY CONSIDERATIONS

Consider the conditions under which a given array of, say, plutonium and other materials will become chain-reacting. The exact configuration or spatial density must be known for each kind of atom present in the system. Thus, criticality depends not only on the amount of fissile material present, but on the size, shape, and material of any containment vessel which may be used, on the nature of any solvents or diluents and on the presence of materials which may act as neutron reflectors. In evaluating the nuclear safety of a given process, consideration must be given to the various in-

plant factors which affect the criticality of an individual unit or quantity of fissile material, and due allowances made for the effects of neutron reflectors. The effects of neutron interaction between associated vessels or units in related operations (storage arrays, interconnected pipes, operations in adjoining hoods, etc.) must also be considered.

Some of the methods which have been used for criticality control are listed below. In some cases combinations of several different methods are used together in order to obtain the most efficient operation consistent with nuclear safety.

i) Process vessels are designed to be safe by geometry, in which case the neutron leakage is so high as to prevent criticality regardless of the quantity of fissile material contained therein;

ii) Operating limits are imposed on material compositions, such as the density of fissile material contained per unit of a solution volume, or on the quantity of diluent or moderating material which may be present;

iii) The amount of material handled in a single operation is limited to a subcritical amount, in which case a safe mass limit or method of batch control is applied;

iv) Restrictions are placed on the amount and form of material permitted in a given storage array, and on the distribution in space of the material contained within the array;

v) Soluble, or fixed, poisons may be used in some cases to increase the mass limit or to make a vessel safe by geometry.

Typical Flow Diagram - Plutonium Solution and Plutonium Metallurgy Facility

A typical flow diagram (Figure 1) has been prepared to illustrate the types of criticality problems which are encountered in handling plutonium and to indicate the areas in which they occur together with the principal methods of criticality control[1]. The preparation of PuO_2 and any Pu metal from a $Pu(NO_3)_4$ solution is not part of the plutonium fabrication plant operations [2, 3]. As far as these operations are concerned, the criticality problems begin with the receipt of the PuO_2 or the Pu metal. The diagram is not intended to be a detailed, all-inclusive flow pattern of each step in the fabrication of the fuel elements.

In examining Figure 1, it is apparent that the criticality problems involve both homogeneous and heterogeneous systems, and include both moderated and unmoderated (fast) systems, as well as lattices or bundles of fuel rods in a moderator (autoclave and etching process).

Further details will be given on the methods used for criticality control in a later Section of this paper.

Criticality Data as a Basis for Nuclear Safety

Data on plutonium criticality, which are used as a basis for establishing nuclear safety, are included and summarized in Reference 4. Recommended nuclear safety limits are given in Reference 5. Criticality data on the plutonium systems referred to therein have been obtained primarily from the results of experiments conducted at the following U.S. sites: data on plutonium metal were obtained from the Los Alamos Scientific Laboratory, the

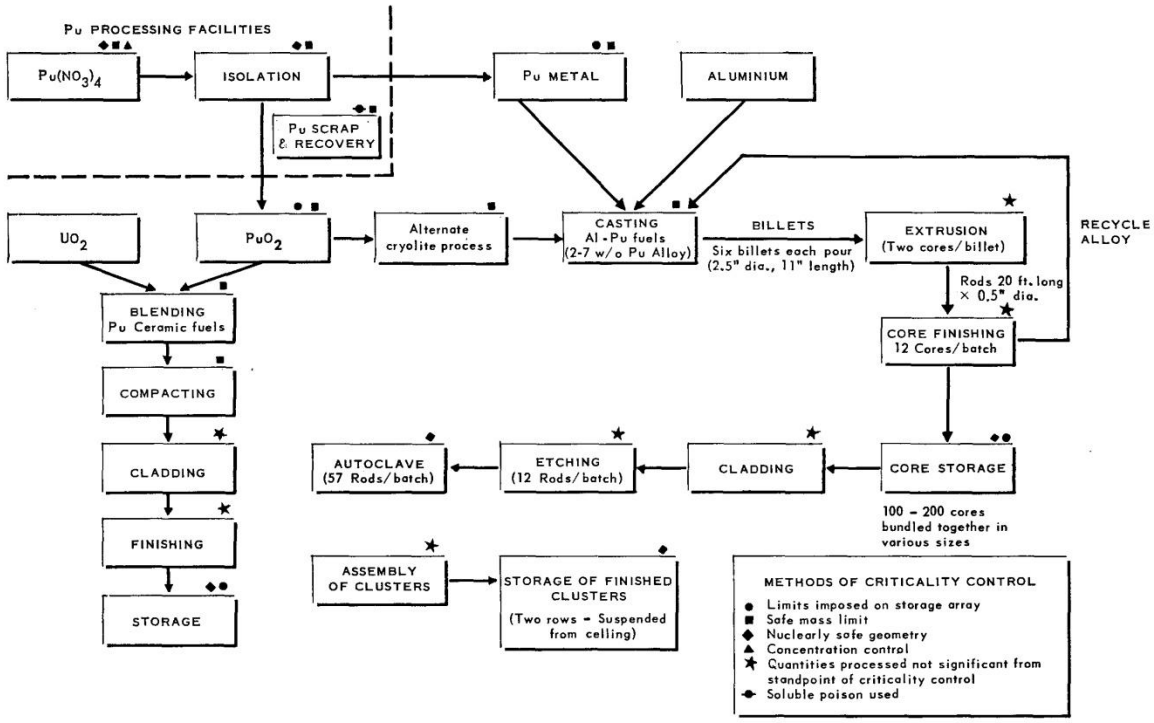

Figure 1. *Typical flow diagram - plutonium metallurgy facility.* Fabrication of aluminium - plutonium fuels and plutonium ceramic fuels.

Lawrence Radiation Laboratory, and some data from the Rocky Flats Plant; data on plutonium solutions were obtained from experiments conducted at Hanford [6]. The nuclear safety of Pu solutions has been based on the results of the Hanford experiments, under which the criticality of aqueous Pu solutions was studied for plutonium concentrations up to 136 g/litre.

Some of these experimental data are presented in Figure 2, which gives the relationship between critical mass and volume for water-tamped spheres and cylinders of aqueous $Pu(NO_3)_4$ solutions. For the data shown, the Pu contained about 3 % Pu 240 and the nitrate concentration was in the range of about 100-170 g/litre depending on the concentration of the Pu. The Pu 240 content of the bare sphere was about 4 %. The critical mass values shown are for the total plutonium, including the Pu 240. The effect of nitrate on the critical mass of water-tamped spheres is shown in Figure 3. The Hanford experiments also included measurements to determine the effect of Pu 240 on the criticality of dilute Pu solutions in the range of 0.54-4.40 % Pu 240.

From the Hanford data, the minimum critical mass for a homogeneous Pu-water system was estimated to be 509 g Pu 239. The minimum value was obtained for a concentration of 33 g Pu/litre in a water-reflected sphere about 12 in. in diameter.

The results of subcritical multiplication measurements have recently been reported in which Pu metal and Plexiglas plates were stacked together in cylindrical geometry [7]. These data may be used to provide a qualitative

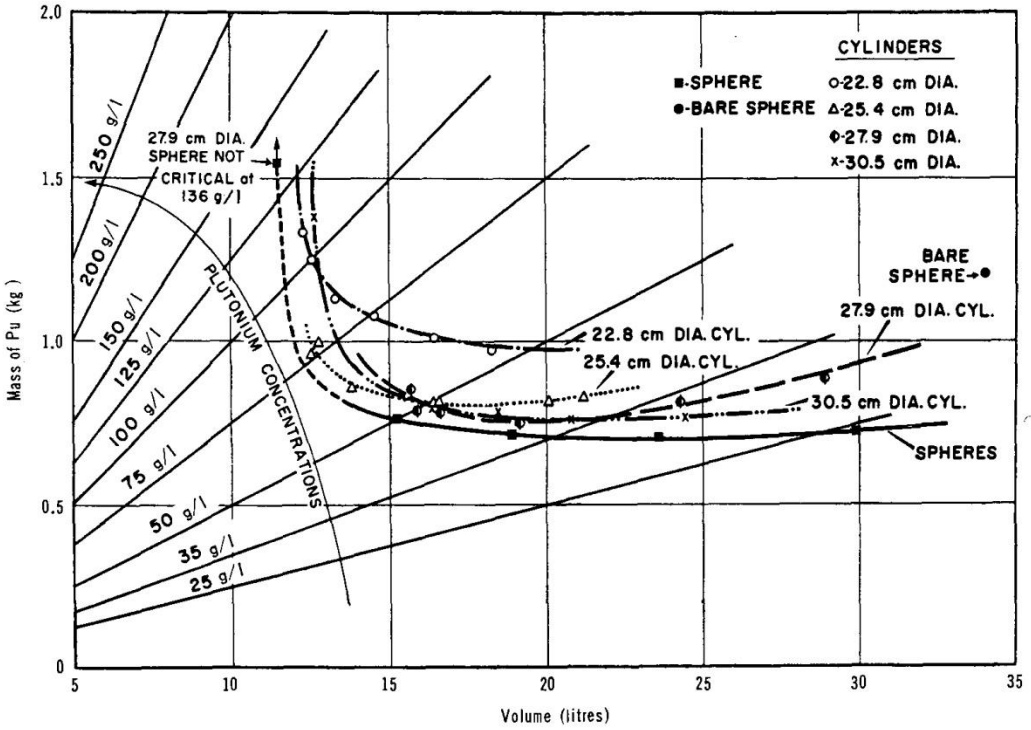

Figure 2. *Critical mass and volume of reflected spheres and cylinders of $Pu(NO_3)_4$ in aqueous solutions.*

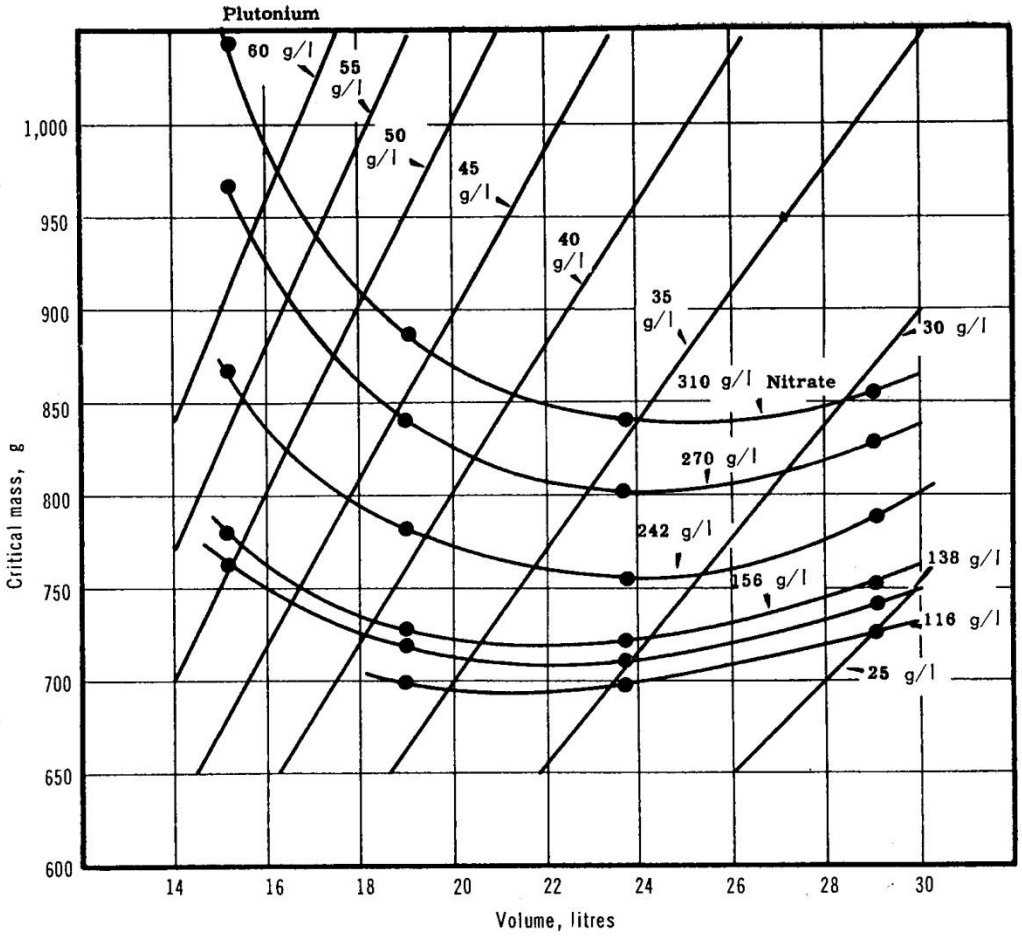

Figure 3. *Critical mass vs. volume water-tamped SS spheres 3.12 % Pu 240, 27C.*

estimate of the critical mass in the intermediate range of moderation between solutions and solid metal.

For unmoderated α phase Pu metal, the minimum water-reflected critical mass is given as 5.6 kg [5].

Criticality of Homogeneous PuO_2-Water System

Criticality parameters for idealized Pu-water mixtures provide the bases for establishing nuclear safety in handling and processing plutonium. However, it is the homogeneous PuO_2 and $Pu(NO_3)_4$ systems which are more frequently encountered. Compounds, such as organic complexes, plutonium polymer and PuO_2, may be formed non-routinely in the aqueous parts of a plutonium nitrate process stream as the result of abnormal operations. For example, plutonium oxide may form if a nitrate solution contacts the wall of a high-temperature evaporator, or the polymer may form if a nitrate solution at the proper pH is diluted or heated [8, 9].

However, information on the density and composition of plutonium polymer is meagre; the density evidently depends greatly on the conditions under which the polymer is formed (i.e., pH, temperature, concentration, etc.). It has been postulated that a density of 1-2 g/cm^3 may be possible, but until there are further laboratory data, the maximum density that might be encountered can only be assumed. The formula for the precipitated polymer has not been firmly established. Experimental work at Hanford indicates that it is of the type $PuO_2 . \times H_2O$ and could probably be $PuO_2(H_2O)_{7^1/_2}$. These 'non-routine' compounds have been found in certain parts of the processes in the past, but there has been insufficient analytical data for positive identification. Identity has therefore been based on the nature of the process and the abnormal conditions encountered.

PuO_2 is a product routinely formed as part of the metal isolation process; the oxide may also be used as a feed material for plutonium metallurgy in fuel element fabrication. Some Pu density-H_2O moderation relationships have been determined by mixing PuO_2 with water [10]. These relationships are shown in Figure 4, together with the H/Pu ratios for Pu-water mixtures. The density of the PuO_2-water mixtures is considerably less than that of the Pu-water mixtures at the smaller H/Pu ratios (for an H/Pu ratio of 4, the density of Pu in the oxide water mixture is $\sim$ 3.5 as compared with $\sim$ 5 for the Pu-water system).

A series of criticality calculations have recently been made for Pu systems using a one-dimensional, multi-group neutron diffusion code [11]. Eighteen energy groups were used in the calculations, which were performed by Dr. W.A. Reardon with the use of the IBM-7090 digital computer [12]. The results of some of these calculations are presented in this and the following Sections. The calculated critical mass for a PuO_2-water system is shown

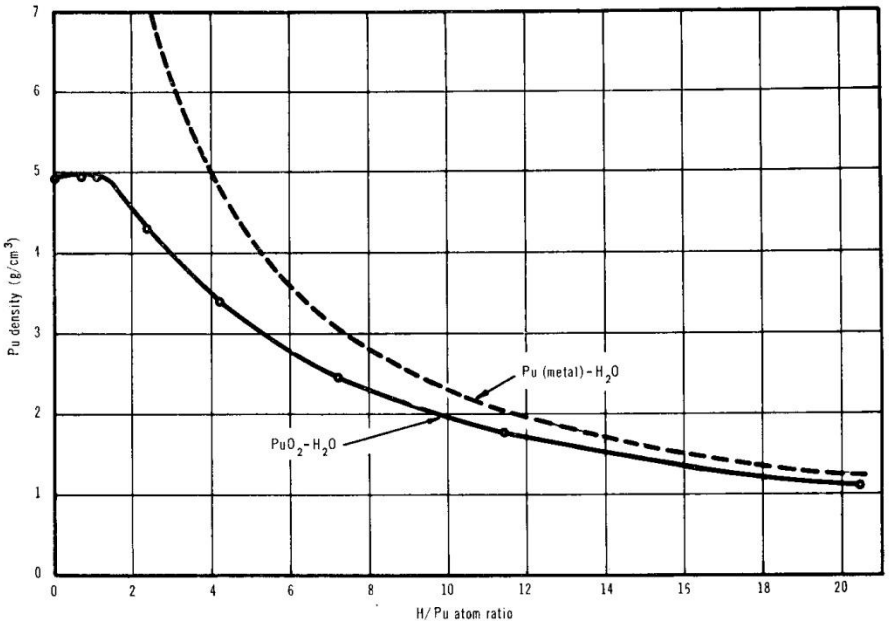

Figure 4. *Plutonium concentration vs. Hydrogen-to-plutonium atom ratio.*

in Figure 5; the density of the PuO_2 for H/Pu = 0 was assumed to have a theoretic value of 11.46 g/cm^3. The critical masses for the measured densities presented in Figure 4 are shown in Figure 6, and in Figure 7 the calculated critical diameters of infinite length cylinders are given for these densities.

The large differences in critical mass between that of a Pu and PuO_2 system is the result of the smaller density for the oxide system at the smaller H/Pu ratios [4]. The effect of density is also quite evident from a comparison of the calculated critical mass for PuO_2 with a theoretic density of 11.46 with that for the PuO_2 with a density of 5.6 at an H/Pu ratio of zero.

It is to be emphasized that the PuO_2 results have not been confirmed by experiments. The calculations were made to indicate the large increase in critical mass which results because of the reduced density of the PuO_2 and to provide improved estimates of the criticality parameters of these systems.

Effect of Pu-240 on Criticality

The Hanford P-11 experiments provided data for the effect of Pu 240 on the critical mass of dilute Pu solutions for isotopic content ranging up to about 4 % Pu 240. These experimental results have been calculated with the 18-group cross section set devised for the 9-Zoom code. Excellent agreement was obtained between the measured and calculated effect of Pu 240 on the critical mass for the 14-in. water-tamped sphere with H/Pu atomic

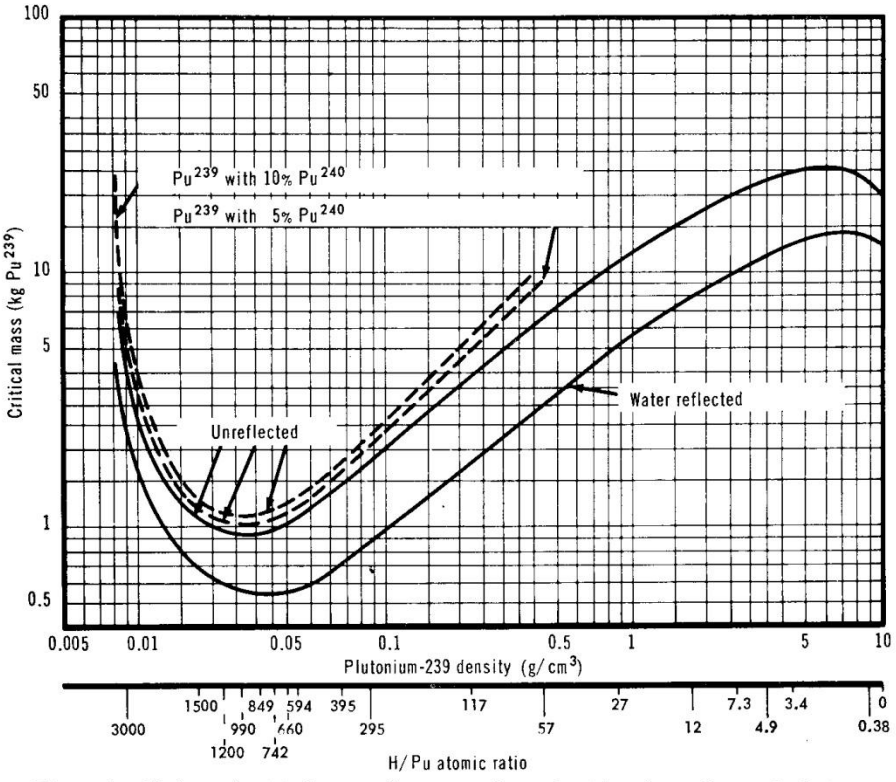

Figure 5. *Estimated critical mass of water-moderated oxide spheres from calculations.*

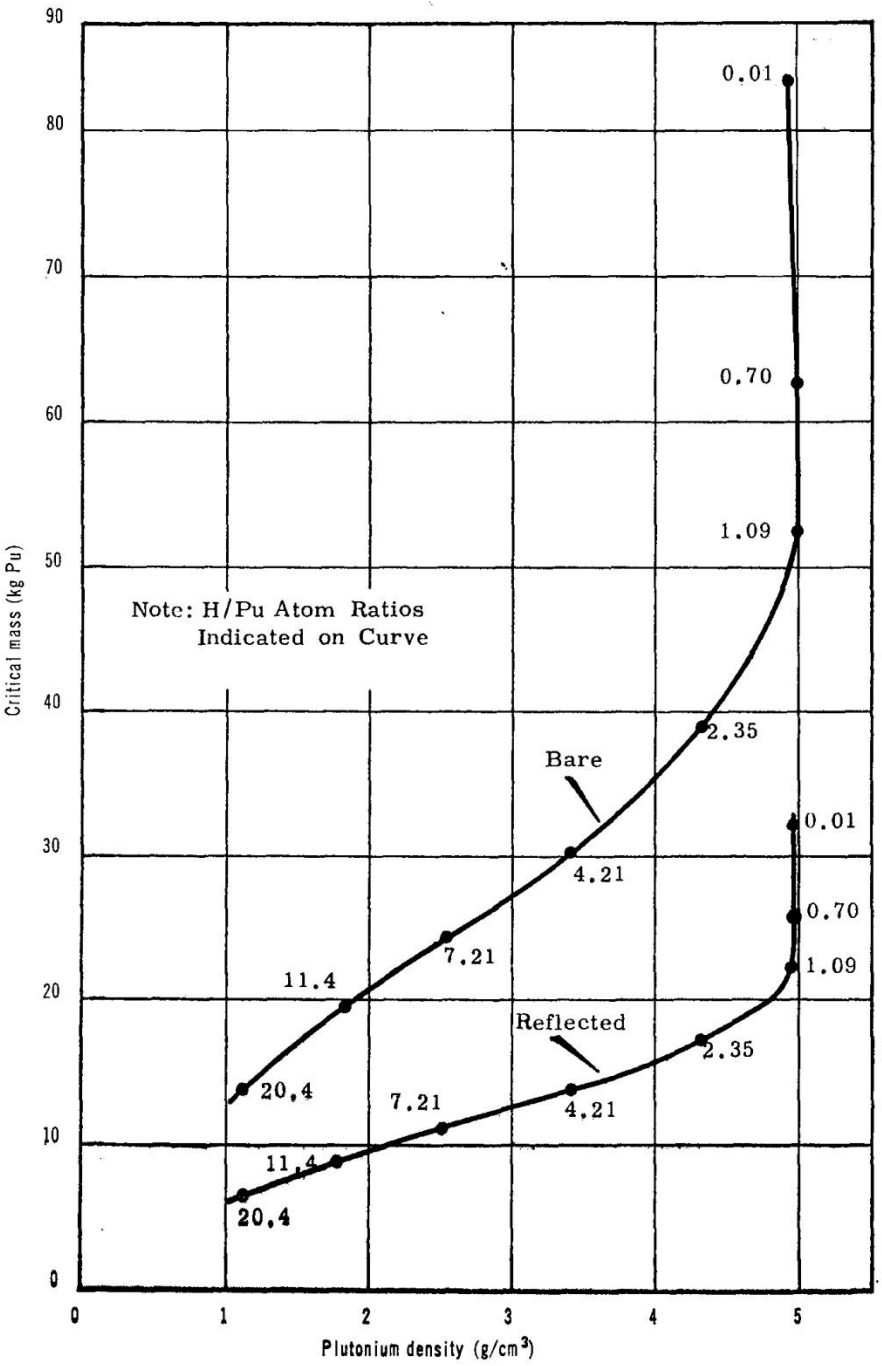

Figure 6. *Estimated critical mass of water-moderated plutonium oxide spheres (based on measured density of PuO_2).*

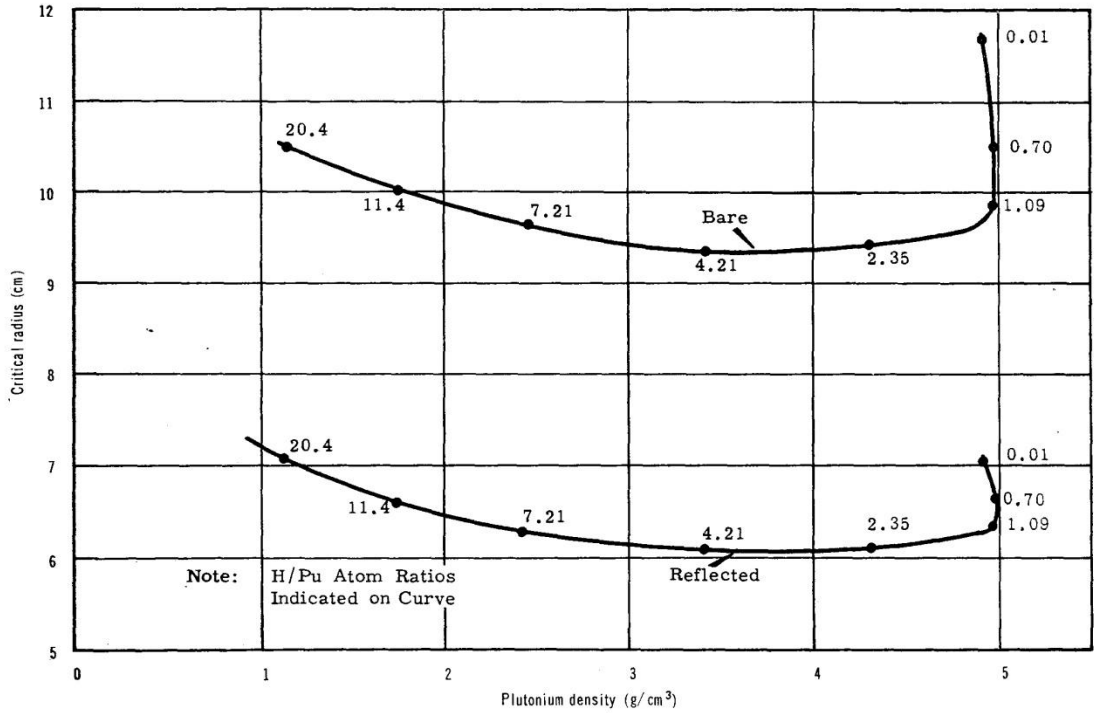

Figure 7. *Estimated critical radius of water-moderated plutonium oxide infinite cylinders (based on measured density of PuO_2).*

ratios in the range of 824-953 [6]. The calculated results for high concentrations, for example H/Pu ratios < 100, are extremely sensitive to the effective resonance absorption cross section for the Pu 240 and are therefore not reliable. The Pu 240 acts as a neutron poison; the calculations indicate the Pu 240 to have the greatest effect on the critical mass for H/Pu ratios of about 20, decreasing for smaller H/Pu ratios. In the case of the Hanford experiments with the 14-in. sphere, the critical concentration of Pu 239 was increased by about 3 % for each per cent Pu 240 present [6].

Measurements made at Los Alamos in fast critical assemblies indicate that the net fission production of Pu 240 is about two-thirds that of Pu 239. It is also reported that the Pu 240 present in Pu 239 metal to less than ~ 10 % increases the critical mass slightly [13].

Experimental data on the effect of Pu 240 will be determined as part of the experimental programme for the new Hanford Plutonium Critical Mass Laboratory (a description of the proposed programme is given later in this paper) [14].

Nuclearly Safe Concentration for Aqueous Plutonium Solution

A quantity of special interest relative to the nuclear safety of solutions is the nuclearly safe concentration limit, i.e., the maximum permissible concentration of fissile material for which $k_\infty \leq$ unity.

Recent measurements have been made of k_∞ for dilute plutonium nitrate solutions in the Hanford Physical Constants Testing Reactor. Some additional confirming experiments are required before the results will be finalized. However, the current data indicate, after corrections are applied for the effects of Pu 240 and nitrate, the limiting concentration to be about 9.3 ± 1 g Pu/litre for a homogeneous Pu 239 water system. This number is about 1-2 g/litre higher than the calculated value. The reasons for the disagreement have not been determined.

Heterogeneous Systems of Pu-Al Fuel Rods in Light Water [15]

A series of critical approach and exponential experiments were conducted at Hanford with Pu-Al alloy fuel elements in light water-moderated lattices. The fuel elements were Al-5 wt % Pu, 0.506-in. dia. and 24 in. long. The work was done in connection with the Plutonium Recycle Programme to provide critical mass data relevant to the processing and handling of this kind of fissile material. The data are of use in connection with nuclear safety specifications in the fabrication of fuel elements and for correlating critical mass calculations with experiments for heterogeneous Pu systems.

The fuel rods were clad in 0.030-in. Zircaloy-2 with 0.020-in. and 0.125-in. thick end caps. This gave a Zr/Pu atomic ratio of 31.92 and an Al/Pu atomic ratio of 168.20. A hexagonal pattern was used for the lattices which were fully water-reflected. The minimum critical mass for the 5 % plutonium-enriched Pu-Al alloy rods in spherical geometry was about 1.5 kg Pu (including Pu 240 and higher isotopes) for an H/Pu ratio of ~ 650. The maximum buckling for these systems was about $11{,}300 \times 10^{-6}$ cm^{-2} for an H/Pu ratio of ~ 450. Data were obtained on the extrapolation lengths for the reflected assemblies by equating the buckling expression for the exponential measurements with the critical buckling as determined from the critical approach measurements.

When the results of the measurements are corrected for the effect of Pu 240, the minimum critical mass is estimated to be about 1.3 kg Pu 239. These data have been used as a basis for extending the theory to calculate mass limits of isolated units of Pu-Al alloy rods in water for several other enrichments and diameters; some of these results are given in the following Table [16, 17].

SAFE MASS LIMITS FOR Pu-Al ALLOY RODS

Enrichment *(Weight % Pu in Alloy)*	Rod Diameter *(in.)*	Safe Mass Limit* *(kg Pu)*
5.0	0.25	0.39
	0.50	0.52
	0.75	0.65
15.0	0.25	0.35
	0.50	0.61
	0.75	1.00

* The safety factor is about 2.3 for double batch allowance.

Plutonium-Aluminium Alloy — Unmoderated System

Allowance factors on mass limits are given in Reference 5 which may be used to determine a 'safe' mass for plutonium when mixed homogeneously with other elements (excluding H, D, Be). The mass limit for Pu-Al alloy is of special interest in connection with the fabrication of Pu-Al fuel elements for the Plutonium Recycle Programme.

To provide better estimates than currently available, the 18-group diffusion code (9-Zoom) was used to calculate the critical mass for Pu-Al alloys [12]. The high energy transport cross sections of plutonium were adjusted to provide approximate agreement with the critical mass of α phase Pu at full density. The results are shown in Figure 8. The calculations are in qualitative agreement with the estimated spherical critical mass for a nominal volume ratio of plutonium to aluminum of 2/1 (Pu density $\sim$ 9.7 g Pu/cm^3) [18].

Plutonium Metal Dissolution

The dissolution of plutonium metal involves a system of one or more pieces of plutonium, which during the course of dissolving are immersed in Pu solution. Such a system is encountered in metal scrap recovery operations.

For the case of plutonium metal pieces in a moderator, it is not possible to give general recommendations concerning nuclear safety, but there are some applicable data which may be used to arrive at answers [4].

A study has also been made, which indicates that a plutonium solution-plutonium metal system might be more reactive than a plutoniom metal-water system. The analysis was based on highly enriched uranium metal immersed in uranium solutions. The analysis indicates that a piece of fissile material, which is subcritical when placed in a solution of nitric acid in which the solution height is safe for the completely dissolved metal, might become

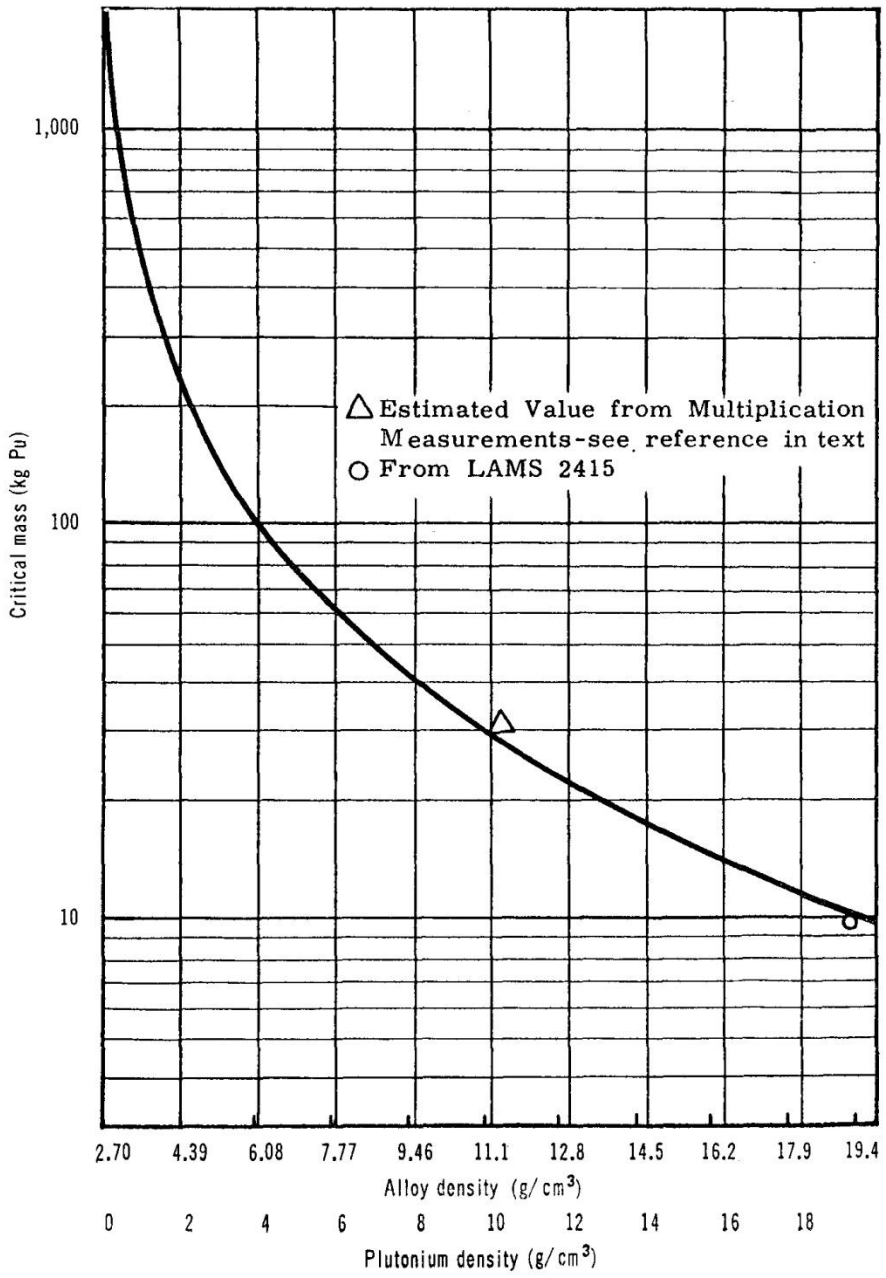

Figure 8. *Estimated critical mass of plutonium aluminium alloy spheres (unreflected).*

critical during the process of dissolution when the metal is partly dissolved and partly in solution [19]. Additional experiments are required to check these conclusions.

Applied Methods of Criticality Control in Plutonium Processing and Fuel Element Fabrication

Some of the methods which have been used for criticality control in plutonium processing and metallurgy operations are discussed below.

Plutonium Solutions

Nuclearly Safe Geometry. The preferred method of criticality control is to design vessels which are safe by virtue of their geometry. Because of the excess neutron leakage, vessels so designed will remain subcritical regardless of the amount of fissile material contained therein. Thus, no further criticality control is required, providing the integrity of the vessels is maintained. In determining the always safe dimensions, allowance must be made for the effects of neutron reflectors which may be proximate, and for possible neutron interaction effects from adjacent vessels which can cause a reduction in the geometrically safe dimensions of an isolated unit.

Safe Batch or Mass Limit. The amount of plutonium that is placed in a container which is not geometrically safe is limited to a safe batch, or mass limit. This amount of plutonium which is less than the minimum critical mass, is determined from analysis and volume or weight measurements. Some examples of batch limits are the following:

a) The amount of Pu is limited to less than one-half (45 %) of the minimum critical mass. (Safe for double batch allowance.)

b) The amount of plutonium contained in the solution is limited to a fraction of the minimum critical mass for the vessel, assuming a uniform water-tamped precipitate layer. (The vessel is then also safe should precipitation occur.)

Limiting Critical Concentration of Plutonium. The limiting critical concentration of plutonium which has been used as a basis for criticality control in aqueous solutions is 7.8 g Pu/litre [5]. For concentrations less than 7.8 g/litre (H/Pu $\geq$ 3,600), excess neutron absorption in the hydrogen prevents criticality even for an infinite system. The maximum plutonium concentration which has been used at Hanford in large vessels for purposes of criticality control is 6 g Pu/litre (23 g/gallon). The degree of safety in using the value of 6 g Pu/litre is larger than implied, since in any finite vessel the critical concentration will be greater than 7.8 g/litre; also the effect of Pu 240, nitrate, etc., would result in a further increase in the minimum critical concentration.

The maintenance of a subcritical concentration is not in itself considered adequate safety against criticality in large vessels. Consideration must also be given to the possibility that plutonium might be precipitated from solution because of operational errors. Limits are, therefore, also established for the total amount of plutonium contained in vessels which are not safe by geometry. Whenever there is a possibility that plutonium precipitation might occur, a mass limit is established for the vessel. This limit is a fraction of the calculated minimum critical mass for the vessel on the basis of a uniform precipitate layer.

In the case of solutions, control is effected through adjustment of the stream ratio to obtain plutonium concentrations corresponding to the storage of no more than an arbitrarily chosen amount (90 %) of the calculated

minimum critical mass when the vessel is filled to the overflow, or the solution transfers are limited so that vessels never contain more than that fraction of the calculated minimum critical mass.

Soluble Poisons. In those cases in which a soluble poison (cadmium) has been used at Hanford, its use has been primarily that of a secondary control to safe plutonium solutions, but not as a primary control. When the poison is used in this manner, criticality will not occur if the poison is removed from the process unless some other operating error were to occur simultaneously, such as double batching or exceeding some pre-set concentration limit.

Plutonium Fabrication Plant

Plutonium Storage. The number of units permitted in a storage array depends on the size of each unit in the array and on the separation between units. Criticality data which may be used as a guide for determining the nuclear safety of arrays is given in Reference 4, and criteria for storage is presented in Reference 5.

Figure 9 is an example of a safe storage system used for Pu metal and unmoderated PuO_2; the centre-to-centre distance between units is ≥ 24 in. This is an interesting method of storage, since two units in adjacent compartments cannot be placed together. The centre-to-centre spacing between units in different compartments (24 in. in this case) is always assured. The maximum unit of Pu per compartment is clearly posted, and detailed records are maintained of all additions and removals from the array.

Casting Operation - Plutonium-Aluminium Alloys. One of the processes used in making Pu-Al alloy is the direct addition of plutonium metal to molten aluminium. With no moderator present, the maximum amount of plutonium which may be alloyed with aluminium in a single batch is 2,600 g. This batch size is required during the alloying process since 100 % plutonium is being alloyed with the aluminium. Safety cannot be based on the alloyed material until after the alloying process is completed.

Cryolite Process. The cryolite process is also used in making the alloy. In this process, PuO_2 is added to the molten aluminium along with cryolite flux. The aluminium reduces the PuO_2 to plutonium metal and the alloying takes place. Larger batch limits would be allowed in this operation, because the PuO_2 has a significantly higher critical mass than the metal.

Core Storage. The cores are stored in wooden boxes. Geometry is given primary consideration in establishing a safe limit for each box and on the number of boxes which may be safely stacked together.

Autoclave. Nuclear safety of the autoclave is based on geometry control. Figure 10 shows a bundle of fuel elements prior to insertion in the autoclave. The diameter of the autoclave is less than the critical diameter for lattices of Pu-Al alloy rods in water for the plutonium enrichments being handled. In this case, the diameter is 6.5 in. (2-7 wt % Pu alloy)*.

* Quite conservative; in view of recent experiments, the 'safe diameter' may now be increased to 9.0 in. with nearly double capacity [15].

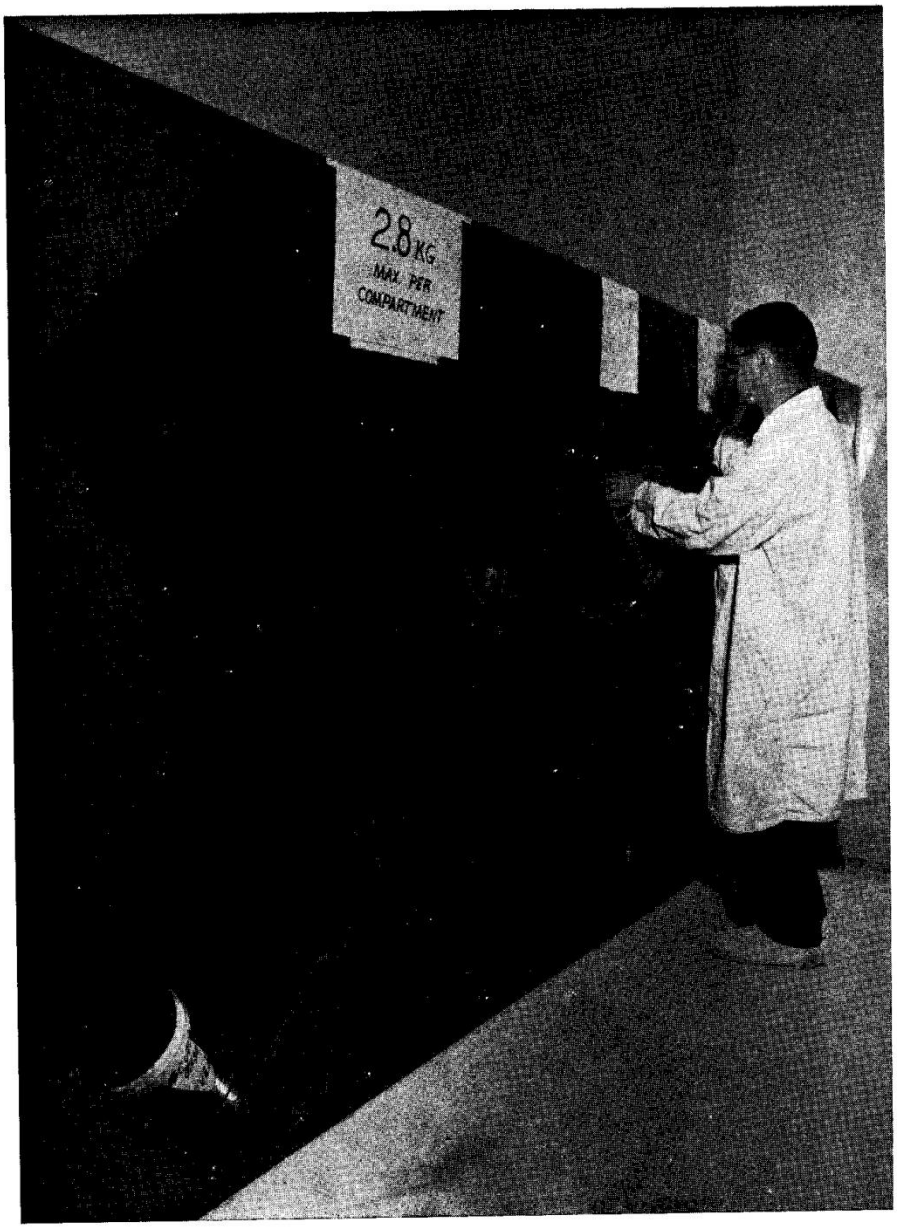

Figure 9. *Safe storage array of fissile material.*

Storage of Completed Fuel Elements. The storage of completed fuel elements is based on geometry control. The fuel elements are stored in a safe slab arrangement, which would also be subcritical in the event of water flooding. Figure 11 shows such an arrangement. Two rows are permitted, although only one is shown in the Figure.

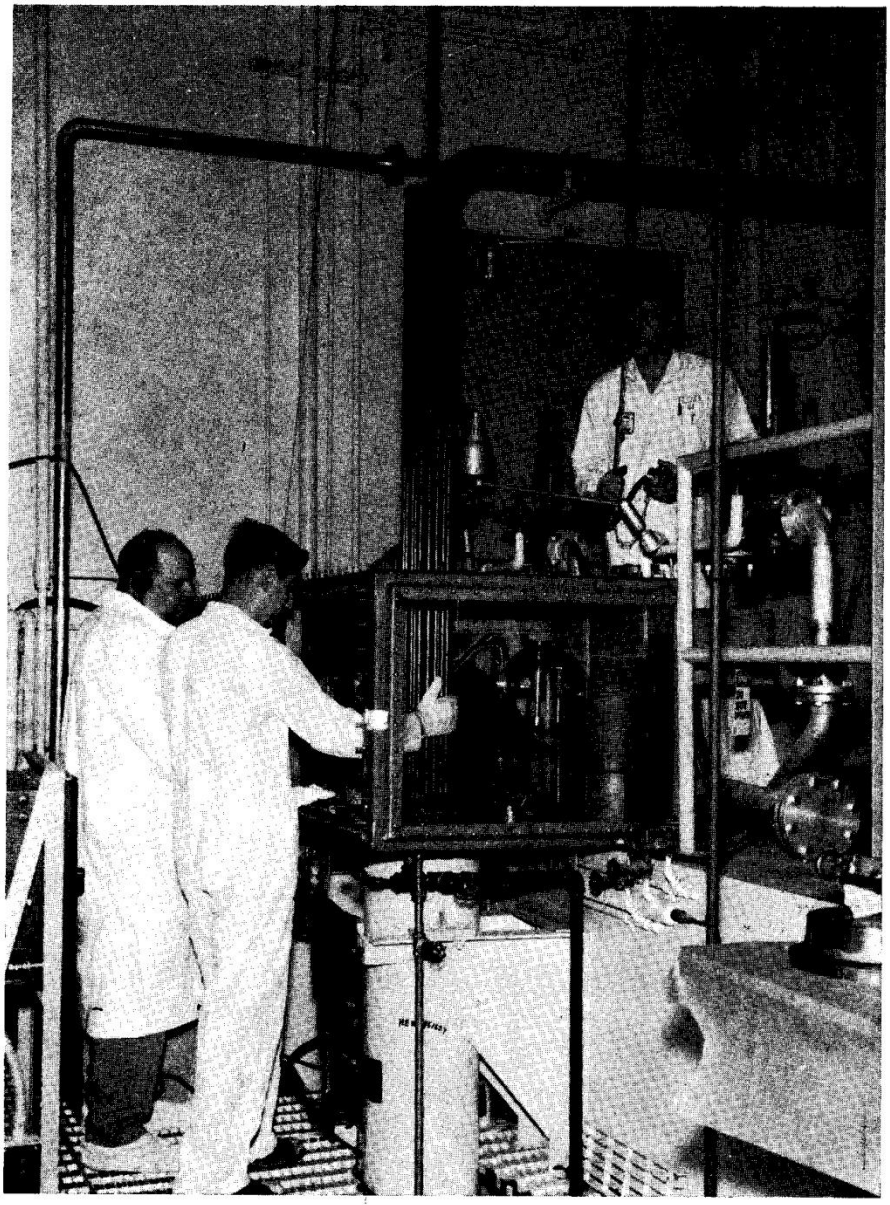

Figure 10. *Fuel tubes being inserted in autoclave which is safe by geometry.*

Hood Limits. Criticality control is effected by setting safe limits on each hood. These limits are usually conservative by more than a factor of two. Further control is afforded by the accountability procedures which require that all plutonium additions and subtractions be noted on the individual hood logs.

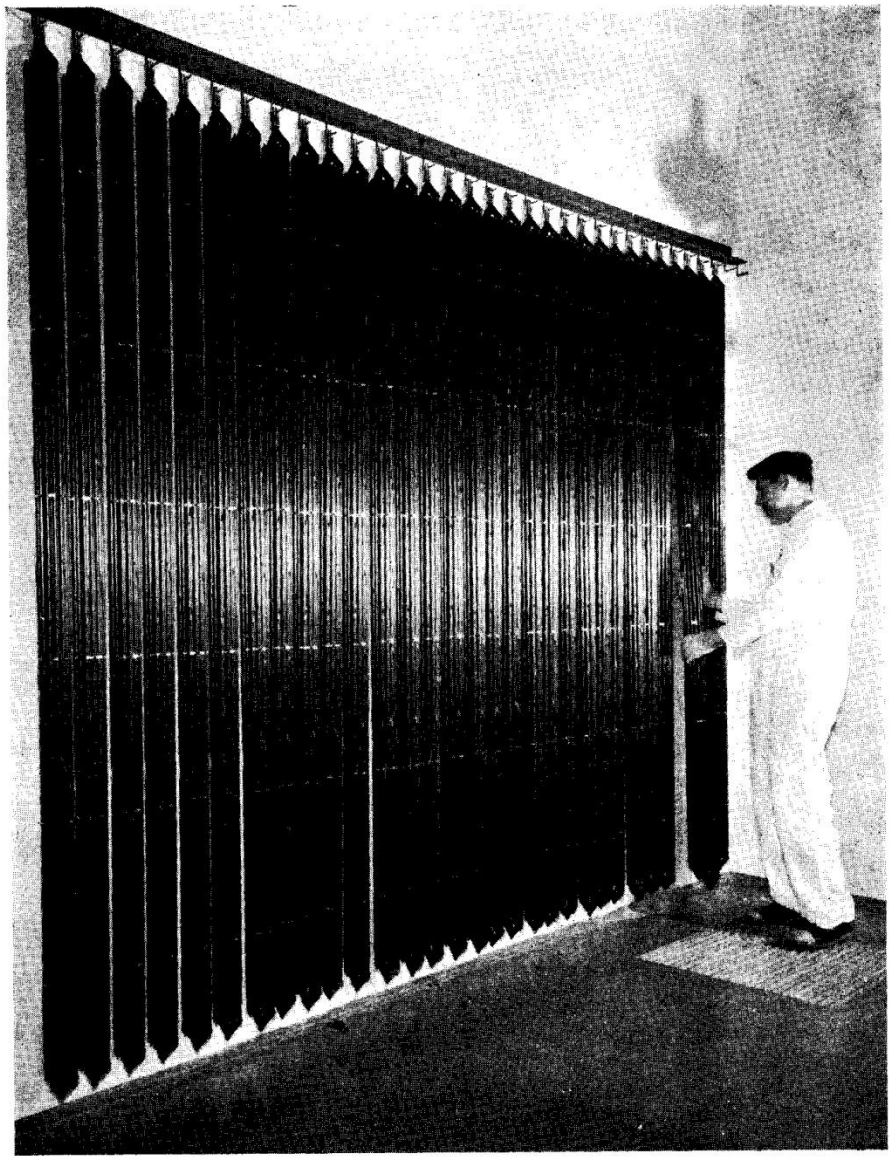

Figure 11. *Method of storing fuel rod clusters in safe slab geometry.*

Some General Types of Criticality Problems Frequently Encountered

A typical list of some general types of problems involving nuclear safety in the chemical and metallurgical processing of plutonium is given below. These problems do not constitute a complete listing, but are representative only of those general types which usually require more than a preliminary examination for solution.

— Pairs or groups of cylinders;
— Arrays of metal parts;
— Arrays of various units (mainly powders and metal);
— Unusual reflectors or reflecting conditions;
— Unusual shapes of solution vessels; unusual spacing and geometries with solid compounds;
— Single units (usually solution cylinders); precipitates or slurries;
— Unusual moderator, or non-homogeneous systems, dissolution of metal;
— 'Safe' ratios or poisons;
— Metal alloys;
— Fuel element fabrication - storage and handling.

ADMINISTRATIVE PROCEDURES

A listing of some typical procedures which have been followed in effecting criticality control are given below:

1. The potential for critical mass incidents is examined at the time designs for new facilities, or for revisions to existing facilities, are developed, and this potential is minimized through appropriate design. Critically safe geometry is the preferred design criterion.
2. In situations where safety cannot be assured by geometry, administrative procedures for control of the process operation within safe limits are established and enforced. Critical mass control under these procedures is based on operational controls such as dilution, neutron poisons, and batch size limits. These procedures are in the form of written specifications for the particular facility under consideration. Adherence to the written specifications is implemented by the incorporation of specific instructions in operating procedures, by the posting of critical mass control limits in appropriate locations and through the training of personnel.
3. When process equipment is altered or is replaced by equipment not identical with the original in design, the effects of the changes on critical mass control safety margins are re-evaluated. If the changes reduce the margins of safety provided by existing control procedures, the latter are revised to re-establish the required margins of safety prior to introducing fissile materials into the equipment.
4. When batch size limits are used as a control method, these limits are such that inadvertent double batching will not result in a critical mass incident.
5. Transfers of unsafe concentrations of fissile materials from geometrically safe vessels to geometrically unsafe vessels are made only in carefully measured, subcritical batches or under controlled conditions of dilution to safe concentrations.
6. At each facility where there is a potential for critical mass incident, there must exist a detection system, equipped with distinctive, audible alarms, which is capable of detecting promptly a radiation burst arising from a critical mass incident. An evacuation procedure is prepared which must be followed if the alarm is sounded or if other warning of a critical incident is received. Personnel are trained to respond in accordance with these procedures.

7. Internal audits will be performed on a continuing basis under the direction of the Operation Manager to assure that administrative controls are adequate and adhered to.
8. A programme for improving the analytical methods and measurements associated with critical mass control is maintained and improvements that are developed will be incorporated promptly into the standard analytical and measurements methods.
9. At each facility where critical mass control is required, training programmes are maintained which keep personnel informed of the general principles on which critical mass control is based and of the operational precepts which must be followed to maintain control.
10. Each facility is required to have procedures for building re-entry following a critical mass incident.
11. When there is doubt that control is within established critical mass specifications, the facility is shut down until control can be assured.

Proposed General Programme of Studies for Hanford Plutonium Critical Mass Laboratory

A new critical mass laboratory has recently been completed at Hanford, Washington. This laboratory was built for the purposes of obtaining needed criticality data on plutonium solutions and precipitates of plutonium in support of chemical processing. The laboratory is fully described in " Hazards Summary Report for the Hanford Plutonium Critical Mass Laboratory " [14].

The programme for the Laboratory is intended to be a general study involving homogeneous and heterogeneous experiments needed for obtaining criticality data on plutonium solutions and precipitates. A combined theoretical and experimental study will be made of ' clean ' cases, i.e., those cases in which a decisive comparison between theory and experiment can be made. With this study as a guide, the more complicated cases often encountered in plant design will be investigated experimentally; the extent to which these types of experiments will be conducted depends on the degree of correlation which can be effected between theory and experiment for the simple geometries.

From a general veiwpoint, the programme will have a several-fold objective:

i) To supply *critical mass data* which will be in direct support of current processes and be of use in the design and operation of new radiochemical equipment;

ii) To obtain a comprehensive set of *critical mass physics data* which will serve as the basic source of information for nuclear safety in the prevention or termination of inadvertent critical nuclear reactions in non-reactor environments;

iii) To provide *basic information on the criticality of plutonium systems* and thereby assist in the development and testing of theory for plutonium reactor systems. As theoretical methods are developed and correlation effected between theory and experiment, an increasing number of plutonium criticality problems can be solved by theory — with significant reductions in the experimental programme.

The initial geometries studied will be spheres and cylinders with eventual extension to slab geometry. A comparison of the critical dimensions for the different geometries will be useful in evaluating extrapolation lengths and for determining if the relationship ('shape factors') between plutonium systems in different simple geometries are consistent with U 235 criticality data which have been obtained by the Oak Ridge National Laboratory group.

The general programme of criticality studies includes the following list of experiments.

Homogeneous Systems

Experiments with plutonium solutions. Plutonium in some form of a water complex is the type of system most frequently encountered. Such systems were studied under the P-11 programme for plutonium concentrations up to 136 g/litre [6]. In view of the current type of separation processes (continuous flow), data are now needed for higher concentrations of plutonium. As the plutonium concentration is increased, the neutron energy spectra become more characteristic of an intermediate reactor system than of a thermal type. Critical mass studies are planned with plutonium solutions and precipitates or hydrogenous mixtures of plutonium with principal emphasis on H/Pu 239 ratios less than 400. The critical dimensions of spheres, cylinders and slabs will be determined and 'shape factors' evaluated for converting from spherical to cylindrical and/or rectangular geometry.

Experiments with precipitates of Pu or facsimiles. Plutonium compounds in the density range 1-4 g/cm^3 and with H/Pu ratios of about 25-5 are encountered in certain operations of the separation plants. Among these compounds are PuO_2, $Pu(C_2O_4)_2$ and $PuO_2(H_2O)_{7\,1/2}$. The latter compound is the plutonium oxide polymer, which forms from plutonium nitrate solution under the proper conditions of acid molarity and temperature. In order to establish the criticality parameters for this plutonium density and H/Pu range, experiments will be made with PuO_2-plastic mixtures or other PuO_2-hydrogenous mixtures. The material will be in the form of solids, or compacts which have been sealed or canned for contamination control. A split table remote assembly machine will be used for these experiments.

Complex geometry. While most of the experiments will be done with geometries for which a decisive comparison between theory and experiment can be made (spheres, cylinders and slabs are examples of such forms), the safe mass limits will be determined for 'one-of-a-kind' odd-shaped vessels which exist in separation processes, for which only crude approximations can be made. These experiments will be done as required to evaluate the nuclear safety of the processes involved.

Interaction experiments. Where more than a single subcritical unit of fissile material is handled, there is always the probability of interaction between units. It must be determined in all instances that there is no criticality hazard. Theoretical calculations do not give reliable answers in many such problems, and thus experiments must be performed.

It is expected that 'interaction' experiments will be conducted for arrays of cylinders and for interacting parallel slabs. In these experiments, work will be done with solutions and precipitates of plutonium with bare and partially-reflected units.

Neutron reflectors. The effect of a given reflector on the critical mass of a system will differ depending on the median energy of neutrons in the system and on the position of the reflector relative to others which may be present. As an example, a thin stainless steel wall of a reactor vessel will reduce the effectiveness of a water reflector, whereas a thick layer of stainless steel may be equivalent to, or even better than, a water reflector [20]. For such reasons, many of the critical mass determinations will be of reflected systems. Of interest are the effects on the criticality of various reflectors, such as concrete, water, polyethyelene, stainless steel, etc., as well as experiments to determine the effective reflector savings for an ' equivalent man '.

Effect of density on criticality. Critical mass experiments will be conducted in selected reactor assemblies to determine the influence of core density change on the critical mass of near-equilateral cylinders with hydrogenous reflector. The critical mass data are of importance in several respects; for application in resin columns, and for application of interaction criterion based on density formulation.

Limiting concentration for aqueous Pu solution and check on value of η. An experiment is considered to evaluate the limiting concentration of Pu in an aqueous solution from critical experiments. The limiting concentration is the smallest concentration which can be made critical in an ' infinite ' system. The value of η will also be determined from the experiment. A large sphere (somewhat smaller than 42 in. dia.) is being considered for the reactor vessel in this experiment.

Effect of higher isotopes of plutonium, Pu 240, Pu 241 on criticality. In dilute aqueous plutonium solutions, Pu 240 acts as a neutron poison; the effect of Pu 240 on the critical mass will vary with plutonium concentration (H/Pu ratio) through dependence upon the neutron energy spectra. In predominantly fast systems, Pu 240 assumes the rôle of a fissile isotope. On the other hand, high exposure plutonium, such as that obtained from power reactor fuels, will also contain non-negligible amounts of the highly fissile Pu 241 isotope in addition to the Pu 240. Experiments will be conducted with both dilute and concentrated solutions containing these isotopes.

Temperature coefficients. Since plutonium has a large fission resonance cross section adjacent to the thermal region, the temperature coefficient for a plutonium system will differ from that of uranium. It is desirable to know this coefficient for evaluating the nuclear safety of those processes where elevated temperatures are encountered.

Soluble poisons. Safety by geometry and batch limits are the most common safety principles observed in the processing of fissile materials. Control through the use of soluble poisons has also been considered, but dropped in some instances for lack of information. Thus, information on the effect of soluble poisons on the criticality of various systems will be developed. The poisonous effect of the standard components in certain systems, such as the nitrogen in plutonium nitrate solutions, will also be studied.

Resonance detector and cadmium ratio measurements. In connection with the above reactor assemblies, resonance detector and cadmium ratio measurements will be performed to gain further insight into the rôle which epithermal absorption plays on the criticality of plutonium systems. A

more detailed knowledge of neutron spectra is desired, since proper values of group-averaged, spectrum-weighted cross sections are not available for calculational schemes.

Reactivity coefficient measurements. The effect upon the reactivity of the system of substituting various materials for a void or a fuel sample will yield a ' reactivity coefficient '. These measurements will be conducted in selected reactor assemblies; the results will be of use to future users of the data, both as known perturbations on the basic systems and as check-points in theoretical treatment of the reactor assemblies. The data will also be of value for evaluating the neutron worth of fixed poisons for use in criticality control.

Measurement of prompt neutron lifetime and k_{off} from pulsed neutron source experiments and from pile noise analyser. The prompt neutron lifetime is an important parameter in the study of reactor kinetics. Measurements will be made of the neutron lifetime in selected reactor assemblies. Consideration will be given to the feasibility of using pulsed neutron source techniques for determining effective multiplication constants of subcritical units.

Minimal critical mass for non-uniform fuel distribution. The minimal critical mass does not occur for a homogeneous uniform concentration of fuel, but rather for that distribution which results in a ' flat flux '. Theoretical predictions indicate a reduction of as much as 30 % over a uniform fuel distribution [21]. Several experiments are planned to check these results for plutonium systems.

Heterogeneous Systems

Plutonium metal-plutonium solution system; plutonium alloy fuel elements in water lattices. The dissolution of plutonium metal in recovery operations, and the manufacture of plutonium-alloy fuel elements for the Plutonium Recycle Test Reactor are examples of a need for critical mass information pertaining to moderated heterogeneous plutonium systems. As with homogeneous systems, the effect on the critical mass of the H/Pu ratio, plutonium isotopic content, etc., will be studied. Plutonium dissolution is of special interest since in this case the plutonium metal (scrap) will exist in a plutonium nitrate solution being moderated and reflected by this solution.

Plutonium-enriched uranium systems. It is anticipated that in connection with the reprocessing of non-production fuels from power reactors and the Plutonium Recycle Programme, a need will arise for critical mass data on heterogeneous and homogeneous systems composed of natural or depleted uranium which has been enriched in Pu; the necessary critical mass experiments will be performed as required.

Initial Experiments for the Laboratory

Current plans for the initial criticality experiments in the Plutonium Critical Mass Laboratory are given below. These plans, similar to the plans of any research and development proposal, are subject to change in the light of new information which will undoubtedly develop as the programme progresses.

The following vessels have been chosen for use in the initial series of criticality experiments with plutonium nitrate solutions:

	Diameter *(in.)*
Cylinders (bare)....	11, 12, and 13
Spheres (bare and reflected)	12, 13, and 14

The first criticality experiments will be conducted with the spherical vessels. Types of reflectors considered for the first spherical assembly are water and concrete with and without Cd sheet interposed between the reflector and the criticality vessel. These measurements will be used as a normalizing point to the P-11 experiments and also to obtain needed data for the further development of theoretical methods of critical mass calculations [6].

Following these measurements, a series of experiments will be conducted with cylinders (bare); the experiments will then be extended to include fully reflected cylindrical vessels. Additional spherical, and cylindrical, reactor vessels will be fabricated; the dimensions of these vessels will depend on the results of the initial measurements.

Experiments with Precipitates of Plutonium or Facsimiles

A split half machine is currently being designed to facilitate criticality studies with plutonium-oxide hydrogen-moderated mixtures which, because of the H/Pu atomic ratio, are not available in liquid form.

Criticality experiments will be conducted with PuO_2-plastic mixtures or other similar materials [22]; small blocks of various dimensions up to two inches on a side, which have been adequately sealed or jacketed for contamination control, will be used. The first experiments will consist of bare and reflected rectangular parallelepipeds (cubical units). The H/Pu atomic ratio of the material for the initial experiments is tentatively 15, with a nominal plutonium density of 1.8 g/cm^3. A series of experiments will be conducted which involve different H/Pu ratios.

The experiments will subsequently be extended to include cylindrical and slab geometry; in the case of cylindrical geometry, the critical assembly will be built up from thin discs or pancakes of plutonium plastic mixtures; both bare and reflected assemblies will be studied.

ACKNOWLEDGEMENTS

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REFERENCES

1. O.Z. Wick and I.D. Thomas. Private communication.
2. W.A. Bailey, " Fabrication of Aluminium-Pu Alloy Fuel Elements by Co-extrusion ", *HW-63151* (1959).

3. J.B. BURNHAM, " Facilities at Hanford for the Manufacture of Plutonium Ceramic Fuels ". Unpublished.
4. H.C. PAXTON, " Critical Data for Nuclear Safety Guidance ", *LAMS-2415* (1960).
5. F.E. WOLTZ *et al.*, " Nuclear Safety Guide ", *TID-7016 (Rev. 1)* (1961).
6. F.E. KRUESI, J.O. ERKMAN and D.D. LANNING, " Critical Mass Studies of Plutonium Solutions ", *HW-24514 (DEL)* (May 1952).
7. G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Plutonium Plexiglas Assemblies, Part II ", *RFP-190* (July 1960).
8. R.A. PUGH, " Notes Pertaining to Recuplex Product Evaporation ", *HW-32100* (April 1954).
9. A. BRUNSTAD, " Polymerization and Precipitation of Plutonium IV in Nitric Acid ", *HW-54203* (Dec. 1957).
10. H.W. CROCKER. Private Communication (October 1960).
11. E.P. STONE, E.T. COLLINS and S.R. LENIHAM, " 9-Zoom... A One-dimensional, Multigroup, Neutron Diffusion Theory Reactor Code for the IBM-709 ", *UCRL-5682* (Aug. 1959).
12. W.A. REARDON, Critical Mass Physics, Private Communication.
13. G.A. GRAVES and H.C. PAXTON, " Critical Masses of Oralloy Assemblies ", *Nucleonics 15*, No. 6, 90 (June 1957).
14. W. A. REARDON *et al.*, " Hazards Summary Report ", *HW-66266* (Aug. 1960).
15. V.I. NEELEY, R.C. LLOYD and E.D. CLAYTON, "Neutron Multiplication Measurements with Pu-Al Alloy Rods in Light Water " in *Trans. Am. Nucl. Soc., 3.* No. 2, 292 (June 1960).
16. N. KETZLACK, " Plutonium-Aluminium Alloy Criticality ", *HW-55173* (Feb. 1959).
17. C.L. BROWN. Private Communication.
18. D.P. WOOD, C.C. BYERS and C.L. OSBORN, " Critical Masses of Cylinders of Plutonium Diluted with Other Metals ", *Nucl. Sci. Eng. 8*, 578 (1960).
19. N. KETZLACK, " Nuclear Safety in Plutonium Metal Dissolution ", *HW-55707* (April 1958), (Class.).
20. A.D. CALLIHAN, J.W. MORFITT and J.T. THOMAS, " Small Thermal Homogeneous Critical Assemblies " in *Proc. First U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva*, 1955, *5*, 145.
21. G. GOERTZEL, " Minimum Critical Mass and Flat Flux ", *J. Nucl. Energy, 2*, 193 (1956).
22. H.W. CROCKER, " Plutonium Oxide-Plastic Mixtures for Critical Mass Physics ", *HW-65207* (May 1960).

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DISCUSSION

R. BAZIRE, France:

Dans le stockage du plutonium métal, à géométrie sûre, il semble nécessaire de prévoir à la fois la possibilité de mettre deux unités de 2,8 kg dans le même casier, et la possibilité pour l'opérateur portant une unité importante de passer à proximité de l'ensemble. Envisagez-vous d'éliminer ces possibilités par des interdictions matérielles ou simplement par la discipline du personnel ?

E.D. CLAYTON, United States:

The limit of 2.8 kg, of course, is less than half the critical mass fully-reflected, so that if an operator were inadvertently to put another unit, it would still be safe.

R. BAZIRE:

Disposez-vous, dans ce cas, d'un double contrôle ? Les opérateurs agissent-ils à deux ou le personnel d'exploitation est-il seul à participer au stockage?

E.D. CLAYTON:

Only operating personnel are permitted in the storage vault which is always locked except for entry, and one man (or alternate in his absence) will have responsibility for maintaining the storage array.

D. BRETON, France :

M. Clayton pourrait-il nous décrire le rôle du réacteur P.C.T.R. dans le nouveau programme qu'il envisage ?

E.D. CLAYTON:

Actually, the Physical Constants Testing Reactor (P.C.T.R.) will play only a minor rôle as far as our critical mass programme for plutonium is concerned. We will do some experiments in the P.C.T.R. with plutonium solutions, but with the P.C.T.R. it is the infinite multiplication constant which we measure, and we cannot get critical masses or critical sizes from measurements in the P.C.T.R. However, we shall do some experiments with the P.C.T.R., in which we insert samples containing the solutions, and then measure k_∞, or the multiplication constants of these solutions. We will then use these data in connection with our critical mass experiments in the Hanford Plutonium Critical Mass Laboratory. But we do not intend to make very much use of the P.C.T.R. for the plutonium work.

J.B. WALFORD, United Kingdom:

You have mentioned the three main types of experimental study which you will be undertaking. Would you say that in the forthcoming programme the greater part of this work will be concerned with slurries, actual or similated, or with solutions?

E.D. CLAYTON:

The greater part of our work with plutonium will be for slurries and similated precipitates of plutonium, because it is in this region where the data is most needed. We will, however, do a considerable number of experiments with solutions. For example, the experiments to determine the effect of various reflectors on criticality and the effect of Pu 240 will be done with both solutions and precipitates — but by and large the biggest emphasis of the programme will be on precipitates of plutonium. I might just mention that it is a rather simple procedure for someone to actually do an *in situ* multiplication measurement in a plant. For example, if one wishes only to know that a given process is safe or that it will not be critical, it is possible to set up counters, and then stack up plutonium or uranium and measure the neutron flux, to determine that, say, 7 kg in the geometry in which you are interested is not critical — and if you are only going to handle some 4 kg, this is sufficient. You may not get a good estimate of the critical mass, but you can handle many plant problems in this way. However, I would

like to caution people about undertaking experiments of this kind, unless they are experienced or have someone with them during the initial phases of the experiments who is fully knowledgable in multiplication measurements and in critical mass physics. The experiments should not be undertaken by anyone who does not know anything about them. This might be an engineer, it could be a physicist, it could be a chemist, it could be anyone, but, if he has no experience in the field, he should not undertake the measurements without having at least someone present who is knowledgable in the field — because the experiment can be hazardous.

Session VI

STORAGE AND TRANSPORT PROBLEMS

PROBLÈMES DE STOCKAGE ET DE TRANSPORT

Chairman - Président :

Mr. A. LARSSON
(Sweden)

METHODS OF CALCULATION FOR INTERACTING ARRAYS, WITH SPECIAL REFERENCE TO STORAGE PROBLEMS

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INTRODUCTION

The interaction between fissile bodies in an array can make them a critical system, even though every body of the system is well subcritical when regarded as an isolated body. This simple fact creates problems of considerable importance when the design and operation are considered of safe and economic plants processing and fabricating fissile material in all but the smallest quantities. Similar problems arise in the transport and storage of fissile material.

The Practical Problems

The practical problems that arise may be classified broadly into two categories:

a) *Well-defined arrays.* In such arrays the composition, density, size, shape and position of the fissile bodies is well known, and the distribution and composition of reflecting and moderating materials can be defined fairly precisely. Examples of well-defined arrays are:

i) Chemical plant, for which the individual units are safe by shape, processing (say) solutions of known composition behind a biological shield, and under closely controlled conditions;
ii) Transport containers which consist of specially designed pockets in blocks of moderating and reflecting material;
iii) A few fabrication shops in which there is a very stringent control of both man and material.

b) *Ill-defined arrays.* In such arrays the composition, density, size, shape and position of the fissile bodies cannot be defined very precisely, and the distribution and position of the reflecting and moderating materials may be variable. Examples of ill-defined arrays are:

i) Open plants such as a gaseous diffusion plant where it is possible for portable plant, personnel and some factory traffic to be in the vicinity of vessels processing fissile material;
ii) Recovery plants processing material such as scrapped crucibles for which the assay of fissile content is only approximate.

There are several methods of finding practical solutions to the first kind of interaction problems, but there are not many which specifically deal with ill-defined problems. A practical solution should err on the side of safety, but not unnecessarily so. At the same time one can hardly call a solution a practical one, if in calculating with great accuracy the safe pitch for units in an array one consumes more money with computing machine time than would be required for the design with a generous pitch for the units which was obviously safe. Thus, in a practical treatment of interaction problems a number of approximations have to be made. This fact and the convenience of some methods in particular circumstances has led to a variety of methods.

Principal Physical Factors

Before reviewing interaction methods, it is worthwhile considering separately the principal factors in any interaction problem. These factors may not be mentioned explicitly in some methods, but they must either be treated implicitly or be negligible for the range of problems considered, for a correct treatment of the problem to be given. An array is regarded usually as a number of separate units with surface sources which are due to interaction with other units. This interaction can take place through air, through intervening material and by reflection. The solution of the general interaction problem requires:

a) An adequate treatment of the neutron economy of each unit for the likely spatial distribution and energy spectrum of the incident neutrons. For a given input the spatial distribution and energy spectrum of the output are required.

b) A method for transforming the output of a unit into the input for any unit. (Note that a fraction of the output of a unit may be transformed by reflection into part of the input to that unit.)

In obtaining transformations of outputs into inputs one has to consider:

a) Reflection back into the originating unit;
b) Coupling of one unit into another by reflection;
c) Direct transmission through air;
d) Transmission by penetration through intervening material, e.g. by diffusion or tunnelling of neutrons;
e) The screening of one unit by another.

During the transmission of neutrons from one unit to another there will in general be a change in the energy spectrum of the neutrons. This change can be neglected in many cases if the outputs of the units are mainly thermal neutrons, for the spectrum is often changed insignificantly by transmission. An exception, for example, occurs if almost thermal units are separated by cadmium sheets; then the interaction is by the minority of neutrons above the cadmium threshold. A case in which the effect of transmission on spectra can be neglected is in a birdcage store of lumps of U 235 set well above any possible level for flooding and remote from walls, etc. Neglecting storekeepers, there is no means by which the spectrum of the transmitted neutrons can be changed. In some applications the neutron spectra for the units in an array are for practical purposes identical, and the amount of moderating material, if any, between the units has a negligible effect on the energy spectrum of the transmitted neutrons. In such cases

the interaction calculations are considerably simplified. In the general case, which can occur frequently in certain types of plant, the neutron spectra for the various units are different, and moderating material between the units modifies the spectra of the transmitted neutrons. In some cases the presence of personnel and portable equipment can make the amount of moderating and reflecting material variable in amount and distribution. A method of solving such an ill-defined problem can, however, be given.

Choice of Method

The best advice that can be given is probably to use (at least) two methods, one to obtain a safe specification for an array, and another to check it. This cautionary note is given mainly for the very simple reason that interaction calculations can be fairly complicated, if full account is taken of the diversity of plant vessels and their contents, and a slip is easily made. The second calculation, if by an independent method, is in our opinion a desirable precaution to take in making safety assessments. The bulk of the interaction problems in the United Kingdom Atomic Energy Authority arises on the industrial side, in the Development and Engineering and the Production groups. The Central Technical Service advising the designers of the Development and Engineering Group prefers its unified theory of interaction for accurate work. Check calculations are made by the Safety Branch, an independent organisation, which uses the interaction parameter method devised by the Atomic Weapons Research Establishment, Aldermaston. At Aldermaston the interaction parameter method is used mainly with checks by detailed calculations on idealized systems. In some respects these detailed calculations are similar to those of the unified theory. The need also arises, before a detailed assessment is made, for approximate answers. The accumulated results of experiment and of interaction calculations made by the unified and interaction methods we fined a great help, but it is also instructive to look at interaction problems by other methods. An interesting empirical treatment of interaction is given in what have been called in the United Kingdom, Kenyon's rules. A somewhat similar approach, which has received considerable attention in the United States, is contained in the density analogue treatment of interaction problems. Another useful approach, which has grown out of experiments, is furnished by the Oak Ridge interaction method.

In the sequel we sketch the methods in turn in sufficient detail to enable them to be readily applied and for a selection to be made. To assist in making the choice we give some views of originators and commentators on the various methods. Finally, we indicate some fields for further interaction experiments and theoretical work.

All methods for interaction problems have to make approximations in calculating the criticality of an assembly, but in making the approximations the following points cannot be too heavily stressed:

a) The approximations should err on the side of safety;

b) In calculating safe spacings for units of an assembly, even major simplifications of a conservative kind for the neutronics of the array are permissible provided the calculated safe spacings are economic and convenient from *other* engineering considerations, e.g. inspection and maintenance purposes.

A Unified Theory of Interaction

This theory [1, 2], as presented, applies to any regular or irregular array comprised of any number of two distinct kinds of units set in either a well-defined or an ill-defined distribution of reflecting and moderating material. The extension of the theory to any array of several distinct units is straightforward, but lengthy, and so it is not given here. In the simplest form of the theory it is assumed that the following conditions are satisfied:

a) The neutrons in the fissile material satisfy the equations of a two-group diffusion theory or the equations of a one-group diffusion theory with albedo type boundary conditions [3].

b) Neutrons in the reflector which are below a certain energy level diffuse according to the equations of a two-group diffusion theory. Neutrons above this energy level either diffuse or tunnel through the reflecting material.

The two-group analysis is used for thermal units and the one-group analysis is used for fast units. More groups can be used [4] (e.g. four groups for thermal units and reflector, two groups with albedo boundary conditions for fast units). In order that one analysis can treat units having different numbers of energy groups and deal also with reflecting and moderating material which can be either well- or ill-defined, the artifice of a ' magic ' shell is introduced. The shell takes the form of a hypothetical thin layer on each unit, and has the property that the diffusion equations for all the units have the same mathematical form, whatsoever the neutron economies of the units and the intervening material.

For any neutron leaving a core there are two possibilities; the neutron either enters or misses reflecting material. Let the probabilities of entering and missing reflecting material be p and q_m respectively. For those neutrons which enter reflecting material we consider three further possibilities. Firstly, neutrons may undergo sufficient collisions with nuclei that their energy is rapidly reduced to levels for which they migrate by diffusion according to condition (*b*) above. Let us assign a probability p_d to this possibility. The other two possibilities are that either the neutrons undergo no collisions at all (i.e. they tunnel straight through the reflector) or they undergo so few collisions that they can merely be regarded as undergoing refraction without appreciable loss of energy. These possibilities apply particularly to fast neutrons. Let us assign a probability q_t to the combination of these two possibilities. Since in the sequel we treat direct interactions and interactions by tunnelling and grazing together, we introduce the symbol $q = q_t + q_m$ for the sum of the probabilities of these events. In any general case it would obviously be extremely difficult to calculate p_d, q_m and q_t since all three probabilities depend on the type, location and thickness of reflecting material. For a fully reflected system p would be equal to unity and here the interaction would be by tunnelling and grazing. For a bare system q_m would be unity, and p zero. However, if personnel can approach any unit of the system the situation would change, and p would contribute to the criticality of the system.

The examples show that p_d, q_m and q_t are not in general fixed but vary with operating conditions. Although $p_d + q_t = p$ and $p + q_m = 1$ for any physical system, i.e. $p_d + q = 1$, it is possible to calculate p_d, q_m and q_t for only a few systems. A safe thing to do when the reflecting material is randomly distributed is to replace the actual system by an idealized one in which $p = q = 1$.

This idealization is not as restrictive as might seem at first sight because the neutrons are at most doubled effectively for a given body only in the solid angles subtended by the bodies interacting with the given body. However, when the distribution of the reflecting material is well defined, e.g. in the case of a pair of spheres immersed in water, one can make more realistic assumptions for p_d, p_m and q_t.

Assumptions

In developing the theory, the following assumptions are made:

a) Identical units have the same neutron spectrum (this is true for an infinite regular array but not necessarily true for a finite irregular array).

b) For the spacings which occur between units in practice, the interaction is the same as that due to taking all the neutrons emerging from a core as being emitted from the centre of the core with a uniform angular distribution (this assumption is common to all currently developed theories).

c) Neutrons emerging from a core which hit reflecting material, partly or wholly surrounding the core, diffuse as if in an infinite medium.

d) All currents incident on a unit, whether they arise through diffusion, tunnelling, or direct interaction of neutrons, are assumed to be uniformly spread over the reception unit. This assumption depends on two subsidiary ones: that the surface multiplication of a unit is independent of the distribution of incident neutrons (this has been proved for one- and two-group diffusion theory [5]); that the outgoing current of neutrons from a unit becomes more uniform as the surface multiplication of a unit increases.

e) Each neutron about to emerge from a core becomes $p + q$ neutrons on emerging; in particular in safety problems involving random distribution of reflecting material, p and q are given the value of 1, and this implies that the number of neutrons leaving a core is doubled. For well-defined reflectors, p and q are calculated (conservatively) for each problem.

The 'Magic' Shell for One and Two Groups

The 'magic' shell is introduced at the boundary of each core (see Figure 1). This shell is assumed to possess the following properties:

a) It is transparent to fast neutrons.

b) It absorbs a fraction ω of an incident beam of thermal neutrons. For each thermal neutron absorbed, 2ρ fast neutrons are produced, ρ inwards and ρ outwards. This property is introduced in order to cover cores containing only fast neutrons, in which case incident thermal neutrons are effectively absorbed at the surface.

c) At the outer boundary of the shell, each emergent neutron multiplies. For each emerging fast neutron, p_1 enter reflecting material and q_1 neutrons are available for interaction. For each emerging thermal neutron, p_2 enter reflectors and q_2 are available for interaction.

By means of this device, using appropriate values for ω and ρ, the one analysis can treat the following cases:

i) Both types of core contain fast and thermal neutrons;
ii) One type of core contains fast and thermal neutrons, the other contains only fast neutrons;
iii) Both types of core contain only fast neutrons.

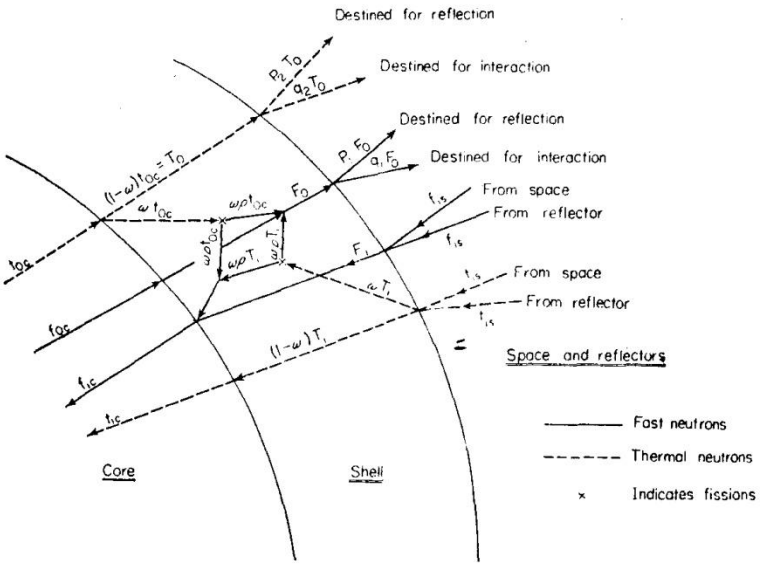


Figure 1. *Sequence of neutron events in the magic shell.*

The sequence of neutron events in the shell is shown diagrammatically in Figure 1. Now consider the balance between the shell currents, F and T, at the outer boundary of the shell, and the core currents, f and t:

$$\left.\begin{aligned} \mathrm{F}_o &= f_{oc} + \omega\rho(\mathrm{T}_i + t_{oc}) \\ \mathrm{T}_o &= (1-\omega)t_{oc} \\ f_{ic} &= \mathrm{F}_i + \omega\rho(\mathrm{T}_i + t_{oc}) \\ t_{ic} &= (1-\omega)\mathrm{T}_i \end{aligned}\right\} \quad \text{boundary conditions} \tag{1}$$

When $\omega = 1$ the core contains only fast neutrons and it is plausible to conclude that $t_{oc} = 0$. This conclusion is justified later.

With the magic shell in place, a generalized form is taken of the two-group equations in the core. These are:

$$\{(1-\omega)\mathrm{D}_{1c} + \omega\mathrm{D}'_{1c}\{\nabla^2\Phi_{1c} - \{(1-\omega)\Sigma_{1c} - \omega\lambda^2\{\Phi_{1c} + (1-\omega)k\Sigma_{2c}\Phi_{2c} = 0 \tag{2}$$

$$\mathrm{D}_{2c}\Delta^2\Phi_{2c} - (1-\omega)\Sigma_{2c}\Phi_{2c} + (1-\omega)\Sigma_{1c}\Phi_{1c} = 0 \tag{3}$$

where $\omega = 0$ or 1.

Solutions are assumed of these equations of the form:

$$\nabla^2\Phi_{1c} + \mathrm{B}^2\Phi_{1c} = 0 \tag{4}$$

$$\nabla^2\Phi_{2c} + \mathrm{B}^2\Phi_{2c} = 0 \tag{5}$$

Substituting equations (4) and (5) in (2) and (3) a quadratic equation is found for B^2 of which the roots are μ^2 and $-\nu^2$. These roots are given by formulae similar to the standard two-group diffusion theory.

Let the solutions for the flux corresponding to the two values μ^2 and $-\nu^2$ for B^2 be X and Y respectively. Then equations (4) and (5) become:

$$\nabla^2 X + \mu^2 X = 0 \tag{6}$$

$$\nabla^2 Y - \nu^2 Y = 0 \tag{7}$$

The general solutions of (2) and (3) are linear combinations of X and Y, namely:

$$\Phi_{1c} = AX + CY \tag{8}$$

$$\Phi_{2c} = S_1 AX + S_2 CY \tag{9}$$

where the coupling coefficient S_1 and S_2 are given by:

$$S_1 = \frac{(1-\omega)\Sigma_{1c}}{(1-\omega)\Sigma_{2c} + D_{2c}\mu^2} \tag{10}$$

$$S_2 = \frac{(1-\omega)\Sigma_{1c} + \omega}{(1-\omega)\Sigma_{2c} - D_{2c}\nu^2 + \omega} \tag{11}$$

It is easily seen that putting $\omega = 0$ reduces the boundary conditions and equations (2) to (11) to the standard equations of ordinary two-group diffusion theory [6].

Putting $\omega = 1$ and $\rho = \frac{1}{2}\eta$ the boundary conditions become:

$$\left.\begin{aligned} F_o &= f_{oc} + \tfrac{1}{2}\eta(T_i + t_{oc}) \\ T_o &= 0 \\ f_{ic} &= F_i + \tfrac{1}{2}\eta(T_i + t_{oc}) \\ t_{ic} &= 0 \end{aligned}\right\} \tag{12}$$

which are those of modified one-group diffusion theory [3].

Equations (2) and (3) become:

$$D'_{1c}\nabla^2\Phi_{1c} + \lambda^2\Phi_{1c} = 0 \tag{13}$$

$$D_{2c}\nabla^2\Phi_{2c} = 0 \tag{14}$$

One of the boundary conditions above is of the Robin type, viz.:

$$\Phi_{2c} + 2D_{2c}\frac{\partial\Phi_{2c}}{\partial n} = 0,$$

and since by equation (14) Φ_{2c} is a harmonic function, it is zero everywhere in the core. Hence, only (13) is left, which is the correct one-group equation of modified diffusion theory. Also, equations (6) and (7) with $\omega = 1$ become:

$$\mu^2 = \frac{\lambda^2}{D'_{1c}} \tag{15}$$

$$\nu^2 = 0 \tag{16}$$

From equations (10) and (11), $S_1 = 0$ and $S_2 = 1$ are found, and therefore

from equation (9), since $\Phi_{2c} = 0$, $C = 0$. Hence the fast flux is $\Phi_{1c} = AX$ where X satisfies the equation:

$$\nabla^2 X + \frac{\lambda^2}{D'_{1c}} X = 0 \tag{17}$$

Thus, it has been shown that equations (2) to (11) reduce to the equations of two-group diffusion theory for $\omega = 0$ and to those of modified one-group diffusion theory for $\omega = 1$ and $\rho = \frac{1}{2}\eta$.

It should be noted that although equations (2) to (11) have been chosen to give the correct results for $\omega = 0$ and $\omega = 1$ only, all the equations (2) to (11) are valid for $0 < \omega < 1$ except (11), where ω has been added to the numerator and denominator in order to make S_2 equal to 1 when $\omega = 1$ and so give $C = 0$.

The General Criticality Condition for One and Two Groups

Due to the use of the magic shell concept, only case (*i*) need be considered in detail, treating cases (*ii*) and (*iii*) as special instances of case (*i*). In the sequel, for brevity, neutron currents which do not enter reflecting material (except by tunnelling and grazing) shall be referred to as space currents, and those which do, as reflector currents.

Firstly, consider either one of the types of core covered by case (*i*). Let the fast neutron space current leaving the core be f_{os}, and let f_{is} and t_{is} be the fast and thermal inward-going space currents to the core respectively. Since by assumption the neutrons $p_2 T_o$, $p_1 F_o$ destined for reflection diffuse into an infinite medium, they are either absorbed or reflected back into the system as $f_{ir} t_{ir}$. In considering an assembly of systems, therefore, each core with its associated magic shell and reflector may be regarded as a box having an input provided by the space currents f_{is}, t_{is} and an output by the space currents f_{os}, t_{os}. It is then physically reasonable to assume that a relation of the form:

$$f_{os} = \gamma_1 f_{is} + \gamma_2 t_{is} \tag{18}$$

must hold, where γ_1 and γ_2 are surface multiplication factors.

Equation (18) can be shown to hold, and expressions for the quantities γ_1 and γ_2 in terms of the nuclear properties of the core and reflector can be derived [2].

Similarly, for the thermal neutron space current leaving the core:

$$t_{os} = y_3 f_{is} + y_4 t_{is} \tag{19}$$

where y_3 and y_4 are surface multiplication factors whose values have been derived [2].

If F_o is the total outward current of fast neutrons from the core, then the outward fast neutron current in the reflector is given by:

$$f_{or} = p_1 F_o \tag{20}$$

and the outward fast neutron space current by:

$$f_{os} = q_1 F_o \tag{21}$$

where p_1 and q_1 are the values of p and q assigned for the fast group. Similar equations may be written for the thermal currents.

Now consider the assembly of case (*i*) containing cores of types (*a*) and (*b*). The space current coming in to a core of type (*a*) consists of the sum of the contributions from all the remaining (*a*) cores and the contributions from all the (*b*) cores. The general balance equations for the space currents in such an assembly are given elsewhere [2]. These, together with equations of the type (18) and (19) for each type of unit, would result in the general criticality condition. However, for practical application of this theory, especially to large assemblies, it is usually adequate to consider only an idealized array in which the sum of the solid angles subtended by one (*a*) core at all the other (*a*) cores is constant, and similarly for the (*b*) cores. In this case, the neutron balance equation for space currents for type (*a*) cores becomes:

$$f_{is}(a) = \Omega_{aa} q_i(a) \mathrm{F}_o(a) + \Omega_{ab} q_1(b) \mathrm{F}_o(b) \tag{22}$$

where Ω_{aa} and Ω_{ab} are the summation of the fractional solid angles subtended by one (*a*) core at all other (*a*) cores and all other (*b*) cores respectively. Using (21), equation (22) becomes:

$$f_{is}(a) = \Omega_{aa} f_{os}(a) + \Omega_{ab} f_{os}(b) \tag{23}$$

Similarly, for thermal neutrons:

$$t_{is}(a) = \Omega_{aa} t_{os}(a) + \Omega_{ab} t_{os}(b) \tag{24}$$

The equilibrium of any unit is given by a set of four equations of the type (18), (19), (23) and (24). As all the currents are space currents, the suffix *s* may be dropped, and the suffix used to distinguish between the types of core (*a*) and (*b*). Then the set of equilibrium equations for a core of a type (*a*) and a core of type (*b*) is:

$$\left.\begin{aligned}
f_{oa} &= \gamma_{1a} f_{ia} + \gamma_{2a} t_{ia} \\
t_{oa} &= \gamma_{3a} f_{ia} + \gamma_{4a} t_{ia} \\
f_{ia} &= \Omega_{aa} f_{oa} + \Omega_{ab} f_{ob} \\
t_{ia} &= \Omega_{aa} t_{oa} + \Omega_{ab} t_{ob} \\
f_{ob} &= \gamma_{1b} f_{ib} + \gamma_{2b} t_{ib} \\
t_{ob} &= \gamma_{3b} f_{ib} + \gamma_{4b} t_{ib} \\
f_{ib} &= \Omega_{bb} f_{ob} + \Omega_{ba} f_{oa} \\
t_{ib} &= \Omega_{bb} t_{ob} + \Omega_{ba} t_{oa}
\end{aligned}\right\} \tag{25}$$

The condition for criticality is that the determinant of these equations vanishes.

If the values of the eight neutron multiplication coefficients are known, and the fractional solid angles expressed in terms of the pitches of the units, the vanishing of the determinant of (25) gives the pitches for which the idealized assembly is critical.

Extension to Multi-group Cores

The accuracy of the interaction calculations can be improved by taking more energy groups in the fissile units, the reflecting material and free space than are used in the simple interaction theory given above. An outline of the necessary generalization is given in Reference 2.

Special Case of Well-Defined Reflectors

When the reflecting material is well defined, one can use fairly precise assessments for the fates of neutrons leaving the core. The procedure can

be illustrated considering interaction of a pair of metal spheres completely immersed in water.

If the distributions of reflecting material were random one would have to use the values of 1 for the values of p and q in the general theory. In the case of the well-defined reflector for a pair of spheres immersed in water, it is fairly easy to arrive at sharper estimates for q than the value of unity. Since there is complete reflection one can take $p = 1$ and $q_m = 0$, i.e. $q = q_t$. A value of q_t, the fraction of neutrons that tunnel through the reflector, has still to be determined.

One can write $q_t = r \times s$, where r is the probability of neutrons commencing to tunnel on entering the reflector and s is the probability of a tunnelling neutron traversing the reflector without collision. A good approximation can be obtained for s by considering the attenuation of fission neutrons in water. From a safety point of view it is desired to overestimate r and s in calculating the tunnelling effect and so the most pessimistic case of all is considered. The neutrons are assumed to be emitted parallel to the line of centres of the spheres, and the attenuation distance is taken to be the surface-to-surface separation of the spheres. To avoid calculating r, the simplest overestimate of $r = 1$ is taken.

A Comparison of the Unified Theory and Experiment

In this Section a brief comparison is given of the unified theory with experiment for a pair of uranium spheres flooded by water, arrays of uranium spheres in a concrete vault and arrays of cylinders containing uranium solutions. The details will be found elsewhere [2].

A pair of uranium spheres in water. Mallary *et al* [7] give the results of some experiments which were performed with a pair of uranium spheres containing 93.5 % U 235, density 18.8 g/cm^3. These results have been converted by Paxton into a plot of the fraction of the single unit mass which is the equivalent of interaction. This fraction is defined as follows. Let M(s, m) be the mass of a single water-reflected sphere which produces the same multiplication as two spheres of mass m immersed in water at a surface-to-surface separation s, then the fraction of single unit mass which is equivalent to interaction is $[\mathrm{M}(s, m) - m]/m$. Experiment showed that two 20-kg spheres were critical at a separation of 1 in. and that the fraction of single unit mass equivalent to interaction was then 0.23. The calculated critical separation of the spheres for the same fraction of single unit mass is 1.2 in. The calculation was made using the interaction theory of this section, and the well-defined water reflector was treated in the way outlined above.

Another comparison which can be made between theory and experiment, in the case of the two spheres, is the surface-to-surface separation at which interaction becomes negligible. The experimental figure is about 7 in. whereas the theoretical one is about 8 in.

Uranium spheres in a concrete vault. The results of interaction experiments on arrays of uranium spheres in a concrete vault have been given by Mallary *et al.* [7]. The spheres were approximately 20 kg of metal (93.5 % U 235, density 18.8 g/cm^3). They were supported on a lightweight steel frame inside a cubic concrete vault with 1 ft thick walls and roof, and a 6 in. thick floor. Up to twenty-seven spheres were used and the internal vault size

varied from 1 ft to 5 ft in 1-ft increments. For the case where the spheres were inside a concrete vault, the theoretical pitches were between 4 cm to 7 cm greater than the experimental ones. The simple assumptions used, namely, $p = \frac{1}{2}$, $q = 1$ with a concrete reflector, are considered to be a fair approximation to the experimental conditions. An extremely simple calculation for the case when the roof and walls of the vault are missing assumed $p = 0$ and $q = 1$. When the number of spheres is less than 16, the theoretical pitches exceed the experimental pitches by not more than 3 cm. The calculated pitches when the number of spheres is greater than 16 are slightly less than the experimental pitches This error can be reduced by allowing for the reflecting effect of the concrete floor. For example, the calculation for twenty-seven spheres with the assumptions $p = \frac{1}{4}$, $q = 1$ gives a pitch of 25 cm which is 2 cm greater than the experimental pitch. However, exact comparison is perhaps not particularly instructive in this case since the experimental results were obtained by making fairly big extrapolations of multiplication curves.

Arrays of cylinders containing solutions of uranium. The results of experiments on the interaction in arrays of three and seven cylinders on a triangular pitch have been given by Fox *et al* [8]. The aluminium cylinders had diameters of 6 in. and 8 in. and contained aqueous solutions of UO_2F_2. The enrichment of the uranium was about 90 % U 235 ratio and the atomic hydrogen to U 235 ratio was 44.3. This ratio results in minimum critical volumes for uranium solutions. Experiments were done with the cylinders immersed in water (but only up to the level of the UO_2F_2 solution) and also with them unreflected.

This problem can be tackled formally using two-group diffusion theory in the core and reflector. This involves the calculation of the surface multiplication factors γ for finite cylinders. The interaction calculations involved are very laborious (a programme for a Ferranti Mercury computer is available for these calculations), but it is not necessary to perform such detailed calculations to get safe pitches for the cylinders. The method adopted here gives for all cylinders safe pitches which are not unduly conservative. For cylinders of height H and diameter D for which $\frac{1}{2}D < H < 2D$, this can be seen from the argument which follows.

The first step in the calculation is to replace each partially-reflected (bare on top) finite cylinder by the equivalent fully-reflected sphere. This can be done by using the reflector savings for water (taken as 1.4 in.) [8], and the results of Prohammer [9] who has compared experimental results for cylindrical and spherical aluminium vessels. The calculation of the γ coefficients for the fully-reflected sphere is now comparatively simple. The critical pitch of the spheres can now be calculated assuming the interaction solid angles to be the same as for the cylinders. Consider the effect of elongating the spheres into the original cylinders. It can be seen that the fraction of diffusing neutrons reflected back to the core is reduced whereas the number of neutrons interacting is the same, because the interaction angles are unchanged. Therefore, the reactivity of each core is decreased and consequently a

TABLE I. SEPARATION OF CYLINDERS *(in.)*

6-in. dia. cylinders			8-in. dia. cylinders		
Number	Experiment	Theory	Number	Experiment	Theory
3	0.15	1.5	3	0.15	1.7
3	3.00	3.1	3	1.00	2.1
7	0.15	1.5	3	2.00	3.2
7	1.00	1.6	7	0.15	1.8
7	2.00	2.1	7	1.00	2.2
7	3.00	2.9	7	2.00	2.9
7	4.00	4.0	7	3.00	4.1

pitch which is calculated to be critical for the sphere becomes a subcritical pitch for the cylinders.

A reasonably simple and safe choice for the probabilities involved, to represent the physical situation, is the following: $p_1 = p_2 = 1$, $q_{1m} = q_{2m} = 0$, $q_{1t} = s$ and $q_{2t} = 0$. Subscripts 1 and 2 refer to the fast and thermal groups respectively, s is the fast neutron attenuation factor and it has been assumed that there will be no interaction due to tunnelling of thermal neutrons (due to their softer spectrum). The calculated separations are compared with the experimental ones in Table 1. It can be seen that for the 6-in. diameter cylinders the results were in reasonable agreement with experiment except at small separations. (It should be noted here that the cylinders had 0.06 in. thick aluminium walls, so that in comparing the results of Table 1, the experimental results should strictly be increased by a thickness of water equivalent to 0.12 in. of aluminium.) For the 8-in. diameter cylinders the agreement was not so good, the theoretical results being ' conservative ' by 1 in. to 1.5 in.

However, as the method of calculation was designed to result in slightly subcritical separations for the equivalent spheres, it will give separations further from critical for the cylinders than for spheres. One can see that the simple treatment used gives good approximate answers which err on the side of safety.

Theoretical results for random reflectors. For two 20-kg spheres of 93.5 % uranium which are critical at a separation of 1 in. in water, the experimental figure for the fraction of single unit mass was 0.234. For a completely flooded pair the critical separation, according to theory for this fraction, is 1.2 in. For a random reflector of air and water, theory predicts a separation of 2.5 in. In Figure 2 the fraction of single unit mass which is equivalent to interaction is plotted against critical surface-to-surface separation. The theory suggests that for high values of the fraction of single unit mass equivalent to interaction there is very little difference for practical purposes between a complete water reflector and a random reflector of air and water. For low values of the fraction of single unit mass, the critical pitches for a random reflector are considerably greater than for the flooded pair. This appears to be reasonable if one considers a reflector of air and water which consists of an infinite water reflector with a central void in which the two spheres are placed. The critical pitch in this case is clearly greater than the critical pitch for two spheres in air alone, as some of the neutrons, which would be allowed to leak away in the former system, are reflected by the water.

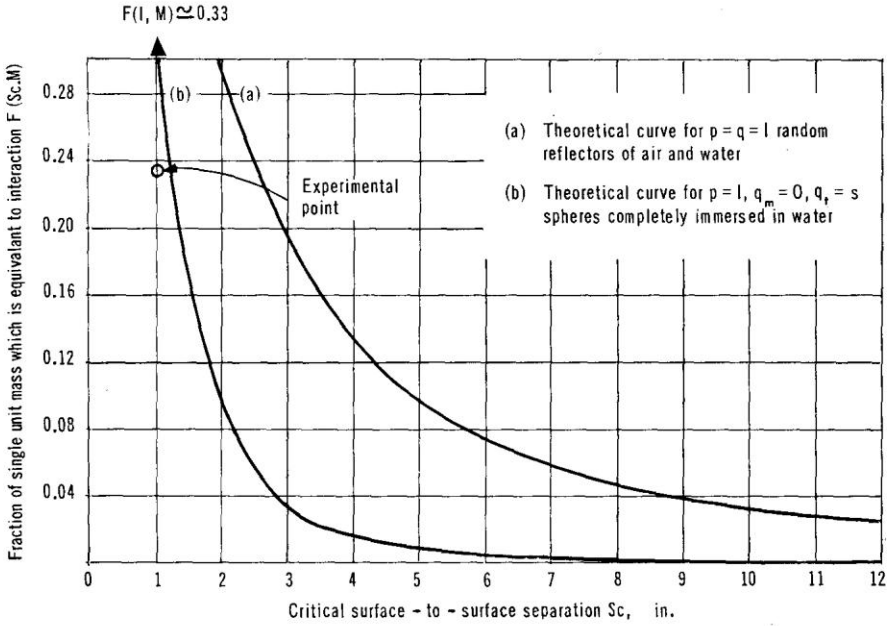


Figure 2. *Interaction between a pair of metal spheres (theoretical).*

For arrays of 20 kg uranium spheres the theory predicts pitches for the spheres which, for a random reflector of water and air, are roughly three times the experimental pitches for the spheres in the concrete vault. The results for the random reflector are possibly very much more pessimistic than for any situation which can arise unless the vault is subject to flooding.

Intermediate Energy Units

In the theory so far, fast and thermal units have been considered. For units of intermediate energy, fast and thermal units a similar analysis can be given. If one energy group is used for fast cores, and two energy groups are used for thermal cores and the reflectors in conjunction with a magic shell, one can use an ingenious analysis due to D.E.J. Thornton [10] which employs two groups in general for intermediate units. As the degree of moderation decreases, the two groups differ in the weight attached to them, and so when the degree of moderation is very slight, the economy of the core is governed by a one-group equation. This type of analysis can be extended with the aid of a magic shell to the case of two groups for a fast core, four groups for thermal cores and reflectors, and two-four for intermediate cores.

When used with simple assumptions for p and q to represent known physical conditions the theory agrees well with all the experimental results known to us. The theory is also a reasonably simple one to use, even with desk computers. The speed of computation is, of course, considerably increased by the use of a medium-size digital computer, such as the Ferranti Mercury. As many as thirty cases of an interaction problem can be dealt with in a computing time of five minutes.

The Interaction Parameter Method

The interaction parameter method [19] was developed by A.F. Thomas for the assessment of the safety of workshop processes and storage arrays involving large quantities of fissile metals, i.e. essentially fast systems. However, it has proved applicable to a wide variety of systems, including mixtures of metal and well-moderated systems. The method expresses the safety of an array of fissile bodies in terms of quantities characteristic of the individual items. These quantities are capable of direct experimental measurement with single bodies or two-body systems, and they can be obtained from other experimental data or by calculation.

The ' interaction parameter ' q_{ij} for one body i as viewed from another body j in the array is defined as the total neutron output induced in i when one neutron leaves j and does not interact with any other body in transit, i.e. if F_j is the neutron output of j, the output induced in i due to j alone is $q_{ij}F_j$. The parameter is expressed as the product of the probability p_{ij} that a neutron leaving j reaches i without interacting with any other body in the array, and the surface multiplication M_{si} of body i for neutrons reaching it from j. The quantity P_{ij} will depend on the geometry of the system, the nature of the bodies (governing their emergent neutron spectrum and angular distribution) and the nature and distribution of any intervening medium. M_{si} will depend primarily on the properties of i and its immediate environment but there may be some secondary dependence on the properties of body j if these introduce major differences in the energy spectrum and angular distribution of the neutrons incident on i.

The Condition for Criticality

The condition for criticality in an array can be derived by writing the equations connecting the outputs of the bodies. Let F_i and F'_i be the outputs of the body i in isolation and in the array, respectively. Then:

$$\left.\begin{aligned} F'_1 &= F_1 + q_{12}F'_2 + q_{13}F'_3 + \ldots\ldots q_{1n}F'_n \\ F'_2 &= q_{21}F'_1 + F_2 + q_{23}F'_3 + \ldots\ldots q_{2n}F'_n \end{aligned}\right\} \qquad (26)$$

The condition for criticality is that positive non-zero values of F'_1 F'_2, etc. can be maintained in the absence of source terms, i.e. $F_1 = 0 = F_2 = 0$, etc. That is:

$$D = \begin{vmatrix} -1 & q_{12} & q_{13} & q_{14} & — & — \\ q_{21} & -1 & q_{23} & — & — & — \\ q_{31} & q_{32} & -1 & — & — & — \\ — & — & — & -1 & — & — \\ — & — & — & — & -1 & — \\ q_{n1} & q_{n2} & — & — & — & -1 \end{vmatrix} = 0 \qquad (27)$$

Criteria for Sub-criticality

In general, the use of the criticality criterion in this form can be tedious for any but the simplest arrays. However, in practical cases, the systems to be assessed are well subcritical and the problem is not so much a full analysis of the situation as a practical demonstration of their safety. It is possible, therefore, to make use of criteria which are more conservative than the above and which are much more simply satisfied. To do this let us define two quantities, S_i and T_j:

$$S_i = \sum_{\substack{j=1 \\ j \neq i}}^{n} q_{ij} \tag{28}$$

$$T_j = \sum_{\substack{i=1 \\ i \neq j}}^{n} q_{ij} \tag{29}$$

Defined in this way, S_i is the induced output in body i when unit source is placed at the positions of each of the other bodies and T_j is the sum of the outputs induced in all the other bodies when one neutron leaves j. The criteria for safety are:

$$S_{max} \equiv \max_i \{S_i\} < 1 \tag{30}$$

and

$$T_{max} \equiv \max_j \{T_j\} < 1 \tag{31}$$

Further and closer approximations to A can be established, e.g.:

$$S'_{max} \equiv \max_i \{\Sigma q_{ij} S_j\} < 1 \tag{32}$$

but the extra effort involved in their application is rarely worthwhile.

Determination of the Interaction Parameter

Experiment:

An attractive feature of the interaction parameter method is that it is possible to make measurements with a single body and a source and thus obtain experimental data which can be used to assess an interaction problem before the numbers of such bodies produced become large enough to present a problem. Such measurements are simple in principle, but since the q values of fissile bodies likely to be considered are small ($\sim 10^{-4}$ to 10^{-2}), they are sometimes difficult to measure accurately, particularly with plutonium where a small increase in output has to be measured against a background of neutrons from spontaneous fission. Care has to be taken that the simulation of a neighbouring body by a small source does not lead to an underestimate of q. It is sometimes necessary to perform subsidiary experiments to correct for this.

When two like bodies either have a useful neutron output or can contain small neutron sources, the problem of measurement is simpler. If three measurements are taken with the first body alone in position, the second body alone in position and both bodies in position, giving count rates C_1, C_2 and C_3, then q is given to a close approximation by:

$$q = 1 - \frac{C_1 + C_2}{C_3} \tag{33}$$

Differences in counter sensitivity and neutron output of the two bodies appear only as second order corrections. This is particularly useful in water, where small errors in position can give large errors in count rates.

Calculation:

a) *Surface multiplication from other experimental data.* Values of the surface multiplication of a fissile system are not easily measured directly.

However, for many systems of interest the central source multiplication M_c can easily be measured. From this an overestimate of the surface multiplication can be obtained for an isotropic source:

$$M_s = 2\theta M_c - 1; \qquad \theta \leqslant 1 \tag{34}$$

where θ specifies the relative importance of neutrons born at the surface and at the centre. For a bare system the one-group diffusion approximation gives θ as $\frac{\sin Kr}{Kr}$ where K is the material buckling. We can use either a calculated value of θ, which is conservative, or take θ to be unity. Then:

$$M_s < 2M_c - 1 \tag{35}$$

For reflected systems, the total number of neutrons which leave the fissile core per neutron entering is M'_s, greater than M_s (the bare surface multiplication) since some of the emergent neutrons will be returned to the core and give rise to further neutrons. If the reflected central source multiplication M'_c is known, then:

$$\frac{M'_s}{M_s} = \frac{M'_c}{M_c} \text{ i.e. } M'_s < \frac{M'_c}{M_c}(2M_c - 1) \tag{36}$$

where M_c is the bare central multiplication.

b) *Direct calculation of surface multiplication.* On the other hand, multi-group methods with fast computers can be used to calculate M_s with some confidence in systems where they can be shown to give estimates of M_c or q in good agreement with experiments and for similar systems where experiment may be impracticable or too time-consuming. Some simple geometrical model of the fissile system is usually required to facilitate calculation; Monte Carlo methods can, in theory, be used for any geometry but the time required to get tolerable statistical accuracy often precludes their use. Calculations using the diffusion approximation (one- or two-group) can be used, provided the constants used are such as to give conservative results in closely allied systems whose behaviour is known from experiment or more sophisticated calculations.

Equation (35) can be used with the value of k, the reproduction constant, to provide an overestimate of M_s for well-moderated systems. It can be shown that:

$$M_s \leqslant \frac{1 + k(1 - 2/\nu_f)}{1 - k} \tag{37}$$

where ν is the number of neutrons per absorption and f the thermal utilization factor.

k, ν, and f may be derived from some simple recipe such as that of Henry, which is conservative for $H/U > 30$ and appears to be conservative for lower ratios.

c) *Calculation of the effective solid angle parameter p.* In air, the fraction of neutrons leaving an isotropically emitting body and which interact with another is taken to be the fractional solid angle subtended by the second body at the first. However, in real arrays, there may be some shielding of neutrons by intervening bodies (reducing p) and scatter from floors, walls, machines, human bodies (increasing p). The way in which these two effects

are taken into account in the interaction parameter method is a matter for the judgment of the user. The reduction of p in ratio with the square of the separation distance cannot be true for distances comparable with the height of the fissile bodies above a concrete floor, as the fraction of neutrons reaching the bodies by indirect paths will increase with distance. It has been the custom at Aldermaston to calculate p for a distance approximately equal to the height from the floor, making an allowance for the scatter, and to allow p to decrease only linearly with separation for greater separations. When the intervening medium is a significant neutron absorber (e.g. water or concrete), a conservative estimate of its effect is made.

Allowance for shielding of one fissile body by another must be made with caution. If the bodies are in immutable positions (e.g. fixed storage arrays or plant) line-of-sight shielding may well be considered, but in a workshop or laboratory it is better to ignore it, since stringent and irksome restrictions on movement would need to be applied to preserve relative positions.

When the bodies consist of fissile material with non-fissile reflectors (e.g. a casting in a graphite mould) some thought has to be given to those neutrons which would have missed the body but hit the reflector and are scattered in. For some simple cases it is possible to show that the total number of neutrons reaching the fissile material is, at most, no greater than the number which would have hit it in the absence of the reflector. A safe overestimate is given by taking:

$$p = \frac{1}{2}(p_1 + p_2) \tag{38}$$

where p_1 is the fraction which would have hit the core in the absence of the reflector and p_2 is the fraction incident on the whole reflector.

The simple value of $q = p\mathrm{M}_s$ is a first approximation. A better one is:

$$q = p(\mathrm{M}_s - \mathrm{F}) \tag{39}$$

where F is the fraction of neutrons incident on the body which penetrate it without collision. The interaction effect of a body is not due to *all* the neutrons which leave it, but only to those which would not have been available to other bodies in the array but for its presence. The fraction F represents the fraction of the total neutrons leaving the body whose path has been unaffected by its presence. In practice for most bodies the distinction is fine, since F is small compared with M_s, but it can be important when considering thin slabs or shells of large superficial area when $\mathrm{M}_s - 1$ is small and F is significant compared with unity.

The Use of the S and T Criteria

To apply (30) or (31) to practical arrays, the value of S_i or T_j ought to be calculated for each body in turn to ensure that the maximum value of one or the other in the array is less than unity. The array is safe even if $\mathrm{S}_{\max} > 1$ but $\mathrm{T}_{\max} < 1$ (or *vice versa*), since either criterion is sufficient. In practice it is usually sufficient to look only at a limited number of bodies since it is obvious that the value of $\mathrm{S}_{\max}$ or $\mathrm{T}_{\max}$ will be at one of them.

For regular arrays of like bodies, each with an interaction parameter q_o at unit lattice spacing, some simple formulae can be derived if some regular variation of q with separation is assumed; for example, if q is allowed to decrease linearly with separation.

For a square lattice,

$$S_{max} < 2\sqrt{\pi}(\sqrt{N} - 1)q_o \tag{40}$$

where N is the total number of bodies.

For a cubic lattice,

$$S_{max} < 2\pi(N^{\frac{2}{3}} - 1)q_o \tag{41}$$

If additions to or variations of an array, which has been shown to be safe, are made, it may be necessary to evaluate S or T for each body *ab initio*. If, however, the array is very safe and all that is required is to produce a demonstration of this for a wide variety of variants of the array (within certain constraints, e.g. minimum separation), it is possible to produce a simple analysis which provides a value of T which is greater than any value that would arise in the array with any rearrangement of bodies. If, for example, we have an array in which it is possible to draw a square of side d_o centred on each body without any square overlapping, then it is always possible to rearrange the bodies with all the squares touching (i.e. with the bodies at the intersections of a square lattice) which is at least as near critical as the real array. If now the bodies are rearranged so that the body with the least value of the interaction parameter at separation d_o is at the centre of the lattice, the body with the greatest parameter at the nearest lattice point, the next greatest at the next nearest and so-on, the resulting array is not necessarily as near critical as the real array but it can be shown that the value of T_{max} for the resulting array is at least as great as, and in the vast majority of cases considerably greater than, any value of T_{max} for the real array. If T_{max} for the hypothetical array is less than unity, the real array is safe.

The calculation of this upper bound to T_{max} for the real array is now reduced to the addition of the products of the interaction parameters and coefficients for the hypothetical array. These parameters and coefficients depend only on the geometry of the regular lattice appropriate to the situation. For instance, if we have n_1 bodies with interaction parameters q_1 at separation d_o, n_2 with q_2 at d_o, etc., where $q_1 \geqslant q_2 \geqslant q_3$, etc.,

$$T_{max} \leqslant q_1 \sum_{1}^{n_1} f_m + q_2 \sum_{n_1+1}^{n_2} f_m + \ldots\ldots$$

where f_m is the ratio of the interaction parameter at distance d_m to that at distance d_o (d is the distance of the mth closest lattice point from the centre). These coefficients f_m can be calculated for any regular lattice and $\Sigma_1^N f_m$ tabulated.

Any rearrangement of the array within the limits of constraint is seen to be safe without further calculation. A change in the number of types of bodies requires only a simple arithmetical process for its assessment.

We have referred only to a square lattice as a model; other models may be more appropriate — if the bodies in the real array can be regarded as at the centre of non-overlapping circles, of diameter d_o, for a plane array, cubes or spheres for a three-dimensional array, regular hexagonal cubic or closed packed spherical lattices are appropriate and the coefficients and calculations made in the same way.

For a plane array a further development of this approach can be made, resulting in an assessment which allows free movement of bodies within certain

restrictions. If a body of interaction parameter q_i at distance d_o is considered as being at the centre of a circle of radius R_i, so that:

$$k_o \pi R_i^2 = q_i$$

where k_o is a constant, then the real array is safe if:

$$\frac{R_i}{d_o} \leqslant 2(\Sigma q_i)^{\frac{1}{2}} q_i^{\frac{1}{2}}$$

provided that none of these circles overlap, i.e. if this relation is satisfied, the bodies can be arranged or moved in any way provided the circles of radius R_i about them do not overlap.

A useful simplification for this type of assessment when one has large numbers of weakly interacting bodies (e.g. swarf bottles) grouped together is the idea of a group interactor parameter q_g. It is easy to show that the total output of the group is less than:

$$q_g = \frac{\Sigma q_i}{1 - (T_{max})_g}$$

where $(T_{max})_g$ is the greatest value of T_{max} for any body in the group due to the other bodies in the group. In other words, if we ascribe a value q_o to a virtual body at the position of the group, we shall overestimate its output due to neutrons leaving other bodies in the main array, i.e. overestimate its contribution to the interaction in the main array.

Field of Application

The interaction parameter method has general application to arrays for which reasonably accurate or conservative estimates of the parameters can be found. The chief drawbacks which arise in practice are:

a) The measurement of q values for systems completely immersed in reflecting material, and in particular water and other hydrogenous materials;

b) The difficulty of obtaining accurate calculations in these cases, although these are to some extent being overcome by Monte Carlo methods on fast computers;

c) The translation of measurements or calculations made with point sources into data for two-body systems when separation distances are small; usually some conservative device has to be adopted.

Cases of awkward geometry can give rise to difficulties but no more so than those presented by other methods.

The Oak Ridge Interaction Method

This method of Geller's was developed by Henry and co-workers [11, 12, 13] at the Oak Ridge Gaseous Diffusion Plant in conjunction with Callihan *et al* of the Oak Ridge National Laboratory. The approach to interaction problems at Oak Ridge was based originally on the interpretation of experiments performed with cylinders containing uranyl fluoride solution at a U 235 enrichment of 93 %. Later, some experiments with slab-sided vessels and unmoderated uranium compounds were also analysed. Basically, a correlation is made between a multiplication factor, K, for a pair of vessels

and the solid angle, $\bar{\Omega}$, subtended by the units. It is our view that this method is best suited to well-moderated systems.

Theory

The derivation of the multiplication factor is based on a conventional two-group approach. For a single vessel, the multiplication factor, k_1, is defined as:

$$k_1 = \eta f U_t U_f \tag{42}$$

where U_t and U_f are the non-escape probabilities for thermal and fast neutrons respectively. These terms are readily calculable for unreflected systems of highly enriched uranium and the analysis is confined to such systems. In considering the effect of interaction between vessels, a multiplication factor, K, is defined by:

$$K = k_1(1 + pV) \tag{43}$$

where k_1 is the multiplication factor for the individual vessels (if identical) or the most reactive vessel, p is a constant depending upon the configuration of vessels, and V is a measure of neutron exchange. Values of p are easily obtained by considerations of solid angle, e.g. $p = 1$ for pairs of vessels, and $p = 1 + \sqrt{7}$ for a 7-cylinder hexagonal array. The value of V is derived as follows. The probability of a fast neutron escaping from a vessel is $(1 - U_f)$. If r is the ratio of the buckling of the interacting surface to the total buckling of the vessel and $\bar{\Omega}$ is the average solid angle subtended by the vessels, then,

$$V = r\bar{\Omega}(1 - U_f) \tag{44}$$

is the probability that a fast neutron will leave one vessel and reach the next vessel. Modifications to this basic approach include an allowance for resonance capture of neutrons in uranium of lower enrichment and an allowance for variations in neutron flux distribution between closely-spaced vessels. This latter was done by introducing a modified extrapolation distance, d, related empirically to an isolated vessel extrapolation distance, d_o, and the solid angle subtended by the vessels. Thus,

$$d = d_o + (d_w - d_o)\bar{\Omega} \tag{45}$$

d_o is an unreflected extrapolation distance (= 2.5 cm),
d_w is the reflected extrapolation distance (= 7.0 cm).

It is noted that when vessel spacings are of the order of 12 in. or more, negligible error is introduced by neglecting corrections to d_o. The error is still small for some separations of less than 12 in.

The derivation above was based largely on experiments with arrays of identical unreflected cylinders containing 93 % U 235 in solution. Since such ideal circumstances are seldom met in practice, it was necessary to test and modify this simple approach by studying interactions between systems of different U 235 assay, different moderation and different shape or relative size. The approach has been to compare known critical assemblies consisting of the distinct types of units A and B with known critical assemblies of type A and type B units only. Thus a cylinder slab interaction experiment (type A-B) is compared with the twin cylinder experiment (type A), and twin slab experiment (type B), using the same cylinders and slabs for the three experiments. In addition, some comparisons have been made between unreflected

and half-reflected systems. The calculation of a multiplication factor has of necessity been based on that of the most reactive vessel. In particular, for a partially-reflected vessel, its multiplication factor is taken to be that for a fully-reflected vessel and made equal to unity. The composite value, K, for a reflected and unreflected pair becomes thus:

$$\left.\begin{aligned} \mathrm{K} = k_{\frac{1}{2}} &= \frac{k_{\mathrm{ref}} + k_{\mathrm{bare}}}{2} \\ &= \frac{1 + k_{\mathrm{bare}}}{2} \end{aligned}\right\} \tag{46}$$

It was demonstrated experimentally, in a number of typical cases, that the value K calculated in this intuitively conservative way always predicted a system to be critical at an interaction solid angle less than the known figure.

In order to present the data calculated by the method in a form suitable for general use, the values of multiplication factor and solid angle found experimentally were plotted graphically as in Figure 3. From these an eye-fit curve (curve A) was deduced as representing safe conditions for use in spacing identical containers. This curve was made to cut off at fractional solid angle $\Omega = 0.48$, to prevent the juxtaposition of two slab-shaped vessels, and also at a $k = 0.9$ since some values of k calculated conventionally showed non-conservative errors of a few per cent for certain closely-spaced arrays of cylinders. For slab-shaped vessels the error in k was found to be slightly larger, and to allow for these uncertainties a second curve (curve B) was drawn, which gives safe conditions for any pair of vessels.

Scope

The Oak Ridge approach seems to have had the primary aim of keeping calculations as simple as possible. To this end the relevant functions were evaluated in graphical and tabular form, thereby making them as widely usable as possible. A price is usually paid for simplicity. In this instance it was a comparative lack of accuracy, though not always reliability, in dealing with small spacings between units; and the inclusion of rather large safety factors in the arrangement of multiple units. Also, since the theory was evolved on interactions between bare assemblies, the possible effect of accidental full reflection or partial reflection could not be adequately allowed for. Thus, for instance, all vessels had to be designed safe against accidental full water reflection. Also the minimum edge-to-edge spacing allowed between units is 12 in. There are also other obvious uncertainties which it is difficult to eradicate in a simple interpretation, e.g. systems in which units may have significantly different neutron spectra, and the difficulty of determining ' average ' solid angles subtended by dissimilar units. If allowance has to be made for the import of unspecified containers whose fissile material content is only partially known, then only the most conservative of alternative approaches may be made to the problem of their storage. In such situations the loads for ' birdcages ' or the pitches for rigid demarcation or positioning of containers would seem to be estimated somewhat conservatively by the Oak Ridge method. However, even in these extreme circumstances the Oak Ridge approach does not become unduly restrictive or inconvenient until the number of vessels or containers becomes relatively large.

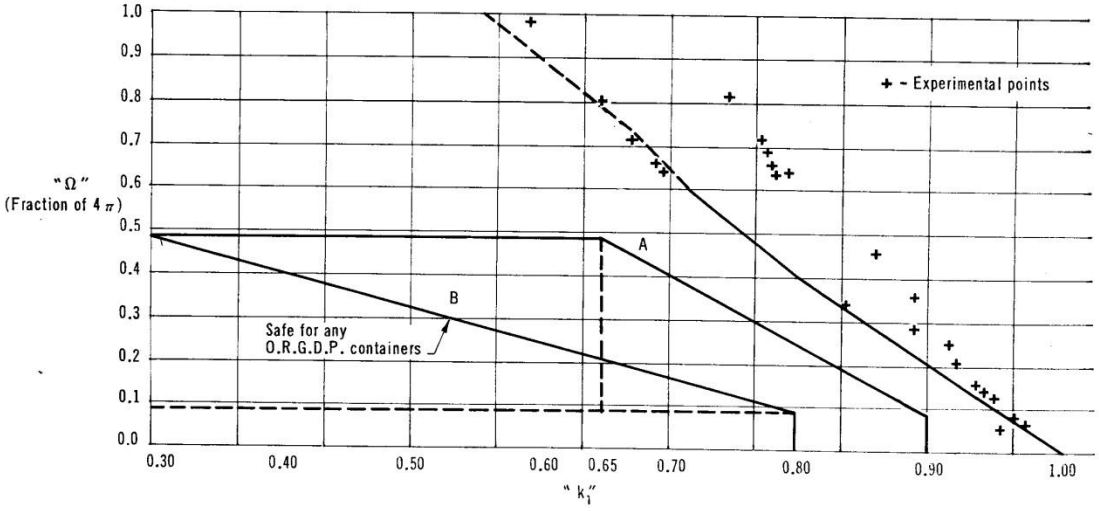


Figure 3. *Interaction solid angle versus multiplication factor.*

The Density Analogue Treatment

It is well known that the critical mass of a fissile isotope can be related to its effective density (in g/cm^3) by an exponent of the form mass $\propto$ (density)$^{-S}$. In general this exponent varies, and for water-reflected systems it usually lies within the range $1.2 < S < 2.0$. As a system becomes large (i.e. the density becomes low) the value of S approaches 2.0 even for reflected systems (the effect of reflector on critical dimension becoming relatively smaller for larger systems). Thus, if a value of S is calculated from a relatively small system, one would expect that value of S to be an underestimate of the exponent for large systems.

Theory

For a given interacting system the value of S is deduced from an effective space density of fissile units (i.e. fraction of space occupied by fissile material compared with the least enveloping volume). This value of S can be used to calculate the critical mass if similar units are occupying space at a lower effective density.

It is suggested that the density exponent will depend not only on the degree of reflection about the array but also on some neutron multiplying characteristic. For lack of precise data on surface multiplication and reproduction constant, a correlation of index S with ' fraction critical ' was made. It was also considered that the surface multiplication for a given ' fraction critical ' was greater for undiluted fissile metal lattices than for lower density assemblies. Hence, values of S deduced from metal assemblies are considered to be conservative for more dilute units. A curve of S *versus* ' fraction critical ' has been constructed (dotted line, Figure 4) for air-spaced units based on metal lattice assemblies. To estimate the critical number of units in any other array it is first necessary to calculate the value of ' fraction critical ' for the units to be stored. The value of S is then obtained from the graph and an effective density can be calculated for the total mass of material to be stored. The volume of space per unit can be estimated from the mass of each unit and hence the pitch of the units can be deduced.

Scope

Some difficulty is encountered in this approach when fissile units of metal have small quantities of hydrogenous reflector at their surfaces (i.e. equivalent moderator between units). For example, in a particular metal lattice it was found that the insertion of 1 in. thick pieces of Plexiglas between units reduced the critical number of units in an array by nearly an order of magnitude. Also the presence of certain materials as reflectors (e.g. concrete) appears to affect the permissible number of units adversely. It is, therefore, necessary to use a pessimistic value of S for a given ' fraction critical ' and to employ a large safety factor to ensure the safety of random, variable reflector, variable moderator units (solid line, Figure 4). This latter type of situation could easily arise in uncontrolled transport and storage of non-isolated fissile materials. The size of the safety factor required to cover such situations makes the density analogue method severely restrictive. This method has been known to predict a number of cases as dangerous which were known experimentally to be safe [14].

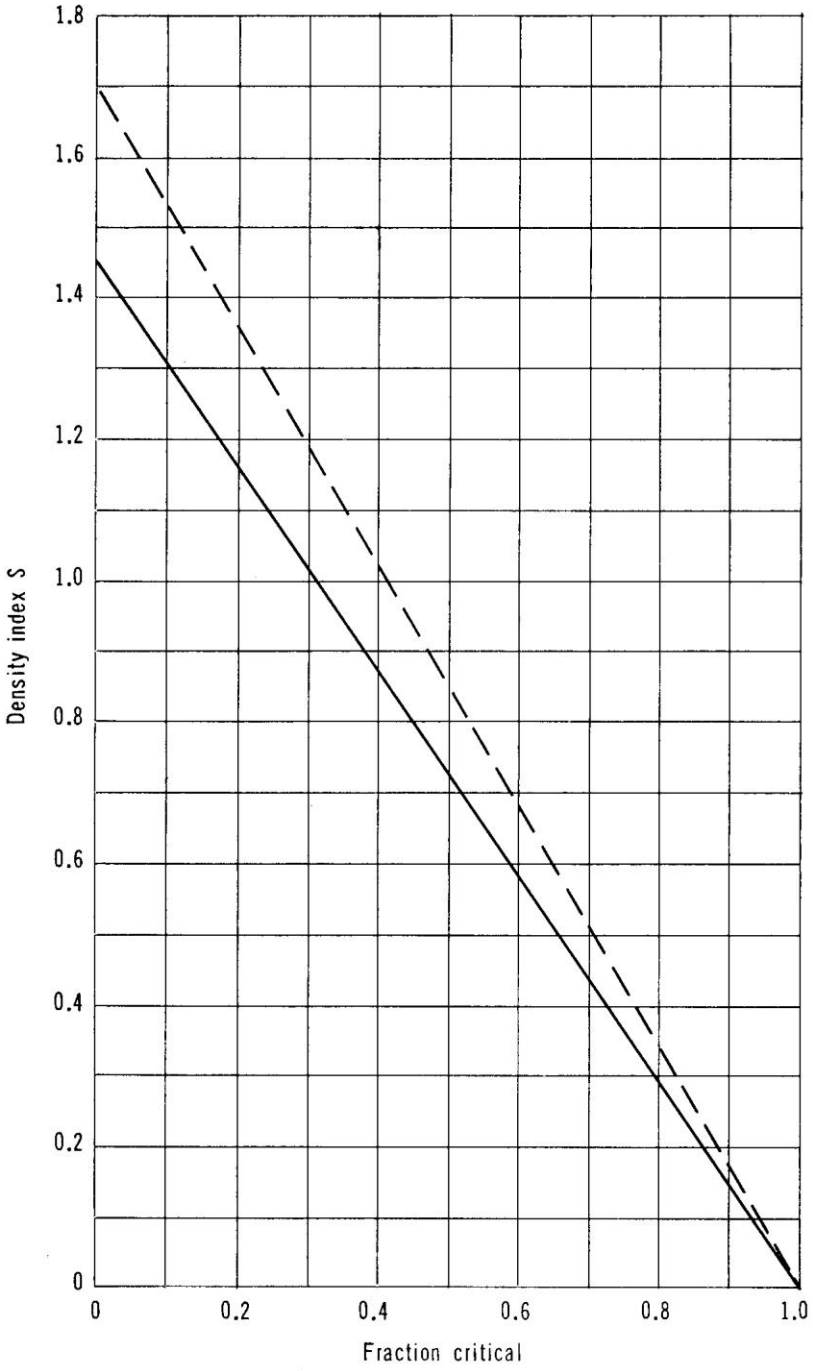


Figure 4. Density index versus fraction critical.

KENYON'S RULES

The important factors upon which the critical size of an enriched uranium system depends are taken to be, firstly, the effective density of the fissile isotope ($\rho_{25} = g$ U 235/cm^3) and, secondly, N the moderating ratio of H atoms to U 235 atoms (H/U 235 = N) [15]. By plotting the available U 235 solution, compact and metal data for spherical, water-reflected systems on a graph with ordinate ρ_{25} and abscissa M_s, the critical mass of U 235, a series of curves for different values of N can be obtained. For practical purposes in the interpretation of an experimental result, the effect of non-hydrogenous neutron scattering materials is regarded as negligible. From these curves can be deduced the variation of M_s with N at constant ρ_{25}.

Basis for the Rules

It was observed from experiments, and can be deduced from considerations of equal buckling conversions, that the diameter of a just critical infinite cylinder D_∞ and the thickness of a just critical infinite slab T_∞, both water-reflected, are almost proportional to the critical sphere diameter for all conditions of ρ_{25} and N. Thence it was inferred that the mass of U 235, M, and the diameter of a finite cylinder D can be related to the mass of U 235 in a sphere, M_s, and the diameter of the sphere, D_s, and the diameter of an infinite cylinder of the same core composition D_∞ by the elliptic formula $\left(Dv\,\frac{1}{M}\right)$.

$$\left(\frac{D_s - D}{D_s - D_\infty}\right)^2 + \left(\frac{M_s}{M}\right)^2 = 1 \qquad \text{for } D < D_s \text{ i.e. slender cylinders}$$

On the other hand, if $D > D_s$ (squat cylinders), then the formula for thickness of a finite slab is used, viz.:

$$\left(\frac{D_s - T}{D_s - T_\infty}\right)^2 + \left(\frac{M_s}{M}\right)^2 = 1 \qquad T < D_s$$

An example of the comparison between theory and experiment is given below for isolated critical vessels, water-reflected on both sides and ends.

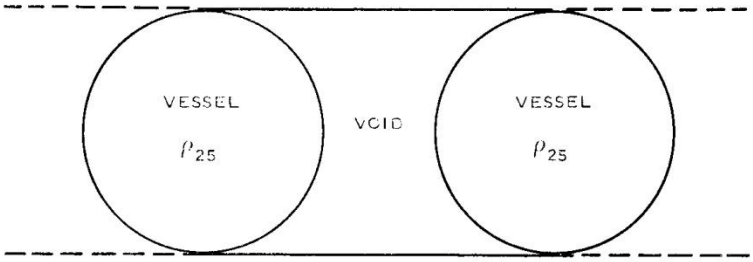
TABLE II

Reactor Dia. *(in.)*	Sol. Conc. *(gU235/cm³)*	Experimental Crit. Mass	Calculated Crit. Mass
6	0.459	5.93	5.7
6.5	0.827	7.91	6.7
	0.212	2.39	1.96
10	0.459	3.12	2.92
	0.0522	0.930	0.88
15	0.459	4.14	4.14
	0.116	1.49	1.42
	0.026	1.31	1.30

As may be seen, the agreement is fairly close and is always conservative.

From a study of the identical pair interaction data for cylinders of U 235 solution it became apparent that the use of a reduced density factor together with the original $\rho - M - N$ curves would give conservative agreement with experiments. Thus, for water-reflected cylinders of varying edge-to-edge separation, a reduced effective density of U 235 was calculated as follows. The water space contained between parallel tangents to the identical cylinders was assumed to be a void and the uranium solution was smeared evenly over the space enclosed by cylinders plus void. For this new configuration an effective density was given by:

$$\rho_e = \rho_{25} \times \frac{\text{total cross-sectional area of the cylindrical vessels}}{\text{cross-sectional area of vessels plus void}}$$



The H/U 235 value, N, is that of the original solution since no addition to the fissile core material is allowed. Using this, the effective density ρ_e and N, a value for M_s is obtained and hence D_s is found. The assumed constant relationship between D_s and the diameter of the infinite cylinder, D_∞, or infinite slab, T_∞, is invoked and finite vessel calculations made as before, using the formulae for D, M and T, M given above.

Scope of the Rules

The example below shows that the method can give conservative results which are in fair agreement with experiment.

TABLE III. TWO PARALLEL CYLINDERS REFLECTED ON ALL SIDES

Reactor Dia. *(in.)*	Edge-to-Edge *(cm)*	Sol. Conc. *(g U235/cm³)*	Experimental Crit. Mass/Cyl.	Calculated Crit. Mass/Cyl.
5	2.9	.566	3.02	2.29
	3.8	.566	3.80	2.40
5.5	2.9	.926	3.60	2.97
6	2.9	.926	3.73	2.87
	5.8	.566	2.64	1.97
	13.0	.566	4.30	2.45
	6.0	.187	1.65	1.19
8	14.7	.566	2.84	2.58
15	5.8	.566	4.00	3.45

The method has also been tried on 7-cylinder arrays, an experimental critical height of 5.4 in. for 6 in. dia. cylinders being predicted as 4.9 in. by

the reduced density formula. It has also been tried on metal sphere arrays, $N = 0$. An extrapolation to critical from an experimental assembly predicted that an array of 40 spheres (20 kg U 235 in each) on a pitch of 11 in. centre-to-centre would be critical. The reduced density calculation yielded an estimate of 32 spheres as being critical.

Some preliminary results of the extension of the method to low-enriched uranium systems (1.4 % in the United Kingdom and 2 % in the United States) indicate that conservative predictions of critical sizes can still be made for homogeneous mixtures of uranium and hydrogenous moderator. At present, the further extension to lattice systems is being studied, though in this instance some preliminary results are not encouraging.

Miscellaneous Methods

The methods to be described should be regarded primarily as providing checks which are complementary to the experimental ones.

The problem of two like spheres and cylinders embedded in finite scattering and absorbing material has been programmed [16], assuming the neutron economy is governed by two-group diffusion theory. Upper and lower bounds for the critical size of regular arrays of spheres and cylinders embedded in scattering and absorbing material can be obtained by applying the mean value theorems of multi-group diffusion theory [17]. These bounds are sufficiently sharp in some cases to serve as checks on interaction theories.

An interesting application of the Monte Carlo method is in checking the safety of the British form of transport container. The principle used in designing this container was to surround the fissile material with a sheet of cadmium, and then at least a 4 in. thickness of wood in such a way that the contents of the container may be limited by mass or controlled by limiting the internal diameter of the wood surround. The permitted contents were calculated by one of us (R.T.A.) on the basis of a form of lattice theory appropriate to fast and thermal systems, and tested experimentally for a few loads in connection with the interaction parameter method by another of us (A.F.T.). A more detailed analysis of the safety of the container has subsequently been made on the following lines by Woodcock [18] for a variety of fissile loads, timber thickness and composition, etc.

Let p_1 be the probability that a neutron entering the container escapes without reaching the fissile contents;
p_2 be the probability that a neutron entering the container reaches the fissile contents;
p_3 be the probability that a neutron leaving the core escapes from the containers;
M be the surface multiplication of the unreflected fissile contents;
R be the probability that a neutron leaving the core is reflected back to the core;

then the condition for the container to be a net absorber of neutrons is taken to be:

$$p_1 + p_2 p_3 \, \mathrm{M}/(1 - \mathrm{RM}) < 1$$

These five parameters were considered separately in the Monte Carlo calculations and their dependence on the fissile contents and the surrounding wood were determined.

Concluding Remarks

A number of methods of dealing with problems of neutron interactions have been outlined and discussed. Fundamentally, there are two approaches to this complex problem by those faced with decisions: the safety advisers, who must decide or confirm spacings and arrangement or safe masses in relation to economics as well as safety. Firstly, there is the empirical approach based on analogies with systems which are known to be safe, exemplified by the Oak Ridge method, the Density Analogue, and Kenyon's rules. Secondly, there is the theoretical approach based on either well-established types of calculation for problems of reactor physics or fundamental experiments (e.g. the unified theory and the general form of the interaction parameter method).

There is no strict dividing line between what we have called the empirical and theoretical approaches. For example there is similarity between the Oak Ridge method and the interaction parameter method. In each there is a geometric parameter and another that describes the neutron-multiplying properties of a unit. The difference, substitution of the familiar k for the more appropriate surface multiplication factor within the interaction parameter, suggests that the one treatment is a special case of the other. Paxton and Woodcock came to the conclusion, " Nevertheless, the solid angle method, though valuable in many contexts for establishing conservatively safe spacings, becomes cumbersome when applied in detail to arrays of large numbers of units such as may be anticipated in transportation. Though it is easily possible that an approximate description based on the solid angle-k method and extended to deal with cubical stacking may be useful for correlating and generalizing the characteristics of large critical arrays, the empirical treatment that results is likely to retain little of the basic concept of a summation of interactions between pairs of units and it would be preferable to use the interaction parameter approach ".

The choice of approach is largely governed by cost, but it is also dependent on the facilities available for computations. For a fixed store with well-defined operating conditions there may be little to be gained by taking the second approach. For example, one can argue by analogy that it would be safe to store more than one half-tonne of U 235-metal as 5 kg billets in a room 10 ft $\times$ 10 ft $\times$ 8 ft, and one would scarcely want to design a smaller room for such a quantity. The limiting factor for the size of the store is not always the closeness of permissible spacing of units; it often is the operational convenience of loading in and taking out of material. On the other hand, in a plant containing uranium compounds in a variety of vessels, and under various degrees of moderation, it would be preferable to adopt the second approach. As a general rule the second approach should be adopted when it is necessary to examine in detail the effect of spacing, shapes, a variety of fissile materials, etc., i.e. when it is not unreasonable to spend time and money on the investigation.

For the second approach one needs a theory which can cope with the significant physics of the interaction problem. The real test that theory is including all the significant physics of the problem, is a comparison of theoretical results with those from a number of well-chosen experiments. There are, or there will be shortly, sufficient experiments on pure metal and pure solution systems of U 235. What is required is a sufficient number of experiments on systems of various uranium enrichments, and various degrees of moderation, particularly for poorly-moderated systems. The effects of

external reflectors and moderators also require to be investigated. There does not appear to be a pressing need for interaction experiments with plutonium, since an adequate knowledge of isolated plutonium systems coupled with a satisfactory theory of interactions for uranium systems is considered to be an adequate foundation for the treatment of interaction problems in plutonium and plutonium-uranium systems.

REFERENCES

1. R.T. ACKROYD, E.J. BURTON and M.A. PERKS, " A Unified Theory of Interaction for Two Arbitrarily Mixed Multi-group Arrays Set in Well-defined and Random Reflectors ", *J. Nucl. Energy, Part A: Reactor Science* V. *11*, 57 (Feb. 1960).
2. R.T. ACKROYD, M.A. PERKS and E.J. BURTON, " A Unified Treatment of Interaction Problems Using the Magic Shell Concept ", *IGR-TN/R.951* (1958).
3. R.T. ACKROYD and J.D. MCCULLEN, " Albedo Calculation Methods " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 38.
4. R.T. ACKROYD and J.M. BALL, " Multi-group Version of the Albedo Method ", *Progr. Nucl. Energy, Series IV, Technology, Engineering & Safety*, V. 3, p. 297, Pergamon Press, London.
5. R.T. ACKROYD and J.M. BALL, " Application of Spherical and Cylindrical Means to Reactor Theory ", *IGR-TN/R.914* (1958).
6. S. GLASSTONE and M.C. EDLUND, *The Elements of Nuclear Reactor Theory*, Van Nostrand, New York, 1952.
7. E.C. MALLARY, H.C. PAXTON and R.H. WHITE, " Safety Tests for the Storage of Fissile Units ", *LA-1875* (1955).
8. J.K. FOX, L.W. GILLEY and D. CALLIHAN, " Critical Mass Studies, Part IX, Aqueous U 235 Solutions, " *ORNL-2367* (March 1958).
9. F.G. PROHAMMER, " A Comparison of One-Dimensional Critical Mass Computation with Experiments for Completely Reflected Reactors ", *ORNL-2007* (1956).
10. D.E.J. THORNTON, " Criticality of Poorly-Moderated Plutonium Systems ", *DEG Report 6 (R)* (1959).
11. H.F. HENRY, J.R. KNIGHT and C.E. NEWLON, " General Application of Theory of Neutron Interaction ", *K-1309* (Nov. 1956).
12. H.F. HENRY, J.R. KNIGHT and C.E. NEWLON, " Self-Consistent Criterion for Evaluation of Neutron Interaction ", *K-1317* (Dec. 1956).
13. H.F. HENRY (Ed.), " Studies in Nuclear Safety ", *K-1380* (Aug. 1958).
14. H.C. PAXTON, " The Density-Analogue Treatment of Arrays " (Private communication to E.R. Woodcock).
15. D. KENYON (to be published).
16. " Interaction Calculations using Diffusion Theory ". Code prepared by M.F. Mitchell.
17. R.T. ACKROYD and J.M. BALL, " Application of Spherical and Cylindrical Means to Reactor Theory " (to be published), abstracted in *Proc. Int. Math. Congr.* Edinburgh, p. 35 (1958).
18. E.R. WOODCOCK. (Unpublished).
19. R.A. SCRIVEN and A.F. THOMAS, " Neutron Interaction in Fissile Assemblies ", *Progr. Nucl. Energy, Series IV, Technology, Engineering & Safety*, V. 3 Pergamon Press, London, 1960.

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DISCUSSION

H.C. PAXTON, United States:

I appreciate the chance to be first to confess his crimes! My motive in developing the really very simple density analogue correlation was that I felt a need for a useful picture of cubic lattices of air-spaced units. This was largely because I had to help designers make some guesses about storage arrays that might be reasonable. I like to be able to make the initial guess, at least, without going to a machine of any sort, without solving any large matrix, and I suppose that was what led me to look upon an array of units as similar to a single low-density unit. The use of essentially the type of density exponent that Mr. Woodcock mentioned in his paper, arose simply from empirical observation. The number of units of the critical array against the lattice density looked very much like what we saw when we looked at a single unit at decreasing density; i.e. a power relationship. The power for cubic air-spaced systems must depend upon some measure of reactivity of the unit that is being used. Surface multiplication would be the ideal thing with which to correlate; unfortunately there is little enough experimental information about this (but there is obviously going to be more). The power must also depend upon the sort of reflector that can be around the array, and this is the basis for the correlation. Now, it is not intended to apply very generally to interaction problems, it is intended to apply simply to the sort of system that I mentioned: cubic, or spherical if you wish, regular arrays of units. With regard to its use, if one takes the correlation seriously one asks the following sort of question: We would like to design a storage vault, say for plutonium metal. We propose to determine how many cubicles there may be, if each kg of plutonium is to be assigned to 10 litres of space, i.e. plutonium metal with a normal density of 20 kg/litre. The reduction in overall density is then 200. The critical mass of plutonium metal — let's take it reflected — is, again for the high-density phase, about 5 kg. Then if the inverse first power relationship between density and critical mass holds, or is conservative, one would say that the storage limit could be perhaps one-half the estimated critical mass of 1,000 kg. For 100 kg we should have no problem. Now the conditions under which the reciprocal first-power law would hold, according to what we have seen so far, would be that the individual unit would be no greater than one-third of the critical mass. The storage problem relates to units well within this range. So we know that there is a flagrant under-estimate of the critical mass in this particular case, where one kg units would be stored, but often we discover that this conservatism leads to a convenient sort of storage array. Of course, we are also looking at an extreme for the entire storage array. Usually cubicles would be arranged along walls, sometimes down aisles, and we are looking at it as though all of these storage compartments were stacked together in a cube instead of being spread out as in an actual vault. It is simply for this sort of convenience that I have pursued the density-analogue method; it is of little value, at least in its present stage, for plant layout problems. I might mention that there is another American method that I suspect must be very similar to the unified treatment, and this is the method that Hugh Clark of Savannah River uses, where he expresses the influence of neighbouring units in terms of reflector savings of an individual unit that he is considering.

A.N. DICKSON, Dragon Project:

I would like to ask Dr. Ackroyd a question about Kenyon's method. Can you always assume that this gives you a safe answer, because it seems to me that if you apply it to the natural uranium graphite system, it would give you an optimistic answer? I believe that a homogeneous arrangement of natural uranium and graphite cannot be made critical, whereas an inhomogeneous arrangement can.

R.T. ACKROYD, United Kingdom:

I don't know whether one can apply this method to graphite systems. I would suspect you could if you had the appropriate data. The reason why I am particularly interested in the density analogue treatment, is that there are some very general principles at work on the criticality of arrays of materials. Some of these deal with the geometrical properties of the system. An array is really only a special case of non-uniform distribution of fissile material. Some effort in the geometrical properties of distributed forms of fissile material was initiated by myself many years ago, and this has recently been taken rather further in the form of geometrical theorems by Mr. Dressner, who has considered the effects of changing the shape of assemblies. Unfortunately, all this work on which, I think, the density analogue methods hinge, is at present confined, because of mathematical difficulties, to the giving of rigorous proofs for one-group systems. One suspects that if one could only find the appropriate mathematical key, one could put many of these rules, like the density analogue method and Kenyon's rule, on a reasonably respectable mathematical basis. At the same time, one could probably sharpen the estimates of safety they give. All I hope is that people will try to apply them, try to find out when they don't work, and see if they can be made to work.

H. DOPCHIE, Belgium:

With regard to this problem of changing geometry, we have been working on this problem in Belgium, and we have, I believe, found a method which in many cases can be applied. It boils down finally to changing, by a very simple method, the albedo of the surrounding material. In Kenyon's method, you change the geometry of the elongated material into spheres or cylinders, or something easier to compute. It can be shown that by applying simple corrections to the albedo of the surrounding water, you can get a fairly good result on the multiplication constant of your material. It's difficult to outline the method here. It needs mathematical demonstrations to show where we are getting at. Let us only say that from the albedo, or the albedo matrix in multi-group theory, one can obtain the admittances. The admittances have then to be multiplied solely by the ratio of the surface area of the body you are studying to the surface area of the equivalent body, when you change the geometry.

R.T. ACKROYD:

I think that a correction factor to the albedo admittance, by bringing in the surface area presented by a body, is implied in the Oak Ridge method. I am very glad to see that some of these ideas are being taken further.

A.J. MALLETT, United States:

The problem of interaction and the great attention it is getting is quite interesting. For spacing control, the Oak Ridge Gaseous Diffusion Plant uses the solid angle interaction method which is considered conservative. I would like to add this comment, that in the past year not only were a considerable number of interaction experiments completed but, in the interim, we improved our interaction method of calculating by the use of a cosine distribution of the flux across the array. This refined version enabled prediction of the critical spacing for the array of test cylinders within approximately 0.25 in.; it also gave quite good results when applied to spacing for metal balls and units immersed in water. In addition to the theoretical work, we are principally interested in simple formulae which can be used by an engineer to calculate, interaction-wise, the layout of his equipment, this being considered as much an engineering problem as calculating heat-transfer or fluid-flow.

G. BLÄSSER, Euratom:

I should like to ask a purely mathematical question concerning the report of Dr. Ackroyd. In the derivation of the two-group formula, you gave two formulae for the two coupling coefficients which hold only in the two limit cases $\omega = 0$ and $\omega = 1$. But this second formula which had this limited generality could be replaced, I think, by the formula without ω appearing separately in the numerator and in the denominator, and still you would have a finite expression for S_2 in the case that ω approaches 1, because then you have an expression of the type $\frac{0}{0}$ which can be calculated very easily and which gives a finite result. You have to find only a finite value for S_2 in order to draw the conclusion that Φ_2 vanishes in the core region. So, I think one can get this formulation quite generally.

R.T. ACKROYD:

The formulation which is quite general in the sense that it covers all cases, was chosen in this form simply because if you do not put in an extra factor, in taking the limit, as ω tends to 1, you will find when you work it out that the limit is not the same as putting $\omega = 1$. If you look very carefully at both the mathematics and physics, you will see that to do the mathematics properly you have to bring in a cross section which changes with the weight of the spectrum. If the spectrum is all fast, then a thermal cross section is not important; and you have to use this concept physically. Now if you do this, you can get a sort of a one-and-a-bit theory where the bit varies between 0 and 0.5, so to speak. Mr. Thornton has done this (I think you saw one of the results of this work on predicting the effect of Pu 240 in a former presentation.) There are other factors taken into this calculation, but when we had formulated this theory and written down our equations, we decided that we didn't want to go back and re-write something like fifty pages of work. It was sufficient from the mathematical point of view to say: put $\omega = 0$, or put $\omega = 1$, and the requisite result comes out. If anyone wants to re-formulate it in a way which looks less artificial, so you can have ω vary continuously, between 0 and 1, then this can be done. You will have to bring in the physical

principle to guide the mathematics. The mathematics is perfectly respectable, but to choose the right key you will have to think physically about the problem first.

J. GUNTHER, France:

Nous désirerions savoir, en premier lieu, si l'on a essayé, au Royaume-Uni ou aux États-Unis, de comparer les résultats de différentes méthodes de calcul qui nous ont été exposées aux résultats qui auraient été obtenus par la programmation sur machines électroniques de la seule équation qui soit rigoureuse en la matière, à savoir l'équation du transport. Cette constatation est particulièrement vraie lorsqu'il y a des vides car, si l'équation de la diffusion n'est certainement pas applicable dans ce cas, elle peut par contre, donner au moins une idée de ce qui se passe lorsqu'on a un système analogue à celui utilisé au Royaume-Uni pour le transport de matières fissiles, c'est-à-dire un système qui consiste à entourer ces matières de modérateur. Lorsqu'on a un vide, seule l'équation du transport permet d'obtenir une solution rigoureuse. Aussi, désirerions-nous savoir si l'on a essayé de comparer les résultats de la programmation TDC, par exemple, avec les résultats des différentes méthodes de calcul approché qui nous ont été exposées.

En deuxième lieu, il convient de remarquer que les différentes méthodes de calcul approché qui nous ont été exposées sont pour la plupart valables surtout lorsque les récipients sont distincts. M. Ackroyd a déclaré, au début de son exposé, qu'on pouvait faire disparaître tout problème d'interraction en éloignant suffisamment les récipients. Cela s'avère toutefois impossible dans certains cas, à savoir lorsque se posent des problèmes d'intersection de tuyaux ou, d'une façon générale, des problèmes de récipients qui ne sont pas convexes, c'est-à-dire, présentant des faces externes qui se regardent à travers le vide. Il me semble d'une part, que plusieurs des méthodes valables pour les récipients séparés, cessent de l'être pour des récipients de ce type et que, d'autre part, on ne peut pas faire disparaître le problème, même au prix d'un sacrifice d'ordre économique, en éloignant les récipients, comme on le fait lorsque les récipients sont complètement isolés. Je me demande, en conséquence, s'il ne serait pas nécessaire de consacrer un plus grand effort à ce type de récipients, qui peuvent présenter un certain intérêt pratique, tel le mélangeur-décanteur en plaques minces, coupé par des cylindres verticaux, dont on a parlé précédemment.

R.T. ACKROYD:

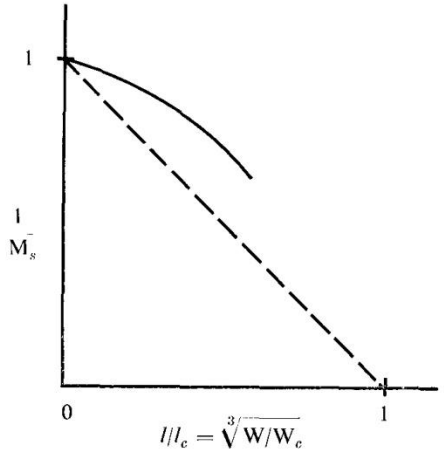
With regard to Mr. Gunther's first question, I would say that some of the details in the crime of the unified theory of interaction have escaped the attention of Inspector Maigret! The first point is that only diffusing neutrons are treated by the diffusion theory. There are two other classes of neutrons, those which go clean through the air, which are treated by the rules of geometrical optics; and those which tunnel and are treated by the transport theory of removal. You will see in the case of concrete and water between spheres, that we do introduce the removal concept. A certain fraction of the fast neutrons is assigned to a removal group. With regard to the other question, I think that we can now go on to a quite advanced field of work which we have only briefly mentioned in the report. Some of this work has been done at Aldermaston, and it concerns the transport theory.

You well know that these transport codes refer to highly specialized forms of geometry, which scarcely arise in big production plants but may arise in very specialized assemblies or units. You can test the unified theory and the interaction parameter methods by these codes, and the results are quite encouraging. It is obvious that they should be, because we know, in the case of isolated vessels, that one can get quite good results by using an appropriate form of diffusion theory. The only additional factors we have introduced are the coupling by direct ray, coupling by attenuation or tunnelling and the effect of reflection. We have used the transport codes, but when we come to the problem of other bodies, say coaxial cylinders, or concentric spheres, one can't use these codes. I'll mention two problems, the problems of two spheres and two cylinders, which have been done at Aldermaston very nicely by noting that in the problem of, say, two spheres, there are only two coordinates: the distance measured along the axis joining the two spheres, and some polar coordinate measuring a radius from that axis. By using the appropriate transformations, we can transform the case of two spheres in an infinite moderator, into the case of two rectangular blocks on an infinite strip. Now, we have done a very similar technique at Risley, by modifying one of our fast reactor programmes which concerns the insertion of control rods of any configuration in any finite medium. We have replaced the fissile material by a moderator, we have replaced the absorbing control rods by fissile material (just by changing constants). In this problem we use a mapping technique again, and we map the whole of the reactor into a strip. Then instead of getting a diffusion type equation, with constant coefficients in each region, you will find that the coefficients vary from position to position. Now, with a digital machine, these equations are fairly easy to solve. It must be pointed out that you can do this effectively if, in the transformation, the operator ∇^2 does produce something like ∇^2; i.e. that mixed derivatives don't appear in your transformed equation. There are also some stray first-order derivatives which are bound to arise, and the terms related to generation or absorption of neutrons. Provided that the second order differential coefficients are of the form of $\frac{d^2}{dx^2}$ or $\frac{d^2}{dy^2}$, you can get quite nice solutions, in the sense that you know that they converge very rapidly on the computor. These kinds of problem have been used to check both the interaction parameter methods and unified theory methods. I think Mr. Thomas might have something to say about a very specialized assembly where, instead of measuring the interaction coefficients, these are actually being computed by a rather advanced method.

A.F. THOMAS, United Kingdom:

We have been in the habit of confining ourselves until very recent years to the use of parameters derived from experiment. The units we wanted to store or to move have been represented fairly easily and without too much conservatism by simple models — spheres in particular. It has been comparatively straightforward to measure the interaction parameters directly and, as Dr. Ackroyd says, nature does its integration for you. However, we have had programmes designed to compute interaction parameters, usually Carlson's S_n on spherical models for bare spheres and for spheres reflected by thin shells of graphite, this in particular reference to casting problems — and

to systems partly or fully reflected by water. This has been compared with our experimental results, and the results are quite pleasing. I would also like to comment on a point Mr. Mallett raised, namely that the engineer or the designer needs a quick method of getting a reasonable idea of what spacing he should use. We can offer one quick way, which is probably conservative but will give the designer some idea of what spacing he should give the lathes or storage arrays, remembering that when you get right down to the heart of the problem, it is not one of bringing the storage array down to the minimum spacing possible; it is showing that the spacing, which is set very often by other considerations — the size of lathes or the fact that people have to wheel trolleys to pick up fissile material in rather large lumps, between storage pits or between storage racks — provides safety. It turns out that what we have to do is to produce an unequivocal demonstration of the safety of the array whose dimensions are essentially fixed, so that conservatism doesn't matter too much. It is possible to derive an interaction parameter by assuming that the reciprocal of the surface multiplication varies linearly with the cubic root of the mass, i.e. with the linear dimension of the system. If we plot the reciprocal of the surface multiplication against the linear dimension divided by critical dimension, or the third root of the mass divided by critical mass, the reciprocal surface multiplication must be unity for zero and must be zero at the critical point. So, if we say that the variation is linear, we can get a quick estimate of the surface multiplication. Mr. Woodcock has done quite a few calculations on objects of various sizes and compositions and shapes, and except possibly for a certain lack of conservatism near M = 1, the systems have the reciprocal surface multiplication curve concave downwards.



Hence we overestimate the surface multiplication. So, given that you know the critical size of, say, the sphere or cylinder of the composition you want, you then get $\frac{1}{M} = 1 - \sqrt[3]{\frac{W}{W_c}}$ where W_c is the critical mass and W the mass considered. Given M_s, you can say that the interaction parameter, i.e. the number of neutrons which leave a body for unit source at the distance of the next object, which we call q, is equal to M_s times the solid angle subtended

by the body at the source, divided by 4π. That is, $2 = M_s \cdot \frac{\Omega}{4\pi}$ and it appears to give an overestimate of q as far as we know, in all the systems we have looked at, and is a good, quick guide.

R.T. ACKROYD:

Apart from these advanced calculations that have been done in calculating the interaction parameters, I think that the unified theory is the only theory that has been produced so far (and which I like best of all) which starts with cross sections that apply solely to isolated vessels and proceeds to interaction problems. I think all the other theories do incorporate certain factors based on, say, one interaction experiment. Whichever theory you wish to use will depend upon the results you get from the various methods. With respect to re-entrant bodies, I think one could genralize the theory to deal with the re-entrant bodies, but I don't think that the effort is generally worthwhile, in the following sense. Provided you keep the same multiplication for the re-entrant bodies in the array, but smooth the surfaces so they are regarded as convex for interaction only, then with a little care you will get quite a safe array, which is not too conservative. This is probably all you would require to do in the few examples of, say, mixer-settler units, which present a re-entrant outline to each other.

D. CALLIHAN, United States:

We internally, in the United States, enjoy a bit of bantering among ourselves on the solid angle *versus* density method. I point out that I believe, with sincerity, that they are both empirical, and that the motivation for each is expedition in a method of practical application. Their similarities are obvious. Whether one speaks of a unit as being such a fraction critical or whether one assigns a value of k — perhaps ill-defined but, nevertheless a value of k — to it, is somewhat incidental; it is also somewhat incidental whether one speaks of volume fraction of an array occupied by the fissile material or whether one speaks of a solid angle, for the two concepts are quite similar. What I really wish to do is add a bit of confusion, at least in the two so-called United States methods, by reciting the results of two experiments. During the second session we commented on a series of experiments being performed with a number of individually subcritical units of fissile material. These were solutions, of some 12 or 14 cm diameter, and 60 cm or more in various cases. These bottles, were arranged in patterns with their axes parallel and vertical. In one experiment, the external dimensions of the array were unchanged, and a fraction of the individual units within the array were rotated through 90°, resulting in a number of units with their axes vertical and a number with their axes horizontal. In a comparison of that array with the one before this simple rotation, the density remained the same, but the measured reactivities were different. For the discussion of the second experiment (I point out that one of the conditions of, or one of the assumptions made in the solid angle method is that effects of vessels shadowed by another vessel can be neglected). In this experiment, it was found that some 18 of these vessels placed in line adjacent to a reflector were subcritical, whereas the addition of the 19th made the system quite critical, an effect for which the reflector is partly responsible. So, I say there are deficiencies in the United

States methods. I do point to something that I think is quite evident, at least from what has been shown and discussed. In my opinion the United Kingdom and the French teams have done considerably more basic analyses of this problem than we have. I think the United States approach, regardless of the name by which it may be called, is very largely empirical and practical, and has attempted to meet the need for plant layout design.

H.C. PAXTON, United States:

I agree that we have not done as much theoretical work as we should have, not nearly as much as in other countries. This does not lessen our interest in their work. I might just mention a practical problem: we asked a man in our group to use something very much like the interaction parameter method with a minimum of distorting approximations, to try not to introduce approximations that would bias data, but simply to try to get a feel about the behaviour of arrays, answer some of the questions about the simple-minded density analogue picture. For example, we expected the assumed constant exponent for a reflected array to break away toward the minus second power, the value for the unreflected array of very small units. Questions such as this we wanted to get some angle on, to see whether we could find a more general basis for the density analogue picture. Well, to put it briefly, we lost the man!

W. SCHÜLLER, Eurochemic:

I should like to come back to the second question which Mr. Gunther brought up earlier, i.e. the problem of interaction of different parts of the same equipment which are not separated but form one body. Unfortunately such problems occur quite often. If I remember correctly, the Oak Ridge Report K 1019 gives a description of the application of the solid angle method to such problems, which I have not entirely understood. Maybe Mr. Mallett could comment on that or say how in practice these problems are solved?

A.J. MALLETT:

Actually, where pipe connections have been proved experimentally to be geometrically safe, we have used two methods. In one, the most reactive position from which to evaluate the interaction effect of adjacent units is considered to be a distance of about 3 times the pipe diameter from the pipe connection; in the other, the centre of the pipe connections is considered to be the most reactive position. It is partly a question as to how close the other units are. Small pipes, such as sample connections or feed lines, which are not larger than 1 in. and are spaced 15 in. apart, are not considered interactionwise when connected to a pipe of safe geometry.

R.T. ACKROYD:

I would like to add just a comment on the body which interacts with itself. Many years ago we studied it but we haven't looked at these problems in any detail since then, because we have not been unduly worried about them. However, Mr. Charlesworth and myself did do one-group analysis of two intersecting cylinders set in a moderator. This method works quite nicely from the point of view of getting a conservative answer which is not

too conservative. All you have to do in the example of a cylinder penetrating a slab, is to replace the actual material by a less reactive material in the region where the crossing of cylinder and slab takes place; then you can do the analysis in the purely classical terms in a finite close solution, and this turns out to be reasonable enough. Since then, the advent of computors allows you to tackle a problem like this without making the 'crossing' approximation, but I don't think it is worth doing a numerical solution. If anybody were interested, I think some of the transformations that have been considered at Aldermaston and at Risley for transforming a homogeneous system of awkward geometry into a system of clean or simple geometry with a heterogeneous core or reflector, could probably be applied to many of these problems.

A.F. THOMAS:

I would like to add two points of caution to those given by Dr. Callihan. Anyone who wants to use the solid angle type of approach, whether the Oak Ridge method or the interaction parameter method or any other such method (i.e. to use some coefficient of reactivity and the average solid angle subtended to one body by another) must be very careful to use the right distribution of neutrons at the surface. Dr. Ackroyd has said that the surface multiplication in one treatment appears to be independent of the distribution of neutrons over the surface. We have shown by experiment that it is not independent of the distribution in angle of the neutrons as they hit the surface. The surface multiplication certainly decreases as the source of neutrons approaches a sphere, that is, as the neutrons striking the sphere become less parallel and in effect become near-isotopic on the surface of the sphere. The other point is to be careful when you have systems which are near large reflectors. A particular case of this occurs in a storage array — bottles or birdcages on the ground or arrays in workshops — where the items are at most a few feet from the ground; you have to be very careful to look at the neutrons getting from one body to another, not by direct but by scattered paths. The proportion of neutrons reaching a sphere of 20 kg of U 235 appears to be about 40 % greater for spheres 2 ft 6 in. apart, when they are about 2 ft above the ground, than when they are about 6 ft above the ground. The moral here is that while we must be prepared always to give the plant designers simple, easy rules for getting a plant which has a very good chance of being approved by specialists, we should not forget that the specialists still want to have a look at it.

CRITICALITY ASPECTS OF THE TRANSPORT OF FISSILE MATERIALS — THE TECHNICAL BASIS OF THE PROPOSED I.A.E.A. REGULATIONS

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INTRODUCTION

The increasing use of radioactive materials throughout the world is making the movement of these materials progressively wider, more frequent and larger in volume. Although regulations for the safe transport of radioactive materials have been in existence in some countries for many years, it has now become necessary to modify or supplement the existing provisions on an international basis. Because of the increasing number of shipments from one country to another, harmonized regulations for safe transport should be adopted and applied uniformly by all countries. It is also desirable that the essential requirements be, as far as possible, common to all modes of transport, so as to simplify the procedures to be complied with by shippers and carriers.

Uniformity can be achieved only by international agreement on the basic conditions of safety. The International Atomic Energy Agency has been engaged in the establishment of safety standards to cover all activities involving ionizing radiations, and transport is one of the first and most important problems being studied.

Considering the different technical and administrative aspects of the problem, the International Atomic Energy Agency convened two panels of experts to draft comprehensive regulations for the safe transport of all types of radioactive materials. One of the panels has examined the safety problems of transporting fissile materials and materials of high activity, while the other has studied the transport of radioisotopes, ores and residues. The work of the two panels has been complementary and the regulations drafted separately by them were put together in a single set of provisions to cover all aspects of the problem.

In September 1960 the Agency's Board of Governors and the General Conference approved the application of these comprehensive regulations to Agency operations and recommended that they be taken as a basis for national regulations as well as for regulations relating to international transport.

The main factor in the safe transport of fissile materials is the criticality hazard. This paper discusses the problems involved in guarding against

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the possibility of a criticality incident during transport, with special reference to the provisions of these proposed regulations.

Criticality Hazard

As a result of the continuing increase in the use of fissile materials, especially uranium enriched in U 235, and Pu 239, the importance of criticality hazard control must be brought to the attention of all users and potential users as well as processors and shippers of fissionable materials. Careless handling of fissile materials can cause — and has caused in a number of cases in the past — supercritical assemblies to be formed, which can dissipate a large amount of energy. If released in a short time, this energy can cause considerable damage and, in any case, is always accompanied by a serious radiation hazard. The burst of radiation could possibly be lethal within a radius of several feet and produce radiation injury to personnel over bigger distances. The prompt evacuation of the personnel could prevent serious radiation exposures from fission products; but both short-term and long-term problems of a decontamination after a criticality incident could prove formidable. Also, the possibility of an explosion-type of energy release, causing considerable mechanical damage, must not be neglected.

Conscious of the need for utmost care in devising nuclear safety rules, the Agency has adopted the criterion of 'double contingency', meaning that not one but two independent and simultaneous accidental events are necessary before the fissile material can become critical. It should be noted that this is a great deal more stringent than the conditions of transport applied to ordinary hazardous chemicals and explosives. For example, with many chemicals, all that is required to create a dangerous situation is the rupture or leakage of a container. Under the Agency's proposed regulations such an occurrence alone would not produce a criticality incident, but it would have to be coupled with a second and normally unrelated accidental event before criticality could take place. The very nature of the hazard and the comparatively casual activities which can initiate it make it necessary to establish and strictly observe practical nuclear safety criteria and procedures.

Basic Considerations in Transport

In the handling of fissile materials a chain-reacting or critical configuration may be achieved inadvertently through carelessness, ignorance or accident. Such configuration is effected by several factors including U 235 enrichment, concentration, density, quantity, geometry, presence or absence of a reflector or moderator, presence of neutron absorbers (nuclear poisons) and interaction between assemblies of fissile material. Nuclear safety is, in general, achieved by limiting the mass or concentration in any one place, or by design of the shipping container so that the geometry for self-sustained fission is unattainable.

Nuclear safety during transport should not depend on the assumption that a reflector will be absent, since so many of the materials encountered during shipment are excellent reflectors; for example, rain or snow, the material from which walls in storage areas are made, personnel working in and around the packages, and other shipments and shipping materials. Further-

more, cans, bottles or other containers must be considered as partial reflectors for the fissile materials which they hold. The presence of any neutron absorbing material should not be taken advantage of as a primary safety control, although nuclear poisons in a system are sometimes practical to supplement other safety measures or to increase an existing safety factor.

To be considered nuclearly safe, a shipment must require the occurrence of at least two unlikely, independent and simultaneous changes in one or more of the conditions originally specified as essential to nuclear safety before a nuclear incident is imminent. Such changes should be unlikely because of positive action taken to reduce the actual chance of occurrence to an insignificant level. This is an application of the double contingency principle.

Where the package cannot be insured against leakage of water, full or partial moderation should be considered. The Agency rules require that all but inherently safe forms and very small amounts of fissile materials be packaged in leak-proof containers and the entire package, including the container and any built-in spacing, must be designed to withstand the maximum credible accident. It was assumed, however, that reflecting materials which are more efficient than light water (e.g. beryllium, pile-grade graphite and heavy water) would not be encountered in transit under conditions which would require them to be considered as full reflectors. Shipments of fissile materials which contain more than trace amounts of such special reflectors require special consideration. It is recognized that some factor of safety must be allowed for uncertainties in the experimental or calculated data used. For instance, if mass is the controlling factor, the permissible value in any single package must not exceed 80 % of the critical mass under the conditions of packing, with due consideration for built-in neutron absorbers. Where geometry is the controlling factor, the permissible value for each controlling dimension must incorporate a 10 % safety factor.

Classes of Shipments of Fissile Materials

First, there are exemptions from the basic mass or dimension limits. Packages containing U 235, U 233 or Pu 239 in small quantities or low concentrations are exempted from the provisions in the regulations for the transport of fissile materials. Those that are covered by the provisions of the regulations are divided into three classes:

Class I: Shipments in packages such that each package is safe from neutron interaction in any arrangement;

Class II: Shipments in packages which are nuclearly safe in any arrangement under special limitations regarding the number and content of packages;

Class III: Shipments under special arrangements.

Exemptions

Shipments in which the only fissile material present is natural or depleted uranium where the U 235 content is no higher than 0.72 % by weight are exempted, provided that such packages do not contain more than trace quantities of beryllium, graphite or heavy water. This provision reflects

the belief that a critical assembly cannot be formed with such packages in any number or arrangement.

Further, shipments of aqueous or other solutions are exempted, in which the only fissile material present is:

a) Uranium, in any enrichment, in which the ratio of H/U 235 atoms is greater than 5,200, corresponding, in common aqueous solutions, to a U 235 concentration of less than 5 g/litre ; or

b) Plutonium, in which the ratio of H/Pu 239 atoms is greater than 7,600, corresponding, in common aqueous solutions, to a Pu 239 concentration of less than 3.5 g/litre.

For exempting small amounts of fissile material from criticality control, requirements were that five times fifty packages should be brought together safely, even under water. No more than fifty packages are, in any case, to be carried in any one vehicle, according to other limitations in the regulations.

Other restrictions on packages for radioactive materials require that they be leakproof and have no external dimension less than 4 in. Because of the minimum size, the volume of each package is at least 1 litre.

Based on the minimum critical mass at the appropriate density for a water-moderated, water-reflected system with a homogeneous core and using a minimum critical number of packages of 250, the maximum content per package was conservatively taken as 16 g for U 233 and U 235 and 9 g for plutonium.

Approvals for Shipment

For other forms, higher concentrations and greater amounts of fissile materials, added restrictions are imposed to ensure safety. First, all such shipments must be approved by the competent authority. This means an independent check of the criticality controls by a nuclear safety specialist who can assess the adequacy of the shippers' controls. Containers for such shipments must be approved by the competent authority and must be capable of maintaining their integrity as to leakproofness, spacing and shape following the most serious accident likely to happen on the chosen mode of transport.

Class I. Safe from Neutron Interaction

A neutron sink, or anything which absorbs more neutrons than it produces, does not contribute to a chain reaction. By properly combining absorbers and moderators, a shipping container can be made a neutron sink.

A type of package meeting these requirements consists of a 30.5 cm (1 ft) diameter cylinder of wood, having a coaxial cylindrical cavity such that this cavity is surrounded on all sides by at least a 10.2 cm (4 in.) thickness of wood. The central cavity is cadmium plated to a thickness of at least 0.38 mm (0.015 in.) giving a minimum of 0.325 g cadmium/cm^2 of surface. A square cross section package measuring 30.5 cm (1 ft) on each side is also suitable, provided the fissile materials are confined within the cylindrical space of specified dimensions. This geometrical limitation represents one way of controlling criticality. The diameter of the circular cylinder depends, of course, on the density and hydrogen content of the wood and also on the chemical composition of the fissile material.

For uranium or plutonium metal it may not always be possible to use the geometrically limited package. For such cases a mass limitation can be imposed.

The fissile material is surrounded by cadmium and the whole surrounded by wood, so that the same outside dimensions as above are achieved. The permissible mass of metal then depends on the density and hydrogen content of the wood.

It is of interest to note that one can experimentally check whether any container is a neutron sink by introducing the filled container into the central cavity in a suitable reactor and determining that the reactivity of the reactor decreases.

Class II. Limited Numbers and Quantities

By limiting the contents of packages and controlling the number of such packages to be collected together, shipments of fissile materials can be made safe from criticality. Class II allows five such loads to collect together, even under accident conditions, without a risk of criticality.

Several methods of establishing criticality control are available. The method on which the proposed Agency regulations are based deal with easily measurable quantities, such as the quantity of fissile material per container, the volume of the container and the total volume of the package, rather than with more specialized quantities like k_{0ff} or solid angle. The amount of fissionable material allowable in a package varies with the ratio H/X, where H represents the number of hydrogen atoms and X the number of fissile atoms. The allowable number of such packages depends on spacing. A curve is developed relating the allowable number of packages to the package volume. It would be desirable to investigate other practical methods. The present method has been developed by the United States group engaged in revising the existing national rules.

In regard to the conservatism of this method, the authors have indicated that the factor of safety varies somewhat with the H/X ratio. For 20-in. birdcages, for example, in the absence of interspersed moderation, the safety factor ranges from twenty for an H/X of less than two to ten at optimum moderation. That is, the minimum critical number for an H/X below two is about 1,000 units, and for optimum moderation is an estimated 462 units.

To allow for the possibility of moderating material becoming interspersed between the packages, a safety factor of 10 has been applied. This is based on experiments with slabs of high assay uranium. For geometries other than slabs, the moderating material would probably have a smaller effect.

Class III. Special Shipments

It is to be recognized that fabricated shapes, alloys and clad fuel elements of different designs are of a very special nature in themselves and must be considered individually. Until experience has been gained in administering the rules of safety in transport and until the transport of such special forms of fissile materials becomes more nearly routine, such shipments should be made as Class III shipments under special arrangements. Also, of course, any shipment which, for any reason, does not fit into the rather restrictive pattern laid down for routine shipments, should be carried under special arrangements.

For special shipments under Class III the safety of the shipment must be demonstrated. The following possibilities must be taken into account, both singly and in combination, in demonstrating that the shipment is safe:

a) Credible accident (with the choice of the alternative leading to the highest reactivity);
b) Flooding;
c) Loss of water;
d) Intermixing with or becoming adjacent to another load of fissile material;
e) Intermixing with or becoming adjacent to other materials which may have neutron moderating or reflecting properties better than water.

The shipper may use many alternative methods of criticality control, provided that he can demonstrate the safety of the shipment and that he can convince the competent authority of the adequacy of his proposed control based upon the ground rules given in the regulations.

Irradiated Nuclear Fuel

The associated problems of heat dissipation, high levels of radiation and contamination control require special care in designing the containers and in handling the material. For example, the possibility of decay heat causing melt-down under adverse conditions requires the total fissile content of a package to be limited below the critical mass of the melted fuel, unless conclusive evidence is given by the shipper that melt-down cannot occur.

Scrap

A very special type of problem is the shipment of scrap fissile material. Scrap includes such things as shavings, filings or borings from machining fissile materials, the coolant filled with chips, etc. With such materials the enrichment, concentration, moderation ratio or density cannot be accurately known and some of the parameters may change considerably during transport. Therefore, the shipment of such residues or scraps should be made under conditions which assure nuclear safety, while assuming that each parameter which is not accurately known has a value which leads to maximum reactivity under credible circumstances.

Conclusions and Future Outlook

The Agency has just issued the proposed Regulations on the safe transport of radioactive materials, including fissile materials, which require criticality control in addition to radiation control. These Regulations make detailed provisions for a wide range of fissile material shipments. However, they give only the basis for considering many others. The Agency invites comments and considered advice on how to improve these rules and expects proposals for additions to them. These Regulations are to be studied and reviewed in the light of experience and new developments and will be subject to amendment as and when needed.

Some proposals are already being considered. Among them is the use of a special label for fissile materials which would list a ' critical value '. That

value would be used to restrict the number of packages containing fissile materials to a specified safe number. The procedure would be similar to that now used for controlling the radiation levels from shipments containing many packages. We would also like to include in the Regulations descriptions of any standard containers found to satisfy the basic criticality controls and designed to withstand a major accident without losing any of their effectiveness.

We encourage those who are working in the field of criticality control to forward any comments, suggestions and proposed additions to the International Atomic Energy Agency.

PRESENTATION OF "CRITICALITY ASPECTS OF THE TRANSPORT OF FISSILE MATERIALS"

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International Atomic Energy Agency

INTRODUCTION

It is rather late in the Symposium to discourse on the need for measures against criticality hazards in transport: let us assume that there is a need. It might be useful, however, to emphasize the need for practical and comprehensive rules for common carrier transport of fissile materials. Such rules must be easily understandable and applicable by shippers and carriers and must not impose upon them restrictions which are out of proportion to those with which they may be familiar in transporting other hazardous materials. They must, as far as possible, apply uniformly to all modes of transport and, if agreed upon, to all countries. The transport of fissile materials across the frontiers of countries will increase in the years to come, and there is obviously, therefore, a need for internationally-harmonized rules. It is on this basis that the International Atomic Energy Agency has gathered together a panel of experts and undertaken the task of preparing and proposing to all its Member States uniform rules governing the transport of fissile materials. These rules, which form a part of the proposed Regulations for the safe transport of all radioactive materials, are just about to be published.

The work of examining the existing national codes of practice — it is fortunate that codes of practice have existed in some countries for many years — and devising international rules so far as possible in line with them, was done by an international panel of experts, some of whom are here to-day. It will be interesting to see how these experts now feel about the work they completed some twelve months ago.

CRITICALITY FACTORS

A critical configuration in transport, as elsewhere, depends on several factors which have been dealt with adequately in the first lectures of this Symposium; the enrichment (if we are dealing with uranium), the fuel concentration, the density, the quantity, the geometry, of any moderating and reflecting materials and the interactions between normally subcritical components. Nuclear safety has, therefore, to be based upon the consideration of all the real and credible possibilities but, in the interests of safety, where there are several alternative possibilities, one must choose that which leads to the highest reactivity.

One important criterion or principle of safety which was adopted in the proposed IAEA Regulations is that of double contingency. Put plainly, this means that the transport arrangement must be so safe that two unlikely and normally independent events must take place concurrently in order to cause a criticality accident. This is a very good principle but it is perhaps not so easy to interpret it quantitively. We use the word " unlikely " because criticality control also consists in taking positive action to reduce the actual likelihood of the occurrence of any of these events to an insignificant level. The IAEA Rules require that all fissile materials excepting those in inherently safe forms and very small amounts, be packaged in leak-proof containers of the Type B which are completely clad in mild steel and so designed as to withstand the consequences of the maximum credible accident and maintain their effectiveness and integrity even under maximum credible accident conditions.

Exemptions

In these Rules the shipments of fissile materials are grouped in three classes; but outside these classes there are exempted quantities and exempted materials, which means that the transport of such materials or such quantities is exempted from the limitations as regards criticality control, but not from the limitations of radiation control. For instance, packages containing U 235, U 233 or Pu 239 in very small quantities or very low concentrations are exempted from the provisions of the Regulations as regards criticality control. Unirradiated natural uranium and depleted uranium are equally exempted, provided that they are so packed as to prevent the ingress of moisture and provided that the package contains no more than trace quantities of special materials such as beryllium, graphite or heavy water. Further, shipments of aqueous or other solutions are exempted if the ratio of hydrogen atoms to fissile atoms exceeds certain specified values. These are: 5,200 for U 235, which corresponds in a common aqueous solution to a concentration of 5 g/litre; 7,200 for plutonium which corresponds to a concentration of 3.5 g/litre. These values of the concentration are about one-half the safe values quoted by many speakers at this Symposium, including Mr. Woodcock and Mr. Gunther. They include, therefore, a safety factor of two.

If in any solution, the maximum concentration — it is better to go by the H/X value — is above 5,200 in the case of uranium or 7,200 in the case of plutonium, such solution is exempted from the restrictions as to criticality. The largest amounts which can be included in a package — we are talking here about small quantities of materials — have also been worked out by Mr. Woodcock on the basis of the minimum critical mass at the appropriate density for a water-moderated, water-reflected system with a homogeneous core and using a minimum critical number of packages of 250. This figure of 250 is five times the maximum number of packages of either radioactive or fissile material which, according to our Regulations, can be shipped in any mode of transport at one time; so five times fifty is a reasonable maximum to take.

Under these conditions, the values adopted for the maximum quantities were 16 g for U 235 and 9 g for Pu 239 and U 233. It should be added that the purpose of these exemptions is to remove certain procedural restrictions for obtaining approval for transport in other classes, but it does not mean that the shipment should automatically be entirely free of any control.

STANDARDS

A shipment which does not qualify for exemption must be packed and transported according to the standards laid down in the Regulations for Class I, Class II or Class III shipments.

Class I

The stipulation for Class I is that a critical assembly will not be formed when any number of packages are put together in any manner, i.e. each package must be a neutron sink in itself; or that in each package the number of neutrons emerging must not be greater than the number of neutrons entering. This is a very simple condition in principle, which is in itself extremely safe; but the multiplication of a reflected core depends on many factors. For instance, the energy distribution and, to a lesser degree, the angular distribution of the neutrons entering, are important. Since thermal neutrons are the most effective in criticality, the fissile material must be completely clad with an absorber such as cadmium in order to shield it from thermal neutrons entering. It remains, therefore, to ensure for all epithermal neutrons the basic condition that not more neutrons emerge than enter. For the calculation of the characteristics of a container satisfying this condition, a fission neutron spectrum was adopted and the angular distribution was also taken as isotropic in angle, which turned out to be more effective — and therefore safer — than any that could arise by interaction between containers. This is the result of the work done in the United Kingdom, especially the theoretical work of Mr. Woodcock.

In the Regulations, we do not specify in detail the type of package which satisfies these conditions, but we do say that a type of package meeting these requirements is a 1 ft diameter cylinder in wood with a cylindrical cavity such that the minimum distance from the surface of the inner container to the surface of the box is 4 in. The central cavity is cadmium-plated to a thickness of at least 15 mills, giving a minimum of 0.325 g cadmium/cm^2 of surface. If so desired, this cylindrical container can be taken as a square cross section container, provided that the largest distance from the surface of the inner container to the outside is kept at 4 in. This size includes a certain safety factor because the package still satisfies the conditions of safety from neutron interaction if a centimetre of layer of wood were lost by some accident such as fire.

The diameter of the circular cylinder in this case depends on the composition of the fissile material and on the density and the hydrogen content of the wood. Wood is used, because it is common material and contains hydrogen (which must be taken into account), and is also a material which ensures positioning and rigidity. Figure 1 shows the values of the diameter of the inner container calculated by a Monte Carlo method. The dashed lines refer to wood with a certain composition by weight of hydrogen (6.5 % weight of hydrogen) and the dotted lines refer to 6.9 % hydrogen by weight; a safety factor of 0.98 has also been included. The horizontal line at the top simply gives the size limit of the cylindrical cavity (4 in.). You will recall that we have a cylinder of 1 ft diameter, of which twice 4 in. would be taken for the thickness of the wood. Therefore the size limit of the cylindrical inner cavity is also 4 in. This would apply to solutions such as uranyl nitrate, uranyl fluoride or plutonium nitrate or to compounds which do not contain hydrogen and in which

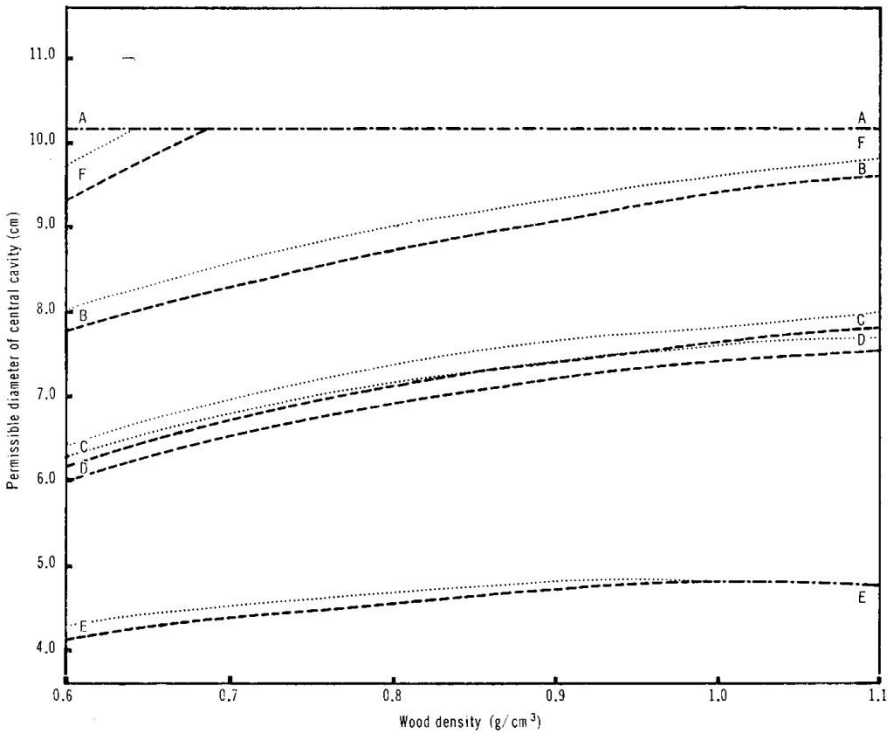

Figure 1. *Packages in which Geometry is the Controlling Factor - Change of Permissible Diameter of Central Cavity with Wood Density. (Dashed lines apply to wood with 6.5 % hydrogen by weight and dotted lines are for wood with 6.9 % hydrogen by weight.)*

the U 235 concentration does not exceed a certain value taken as 4.8 g/cm³. It would also apply to metallic uranium with a U 235 enrichment below 25 %.

The curve B applies to mixtures or compounds with a higher U 235 concentration (9.6 g/cm³) or metal with a higher enrichment (50 %). Curves C below apply to metal enriched up to 92.5 % or to any material in which the U 235 concentration is below 17.3 g/cm³, provided that U 235 is the only fissile material. Curves D (which are just below the curves C) apply to plutonium compounds or mixtures in which plutonium concentration is below 10 g/cm³. Curves E (still lower) apply to any fissile material, including plutonium metal. At the top, just below the horizontal line A, you have curves marked F; they refer to U 233 solutions and they show the difference between the U 233 and the U 235 behaviour. These calculations show the dependence of the size of the diameter plotted against the density of the wood, which can vary from one to another over a factor of two.

For uranium or plutonium metal, a mass limitation is more practicable than a volume limitation of the cavity. In this case the fissile material is still surrounded by cadmium and the whole thing is surrounded by wood, so that the same outside dimensions as I have just mentioned are achieved.

Then we have the permissible mass of metal which again depends on the density and hydrogen content of the wood. Figure 2 shows again the values

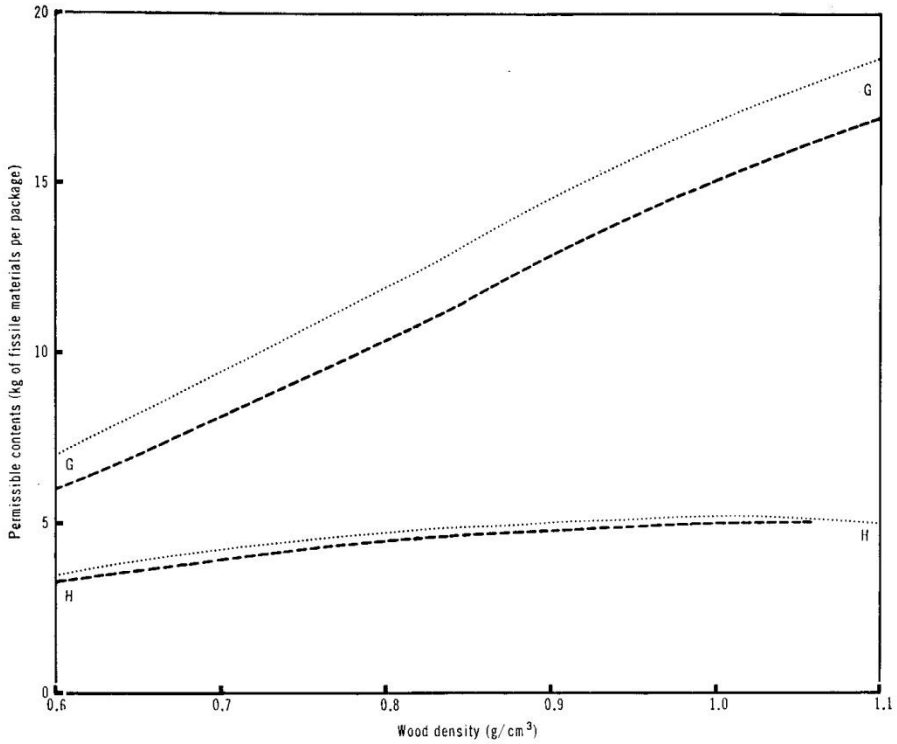

Figure 2. *Packages in which Mass is the Controlling Factor - Change of Permissible Content with Wood Density. (Dashed lines apply to wood with 6.5 % hydrogen by weight and dotted lines are for wood with 6.9 % hydrogen by weight.)*

calculated by Woodcock by a Monte Carlo method and there the curves G are applicable to uranium metal with an enrichment below 92.5 % and with no U 233, and the lines H refer to plutonium metal. Again, the dash lines refer to a certain wood with a certain hydrogen content and the dotted lines to a slightly higher hydrogen content: in this case also a matter of safety of 0.95 has been included.

We do not say in the Regulations that these are the packages which should be used universally, but they are presented as a well-known and well-investigated type of package which meets the specifications of Class I shipment. It is of interest to note that as a type test check such a package could be tried experimentally on a reactor, provided that allowance were made for the right conditions of neutron energies and distributions. Class I shipment can be effected fairly freely, since any number of packages so designed can be transported in any manner. This means that there are no special restrictions as to their number.

Class II

Where this criterion is too onerous for reasons of economy, we can pass to a Class II shipment. In Class II, the content of the packages is limited,

as is also the number of such packages which are allowed together. The sizes and the numbers are estimated somewhat conservatively so as to allow a total of five such loads without a criticality hazard.

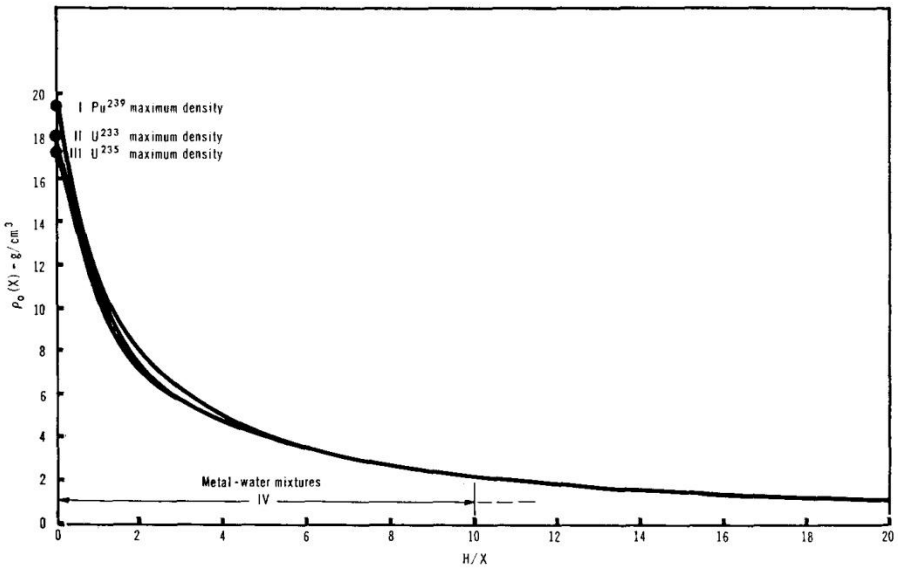

Figure 3. *Assumed Densities of X (U 235, U 233 or Pu 239) at H/X less than 20.*

Several methods of establishing criticality control are available and in use, but that given in the proposed IAEA Regulations is perhaps concerned with quantities that are more easily understood by laymen such as the shippers and the carriers. It deals with quantities of materials and with volumes,

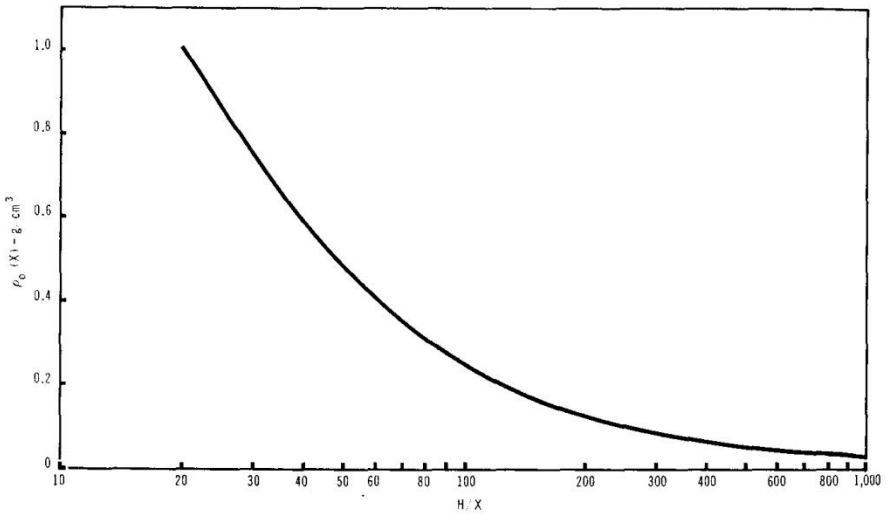

Figure 4. *Assumed Densities of X (U 235, U 233 or Pu 239) at H/X greater than 20.*

rather than with the more specialized quantities such as solid angle and interaction parameter of K effective. The amount of fissile material which is allowed in a package is shown to depend on the H/X ratio where H is the number of hydrogen atoms and X the number of atoms of the fissile material. Table I shows the allowable quantities, expressed in kg, for metals, compounds

TABLE I. MASS AND VOLUME LIMITS

	Maximum Unit		
	U 235	Pu 239 [b]	U 233
Metals, compounds, or mixtures $H/X \leq 2$ [a] mass limit, kg	5.0	1.7	2.0
Hydrogenous compounds or mixtures, $2 < H/X < 20$ [a] mass limit, kg	1.0	0.7	0.7
Solutions, or hydrogenous mixtures, $H/X \geq 20$ [a] volume limits, litres	2.0	1.3	1.3

a) H/X signifies H/U 235, H/Pu 239 or H/U 233 atomic ratio.
b) This limit holds for Pu metal at $\rho = 19.6$ g/cm³; for the alloy at $\rho = 15.8$ g/cm³ the corresponding limit is 5.0 kg.

or mixtures and, in litres, for solutions. They are given for a range of H/X less than 2, between 2 and 20, and above 20, and for the three fissile materials. These mass or volume limits give the maximum amount of material that can be put in a package. The material density must also be taken into account, should this exceed the indicated values for such materials. Table I shows the relationship between the density of the material unit against H/X for H/X below 20. Table II shows the same for H/X up to 1,000. You will remember that curves shown by Mr. Paxton indicated the density exponent which varies around 1.4, 1.6 or approaches 2. In the case of the IAEA Regulations, we say that the exponent 2 is safe; this practice is similar to that mentioned by Mr. Mallett earlier in this Symposium. If the material density exceeds the indicated amount by ratio N, the mass limits must be reduced by a factor $1/N^2$ or the volume limits by a factor $1/N^3$.

Since the outside dimensions of the containers are known, the overall volume can be calculated. This volume is now expressed in cubic feet and divided by the kilogrammes or litres we had earlier. The ratio then gives us a number. This number does not really mean anything, except that it is used in establishing the number of similar packages which can be safely put together. Figure 5 shows the relationship between this value (the package volume divided by the maximum unit of the previous table) against the allowable number of packages. This gives the maximum allowable number of such packages which may be introduced on one mode of transport during any 24-hour period. It must be remembered that for radiation control as well as criticality control 50 is, in any case, the maximum number of packages which should be put on one mode of transport.

As already mentioned, the packages for Class II are of the so-called Type B, which are so designed to withstand the consequences of the maximum credible accident and to be secure against leakage. There is a point here which could perhaps be further elaborated, namely how much leakage and how

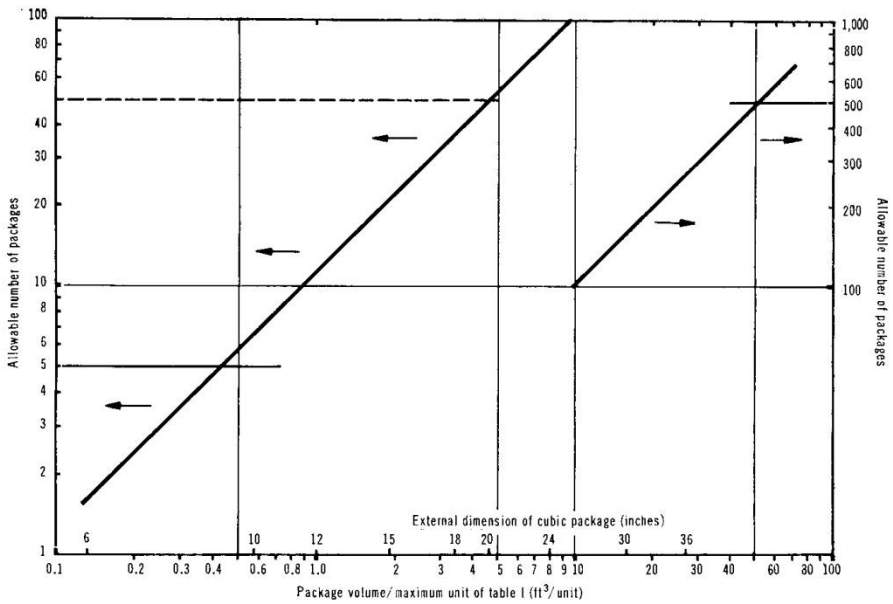

Figure 5. *Allowable Number of Packages in a Class-II Shipment.*

many packages leaking or flooded could be allowed in a shipment and still be safe. If amounts less than those shown in the first table are put in each container, then the allowable number of packages can be increased proportionally. This method is, I believe, somewhat similar to that adopted in the revised form of the " Nuclear Safety Guide " (TID 7016). However, we must not neglect one further check to ensure that the individual packages are within safe limits as regards volume or dimensions. Table II shows, for uranium (perhaps it might be interesting to add a similar table for plutonium in the future), against the enrichment, the maximum volumes expressed both in litres and gallons, and the safe diameter of the infinite cylinders and safe thickness of the infinite slabs.

To recapitulate briefly, we first determine the maximum amount of the material and then determine the outside dimensions of the container, divide the volume by this maximum amount, obtain a number, look up a curve and obtain the number of such packages which can safely be shipped together. This further check on the safety of each individual package completes the procedures recommended for a Class II shipment.

What is the safety factor or the conservatism of this method? It depends, of course, on the H/X ratio which has been somewhat arbritarily — albeit adequately maybe — divided into three ranges. The safety factor starts with rather low value at low H/X and goes up to a high value at high dilutions. This is for the case of no interspersed moderating materials but it includes a certain factor of safety of ten, to allow for the possibility of a moderating material being interspersed among the component packages of the shipment. For slab geometries of high enrichment at least, this has been checked experimentally, but for other geometries the factor of safety would probably be even better because the moderator may not have as much effect.

TABLE II. VOLUME AND DIMENSION LIMITATIONS

Tabulated values give the maximum permissible inside dimensions or the maximum permissible internal volume of the receptacle.

U 235 Assay (Wt. % U 235)	Volume		Diameter of cylinders of infinite length		Thickness of slabs of infinite area	
	litres	*gallons*	*cm*	*in.*	*cm*	*in.*
100.0	4.4	1.16	12.7	5.0	3.8	1.5
75.0	5.0	1.32	13.2	5.2	4.0	1.6
50.0	6.0	1.58	14.4	5.7	4.8	1.9
40.0	6.7	1.77	15.2	6.0	5.0	2.0
30.0	7.7	2.03	16.0	6.3	5.5	2.2
20.0	9.5	2.50	17.5	6.9	6.8	2.7
15.0	11.0	2.90	18.8	7.4	7.8	3.1
12.0	12.5	3.30	19.8	7.8	8.6	3.4
10.0	14.0	3.69	20.8	8.2	9.1	3.6
8.0	16.0	4.22	22.0	8.7	9.9	3.9
6.0	20.5	5.41	24.3	9.6	11.4	4.5
5.0	27.0	7.13	26.0	10.25	12.7	5.0
4.0	33.8	8.92	28.4	11.2	13.9	5.5
3.5	40.0	10.6	30.4	12.0	15.2	6.0
3.0	49.2	13.0	32.5	12.8	16.5	6.5
2.5	64.6	17.1	35.5	14.0	18.0	7.1
2.0	95.1	25.1	40.6	16.0	22.0	8.7
1.75	126.0	33.3	44.7	17.6	24.3	9.6
1.5	186.0	49.1	50.8	20.0	27.9	11.0
1.25	308.0	81.3	60.9	24.0	34.2	13.5
1.0	731.0	193	83.8	33.0	49.5	19.5
0.8	3,917.0	1,040	147.3	58.0	91.4	36.0
0.72	∞	∞	∞	∞	∞	∞

Class III

Those who are used to the United Kingdom methods will easily recognize Class I and those familiar with United States methods will recognize Class II. Now we come to Class III which is the field of anyone who is brave enough to demonstrate the safety of his shipment. Class III shipments should cover any fabricated shapes, alloys or clad fuel elements. Until experience has been gained in administering the rules of safety in transport, and until the transport of such special forms of fissile materials becomes more routine, such shipments, according to our proposed Regulations, are to be made under Class III as special shipments for which permission for transport will be obtained upon demonstration, by any method, that criticality control is actually achieved. For special shipments which fall into this Class, each must be considered individually and, in the Regulations at least, we list, for guidance, the possibilities which must be taken into account, singly and in combination, in order to demonstrate that a shipment is safe. That is to say, account must be taken of the credible accident, with the choice of the alternative leading to the highest reactivity; the possibility of flooding or the loss of water; the possibility of intermixing with another load of fissile materials or a load of special materials with better reflecting properties than water. The shipper may use any alternative method of criticality control

provided that his criticality expert can by demonstration convince the competent authority of the adequacy of his proposed control.

The case of irradiated fuel is a special one in the sense that, in addition to criticality considerations, the heat dissipation owing to the decay heat, the high levels of radiation and the necessary contamination control must be taken into account in container design and materials handling. The possibility, for instance, of the decay heat causing a melt-down of the fuel will require that the total fissile content of the package be limited below the critical mass of the melted fuel, unless it can be shown conclusively that melt-down cannot occur. We have also made certain points of caution with regard to irradiated fuel; we have said that if the reactivity of the irradiated fuel is on the increase with irradiation — that is, if you are in the early stages of burn-up of natural uranium — the fuel should be taken as irradiated up to that value of burn-up which corresponds to the maximum reactivity. If on the other hand, the reactivity is on the decrease with irradiation, the fuel can, for the purpose of criticality control, be treated as fresh fuel. I hope this will not cause any confusion, because it only happens in the case of natural uranium, and we do say, somewhere else, that natural uranium is exempted. Perhaps we should add here a reminder that irradiated natural uranium is no longer natural uranium.

As regards odd items such as scrap, which includes shavings, filings and borings from machining fissile materials or coolants filled with chips, etc., these are materials for which the enrichment, concentration or density cannot be known with any accuracy, and some of the parameters may conceivably change during transport. We do recommend, rather loosely, that when any parameter is not known accurately, a value should be taken which eads to a maximum reactivity under credible circumstances.

Concluding Remarks

In conclusion may I say that these proposed Regulations are being issued in the hope that they will be given a trial by Member States as a basis for both national and international transport of fissile materials. We hope that after a period of trial of, let us say, two years, we will have enough experience to enable us to revise and improve on them.

Some proposals for revision are already being considered. Among these, for instance, is a special label for fissile materials which would list a critical value in terms of an arbitrary unit, " This package is worth so-many units of criticality " and the carrier knows that he must not have more than so-many units together. Such a procedure would be rather similar to that now used for controlling the irradiation levels of shipments containing many packages. Another proposal concerns any standard containers which are found in practice to satisfy the basic criticality control and designed to withstand a major accident without losing any of their effectiveness. We have mentioned in some detail a type of package or container which meets the conditions of Class I, but it would be interesting, as a result of experience, to arrive at some kind of standard container.

Since these proposed regulations are at present very incomplete, we should like to encourage those who are working in the field of criticality control to keep them up to date by suggesting amendments and additions.

DISCUSSION

R.T. ACKROYD, United Kingdom:

I would just like to comment on one technical aspect of the basis of these Regulations. When interaction takes place we have to consider, on the surface on which neutrons are emitted, a distribution round the surface and an angular distribution at each point of the surface. In some of these calculations I see that Mr. Woodcock has used an isotropic source. This distribution intrigued me at the time it was proposed, for a number of reasons. If you send out a neutron in free space, its effect is overestimated by allowing it to go normal to the surface rather than by giving it an isotropic distribution; this effect would also apply to very high energy-neutrons, of the order of say 3 up to 10 MeV (or something like that), which would just tunnel through the material. I think in most of these problems, the bulk of the neutrons that are going from one package to another will have suffered a collision or two, and are such that when they strike the surface, they are almost bound to have an isotropic distribution. The actual spectrum is probably softer than a fission spectrum, so that the fact that the fission spectrum is being taken forms a nice counter-balance. There is just a warning that there may be one or two special assemblies where you can get higher rates of multiplications by assuming a spectrum which is not a fission one, and which is not isotropic; but I don't think these are likely to arise if the bulk of the neutrons have been into some scattering material before they get to another container.

E.R. WOODCOCK, United Kingdom:

The philosophy behind these Class I packages is that the surface multiplication of the outside of the package must be less than unity: that is to say, that if neutrons in any angular distribution or spectrum distribution that can arise, enter the package from other packages in the same Class, there must be fewer neutrons come out than go in. Now, we were in the fortunate position in making the calculations for these particular wooden transit containers that have been described, that we were designing the first types of Class I packages to be made, so we could find out what was the worst spectrum and angular distribution. If we had taken the most effective distribution from one of these wooden packages, we would have gone to the extreme limit of what could be transported in any one of them and this would have probably meant that any future packages would have had to be substantially of the same design. It would have been extremely difficult to design containers made of other than wood and cadmium plate. So we did not go to the extreme and the calculations were made with a fission spectrum, which is in most cases the hardest spectrum that can come out of any package, and with an isotropic distribution, which proved to be convenient for the calculations. We have been able to demonstrate by a number of subsidiary calculations which it would take too long to quote now, that the input we took was, in fact, more effective than anything which would come out of any of our packages. It turned out that the hardness of the spectrum was more important than the angular distribution. When anyone goes to the next stage of designing Class I packages they may make them with some different shielding material. It will be necessary for them

to allow for the input of neutrons into the new packages being the worst distribution that can come out of either the old ones, or the new ones, so it is going to be a somewhat more difficult task to add to the types of Class I container. I think it should be emphasized that the new containers must be thought of as being possibly mixed with the old ones and that the degree of safety we are after must remain unimpaired.

D. CALLIHAN, United States:

Shippers in the United States are meeting with objections from certain regulatory bodies in the use of combustible materials for transport purposes. I wonder if the IAEA has any comments on the use of wood in this connection?

C. ERGINSOY, IAEA:

Of course wood is used in this case as the material of the inner container, but this container itself is completely covered with a sufficiently thick shell of mild steel, which would be designed so as to withstand the maximum credible accident including the hazard of fire. However, as I have mentioned, if this mild steel envelope would in any way be damaged and fire be started in the wood, which is the inner material, at least a centimetre of wood would have to be charred away or burn out and disappear in order to arrive at any hazardous condition.

R.T. ACKROYD:

Many years ago we were interested in this and I think one of our engineers arranged a demonstration whereby we had a wooden package container put in an old truck filled with straw which was set ablaze. It burned merrily for quite a long time until the package fell on the ground. The only damage to it that I could see was about a quarter of an inch of charring. The wood being of high density didn't seem inclined to burn — I think we have all found this problem at times with domestic grates. The other point is that these high-density woods are impregnated with a fire-resistant compound. Before the woodwork could be consumed to any significant extent the temperatures in the containers would have risen and something would have happened to the contents, namely — in the case of solutions — they would have vaporised.

E.R. WOODCOCK:

I was involved in analysing the result of a fire test which was intended to represent the worst credible fire. The result was that the equivalent of one centimetre of wood had been lost from the outside and this, in fact, was the reason why these packages were designed to remain safe in any arrangement even if one centimetre of wood is removed from the outside.

A further point that also came out of the test was that the wood proved to be a very good insulator and ensured that the fissile contents of the container did not reach a dangerous temperature. This is, I think, one further advantage in making the containers of wood: it shields the contents from the effect of a fire.

A.N. DICKSON, Dragon Project:

I wondered if in the fire hazard the contingency of a melt-down of the cadmium had been considered and whether you had to show it was safe without the cadmium. In which case I would question why it was necessary to put it there in the first place?

E.R. WOODCOCK:

The answer is that in this fire test, the cadmium was not raised to a temperature anywhere near the melting point.

A.N. DICKSON:

Have you postulated a standard fire which put the maximum heat input into the package? I could think of a fire, say in a ship at sea, that could go on for a very long time, and would not like to guarantee that, whatever it was made of, you wouldn't utlimately melt the cadmium.

C. ERGINSOY:

I don't think a standard fire was really specified. Pershaps Dr. Ackroyd or Mr. Woodcock would tell us about what kind of fire they had in the demonstration.

R.T. ACKROYD

It's many years since I saw these tests but I formed the impression from the sort of temperature measurements made inside the chamber of the containers, that if the cadmium had gone the thermal solution would have evaporated, and so there would be no real problem: the moderator would have been lost. In the case of material in the form of metal held inside, the cadmium would be having very little effect, so perhaps these things balance out.

H.C. PAXTON, United States:

This whole problem of transportation is one that can drive a person crazy if he thinks enough about details. With enough imagination I believe anybody can convince himself that only minute amounts of fissile material can be shipped safely, so one is immediately driven to some degree of arbitrariness about what is sufficiently safe, and of course this arbitrariness involves pictures of perfectly regular arrays, perfect reflection and such. Particularly in connection with Class II shipment, different factors of safety have evolved in the minds of various people who have either pronounced about such shipments and been listened to, or are in a position of authority so that one can't help but listen to them. There are going to be arguments about the sizes of units represented in Table II. There are other problems too; it has become quite apparent during the last few years that Table II is not sufficiently general to fit the needs, at least in the United States; a real difficulty arises for those who must ship slightly enriched uranium-oxide batches, for example. The extension of this Table to include uranium in various degrees of enrichment is a complicated one. One is working with a good many possible parameters here — degrees of enrichment, size of unit, package size, allowable number of units, etc. That is quite a number to correlate and if one includes all the possibilities I'm afraid the result will be complicated. In the first place, some

of these possibilities cannot be defined precisely and would not be interpretable if written down. So there is a problem of selecting relatively simple rules that are general enough to satisfy most purposes, and behind this is the problem of finding out what people need before establishing an extension of these rules. Table II actually grew out of types of limits that were originally set in the old days when no fissile material was shipped by common carrier, where exclusive use was essentially the only means of shipment and when the character of shipments — at least bulk shipments — was generally quite different from what it is to-day. I point this out as an indication that what was proposed two years ago is now quite out-of-date. There is another problem, that the volume limitation on solutions is much too restrictive for certain classes of solution, and, presumably, there should be an extension there. We can foresee other problems. People try to make money out of their production of uranium compounds for use in fuel elements and object violently to the cost of transportation. There is much pressure for a cheap sort of container, though there probably is no real cheap container that could be guaranteed to withstand crushing in whatever is regarded as a reasonably defined maximum credible accident. The question that arises is whether we must expect more than a few containers to crush in an accident — one can certainly conceive of an accident in which everything is crushed together though this is not usually what happens. There are signs that pressures will be brought for further compromises. Some authorities in the United States who are concerned with transportation, and in a position to be listened to, claim that there is no such thing as a leakproof container; they will admit that containers can be tight enough so that the material inside won't leak out, but they won't admit that they will be tight enough so that water can't leak in and, unfortunately, they insist upon the possibility of this happening to all containers in a given shipment. Here is something that differs rather from the philosophy behind the original proposal for Table II. I do not myself agree with this particular point of view. Not the least of the problems in this connection is the lack of basic information. Another question has to do with the fineness with which specifications should be given. Table II, we now realise, is much too coarse and it does not cover things adequately. On the other hand, I wonder if the figures that distinguish between the effects of wood of the two different hydrogen contents aren't cutting things a little too fine? Generally the magic curves were very close together and this implies, I believe, amazingly precise computation if the difference is to be taken seriously — this might be overdoing the fineness just a little.

C. ERGINSOY:

Dr. Paxton has admirably amplified what I have attempted to express, namely that the proposed Regulations now need a period of confrontation with actual needs and actual experience. The table giving the maximum quantities, whether mass or volume, and the graphs illustrating the characteristics of a type of container which meets the specifications of Class I are not really in the same category. Whereas one imposes certain restrictions, however coarse, the other is just for illustrative purposes. It is perhaps just as well that we maintain a uniform level of coarseness rather than try to achieve at the very beginning a high level of precision which we would have to change many times later on.

H. DOPCHIE, Belgium:

My question has to do with temperature limitation in designing fuel element shipment casks, which was only briefly mentioned in Dr. Erginsoy's presentation. In the United States Regulations the maximum temperature which can be attained in any conceivable accident is, I believe, 100° C below melting. Of course, quite a few things might happen before this temperature is reached: metallurgical changes and fission product diffusion can happen at quite low temperatures. This is the case with natural uranium but it is also true of all types of metallic fuel elements. I wonder if some attention has been paid to the need to establish a regulation to the effect that under normal circumstances the temperature of the fuel element should not rise above the normal operating temperature of the fuel element in the reactor? I wonder if the IAEA or someone associated with the United States Regulations could comment on that?

C. ERGINSOY:

I would really like to leave this question to those who have experience in the transport of irradiated fuel, but I could try to guess at the thoughts of the people who devised the conditions for Class III, since it is Class III under which, I presume, the transport of irradiated fuel would fall. You will notice that in this Class we have not specified any quantities and it might perhaps have been out of place to specify a quantity such as temperature before specifying other quantities. We have drawn attention to the question of melting and tried to list the considerations to which attention should be given, but have left the safety of special shipments to be demonstrated by any method.

A.N. DICKSON:

With regard to fixing the maximum temperature as not greater than that in the reactor, it certainly would not meet the case of a Dragon-type reactor where the maximum fuel temperature may be as high as 2,000° C.

E.R. WOODCOCK:

Perhaps I should add to Dr. Erginsoy's answer that when the new Regulations are out and have been read, you will see that quite a number of provisions are made for irradiated fuels. There is a provision on temperature which is really to ensure that the fuel remains in its original shape and doesn't melt and run into a more reactive shape. There are provisions about the escape of poisons; I do not recollect the exact wording but it is to the effect that any poison which may come out of the fuel during transport should be assumed not to have been there when the conditions for transport were determined.

Session VII

OPERATIONAL PROBLEMS
PROBLÈMES D'EXPLOITATION

Chairman - Président :

Dr. H.K. SAUER

(Germany)

ASPECTS ADMINISTRATIFS DU CONTROLE DE LA CRITICALITÉ

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LE CONTROLE ADMINISTRATIF DE LA CRITICALITÉ AU COMMISSARIAT A L'ENERGIE ATOMIQUE

La sécurité dans les installations nucléaires du Commissariat à l'Énergie Atomique est étudiée par la Commission de Sûreté des Installations Atomiques (C.S.I.A.), qui délivre les autorisations de fonctionnement. Son autorité s'exerce aussi bien pour autoriser la mise en route d'installations nouvelles, que pour la remise en route d'installations existantes, après modifications.

La C.S.I.A. est assistée de sous-commissions spécialisées disposant de groupes de travail. L'une de ces sous-commissions, la « Sous-Commission des Masses Critiques », est particulièrement chargée de l'étude des problèmes de criticalité.

Le processus est le suivant : lorsqu'une installation nouvelle ou des modifications à une installation existante doivent être entreprises, l'avant-projet correspondant est soumis à la C.S.I.A. Celle-ci charge chaque sous-commission de l'étude des problèmes relevant de sa spécialité.

En ce qui concerne les problèmes de criticalité, la Sous-Commission des Masses Critiques charge son Groupe de Travail d'étudier, avec les auteurs de l'avant-projet, les moyens propres à assurer le contrôle de la criticalité de l'installation. Ce premier stade des études permet de définir les parties de l'installation où la sécurité reposera sur la géométrie, sur la concentration ou sur les masses mises en jeu.

Après accord de la Sous-Commission sur ces définitions, les réalisateurs peuvent passer à l'établissement du projet définitif, qui est à nouveau soumis à la Sous-Commission. Celle-ci s'assure que ses recommandations ont bien été suivies, et signale éventuellement aux réalisateurs les points où des précautions particulières devront être prises. Les plans détaillés d'aménagement et les commandes de matériel peuvent dès lors être entrepris.

Les plans sont vérifiés par le Groupe de Travail qui s'assure que les risques de criticalité sont bien exclus (en particulier, risques d'interaction).

La C.S.I.A. est régulièrement informée du déroulement de ces études, afin de pouvoir intervenir le cas échéant, en particulier pour imposer un renforcement de la sécurité nucléaire.

Lorsque l'installation est terminée, la Sous-Commission la visite et vérifie sa bonne conformité aux plans. Elle assiste éventuellement aux essais en inactif.

Un rapport de sécurité est établi en commun avec les réalisateurs et les responsables de l'exploitation. Il indique toutes les mesures prises pour éviter les risques de criticalité, en particulier toutes les règles administratives qui devront être strictement appliquées.

Ce rapport est adressé à la C.S.I.A., accompagné d'une proposition de la Sous-Commission des Masses Critiques d'autoriser la mise en route de l'installation.

La C.S.I.A. est alors en mesure de délivrer l'autorisation de fonctionnement, éventuellement après avoir demandé à la Sous-Commission des compléments d'information.

Une fois l'autorisation de fonctionnement accordée, il incombe au responsable de l'exploitation de prendre toutes dispositions pour s'assurer que tous les impératifs liés à cette autorisation, et en particulier les règles administratives, sont bien respectés. Aucune modification, même minime, de l'appareillage ou des consignes d'exploitation, ne doit être effectuée sans accord préalable de la C.S.I.A.

Cette procédure, évidemment assez lourde, donne satisfaction quand il s'agit de constructions neuves. Elle demande certainement à être allégée quand il s'agit de modifications relativement peu importantes, pouvant être réalisées dans un délai court. Il semble souhaitable que, dans ce cas, l'exploitation puisse faire appel à un spécialiste « Criticalité ». Celui-ci jouerait sur place le rôle dévolu au Groupe de Travail de la Sous-Commission des Masses Critiques. Il rendrait compte à celle-ci du résultat de ses travaux, afin qu'elle puisse proposer à la C.S.I.A. d'autoriser les modifications.

Conception des installations du point de vue des risques de criticalité — Règles générales

Nous étudierons ici successivement le cas des installations industrielles ou semi-industrielles où sont traitées des quantités importantes de matières fissiles, suivant des procédés bien définis, dans des appareillages fixes, et celui des installations du type laboratoires où sont effectués des recherches ou des traitements spéciaux, portant sur des quantités restreintes de matières fissiles, dans des appareillages généralement mobiles.

Installations industrielles ou semi-industrielles

Chaque fois que cela est possible, on recherche la sécurité géométrique. Malheureusement, celle-ci n'est pas toujours compatible avec les nécessités de la fabrication (rendement, mise en œuvre du procédé chimique, etc.). Il devient alors nécessaire de faire reposer la sécurité sur le contrôle de la concentration ou de la masse, ces deux données pouvant être éventuellement liées entre elles.

Le contrôle de la concentration s'applique plus généralement à des procédés de fabrication continus utilisés dans les usines chimiques (par exemple, extraction au solvant dans des batteries de mélangeurs-décanteurs). Il est évidemment nécessaire que la concentration soit suffisamment stable pour qu'un engorgement soit décelé bien avant qu'elle ne devienne critique. Le contrôle de la criticalité par la concentration ne pourra donc s'appliquer que si la rapidité d'obtention des résultats d'analyse est suffisante. Les règles administratives à appliquer dans ce cas devront définir la fréquence des analyses et les mesures à prendre en cas d'augmentation anormale de la concen-

tration. Elles devront si possible indiquer, en outre, les contrôles périodiques à effectuer pour s'assurer de l'absence d'accumulation de matière fissile (précipités insolubles, par exemple).

Le contrôle de la masse s'applique plus généralement à des procédés de fabrication discontinus utilisés dans les usines chimiques (par exemple, chargement de pots de précipitation), ou à des chaînes de fabrication dans des usines métallurgiques (travail sur métal ou sur sels secs). Ce contrôle devrait toujours être double. Par exemple, le chargement d'un pot de précipitation ne doit pouvoir s'effectuer que si l'on est certain que le pot est préalablement vide. Le chargement pourra se faire par l'intermédiaire d'un bidon mesureur, des verrouillages mécaniques ou électriques interdisant la vidange de ce bidon si son remplissage n'est pas terminé, et si le pot de précipitation n'est pas vide. En outre, le pot de précipitation étant réalisé en matière transparente, l'ordre de chargement ne sera donné qu'après s'être assuré visuellement que le pot est bien vide.

Un autre exemple peut être le suivant : le transfert d'un récipient d'ajustage dans un récipient de volume fini où la sécurité nucléaire repose sur la masse, ne doit s'effectuer que si la concentration de la solution à transférer est inférieure à une certaine valeur. Il est donc nécessaire de s'assurer au préalable du résultat de l'analyse. La commande de transfert comporte un verrouillage à double serrure, une clé étant détenue par le responsable de la fabrication, l'autre par le responsable du laboratoire d'analyses.

Installations du type laboratoires

Ce sont généralement soit des installations de recherches, soit des installations de récupération de déchets divers. Il s'ensuit que les procédés utilisés dans un même laboratoire peuvent être très variés. Évidemment, chaque fois qu'un appareillage peut être conçu pour assurer la sécurité géométrique, il est recommandé d'utiliser ce moyen de contrôle de la criticalité. Cependant, la mobilité des installations et les modifications fréquentes qu'elles subissent, conduisent à adopter en outre le critère de sécurité par la masse.

C'est ainsi que le contrôle par la masse a été retenu pour assurer la sécurité nucléaire dans deux installations d'un de nos Centres d'Études Nucléaires.

a) *Installation de traitement du plutonium*

Le bâtiment est divisé en tranches, chaque tranche comportant deux laboratoires et un hall. La quantité totale de plutonium stockée ou mise en œuvre ne doit pas dépasser :

— 200 g, par laboratoire ou hall,
— 500 g, par tranche.

Des règles administratives très strictes ont été établies. Elles définissent en particulier les conditions dans lesquelles doivent s'effectuer les mouvements de plutonium, et les responsabilités de chacun.

C'est ainsi que tout mouvement de plutonium, intérieur au bâtiment et inférieur à 100 g, peut être fait librement, à condition de respecter la règle : « 200 g par laboratoire ou hall », ce qui implique l'accord préalable des responsables des laboratoires intéressés par ce mouvement. En outre, les responsables du laboratoire doivent s'entendre pour ne pas effectuer plusieurs transports à la fois.

Par contre, pour tout mouvement de plutonium intérieur au bâtiment et supérieur à 100 g, l'autorisation doit en être demandée préalablement au responsable du bâtiment qui en informe le Service de Protection contre les Radiations, et peut imposer un horaire et un itinéraire déterminés.

Les responsables du laboratoire, du hall ou du bâtiment sont personnellement responsables des quantités de plutonium abritées par leur laboratoire, hall ou bâtiment.

b) *Installation de traitement de l'uranium enrichi*

Cette installation assure le traitement (récupération, purification, mise sous une forme chimique donnée) de petits lots d'uranium enrichi non irradié, compris dans la gamme de 3,5 à 90 % d'enrichissement. Elle comprend plusieurs laboratoires non isolés neutroniquement les uns des autres. Les opérations de traitement sont effectuées en discontinu.

Le contrôle de la criticalité est assuré en limitant à 350 g la masse d'U 235 contenue dans l'ensemble de l'installation.

Si un doute subsiste sur la teneur isotopique d'un lot à traiter, celui-ci est considéré comme constitué d'U 235 pur jusqu'à ce que le doute ait été levé (en particulier, prélèvement des échantillons destinés à l'analyse par spectométrie de masse). Un appareillage est à l'étude pour permettre un contrôle grossier rapide de la teneur isotopique. Nous en reparlerons plus loin.

Le principe de contrôle par la masse d'U 235 dans l'installation implique un contrôle administratif de toute entrée ou sortie de matière fissile.

Dans ce but, la comptabilité matière est tenue à la fois par l'ingénieur responsable des laboratoires, et par le responsable d'un magasin où transitent obligatoirement des lots d'uranium enrichi. C'est seulement après vérification de la concordance des deux comptabilités que les autorisations de transfert sont accordées par l'échelon hiérarchique supérieur.

INSTRUMENTATION

Nous mentionnerons ici deux types d'appareils de contrôle que nous étudions actuellement en France et qui, sans évidemment contrôler la criticalité elle-même, devraient permettre de vérifier certains paramètres sur lesquels repose la sécurité nucléaire.

Contrôle de la concentration en plutonium de solutions aqueuses de nitrate de plutonium par comptage neutronique

Il a semblé intéressant de chercher à contrôler la concentration en mesurant le flux de neutrons dû en particulier aux réactions (α, n) des α émis par le plutonium sur l'O 18 de l'eau. La mesure devant être faite en présence d'une forte activité γ, on a utilisé des chambres à fission (chambres CFU 1, construites par la radiotechnique). La sensibilité de ces chambres est malheureusement très faible (0,05 coups/s pour un flux de 1 neutron/cm^2 s).

En plaçant la chambre dans un doigt de gant accolé au récipient contenant la solution, on a cependant pu constater qu'un contrôle qualitatif de la concentration était possible pour des teneurs en Pu de l'ordre de 10 g/l. Il a été ainsi possible de déceler très rapidement un engorgement dans une batterie de mélangeurs-décanteurs.

Les études de tels dispositifs se poursuivent actuellement, et l'on cherche en particulier à utiliser des ensembles de comptage plus sensibles. Il semble en outre qu'un tel dispositif devrait permettre, lorsque la concentration est connue, de contrôler le point de remplissage d'un récipient.

Dosages approchés de l'uranium 235 *par comptage* γ

Avant d'effectuer un transfert d'uranium enrichi du magasin de stockage à l'installation de traitement mentionnée sous (*b*) ci-dessus, il serait utile de pouvoir vérifier de façon approchée la teneur isotopique du lot à transférer.

Le principe d'une méthode actuellement en cours d'étude, et dont les premiers résultats sont encourageants, est le suivant :

L'uranium 235 est émetteur γ d'énergie 0,184 MeV. Les énergies des γ émis par les autres isotopes de l'uranium sont plus basses. En éliminant par voie chimique les descendants de l'U 238 émetteurs γ de forte énergie, on peut doser l'U 235 par comptage des γ de 0,184 MeV.

On prépare par voie chimique de l'U_3O_8, dont on utilise pour la mesure 500 mg, placés dans un porte-échantillon de géométrie bien définie.

Un premier comptage de dégrossissage permet de choisir parmi une gamme d'étalons, ceux de teneur immédiatement supérieure et immédiatement inférieure à celle de l'échantillon. On effectue alors un nouveau comptage de l'échantillon et un comptage de chacun des étalons. La teneur isotopique est déterminée par intrapolation linéaire.

Conclusion

Il n'est malheureusement pas toujours possible de concevoir des installations géométriquement sûres. Dès que le contrôle de la criticalité repose sur une limitation des concentrations ou des masses mises en jeu, un contrôle administratif strict s'avère nécessaire. Il implique une application d'autant plus rigoureuse des consignes par le personnel d'exploitation, qu'il n'existe pas à notre connaissance d'appareils capables de donner l'alarme avant que l'excursion critique se soit produite. C'est pourquoi il nous semble indispensable de chercher à développer des appareillages qui permettent de contrôler au moins qualitativement certains des critères sur lesquels repose la sécurité.

ORGANISATION FOR OPERATIONAL CONTROL OF CRITICALITY

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INTRODUCTION

It is important that before considering the principles and details of criticality control or nuclear safety, it should be clear what we are trying to achieve, and why we wish to achieve it. Why must a criticality incident be avoided? There are three principal reasons:

To prevent injury to personnel. Personnel who could be involved in the effects of a criticality incident are both those who are operating the plant and those members of the public who may be close enough to be exposed either to the direct radiation hazard or to any airborne activity that may arise. There is a prime responsibility to see that any operations in which we may be engaged are conducted in such a manner as not to harm any individual. In addition, most countries possessing nuclear research facilities, and particularly nuclear power plants, have statutory legislation which requires the operators of all plants capable of causing the emission of ionizing radiations, to ensure that nobody is harmed by their activities. This requirement is both moral and, in some cases, legal.

To avoid damage to plant. A criticality incident can be of sufficient violence to damage, and perhaps destroy, sections of a chemical or metallurgical plant. Not only the fission products from the excursion are released, but processing of irradiated fuel elements involves highly radioactive solutions which could be expelled from their vessels as a result of the incident. These would contaminate both plant and buildings and produce a decontamination problem that could take many months to rectify. There are three economic factors involved: the cost of re-instatement of the damaged plant; the cost of decontamination of plant and buildings; and loss of the use of the facility whilst these remedial operations are carried out.

To avoid adverse publicity. Although in civilised society there is a great respect for human life, the majority of our populations have become inured to the toll of lives taken daily by conventional industrial accidents. Many fatal and serious accidents occur in industry, but this has always been so and there is little public reaction. However, should a man be injured or killed as a result of exposure to ionizing radiations, there would be a considerable adverse reaction, from the public in general and from the press in particular. This, undoubtedly, is due to the esoteric dangers of radiation which, to the

layman, are incomprehensible, whereas conventional hazards are recognizable and would appear to be more easily avoided by personal action. There is therefore a duty to the public to conduct operations in such a way as not to cause alarm and disquiet. Any adverse public reaction, unjustified as it might be when compared with the safety record of other industries, would have a detrimental effect on the progress of the atomic energy industry.

Summarizing, there are three main reasons for avoiding a criticality incident: humanitarian, economic, and public relations.

Responsibility for Safety

It has long been the practice, in the United Kingdom, to hold the 'occupier' responsible for the safety of all work carried out on his premises. This principle has been extended into the field of nuclear safety and it is believed that the responsibility for the safety of an installation must always rest with the operator. By operator, is meant the Head of an Establishment when considering the whole site, or his Heads of Department and Sectional Managers when considering specific processes. Managers at all levels have a duty to consult their various specialists and obtain the best advice available: the ultimate responsibility, however, for ensuring that the operations under their control are carried out safely must always be theirs. No clearance certificate or advice can make a process safe if sufficient accent is not placed on criticality precautions by responsible management. It is well recognized that both workmen and staff are inclined to take short cuts, especially if safety requirements are onerous and appear to be unnecessary. Other papers will show that mistakes which afterwards seemed to have been utterly foolish have been made by men who were well qualified to understand the hazards of what they were doing. Their familiarity led them to overlook simple safety precautions that in some cases would have saved their lives. Only a thorough understanding of the hazards and a strict adherence to procedures which have been carefully worked out beforehand can ensure that mistakes are not made that are costly in men, materials and time.

The operator who takes a formal clearance certificate and interprets or misinterprets it according to the letter is adopting a very dangerous course. He must accept the responsibility that is his and trust his criticality safety colleagues to be realistic and have confidence in their judgment. The converse also applies in that the criticality specialist who seeks to cover himself by writing unrealistic terms into his clearances soon establishes a reputation for being over-restrictive and by this means encourages operators to disregard not only his restrictive conditions, but also others that are essential for the safety of the process.

Formal Clearance

It is considered necessary to lay down in a formal manner conditions which, if complied with, will result in the safe operation of an installation. Clearly, the safe operation of a plant with a criticality hazard cannot be left to chance. The desiderata are such that, whilst the operator might be aware of them, they can only be fully understood by a specialist. We can postulate three persons who can participate in a criticality assessment: the designer, the operator, or the independent specialist.

The Design Engineer is given a specification and a flowsheet for the

plant he is to design. His object is to design and construct this plant to be as efficient as possible, consistent with economy of design effort and construction costs. Although he might devote considerable effort to incorporating features in his design to ensure criticality safety, he can never be detached enough to place himself in a position to make an independent assessment of the effectiveness of his efforts. His thoughts must always be conditioned by the conviction that he has designed a plant to meet the specification he has been given and that it will operate as intended.

Similarly, an operator is concerned with making his plant work as efficiently as possible to meet a production commitment for a product of a specified quantity and quality. The problem that is continually before his eyes is that of making his process work smoothly and efficiently. He is too closely involved to have, at the same time, an independent approach to the measures of criticality control that are required and which must inevitably restrict his freedom of action in achieving his production schedule.

We thus come to the need for someone whose sole purpose is to see that a plant cannot become critical during all foreseeable conditions of both operation and maloperation. Accident conditions that must be considered credible by the criticality specialist are often ruled out by the operator. Valid assessments can only be made after close association with operational safety problems. It is thus the rôle of criticality specialists to define the limits within which plant may be operated to avoid a criticality incident. It is still the operator's responsibility to devise his own method of operating to meet these requirements. He must interpret the requirements of the criticality clearance certificate into working procedures so that his workmen understand exactly what they must and must not do.

Methods of Effecting Clearance

In outlining some of the ways in which a clearance procedure may be established, it is useful to record briefly the history of criticality clearance in the United Kingdom Atomic Energy Authority. The Authority has gone through the stages of an embryo, a rapidly-expanding, and currently an established industry. The procedures used at these various stages for obtaining criticality clearance can serve as case examples of one way in which this job can be done.

Clearance by Committee

In the early days of the Authority there was little background of experience to build on and it was necessary to give clearance for each and every activity by means of Criticality Clearance Committees. Gathered together on these committees were those few experts who could be found at the time to consider each proposal involving fissile material. This was necessary because the fund of experience was small and it was a possible mode of operation because the scale of activities was also small. The number of applications for clearance was such that a Committee had sufficient time to consider applications thoroughly.

Clearance by a Criticality Inspector

As the scale of activities increased, the need was felt for an independent system of inspection and a post was created with the title of Authority Cri-

ticality Inspector. Although the Criticality Inspector's interests covered the whole of the Authority's work, his attention was focussed on what was then the Industrial Group. In this group of factories located in the North-West of England and the North of Scotland, there was a rapidly expanding process industry which, by its very nature, handled large quantities of fissile material. The work of the committees continued, but as the Criticality Inspector gained in experience, as their chief executive officer, he gradually took over their clearance functions and the various committees became mainly policy-making bodies. A small Inspectorate of supporting staff of varied disciplines was formed to assist the Inspector in discharging his duties.

This type of organisation with the executive duties invested in one man, supported by a small body of specialists and backed by a system of committees concerned with overall policy, worked very well and enabled a large number of clearances to be granted in a comparatively short time. The committee structure gradually became redundant and with the formation of an independent Authority Health and Safety Branch which embodied the Inspectorate, some of them were disbanded. The committees concerned with the Research Establishments of the Authority continue to exist, however, as in these cases the number of applications for clearance is not excessive and there is a reserve of qualified staff familiar with the fundamental concepts involved.

In each of the process works, there is a Health and Safety organisation which is responsible to the Head of the Establishment, advising him on all the safety aspects of his operations. Whilst criticality clearance certificates were being issued by the Criticality Inspector, the criticality specialists in the Works Safety teams were playing an increasing rôle in the preparation and drafting of certificates. Eventually a state was attained when the draft clearances submitted by the works normally required little alteration by the Inspector and his action was largely that of independent verification and final ratification.

Clearance by Establishment Staff

In keeping with the principle that all staff are responsible for the safety of operations under their control through the normal management chain of command, the responsibility for the issue of criticality clearance certificate now rests with the Head of each Establishment. This change was considered to be justified at the stage of development when the works' criticality staff had sufficient experience and confidence to allow them to take the full technical responsiblity for the issue of a clearance. It is a well-tried and accepted principle of safety practice that staff who are responsible for safety advice should have a chain of command deriving directly from the man who carries the ultimate responsibility for the safety of the establishment. They must have a right of appeal, if difficulties arise, to a level of authority that is not primarily concerned with production or operations. In any organisation where the Safety staff is answerable to production management, they will inevitably be subjected to pressures to clear plant against production requirements.

Criticality Inspectorate

It is incumbent upon the Works Safety staff to carry out inspections to satisfy themselves that the conditions of clearance are being met. The rôle

of the Criticality Inspectorate of the Authority Health and Safety Branch is to provide an independent inspection service. The use of the term 'inspection' is perhaps an unfortunate one as it implies a rather rigid and formal operation. This is not so. The Inspectorate conducts its work in the rôle of an independent observer, uncomplicated by the details associated with the day-to-day operations, and so better placed to observe the overall principles involved.

On a long-term basis, complete reviews are made of processes to re-evaluate their safety. In this way it is expected to make an up-to-date assessment of the safe operation of a complete plant and so ensure that clearances are not only in force but are currently applicable and mutually consistent. Many small alterations can be made to a plant over a period of years and although each one is assessed and cleared at the time, an insiduous change can take place which can have a cumulative detrimental effect on the integrity of the initial safeguards. The Inspectorate also advises works' staff on the techniques and data available for clearance purposes. Whenever works' staff feel that they are in need of further information or that they require a second opinion, then they are free to consult the Criticality Inspectorate. Day-to-day contact is also maintained with the Design Offices so that criticality advice is available when required. At a later stage of a design project, formal assessment of criticality safety is made, together with an assessment of other hazards. In this way the Inspectorate fulfills three major functions: inspection, assessment and liaison.

The three functions are interdependent. Inspections cannot be made effectively without familiarity with assessment problems, nor can a valid assessment be made without the operational experience gained from inspection. There is a common area between these two functions which envelopes the third aspect of liaison.

Theoretical Support

The work of criticality clearance that has been outlined above is largely of a practical nature, using technical data that has been evaluated beforehand. Much work has been done on criticality experiments, particularly in the United States and the United Kingdom, and a considerable effort has been put into the correlation of the results of this work, together with theoretical predictions. These have been used to compile complete sets of data for the various systems encountered. There are teams of scientists working in this field at the Atomic Weapons Research Establishment, in the Reactor Group and in the Authority Health and Safety Branch. The Inspectorate is concerned that only the best available advice and knowledge should be used in making an assessment, and advice is frequently sought from these bodies.

Design

In the preceding sections of this paper the broad principles of the organisation needed to ensure the criticality safety of process equipment have been discussed. To explore further the detailed mechanisms that are used in effecting clearance in the Authority, let us consider the design and commissioning of a new plant. It is in the design stage, where there is the greatest freedom of action, that the most important safety decisions should be

made. Even with conventional plant it is very much easier to make alterations on the drawing-board than on the finished plant, but in plants handling active materials, post-commissioning modifications are even more costly and often impracticable. Processes involving new techniques are subject to exhaustive development programmes to investigate the major problems which are high-lighted during the design phase. Smaller schemes with no new concepts involved are liable to be taken for granted if a conscious effort is not applied to see that criticality considerations are borne in mind. The stages of a new project outlined below are not strictly applicable to all parts of the Authority, nor are they applicable to all projects, but, in general, the broad principles are followed. The small-scale modification or extension of a plant is not subjected to the full rigour of formal documentation as is a major project.

Specification. In the first place the organisation sponsoring the design and construction of the plant must draw up a specification which may include the method of criticality control to be adopted.

Preliminary Document. As the design work proceeds, a document is prepared giving the principles which are to be employed to meet the design specification. This is called either the Preliminary Safety Document or the Safety Principles Paper. It can be prepared by either the designer or the operator, according to where the expertise lies, but it must always meet the operator's approval.

Acceptance. An independent appraisal is made of the Preliminary or Principles paper either by a Criticality Committee or by the Authority Health and Safety Branch. This assessment is reported to the sponsoring officer who then knows that the design may proceed according to these principles.

Advice. Whilst the design is in progress, advice is sought on the criticality aspects of each section of the plant. This advice may be obtained from wherever the designer thinks best. Where it is given by the approving body, it is always without prejudice to the independence of a final assessment.

Liaison. In parallel with the stage " Advice ", consultation takes place with the operator to ensure that his views are brought to bear as the design develops.

Final Safety Document. As the design is nearing completion, a formal submission is made in the form of the Final Safety Document. Once again, this can be written by the designer or the operator but it must carry the operator's sponsorship. On a major project this may be preceded by an Interim Safety Document.

Assessment. The final safety submission is assessed by either the Authority Health and Safety Branch or an appropriate Working Party of the parent Criticality Committee and their findings are communicated formally as a written appraisal. By this stage the plant will be under construction and perhaps nearing completion.

Pre-Commissioning Trials

When the plant is handed over to the operator, it is necessary for him to undertake proving trials to evaluate the plant's characteristics. The ope-

rator will calibrate his vessels and metering tanks, measure flow rates and temperatures, observe control characteristics of instruments and test his sampling facilities. In general, he will explore every possible facet of the operation of his plant whilst there is no criticality hazard.

Stages of Clearance

The procedure which has been outlined above is that which is followed for a project which has gone through a formal process of design. Many requests for clearance arise, not from the commissioning of new plant but from the change in method of operation or the incorporation of minor modifications of existing plant. In order to present as complete a picture as possible, the following stages which are outlined are those which would be followed by the Works Safety staff in considering a new proposal. The fact that a design has received approval from a parent body does not absolve the works' staff from satisfying themselves that it is safe to operate.

To ensure that a clearance procedure is effective, it is necessary to define carefully the responsibilities of the various persons taking part. There are five main stages in a full and vigorous clearance system, and it has been found desirable to formalize these steps by a system of documentation and signature. The act of signing a document of this nature is a salutary one, since it emphasizes the responsibility that each person is carrying.

Application

Before a clearance can be granted for an operation, it is essential to define, with as much precision as possible, what is to be done, where it is to be done, and by whom it is to be done. This is clearly the operator's responsibility since he will know how he intends to operate his plant from his proving trials. It is at this stage that the operator must use the information to make his application comprehensive; otherwise he will receive a restrictive clearance. Given inadequate information, a criticality specialist must always specify conditions of clearance restricting the process to the use of a minimum mass, a minimum volume or a limited geometry, assuming the maximum theoretical density, full reflection and optimum moderation. If the details of the process and plant which are given are such that chemical and process guarantees can be accepted for the maximum credible value of the main variables, then due allowance can be made in the clearance. All applications for clearance should be made in writing and with as much fore-warning as possible. It is wrong for a criticality specialist to be asked to consider clearance against a restrictive time scale. Very few operations are such that good planning and management cannot provide ample warning of the need for a clearance. If a clearance has to be granted without full and mature consideration, the only safe course is to specify restrictive limits which can be guaranteed safe under all known conditions.

The action of making an application in writing is again often a salutary one. In setting down his requirements in a formal manner, the operator must concentrate upon the problem and consider logically all aspects of his process.

Submission

A submission is a developed argument substantiating the safety of the application. Where there is sufficient knowledge and experience, the sub-

mission can be written by the applicant. Where this is not so, then specialist help must be sought. The submission must contain nuclear safety arguments which give detailed reasons justifying the safety of each and every facet of the operation. It must give the source of its information on nuclear parameters and, if reference is made to previous submissions, it must examine their viability in the new context. It should consider the safety of each vessel or stage separately for normal operation under flow-sheet conditions. Each operation should then be examined for all credible maloperations. Having established the safety of individual items of plants, it is then necessary to consider their safety in relation to each other. Thus the submission must consider interaction between vessels, first under normal operating conditions and secondly under conditions of maloperation. The contingencies which must be borne in mind when making this examination are developed further in the following Section. The submission should establish quite clearly that under all credible conditions the plant will be safe. Where guarantees are put forward that preclude certain conditions as incredible, then arguments should be included which establish the validity of these guarantees.

Appraisal

The independent appraisal of a submission is the stage at which a check is made that the arguments advanced are valid. It is the act of taking a second opinion. Specialist staff must be consulted at this stage, or if a committee is being asked to give clearance, the propositions are put before them. It is essential that the person or persons making the appraisal shall be as independent as possible of the person who has made the submission and should preferably have no connection with the proposals. If the submission is not acceptable, then there must follow discussions and interchange of ideas between the various parties to resolve these differences. If the arguments are still unresolved, advice should be sought from an outside body. It may be that a satisfactory solution cannot be found and the answers to the problems can only be supplied by experiment or detailed study involving sophisticated calculation techniques. This type of work is normally beyond the scope of those practising criticality clearance and the ease with which such a solution can be obtained will depend very largely upon the facilities available.

The end product of the appraisal stage should be a draft operating clearance certificate. This certificate must do two things: it must clearly define the plant and operations to which it applies, and it must lay down in unambiguous terms the conditions of clearance. There is no place in this final certificate for nuclear safety arguments or any extraneous detailed information that may confuse the issue.

Approval

Before a certificate is finally issued, it must carry the authority of the person who is ultimately responsible. This implies that the process operator has accepted the conditions of clearance as being realistic and workable. It is possible to draw up clearance certificates that are technically correct in their definition of safe values but unacceptable as working documents. The parameters defined must therefore be meaningful and expressed as plant variables. If full consultation has taken place at the application, submission and appraisal stages there should be no difficulty in doing this. The signa-

ture of the Head of the Establishment or that of his authorized deputy gives the certificate its full status as a working document. By having certificates signed at this level, attention is drawn to the fact that the terms of the certificate are known to the Head of the Establishment and that his executive staff are expected to comply with them.

Commissioning Inspection

Very few, if indeed any, chemical or metallurgical plants start up and work exactly as designed. This is especially true of the atomic energy industry in which many of the problems are new and there is often a margin of uncertainty. It is therefore most important that both operators and safety staff should be extremely vigilant during the start-up period, and full and frank co-operation must exist. A certificate may be granted on the basis of a limited start-up under restrictive conditions and close supervision until the plant's performance can be evaluated. A reappraisal can then follow to give the maximum freedom of operation in the light of new information. Although this final step of commissioning control and start-up evaluation is not necessarily a formal and documented step, it is in every way as important as the formal measures.

Contingencies

In the aforegoing Section it was stated that consideration should be given to the contingencies which must be allowed for in the preparation of clearance certificates. It is impossible to be comprehensive on this topic because, by their very nature, contingencies will depend upon local plant conditions.

Since this is a most important aspect of criticality control it is necessary to mention at least some of the more important examples:

Flooding

The flooding of a system containing fissile material could cause increased neutron reflection and moderation and sometimes interaction between contiguous sections of plant. Such changes could markedly reduce the nuclear safety of the system. Possible causes of such an effect could be:

a) Leakage or breakage of vessels and pipework;
b) Unintentional transfers of liquors via extract or vacuum lift systems;
c) Fire-fighting operations;
d) Flooding from natural causes.

These factors should be taken into account when defining safe parameters, or alternatively, positive provision should be made to exclude the possibility of such events.

Interaction

During the design stage of a plant, the problem of neutron interaction between units will have been considered in some detail. This will have included interaction effects both under conditions of normal and abnormal operation.

Where interaction is possible, adequate spacing or neutron shield blocks between units should ensure nuclear safety. If use is made of neutron shield

blocks, then strict control should be exercised during manufacture, installation and maintenance.

Particular attention from the interaction aspect should be paid to the movement of fissile material into or from the process lines and it is essential that a formal clearance procedure should be adopted whenever non-standard movements of such materials occur.

Maloperation and Deviation from Specified Flow-sheet Conditions

At the design stage the possibility of quantities and concentrations of fissile material exceeding specified flow-sheet values must be taken into account in choosing the safe operating parameters for the plant. Deviations may arise from over-evaporation or the formation of unexpected sludges and precipitates. The concentration of solutions may increase with the result of changes in flow rates, feed solution concentrations and the normality of solutions.

Failure of operational controls to ensure nuclear safety must always be considered in the assessment of a plant. Typical examples of such occurrences are incorrect setting of diverter units, errors in operation of valves, cooling and heating system faults and inadvertent double batching.

Whenever possible the principle of avoiding transfer from geometrically- to non-geometrically limited systems should be maintained. If such transfers become necessary, then strict control of the operation must be exercised since such transfer is potentially dangerous.

Interconnection with Other Plant

If there is an intentional interconnection between plants, then the possible implications of this transfer must be considered, together with accidental interconnection. Inadvertent transfers could arise, due to failure of equipment such as valves, siphons or maloperation on the part of an operator.

Waste

In any process there will be some wastes arising. Under normal operation these will be of low fissile material content. If, however, a fault does arise in the process, then there will be the possibility of a marked rise in the fissile material content of the waste. Consequently, all waste, whether in liquid or solid form, should be taken into consideration in assessing the nuclear safety of a plant, attention being paid to the ultimate fate of such material.

Vessel Extract Systems

Since the processing of high-quality fissile material involves the handling of large quantities of highly radioactive substances, the process plants will have complicated vessel ventilation and air extraction systems. These extraction lines should be considered in the nuclear safety assessment, attention being paid to catchpots and filters in the lines. Consideration must also be given to the eventual disposal of the material which will accumulate in these catchpots and filters.

Accident Safeguards and Procedures

Despite all the precautions of design and the control by plant supervision, if large quantities of fissile material are handled, there will always remain a finite possibility of an incident. Both the experience of actual incidents which have occurred and theoretical assessments show that such incidents will cause the release of energetic penetrating radiations together with a possible loss of containment of large quantities of highly radioactive materials. It is possible, too, that these effects may occur without being immediately appreciated, the effects being insidious and not detectable by the normal senses of the operators. It is, therefore, necessary to have a detection system which will give a rapid and immediate warning of accident conditions, and a ' follow-up ' procedure to deal with the incident.

Warning Systems

It is the practice in the United Kingdom to provide criticality detection systems for processing plants handling fissile material in quantity. These systems have two functions: primarily to give immediate warning to plant personnel that a criticality accident has occurred; and also to present a display of the radiation conditions existing in the plant.

To achieve these aims, simple gamma radiation detectors are sited around the plants. These detectors respond to the prompt gamma radiation emitted when an incident occurs. This is not quite as simple as it sounds since in large-scale plants it requires the installation of a large number of detectors which must be strategically located to provide adequate coverage of the operating area. Before installing such a system, a definition of ' adequate ' coverage must be given, so that the instrumentation may be designed and installed to meet this requirement. The Authority has decided that such a system must be capable of reacting to a dose of 25 rads of prompt gamma radiation at a distance of 5 ft from any possible accident source. This is based on the assumption that an operator may be stationed at this distance from the incident when it occurs and will need an immediate warning. This criterion represents a minimum requirement, since it is appreciated that the operator, in the event of such an incident, could be subjected to neutron irradiation in addition to gamma exposure.

The instruments used for this type of work must be designed to a high standard of reliability, the circuitry should provide self-checking facilities and failure of any part of the system should be ' safe ' in that the alarm would sound. Essentially, this requires a robust, simple, basic design. It will be appreciated that false alarms due to failures of the system could have the following effects:

a) Frequent false alarms will quickly bring it into disrepute by crying ' wolf ' unnecessarily;
b) The evacuation drill which follows the false alarm could initiate an incident by leaving complex plant unattended;
c) Since evacuation under these circumstances must be rapid, there is always the possibility of injury occurring while leaving the plant.

The alarm itself must be an audible one, urgent, distinctive and clearly heard throughout all the working areas. It is most important that it should not be confused with other alarms, such as process alarms, radiation or fire-alarm systems.

Emergency Organisation

Sounding of the criticality alarms initiates immediate evacuation of the plant by all personnel, without any attempt to close down the plant. The urgent need is to get personnel away from the source of the incident as quickly as possible. To achieve this, direction arrows are displayed on the plants showing the nearest exit from the building. Efficient evacuation can only be achieved by organizing periodic practice so that personnel become accustomed to rapid response to the alarm and familiarize themselves with emergency exits from the plant.

Post-evacuation Procedure

After evacuating the plant there are then two urgent problems. The first of these concerns the evacuated personnel and the other is an assessment of the state of the plant. These are dealt with separately from two control points, the Assembly Point and the Criticality Control Centre.

All personnel will, after evacuation, go immediately to the *Assembly Point*. The first action by the staff manning this control will be to make a roll-call to establish that the building concerned has been completely evacuated and to establish, as far as possible, the location of personnel on the plant at the time the alarm sounded. All the personnel on these plants are provided with personal dosimeters which respond to the radiation incident upon them. Consequently at the same time that the roll-call is being taken these dosimetric packs will be collected and arrangements made for their assay. During this time a small Health Physics team will approach the building concerned with survey meters. The function of this team is to establish whether the incident is a real nuclear excursion or if the alarms have been triggered by an instrument fault or other extraneous causes.

At the *Criticality Control Centre* there will be a display of the radiation field in the plant, provided by remote instrumentation. The emergency team who will have to deal with the incident, will assemble in this Centre. All radiological data will be supplied to this control and the team consisting of Process Control, Engineering and Health Physics staff will assess the situation and decide on the action to be taken.

Training

Various procedures have been discussed whereby the safety of an intended operation can be established and formally certified. Effort put into the formal procedure of clearance is circumscribed, however, if workmen are ignorant of the restrictions of the process they are operating. There must be an overall awareness of the hazards of criticality. Each man must understand the consequences of irresponsible action on his part and must be trained to observe and report anything unusual. To achieve this, a training programme is regarded as an essential part of the safety structure. It is necessary for workmen to have a basic knowledge of the hazards involved so that they will appreciate the precautions that have been taken for their safety. Ignorance invariably leads to worry and fear, which will ultimately have an adverse effect on labour relations. It is emphasized that criticality safety is not any one man's job, but is the concern of everyone who has any contact with operations or equipment, from the Head of the Establishment to the man on the shop floor.

ADMINISTRATIVE AND OPERATIONAL NUCLEAR SAFETY CONTROLS IN A CHEMICAL PROCESSING PLANT

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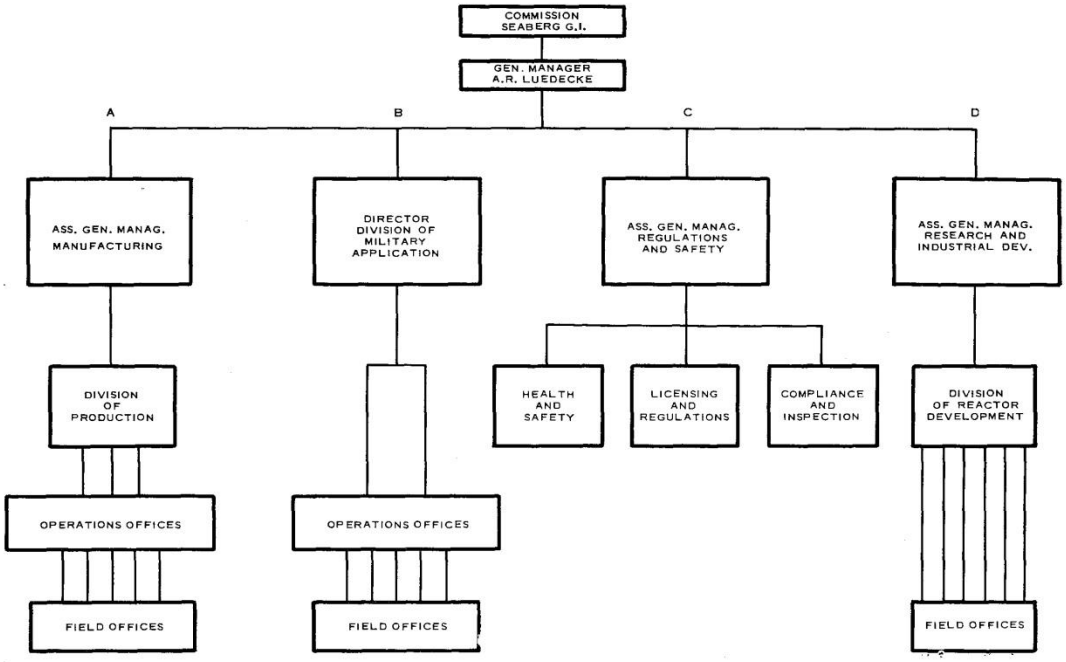
INTRODUCTION

Chemical processing plants for fissile materials in the United States can be grouped under two basic types: the Prime Contractor plants owned by the U.S. Atomic Energy Commission and operated for the Commission by private industry; and the Licensee plants which are owned and operated by private industry. Figure 1 presents the portion of the U.S.A.E.C. line organization which provides nuclear safety guidance for the chemical industry.

Several Prime Contractor plants were established during the days of the Manhattan District (13th August, 1942) about four years before the formation of the U.S.A.E.C. Nuclear safety policies and know-how were developed and successfully administered by the Prime Contractors, and to a great extent this is true today; this includes administrative and operational knowledge as well as technical information. After the establishment of the A.E.C. (1st August, 1946), the Prime Contractors reported to branches of the A.E.C. which today are known as the Division of Manufacturing, the Division of Military Application and the Division of Reactor Development. These are shown in Figure 1 as branches A, B and D of the A.E.C. line organisation.

Nuclear safety controls such as mass and geometry limits are established by each Prime Contractor from experimental and theoretical work done in Prime Contractor facilities, in much the same manner as they were under the Manhattan District. However, Prime Contractors are under the general surveillance of their local A.E.C. offices, who review the nuclear safety operations as part of their general responsibility.

Until the advent of the Licensee, about 1955, the A.E.C. relied upon its Prime Contractors for know-how and had not provided much guidance in the field of nuclear safety as applied to the chemical processing plant. However, with the advent of the Licensee, a group which has evolved into the Division of Regulations and Safety provides guidance and reviews the applications of the Licensee. This A.E.C. Division develops guide lines and rules based upon information or data supplied by the Prime Contractor for nuclear safety control and these rules are contained in Title 10, Code of Federal Regulations, Part 70, " Special Nuclear Materials ".


COMMISSION
SEABERG G.I.
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A.R. LUEDECKE
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DIVISION OF
MILITARY
APPLICATION
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REGULATIONS
AND SAFETY
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AND
INSPECTION
DIVISION
OF REACTOR
DEVELOPMENT
OPERATIONS OFFICES
OPERATIONS OFFICES
FIELD OFFICES
FIELD OFFICES
FIELD OFFICES

Figure 1.

As a case study, I shall orient my discussion in the direction of the Prime Contractor, mainly because of my knowledge of this type of operation.

Policy

We have found in our conventional safety programmes that it is insufficient to have a responsible supervisory service unless top supervision and management are actively concerned in prosecuting safety. This principle extends to the nuclear field. It is imperative that the top management of any installation be completely convinced of the importance of nuclear safety and give it full support if the line supervision are to carry out fully their responsibilities as outlined herein.

Responsibility for nuclear safety lies directly with the supervisor under whose direction the fissile material is being processed. This policy is in keeping with the general policy of assigning primary responsibility for all plant safety to the supervisory service in charge.

The nuclear safety group is established to give the supervisory service the necessary technical assistance to help them fulfil their responsibility for nuclear safety. The location of the nuclear safety group in the supervisory structure varies from plant to plant. However, there is a tendency in the United States to avoid direct supervisory coupling between production and nuclear safety groups. The nuclear safety group functions in an advisory capacity and reports to the director of research or the technical director, who in turn reports to the plant manager: in one case the attachment was through the industrial relations manager. Production supervision reports to the production manager who, in turn, reports to the plant manager.

Responsibilities

The production or other supervision service entrusted with fissile material has the following specific responsibilities with respect to nuclear safety:

a) To be adequately acquainted with all aspects of operations which involve nuclear safety, and to ensure that all operations are carried out according to established and approved safe procedures.

b) To request the advice of the Criticality Group concerning any unusual circumstances or conditions which might affect nuclear safety, for example:
- — A large increase in plant throughput;
- — Receipt of quantities of fissile material in excess of approved storage capacities;
- — A major production line tie-up due to equipment failure;
- — Receipt of fissile material of unknown composition;
- — Receipt of fissile material in non-standard containers.

c) To request the advice of the Criticality Group concerning contemplated alterations to existing facilities or changes in operating techniques where factors affecting the nuclear safety of the equipment or operation may be involved, and to request its advice concerning new equipment or procedures under consideration for handling fissile material prior to putting such equipment or procedures into operation. This implies that production supervision must be aware of many of the factors which influence criticality.

The *Criticality Group* also has certain obvious responsibilities, such as:

a) To review all known sources of relevant experimental and theoretical data, including all reports of critical mass incidents at other installations.

b) To pursue experimental and theoretical programmes and to provide original criticality information when necessary.

c) To advise operating supervision, from a nuclear safety standpoint, on the design, installation, alteration, operation, and maintenance of equipment and facilities for processing, storing and transporting fissile materials; and to assist operating supervision in recognizing and eliminating practices which are undesirable from the point of view of nuclear safety.

d) To assist in the investigation of non-routine incidents, such as near misses and infractions of criticality rules, and to suggest possible methods of eliminating such incidents in the future.

e) To assist supervision in the design and layout of instrumentation for the detection of radiation from a critical mass incident, should one occur.

f) To provide a programme of nuclear safety lectures and demonstrations to those groups requesting such a service.

From this, one can see that the nuclear safety group has two prime functions. The first is to generate new criticality data, and the second is to translate these data to the needs of the design engineers and production supervision. The nuclear safety engineer must be well acquainted with, and appreciate, the problems confronting the design engineers and production supervision. For this reason, it is often found desirable to assign a nuclear safety engineer to a specific production area so that he may become adequately acquainted with the various chemical processes, as well as with the personnel who may need his services.

Procedure and Methods

It is appropriate at this point to discuss a method used in arriving at a nuclear safety limit in a given process area. Nuclear safety limits are usually mutually agreed upon by both parties concerned, production or operations, on the one hand, and nuclear safety on the other. Most of this discussion occurs at lower supervisory levels. If agreement cannot be reached at the lower levels, then higher supervision, but still below that of the production manager, becomes involved. During my experience in the field and many thousands of recommendations later, I can recall only several reaching the level of the production manager, and none officially reaching the project manager.

In some United States companies, nuclear safety committees are established to review nuclear safety practices throughout the plant. Meetings are held periodically and are attended by the supervisors in charge of production operations, the technical directors, and a nuclear safety representative. The agendae of the meetings are decided upon by the chairman and other members of the committee. The commitee concerns itself with phases of nuclear safety such as plant policy affecting nuclear safety, nuclear safety experience gained at other companies, near criticality incidents referred to as ' near misses ', and so forth. The committee may also help in solving

knotty problems on criticality limits arising in various areas, though it has in fact rarely been used for this purpose.

Nuclear safety audits of production areas are conducted to give production supervision a measure of the effectiveness of their control methods. The audits range in scope from informal non-scheduled checks by a nuclear safety engineer to elaborate schemes involving outside consultants. The size of the audit team, the type of representation on the team and the frequency of the audits vary greatly from company to company. Small Licensees tend to have informal non-routine audits, while the large Licensee and Prime Contractors generally have more elaborate schemes.

Administrative Aids

Other nuclear safety administrative aids are criticality manuals and posted limits. The manuals contain a compilation of limits for specific work areas. These manuals are revised and kept current. This requires considerable work on the part of the nuclear safety engineer but is considered well worth the effort. The posting of criticality limit signs in specific work areas is widely used.

Let us now follow through with an example to illustrate how nuclear safety controls function in an actual plant situation. The problem is as follows:

A supervisor in a chemical salvage operation desires to increase, for operational reasons, a previously-agreed 400-g batch limit. The equipment involved is a geometrically unsafe, 24-in. dia. dissolver. The original 400-g limit was arrived at by assuming the worst possible conditions, namely: a UO_2F_2 aqueous solution in the tank in the shape of a sphere completely reflected by the remaining water in the tank. On this basis the 400-g limit would allow a safety factor of slightly greater than two for an inadvertent double batch possibility.

This crude empirical approach ignores certain important information such as a reasonable critical configuration and, secondly, makes no allowance for human error in estimating a batch size. The first problem is purely mechanical: the second requires operational data which has been accumulated over a period of time (one or two years). It should be borne in mind that the salvage materials involved cannot be representatively sampled for active material content prior to complete dissolution of the waste material. Waste materials might include contaminated paper waste, sand, rubber gloves, and other materials commonly met with in salvage operations.

Therefore, a scheme for estimating the material content of salvage wastes, and consequently a batch size, is necessary. The estimate is based upon the difference between what goes into a process and what comes out: thus the difference should be found in the salvage materials, but this still leaves considerable room for error. The data given to the nuclear safety engineer consist of these batch estimates accumulated over the year or two and the corresponding actual content determined by analytical means after dissolution of the waste materials.

The difference between the estimated batch size and the actual or analysed batch size as a function of the frequency is found to be normally distributed about the mean, for large numbers of samples. With this information and knowledge of the critical mass of the dissolver, one can calculate the approximate probability of an inadvertent critical incident in the dissolver as a function of the estimated batch size. After these quantities have been

determined, then an administrative decision is made as to whether or not the previously acceptable batch size can be increased. The final decision is made by the supervisory service in charge of the operation, but I must admit the supervisor in charge is heavily influenced by the nuclear safety engineer.

Obviously one cannot actually calculate the probabilities of an incident with any precision because of a number of uncertainties, such as the distribution of the fissile material in the dissolving vessel; whether or not a quantity of active material from a previous dissolution remains in the dissolver; or, for example, the effect on the critical mass of quantities of other ions present such as iron, silicon, nitrogen, etc. However, I feel that the value of this technique lies in giving an approximation of the magnitude of the probability as a function of the batch size. For example, it was found in one case that increasing the batch size from 400 g to 500 g increased the probability by a factor of more than 100.

It is difficult to say what probability would have been considered as a sane or reasonable objective in this operation. But it must be remembered that a probablitity exists whether a calculation is made or not made. Although geometrically unsafe dissolving vessels are in use in many plants, they are being replaced by vessels of safe dimensions: soluble nuclear poisons are being used in this type of operation but there exists a finite probability that the poison may not be added.

Other Aids

Mechanical and electrical aids for nuclear safety controls are numerous and varied and limited in scope only by the design engineer's ingenuity or lack thereof. Devices which have found a use in process plants include spring-loaded drybox floor drains which open when the water level reaches a given height and which are thus of value in preventing flooding should a water line break in an area where the criticality limits are based on non-flooded conditions; electrical equipment which is sensitive to water vapour and sounds an audible alarm when water finds its way into areas where flooding could be a hazard; nuclear poisons in the form of Pyrex Raschig rings, boron steel and soluble cadmium salts for use in geometrically unsafe vessels where build-up may occur.

These are but a few of the devices and methods used to help in assuring nuclear safety. However, none of them can take the place of well-trained process personnel. In the end, safe operations can only proceed when all those handling the materials, down to the operator levels, are frequently instructed in the correct handling of process materials. It has been said that a safety device can be made foolproof but not idiot-proof. In most United States plants the chemical operator moves material from one location to another and performs the necessary processing under the general direction of his supervisor.

Instrumentation for Control

In the last two or three years, considerable thought and effort has gone into the problem of detecting a dangerous build-up of fissile materials in geometrically unsafe process vessels. To my knowledge, no one has developed a reliable piece of equipment which can ascertain the nearness to the critical

TABLE I. IN-LINE INSTRUMENTS FOR PLUTONIUM AND URANIUM CONCENTRATION CONTROL

Instrument	Principle	Conc. Range	Interferences	Block Diagrams	Cost
Gamma absorptometer	Absorption of Am 241 60 keV by U 235 and Pu-NaI(Tl) detector	1-300 g/l	Other heavy ions or Other gamma emitters		~1,000 dollars
Cd sulphide flow colorimeter	Absorption of specific wave lengths due to Pu or U ions	0.3 to 50 g/l	Ions with colour absorption near the wave length being measured		~ 400 dollars
Gamma-alpha counters	Measurement of alpha activity of Pu and Am and gamma activity of Am subtraction gives Pu concentration ZnS for alpha NaI(Tl) for gamma	10^{-4} to 10 g/l	High Am conc.		~1,500 dollars
Neutron counter	Measurement of neutron flux of Pu solutions B 10 coated chamber 2528 photo tube O 16(αn)	0.1 to 100 g/l	External neutron flux, other alpha, neutron ions		~1,000 dollars
Gamma counter	Measurement of U 235 184 keV	.005-150 g/l	Other gamma emitters		~1,000 dollars

TABLE II. COMPARISON OF SEVERAL CRITICALITY ALARM SYSTEMS, COMMERCIALLY AVAILABLE

	Cost estimate 10 locations	Detector	Range	Circuit	Response	Readout, controls, and remarks
A	6,000-10,000	*Ionization Chamber* Sensed to gamma dose (80 keV to 2 MeV) Sensed and pressurized (5 atm.)	Any three consecutive decades, from 0.01 mr/hr to $1\ 10^6$ r/hr (six on special order).	One electrometer tube in each detector. Low voltages, 1 VR tube in each power supply. Zener regulation in each plug-in.	Response time is inversely proportional to intensity. Approximately 10 s at 1.0 mr/hr. Accuracy to $\pm$ 2 %.	Remote calibration available (Sr 90). Remote meters and lights remote reset buttons. Automatic alarm reset available.
B	6,000-10,000	As above.	Three or six consecutive decades, from 0.01 mr/hr to $1\ 10^5$ r/hr.	Probably as above.	Inversely proportional to flux. Approximately 12 s at 10 mr/hr, 4 s to 1 r/hr.	Remote calibration available (Sr 90).
C	5,500-8,000	*Scintillation* Sensed to gamma flux	0.05 to 50 mr/hr log, others available.	PM tube w/HVPS, two vacuum tubes and VR's per detector. Not sensed to line transients. Fail light.	Probably approximately 2 s at 1 mr/hr.	Will not saturate in high fields. Meter relay bell on each unit.
D	5,000-8,000	*GM Counter* Halogen quenched. 10^{10} counts minimum. Gamma flux sensed. Not damaged by overvoltage or high intensity.	0.01 mr/hr to 10 r/hr, six decades.	Transistorized ratemeter.	Time constant, 1.8 s.	Check source Sr 90 remote. Has memory to cover short bursts. At 6 s intervals motor driven ckt. resets meter-relays.

state of a process vessel in which the distribution of the fissile material in the vessel is not known. In principle, the pulsed neutron source and time analyser method can be used to measure the nearness to the critical state with a single measurement, provided that the solution contained in the vessel is homogeneous and provided that an estimate can be made of the buckling of the vessel.

In spite of this, a number of devices known as in-line instruments have been put into use to provide accurate measurements on solution concentrations and to monitor the changes in concentration of various process streams. The by-product of these measurements is also nuclear safety control. Table I includes several of these devices and lists information on principles of operation and types of process streams in which they are most applicable.

All the above policies, procedures, and aids have been aimed at preventing nuclear incidents with their very real effects and also their intangible effects on world opinion which reflect on, and tend to restrict, the nuclear industry. However, as one does not ignore fire-fighting equipment and fire departments just because one is working for fire safety, so too, in the field of nuclear safety, it has been the policy of the various organisations to provide equipment, staff and training to cope with nuclear incidents.

Radiation Alarms

Gamma-sensitive radiation devices which energize audible alarms in the event of an accidental criticality can be found in nearly all United States processing plants. Mosts of these instruments fall under three types: the ionization chamber, the NaI(Tl) scintillator; the Geiger.

The Licensee is required by law (10 CFR-70) to provide an instrument that will give an alarm for a steady-state gamma field of not less than 5 mr/hr. In no event should this device be farther than 120 ft from the areas handling special nuclear material; intervening shielding will lessen this distance. The Prime Contractors also have radiation alarms.

The value of this type of instrument is that it enables personnel to evacuate the area promptly, and prompt evacuation is advisable in order to minimize exposure due to continuing types of excursion and to reduce exposure to delayed gamma. Several commercially available devices are listed in the Appendix.

In addition to the radiation alarms, gamma and neutron films are used extensively to arrive at personnel dose. Threshold detectors containing sulphur, gold, plutonium, uranium and other foils help in evaluating the neutron spectrum for a given incident. This information is used, in conjunction with the Na 24 content of a victim's blood, to arrive at a dose value. Other methods are also used.

Concluding Remarks

There are two distinct types of plant in the United States: the Prime Contractor plants owned by the U.S. Atomic Energy Commission and operated by private industry, and the Licensee plants owned and operated by private industry. The difference between these two from the nuclear safety control point of view lies in the method used in arriving at nuclear safety limits. The Prime Contractor develops and is responsible for his own nuclear safety limits, whereas the Licensee receives approval from the Commission.

Supervisory personnel in both plants are responsible for nuclear safety. The nuclear safety groups are advisory in nature and are established to provide the supervisory services with advice which will enable them to perform their job safely.

Personnel down to the chemical operator (non-supervisory) receive nuclear safety training in how to handle and work safely with fissile materials. Each operator may be given the responsibility of moving fissile materials from one work area to another by his immediate supervisor.

Practically all plants contain instrumentation which will warn personnel of an inadvertent nuclear excursion. Although considerable thought has been given to devices which warn of a potential criticality, results as yet have not provided reliable instrumentation.

ACKNOWLEDGMENTS

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In addition to the above Licensees, personnel from eight Atomic Energy Commission Prime Contractor plants were previously interviewed concerning their nuclear safety policies.

APPENDIX

MANUFACTURERS OF GAMMA ALARMS

1. Eberline Instrument Corporation, Box 279, Santa Fe, New Mexico.
2. William B. Johnston and Associates, Incorporated, Post Office Box 415, Mountain Lakes, New Jersey.
3. Nuclear Materials and Equipment Company, Apollo, Pennsylvania.
4. Nuclear Measurements Corporation, 2460 North Arlington Avenue, Indianapolis 18, Indiana.
5. Riggs Nucleonics Corporation, 717 North Victory Boulevard, Burbank, California.
6. The Victoreen Instrument Company, 5806 Hough Avenue, Cleveland 3, Ohio.
7. Tracerlab, Incorporated, 2030 Wright Avenue, Richmond 3, California.

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REFERENCES

1. R.E. CONNALLY, U.L. UPSON, P.E. BROWN and F.B. BRANER, " Uranium Analysis by Gamma Absorptometry ", *HW-54438* (1958).
2. G.J. ALKIRE, " Information for Application of Gamma-Absorptometers for Chemical Processes ", *HW-45664* (1956).
3. D.G. MILLER and R.E. CONNALLY. " A Gamma Absorptometer for the In-Line Determination of Pu or U ", *HW-36788* (1955).

4. D.G. MILLER, " Am^{241} as a Photon Source for the Gamma Absorptometric Technique ", *HW-39971* (1955).
5. R.E. CONNALLY, " Uranium Analysis by Gamma Absorptometry ", *Nucleonics, 17* No 12, 98 (1959).
6. M.B. LEBŒUF, D.G. MILLER and R.E. CONNALLY, " Gamma Absorptometry — A New Tool for Analytical Chemistry ", *Nucleonics, 12* No. 8, 18 (1954).
7. W.L. MADDOX and M.T. KELLEY, " A Gamma Absorptometer for Laboratory Analysis of Heavy Elements ", *Talanta, 3*, 172 (Dec. 1959).
8. G.A. ROST, *RFP-180* (All On In-Line Filter Photometer).
9. W.N. CARSON and C.E. MICHELSON, " An Automatic Photometer ", *HW-27744*(1953).
10. A.H. BUSHEY, " The Less Familiar Elements in the Atomic Energy Program ", *HW-29599*, p. 25.
11. W.H. REAS, " Automatic In-Line Monitor for pH Gamma Activity, and Uranium in the Metal Recovery Plant ", *HW-35291* (1955).
12. N.T. HILDRETH, " In-Line Alpha Monitor Prototype Performance ", *HW-37272* (1955)
13. E.C. WINGFIELD, " Alpha Monitor for Waste Streams ", *DP-197* (1957).
14. G. FRIEDLANDER and J.W. KENNEDY, *Nuclear and Radiochemistry*, p. 190, John Wiley and Sons, 1955.
15. W.L. PILLINGER, " An Alpha Monitor for Process Streams ", *DP-302* (1958).
16. R.E. CONNALLY, " Design Notes and Component Recommendations for Gamma Scintillation Monitoring Systems ", *HW-38636* (1955).
17. " Manual for In-Line Analytical Instruments ", *HW-41093* (1956).
18. G.J. ALKIRE, " Technical Specifications for Gamma Scintillation Monitors ", *HW-39821* (1956).
19. E.C. WINGFIELD, " A Gamma Monitor for Liquid Streams ", *DP-342* (1958).
20. W.A. KROPP, " Neutron Monitor for Pu Inventory Control ", *DP-267* (1958).

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DISCUSSION

H. DOPCHIE, Belgique:

Dans le cadre de l'exposé de M. Lecorché relatif à l'instrumentation, j'aimerais signaler un appareillage existant au Centre de Mol et permettant de déterminer le rapport des quantités d'uranium 235 et de plutonium qui se trouvent dans un récipient. Cette détermination est non destructive ; en d'autres termes, elle ne demande ni l'ouverture du récipient ni la destruction de l'élément combustible éventuel et est également indépendante de la géométrie du récipient. Elle est fondée, en fait, sur la mesure des neutrons rapides produits par des neutrons thermiques incidents et du nombre de neutrons retardés. Malheureusement, cette détermination exige la présence d'un faisceau de neutrons important et, par conséquent, la présence d'un réacteur qui puisse fournir ces faisceaux de neutrons. Nous avons obtenu une précision de 5 % dans le rapport d'uranium 235 au plutonium; nous nous efforçons encore d'obtenir une meilleure précision. Le rapport final paraîtra d'ici trois mois.

A. J.MALLETT, United States:

Mr. Lecorché has mentioned checking by instrumentation up to 35 % of criticality in a plutonium mixer-settler equipment where the operator is warned by alarms. Do you have automatic valve closures to stop flows and are they in turn backed up by a manual closure valve?

P. LECORCHÉ, France:

Comme je l'ai fait remarquer, l'installation en est encore au stade des essais et l'on contrôle actuellement la concentration par l'analyse chimique. Cependant l'appareillage donne effectivement satisfaction et, nous avons été amenés, il y a quelque temps, à la suite d'une mauvaise opération, à arrêter l'installation manuellement sur alarme donnée par l'appareil. Il est certain que, si l'appareillage s'améliore encore et se révèle sûr, nous envisagerons alors d'utiliser des automatismes.

J. GUNTHER, France:

Je voudrais poser une question relative à un appareil servant à détecter les accumulations de matières fissiles dans des installations chimiques ou métallurgiques. Le principe de cet appareil est le suivant : une source de neutrons qui n'émet que des neutrons thermiques est disposée à proximité de l'appareillage dont on recherche le contenu et l'on détecte les neutrons rapides qui pourraient se produire à la suite de fissions induites par ces neutrons thermiques. Ces neutrons rapides sont détectés au moyen d'une chambre à fission dont les dépôts sont constitués d'uranium 238 ou de thorium qui ne sont évidemment sensibles qu'aux neutrons rapides, lesquels ne peuvent pas provenir directement de la source. Je voudrais savoir si quelqu'un a étudié ces dispositifs et est arrivé au stade de la réalisation.

H.C. PAXTON, United States:

I would like to take advantage of Mr. Howells' very appropriate reference to the work of nuclear safety specialists as an art.

I claim that it is not until the work of the specialist can become more of a technical matter than an art that nuclear safety information can approach the point of adequacy. We have noticed that there are many hidden factors of safety in the judgment of criticality matters and I believe that until these hidden factors can be brought into the open, there is need for more basic information. A point that I would like to address particularly to administrators is again this economic factor: it seems deplorable that nuclear safety studies must be gauged in terms of this year's operations; I know this is the way budgets usually are made out and this may be a fact of life, but I feel that such work should be considered rather as an investment in the future.

H. HEIN, Germany:

The UKAEA has decided that the warning system must be capable of reacting to a dose of 25 rads over a gauge at a distance of 5 feet from any possible accident source. Is it right that this warning system has to give an alarm on only one detector?

H. HOWELLS, United Kingdom:

The system is based broadly on the same philosophy as that in reactor control: two such instruments out of a group of three have to react before an alarm sounds. The instruments are very carefully designed on the basis that if there is a failure in the system it tends to fail towards safety, in other words, it will operate the alarm rather than not respond. Also, built into the electronics of the systems are self-checking arrangements.

I was interested in Mr. Schuske's remark that the medical personnel would treat patients involved in a radiation accident on the basis of clinical symptoms. We find, at the moment, that due to the extreme difficulties in dosimetric analysis as carried out by the health physicist large uncertainties can occur in the estimated exposure. Consequently, treatment must be decided on the basis of clinical symptoms, though the health physicist is strongly pressed by medical personnel to improve his dosimetric techniques.

C. SCHUSKE, United States:

The trend certainly may change as time goes on, but the medical profession, as Mr. Howells points out, is not too happy with the present methods of determining dose by dosimetric means.

A.J. MALLETT:

I might mention that, in conjunction with the other Oak Ridge facilities, there were completed recently a rather large number of experiments with various foils with reference to measuring radiation effects. The data are so numerous that they have not been evaluated and tabulated properly, but we should know considerably more in the near future.

H. HOWELLS:

I should mention that we also use an indium strip detector as a method of making a quick segregation of personnel indicating those who have received significant radiation exposure from those who haven't. On plants which handle fissile materials in quantities we also issue a special dosimetric pack containing a tantalum disc for measuring the slow neutron exposure and a sulphur disc which will make measurement of the more energetic neutron dose, together with a phosphate glass to indicate the gamma radiation exposure. While this is not as comprehensive as one might wish, since intermediate range neutron exposure will not be recorded in this type of pack, at least it helps to give an indication of the levels of exposure involved. We are at present still in the phase where we have to try a variety of indicators to give a number of estimates of the exposures incurred.

G.W.K. FORD, United Kingdom:

Has it been found practicable to devise criticality alarm systems which are directional, giving those evacuating a building a chance to run in the correct direction rather than pass the incident which may be producing successive pulses?

C. SCHUSKE:

Some thought has been given to this, but thus far we do not have instrumentation of this type.

H.C. PAXTON:

I might just mention that there have been some developments along this line. One such item was brought to Los Alamos for a test, but moving it was about the same as moving a reactor.

G.W.K. FORD:

Does one consider that there is a difference in regard to the likelihood of a succession of pulses arising on the one hand from solution systems, which experience has shown can produce a series of pulses, and on the other hand from stored solid materials like metal and oxide fuel elements? Do they feel that stored solids present the possibility of a succession of pulses, and should one adopt a different philosophy in regard to alarm systems in these two cases?

W.R. STRATTON, United States:

It is hard to see how a metal system would give a series of pulses. Generally the history is a sharp spike followed by steady power at a much lower and usually decreasing rate. The pulses found in solution systems are usually such that the first pulse is much larger than any succeeding pulses by an order of magnitude, and any solution system, of course, will eventually start to boil, reducing the power to a lower level.

A REVIEW OF CRITICALITY INCIDENTS*

W. R. STRATTON
Los Alamos Scientific Laboratory, United States

INTRODUCTION

Since the beginning of the atomic energy industry there have been no less than twenty-four occasions when the power level of fissile systems became uncontrollable because of unplanned or unexpected changes in the reactivity of the system. At least three of these power excursions were planned as such, but for various reasons the energy release was significantly larger than expected. Of these twenty-four cases, five have caused seven deaths, two of which occurred in the early, rushed period towards the end of World War II. It is hoped that this review will illuminate some of the causes of accidental excursions and aid to some extent in understanding the physical phenomena that control their behaviour. Such a study is of interest in itself, but of more importance is the possibility that in time we may be able to limit the magnitude of such excursions and minimize or eliminate radiation injuries and damage to property.

Of these twenty-four unexpected and often very complicated events, we may ask how many can be understood in a satisfactory, quantitative fashion. A brief answer is that it can be done in several cases: in others we can fix certain limits and in a few we must restrict ourselves to qualitative statements of the events. A complete analytical description of all the events would be interesting and would satisfy our scientific curiosity, but for some this probably never will be possible because of inadequate data.

This review attempts to include all those incidents where, for some short period of time, the reactivity and fission power level were uncontrolled and increased independently of the efforts of the operators or experimenters. During many of these excursions the reactivity exceeded prompt critical**, a most dangerous situation because of the possible very short periods for power generation, but as will be seen, this criterion is neither necessary nor sufficient to guarantee personal injury or extensive damage to property. Wherever possible, an estimate of the maximum reactivity will be made. It is realised that the list may not be complete; in particular, near-misses — cases where

* Work performed under the auspices of the U.S. Atomic Energy Commission.

** Much of this review is taken from a similar article by W. R. Stratton, "A Review of Criticality Accidents" (*Progr. Nucl. Energy, Technology, Engineering and Safety, Series IV*. Vol. 3. Pergamon Press, London, 1960). In particular the description of eighteen incidents thought to have been prompt critical is taken nearly verbatim and the analysis of these (and some other sections) is condensed. Six incidents not examined in the earlier review are discussed here and only references for these incidents are included hereinafter. To complete the list of references the reader should consult the earlier review.

a power excursion was narrowly averted — have not been discussed at all. The causes and results of the several excursions will be discussed qualitatively and arranged according to type, with some mention of their analysis and a brief mention of the large amount of experimental and theoretical work that has been done in this field, while the contents of the review will be summarized to some extent in an attempt at applying them to production plant problems.

The Incidents [1]

Twenty of the systems in which the excursions occurred can be classified according to three major types: eight water solutions of U 235 or Pu 239, five metal assemblies in air, and seven inhomogeneous light or heavy water-moderated systems. The remaining four assemblies are unique (in one case, unknown) and will be discussed under ' Miscellaneous '. A summary of the data concerning these incidents is presented in Tables I to III. For easy reference, the date, location, active material, geometry, number of fissions and resulting damage are listed.

Water Solutions of U 235 or Pu 239

The Los Alamos Scientific Laboratory — December 1949 (water boiler reactor; control rods removed by hand). This incident occurred while two new control rods (poisons) were being tested in the water boiler reactor. The water boiler is a 12-in. dia. stainless steel sphere containing 13.6 litres of a water solution of uranyl nitrate. In 1949 it was reflected by a thick graphite shield.

The rods had been installed and the operator was checking manually their dropping times. After several tests of each rod individually — a safe procedure since one rod was sufficient to maintain a subcritical reactivity — both rods were pulled, held for about five seconds and then dropped simultaneously. A short time later the rods were pulled again and dropped together a second time. The removal of the two rods increased the reactivity to about 3 cents* over prompt critical, corresponding to a period of 0.16 s. The power probably rose on this period to a very broad peak of 2 or 3×10^{16} fissions/s and remained close to this value for about 1.5 s. The excursion was not detected immediately because all equipment was turned off except for a direct reading thermometer that showed a temperature rise of 25° C, equivalent to a yield of 3 or 4×10^{16} fissions.

In this incident the operator received 2.5 r of gamma radiation. No damage was done to the reactor.

The Hanford Works, Richland, Washington — 16th November, 1951 (plutonium solution assembly; cadmium rod removed too rapidly, remote control). The critical assembly in which the excursion occurred was a water solution of 1.15 kg plutonium in the form of plutonium nitrate contained in an unreflected aluminum sphere of 20-in. dia. The purpose of the experimental programme was to determine the maximum safe mass of plutonium for various container geometries and solution densities.

* The dollar, or 100 cents, is defined as the reactivity increment between delayed critical and prompt critical.

The excursion occurred during the approach to critical when the sphere was 93 % full, as a result of withdrawing a remotely-controlled hollow cadmium safety rod too rapidly in a series of steps with insufficient waiting time between the steps. The excursion yield was 8×10^{16} fissions and a small amount of fuel was forced through gaskets at the top of the reactor assembly. As these gaskets sealed a volume of air of about 18 litres above the fuel level prior to the incident, pressures considerably in excess of atmospheric must have existed in the assembly during or immediately after the incident. The published data suggest that the reactivity insertion rate resulting from the safety rod withdrawal must have been about 4.7 dollars/s which would lead to a fission yield about twice that of the observed value if known values of temperature and void coefficients of reactivity are used. In this case, however, the action of the scram circuit was sufficiently fast that the cadmium rod most probably contributed to the shut-down of the excursion. A slight reduction in the assumed reactivity insertion rate would lengthen the time scale, making it even more certain that the excursion was stopped by the falling poison rod.

In this excursion there was no injury to personnel, but plutonium nitrate solution contaminated the experimental area. The rest of the building was successfully decontaminated within a few days, but before clean-up of the test area was completed a fire occurred and the building was abandoned.

The Oak Ridge National Laboratory — 26th May, 1954 (uranium solution assembly; central poison cylinder tilted to a less favourable position, shielded and remote operation). The experiment in progress was one of a series in which the critical properties of aqueous solutions in annular cylindrical containers were being investigated. The outer cylinder was of 10 in. dia. and an inner cadmium-clad cylinder was of 2 in. dia. The system was unreflected and consisted of 55.4 litres of a water solution of UO_2F_2 which contained 18.3 kg enriched (93 % U 235) uranium.

The excursion occurred while the liquid level was at 40 in. and more solution was being added slowly to approach a delayed critical configuration. Figure 1 illustrates the experimental situation before and after the incident. The inner cylinder was essentially a poison rod, and when it became detached from its connection at the top and tipped to the side of the outer container, it fell to a less effective position, raised the system reactivity well over prompt critical, and caused a power excursion of 10^{17} fissions. The reconstruction of this incident was most thorough. The tilting inner cylinder added reactivity to the system at a rate corresponding to 3-1/3 dollars/s which continued well into the prompt critical region. Using known coefficients and generation times, an initial power spike* of 5.1×10^{16} fissions can be calculated. Since this power spike required only ~ 0.07 s after the system reached prompt critical (0.43 s after the cylinder began to tip), the cylinder was still falling, and it is characteristic of such incidents that after such an initial spike, the power will adjust to a value that will cancel the reactivity insertion rate. For this solution, the required power was a few MW, which must have been fairly constant until the inner cylinder reached its maximum displacement of 0.91/s after inception of the transient. At this time the power dropped sharply and when the liquid commenced to drain, the system became safely subcritical.

Because of thick shielding, no person was irradiated in an amount greater

* The terms 'power spike' and 'plateau' are defined and discussed on page 516

than 0.9 rem, only a few tens of cubic centimetres were displaced from the cylinder, and the area was returned to normal experimental procedures within three days.

The Oak Ridge National Laboratory — 1st February, 1956 (uranium solution assembly; wave motion created by falling cadmium sheet, shielded and remote operation). In this experiment certain reactor parameters were being investigated by measuring stable reactor periods. The system was a deep tank of 30 in. dia., filled to a depth of 5 in. with 58.9 litres of water solution containing 27.7 kg U 235 in the compound UO_2F_2. Transfer of solution from storage to the test cylinder was effected by the application of air pressure to the storage vessel, and flow was controlled by a remotely operated valve in a 0.5-in. dia. line. With the control switch in the ' feed ' position, this valve was open and the air pressure was applied; with the switch in the ' drain ' position the valve was also open, but the air supply was turned off and the storage vessels were vented to the atmosphere. When the switch was in the intermediate ' neutral ' position, the valve was closed and the storage vessels were vented.

The situation was one in which the solution volume was about 100 ml less than the critical volume. An increment of solution was added and the transient period decreased rapidly to approximately 30 s when it seemed to remain constant. Shortly thereafter the fuel control switch was placed in the ' drain ' position and the period meter indicated a rapid decrease in period, so that the safety devices were actuated more or less simultaneously by both manual and instrument signals; the instrument trip-point had been set at a 10-s period. Immediately thereafter, the excursion occurred. The burst yield was 1.6×10^{17} fissions and in this case a " considerable volume " of solution was forcibly ejected from the cylinder. Post-excursion tests showed that addition of solution to the reactor could have continued for several seconds after the control switch was placed in the drain position if insufficient time were allowed for the operation pressure to be vented. This addition of solution accounted for the decrease in period that precipitated the scram, but the increment of solution could not have added reactivity enough to account for the excursion.

It had been observed that the reactivity of such shallow large-diameter assemblies is very sensitive to the solution depth, but quite insensitive to changes in the diameter. For this system, the estimated difference between delayed critical and prompt critical is only a millimeter in depth. If the effective diameter were reduced to 20 in., the depth would have to be increased by only 0.5 in. to maintain delayed critical. It is thought that the falling scram, a cadmium sheet slightly deformed at the bottom, set up a wave system which must have converged at least once and created a super prompt critical geometry. In this case the analysis was directed to finding what reactivity insertion rate would cause a power spike in the system of the required yield. The problem was then examined to see if it contradicted any known facts. It was found that a rate of 94 dollars/s was adequate to cause a power spike of duration over prompt critical of only 8 ms and accounting for the observed yield. The maximum excess reactivity would be about 2 dollars over prompt critical and the void volume could be twelve times that of the previous case, easily accounting for the splashing of the solution.

A considerable volume of solution was ejected from the cylinder, requiring a laborious chemical decontamination of the assembly room. No damage

was done except for distortion of the bottom of the cylinder. No person was irradiated in an amount greater than 0.6 rem.

The Y-12 Chemical Processing Plant, Oak Ridge, Tennessee — 16th June, 1958 (uranium process solution combined with wash water in 55-gallon drum; unshielded operation). The nuclear incident occurred in a processing area in which enriched uranium is recovered from various materials by chemical methods in a complex of equipment. This recovery process was being remodelled at the time, and the situation was further aggravated by an inventory then in progress, which required disassembly, cleaning, reassembly, and leak testing of certain pieces of equipment, particularly several long 5-in. dia. pipes used for storage of aqueous solutions of U 235. The shape and dimensions of these pipes were such that contained solutions could not become critical. The inventory procedures extended over several days, and operations had been re-established in the step immediately ahead of the area in which the incident occurred. As a consequence of this close overlapping of operations, and irregularities in the function and operation of some valves, a quantity of solution was inadvertently transferred from the area already in operation into the one still undergoing leak testing. It has been established subsequently that the flow pattern from these pipes into a drum intended to receive water that had been used to test for leaks was such that the accumulated solution preceded the water. The dimensions of the 55-gallon drum (about 22 in. dia.) permitted the solution to become critical. Further flow of water first increased the uncompensated reactivity* for about 11 min, then decreased it, and the solution became subcritical after a total of about 20 min.

When the system became critical, the solution volume was thought to be 56 litres, a cylinder 23.45 cm high and 55.2 cm in diameter. The U 235 mass at this time was 2.1 kg; 0.4 kg was added later during the time when water was diluting the system. During the excursion a radiation detection instrument consisting of a boron-lined ionization chamber, amplifier and recorder was operating about 1,400 ft distant and cross-wind from the area of the incident. The trace shows that the radiation intensity first drove the pen off scale and within about 15 s drove the pen off scale a second time. During the next 2.6 min, the trace oscillated an indeterminate number of times. It is possible that these oscillations were ordered such that they were decreasing in amplitude, but this cannot be proven by examination of the trace. This average high intensity field was followed by a slowly decreasing level for 18 min, described as about five times background.

The power history can be reconstructed only qualitatively. The most likely source of initiation was neutrons from the reaction O^{16} (α, n) Ne^{19} between U 234 alpha particles and the oxygen in the water, so that it is possible that the system was over prompt critical before the first excursion started. The reactivity insertion rate was about 0.17 dollars/s at this time, a relatively low value, and the first spike must have been determined by the reactivity attained when the chain reaction started. Though there is no way to estimate this, a reasonable guess is that the first spike contributed about 10^{16} out of the total yield of 1.3×10^{18} fissions. The second oscillation or spike (which also drove the recording pen off scale) occurred in 15 s, a quite reasonable

* It is convenient occasionally to discuss the reactivity which would pertain to a fissile system if the state of the system were not altered by its power: this is referred to as the 'uncompensated' reactivity.

value of time during which existing bubbles could have left the system. The oscillations for the next 2.6 min appeared to be no greater than about 1.7 times the average power. The power trace suggests that most of the fissions occurred in the first 2.8 min, in which case the average power required to account for the observed yield is about 220 kW. After this period, the system probably started to boil, causing a sharp decrease in density and reactivity, and reduced the power to a low level for the final 18 min.

During this incident, 1.3×10^{18} fissions occurred. There was no damage or contamination. Eight people were irradiated in the amounts of 461, 428, 413, 431, 298, 86.5, and 28.8 rem. At least one person owes his life to the fact that prompt and orderly evacuation plans were followed.

The Los Alamos Scientific Laboratory — 30th December, 1958 (separated phases in plutonium process tank; unshielded operation). The operations underway at the time of this incident were those chemical steps used to purify and concentrate plutonium from slag, crucible and other lean residues resulting from recovery processes. Typical and expected solutions contained less than 0.1 g/litre plutonium and traces of americium.

At the time of the incident, a physical inventory was in progress so that the normal flow into the area was interrupted and residual materials in all process vessels were to be evaluated for plutonium content. Reconstruction of significant events indicates that plutonium-rich solids, which normally would have been handled separately, were washed from two other vessels into a large vessel that contained dilute aqueous and organic solutions. After removal of most of the aqueous solution from this vessel, the remaining ~ 52 gallons of material, including nitric acid wash, were transferred to the 225-gallon, 38-in. dia. stainless steel tank in which the incident occurred. This tank already contained ~ 78 gallons of a caustic-stabilized aqueous-organic emulsion and the added acid is believed to have separated the liquid phases. The bottom layer (87.4 gallons) is thought to have contained 60 g Pu; the top layer (42.2 gallons) contained 3.27 kg Pu. Estimates indicate that the 8-in. thick layer was perhaps 5 dollars below delayed critical and that the critical thickness was 8.25 in. When the motor drive of a stirrer was started to mix the solutions, the initial action was to force solution up along the wall, displacing the outer portion of the upper layer and thickening the central region. This motion changed the reactivity of the system from about 5 dollars subcritical to super prompt critical, and a power excursion occurred. None of the gamma-sensitive recording meters within range of the incident showed an ideal trace; they suggest, however, that there was but a single burst. The excursion yield was 1.5×10^{17} fissions.

From post-excursion experiments in a similar geometry it was observed that (*a*) there was no apparent time delay between start and full speed at 60 rpm, (*b*) after 1 s (i.e. one revolution) there was a visible movement or disturbance on the surface, and (*c*) within 2 to 3 s the system was in violent agitation. From these observations it can be concluded that the system could have been made critical within about 1 s and within no more than 2 to 3 s it must have been far subcritical, and the burst terminated.

Since it was estimated that initially the system was 5 dollars subcritical, the reactivity insertion rate must have been about 5 dollars/s. This, with coefficients consistent with the solution, leads to a burst yield of 2.2×10^{17} fissions with the burst completed in a time of 1.65 s, 0.45 s after reaching prompt critical. To obtain the observed yield (1.5×10^{17} fissions) in a single

spike, the reactivity insertion rate must be reduced to about 2 dollars/s. As this is inconsistent with the time scale, namely complete mixing in 2 s, the only remaining alternative is to assume that the rate was somewhat less than 5 dollars/s and that the excursion was terminated in about 2 s by the stirring action. One can guess that the initial action was to thicken the upper layer, followed almost immediately by distortion into a less critical, vortex-like geometry by the action of the stirring blades.

This incident resulted in the death, thirty-six hours later, of the operator who was looking into a sight glass when the motor was turned on. The radiation dosage was estimated to be 12,000 $\pm$ 50 % rems. Two other persons were irradiated in the amounts of 134 and 53 rems and suffered no ill effects whatsoever. There was no contamination or damage to equipment, even though the shock displaced the tank about 3/8 in. at its supports.

The Idaho Chemical Processing Plant, Idaho Reactor Testing Area — 16th October, 1959 [2] *(enriched uranium solution siphoned from a safe to an unsafe geometry; shielded operation).* This incident occurred in a chemical processing plant which accepts, among other items, used fuel elements from various reactors. The active material involved (34 kg enriched uranium, 93 % U 235) was stored in a bank of safe containers in the form of uranyl nitrate concentrated to about 170 g U 235/litre. During an air-sparging operation a siphoning action was initiated which transferred about 200 litres of this solution to a 5,000-gallon tank containing about 600 litres of water. The resulting power excursion created 4×10^{19} fissions, sufficient to boil away nearly one-half the 800-litre solution volume. The siphoning rate was equal to 13 litres/min, but the reactivity insertion rate would depend on the degree of mixing; it could have been as high as 0.25 dollar/s. Since the 9-ft dia. tank was lying on its side, the solution configuration would be a near-infinite slab and waves in the solution could have caused violent fluctuations in the reactivity of the system.

The power history is a matter of conjecture — one can guess that it was similar to the Y-12 incident. It is not unreasonable to assume an initial spike of the order of 10^{17} fissions, followed by power oscillations and finally boiling for 15-20 min since much of the uranyl nitrate was found to be crystallized on the inner walls of the tank and most of the water had left the tank. The very large yield is connected with the large volume of the system and the time scale rather than with the violence of the excursion.

Personnel received no significant gamma or neutron doses because of the thick shielding, but beta dosages were 50 r (one person), 32 r (one person), and small amounts for seventeen persons, all obtained while the building was being evacuated. The physical equipment involved in the excursion was not damaged.

The Idaho Chemical Processing Plant, Idaho Reactor Testing Area — 25th January, 1961 (uranium process solution transferred from a safe to an unsafe geometry; shielded operation). This most recent chemical plant incident is thought to have been caused by a bubble (residuum from an earlier line unplugging operation) of high pressure air forcing about 40 litres of uranyl nitrate up a 5-in. dia. pipe into a cylinder of 2 ft dia. and 4 ft high. The excursion occurred in this latter volume when the level of the 200 g U 235/-litre solution was about 5 in. The yield has been determined to be 6×10^{17} fissions but no estimates have been made of the reactivity and power history.

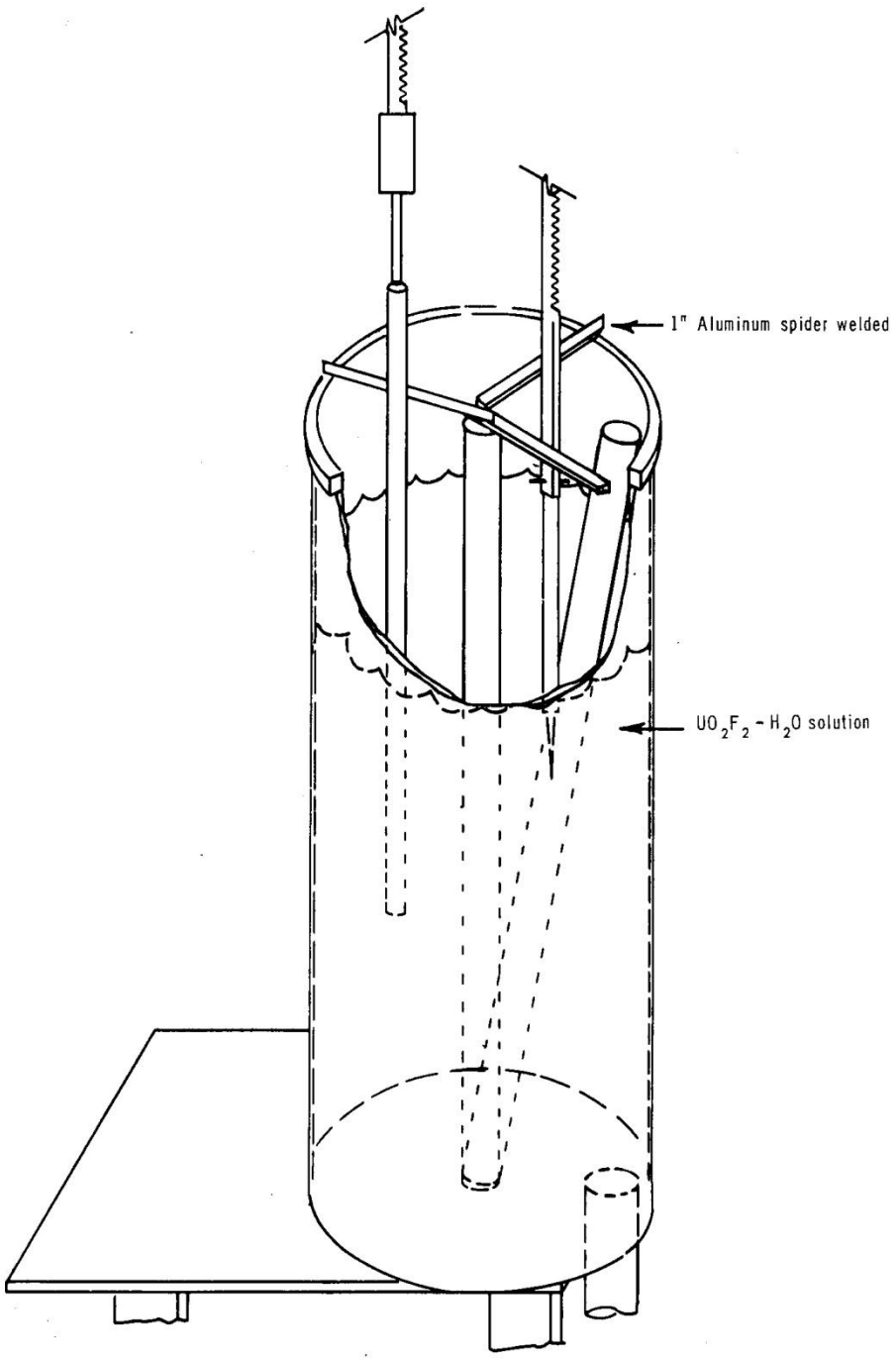

Figure 1. *Schematic of the ORNL uranium solution assembly indicating the normal and detached positions of the central poison rod.*

Prompt gammas created during the excursion activated an alarm system which caused the plant to be evacuated. However, full operations were resumed in less than an hour, irradiations were trivial and there is no reported damage. A full report on this incident has not yet been issued.

Metal Assemblies in Air

Los Alamos, New Mexico — 21st August, 1945 (plutonium core reflected with tungsten carbide; hand assembly) : 21st May, 1946 (plutonium core reflected with beryllium; hand assembly). These two accidental excursions occurred with the same core and were, in several respects, quite similar. The core consisted of two hemispheres of delta phase plutonium coated with 5 mils of nickel. The total core mass was 6.2 kg and the density about 15.7 g/cm^3.

In the earlier incident a critical assembly was being created by hand stacking 4.4 kg tungsten-carbide bricks around the Pu core. The lone experimenter was moving the final brick in his left hand over the assembly for a total of 236 kg when he noticed from the nearby neutron counters that the addition of this brick would have made the assembly super-critical. As he withdrew his hand, the brick slipped and fell into the centre of the assembly, adding sufficient reflection to make the system super prompt critical and a power excursion occurred. As quickly as possible, the final brick was pushed off and he proceeded to unstack the assembly. He died twenty-eight days later as a result of radiation injuries. The exposure was about 800 rep. The yield of this incident was 10^{16} fissions*. An Army guard assigned to the building, but not helping with the experiment, was irradiated in the amount of 20 rep. The nickel canning on the Pu core did not rupture.

In the later incident the techniques involved in creating a metal critical assembly were being demonstrated to several people. The system consisted of the same Pu sphere reflected in this case by beryllium. The top and final hemispherical Be shell was being lowered slowly into place; one edge was touching the lower beryllium hemisphere while the edge 180° away was resting on the tip of a screw-driver. The person conducting the demonstration was holding the shell with his left hand, with his thumb placed in an opening at the polar point, while slowly working the screw-driver out with his righth and. At this moment the screw-driver slipped from under the shell and the shell fell completely into the lower hemisphere. A burst occurred at once, the shell was thrown to the floor and all personnel left the room. The yield of this excursion was 3×10^{15} fissions and again there was no plutonium contamination. The eight people in the room were irradiated in the amounts $\sim$ 900, 185, 116, 93, 41, 26, 18, and 18 rep. The man who performed the experiment died nine days later as a result of radiation injury.

An analysis of these two incidents can be done in a semi-quantitative manner. By analogy to similar near-solid metal critical assemblies a rough prompt negative temperature coefficient of reactivity can be estimated ($\approx 3.6 \times 10^{-3}$ dollars/°C), and by comparisons with the behaviour of the Los Alamos GODIVA reactor (discussed below) the fact that the nickel coating

* The Los Alamos archives include some data and comments about a re-run of this incident on 2nd October, 1945, performed to determine the radiation dose received in the incident of 21st August, 1945. The yield of this re-run was about 6×10^{15} fissions but the prompt critical state was not reached. The maximum reactivity of the system during this experiment was about 60 cents above delayed critical.

did not rupture limits the excess reactivity over prompt critical to not more than 10 or 12 cents.

In the earlier incident, if the excess reactivity were 10 cents, the yield in the power spike would be 1.8×10^{15} fissions and the remaining fissions could have been created in the plateau in which region the power would be close to 10^{16} fissions/s. The time for this power to produce the remaining fissions would be 0.8 s, a not unreasonable value for the reaction time to remove a brick. Clearly, the observed yield can be satisfied with slightly different assumptions. The later incident can be examined in the same manner. If the excess reactivity over prompt critical were, for example, 0.01 dollar, the spike yield would be 18×10^{14} fissions and the remaining 2.9×10^{15} fissions could have occurred in the 0.28 s before the shell was removed. Again, the assumptions are not unique.

The Los Alamos Scientific Laboratory — 18th April, 1952 (JEMIMA, a cylindrical unreflected U 235 metal assembly; control one-quarter mile away). The system in which this excursion took place was a cylindrical, unreflected, enriched (93 % U 235) uranium metal assembly made up of a number of plates each of which was of 26.7 cm dia. and 0.8 cm thick. A photograph of the assembly (commonly called JEMIMA) in the scrammed condition is shown in Figure 2.

Complete assembly of the two components had been made previously with six plates on the lower component but first with three and then with four plates on the upper component. A plot of the reciprocal multiplication* *versus* number of plates or total uranium shows clearly that the system should not have been assembled with eleven plates. Nevertheless, such an assembly was attempted following a computational error made independently by two people. Contrary to operating regulations, a graph of the data had not been plotted. The burst yield was 1.5×10^{16} fissions.

There is no way of determining uniquely the power history experienced by the 92.4 kg mass without reproducing the experiment. At the time, the system was near prompt critical, the lift was coasting and probably inserting no more than 2 or 3 dollars/s, a rate which could cause a power spike of about 10^{15} fissions. The power would then stabilize at about 10^{17} fissions/s, just enough to compensate the reactivity insertion rate, so that most of the 1.5×10^{16} fissions occurred in this plateau. The power dropped to zero when the automatic scram system separated the two masses of metal.

During this remotely-controlled operation no damage was done to the system, even to the fissile material, no one was irradiated, and the experimental area was not contaminated. The apparent self-terminating property of this excursion stimulated study of GODIVA-I, which became a facility for generating large bursts of fission spectrum neutrons in times less than 100μ s.

The Los Alamos Scientific Laboratory — 3rd February, 1954 (GODIVA, a bare U 235 sphere; control rod maloperation, control one-quarter mile away): 12th February, 1957 (GODIVA, a bare U 235 sphere; added reflection, control one-quarter mile away). These two excursions occurred in the LADY GODIVA assembly, an unreflected metal reactor fabricated in three principal

* The multiplication is the ratio of the leakage neutron flux from the enriched assembly to the leakage flux from an identical normal uranium assembly each containing the same neutron source. Clearly, the reciprocal multiplication goes to zero at delayed critical.

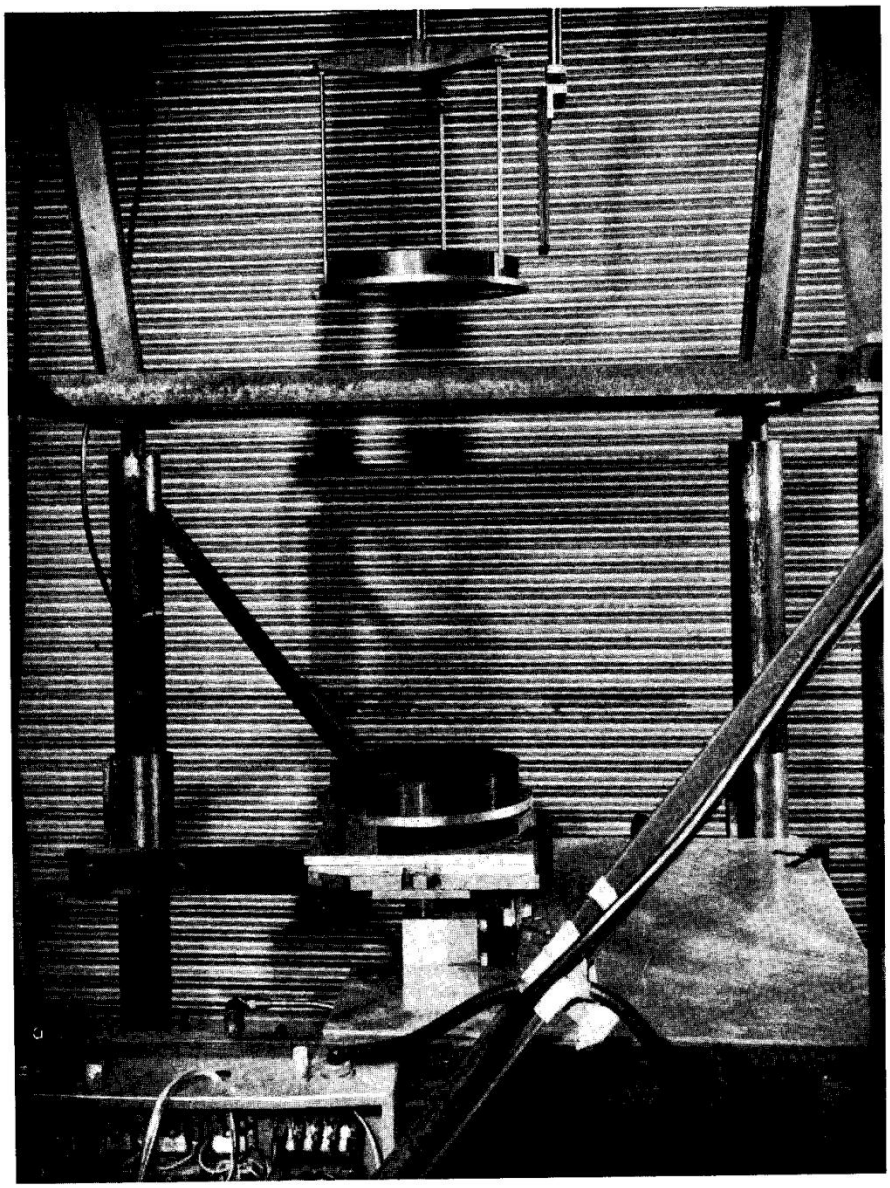

Figure 2. *LASL JEMINA enriched uranium metal assembly in the scrammed configuration.*

sections which assembled to form a sphere. Figure 3 shows GODIVA in the disassembled state. The central section was fixed in position by small tubular steel supports while the upper and lower sections were retractable by means of pneumatic cylinders, thus providing two independent scram mechanisms. The critical mass was about 54 kg uranium enriched to 93.7 % U 235. It was operated remotely at a distance of one-quarter mile.

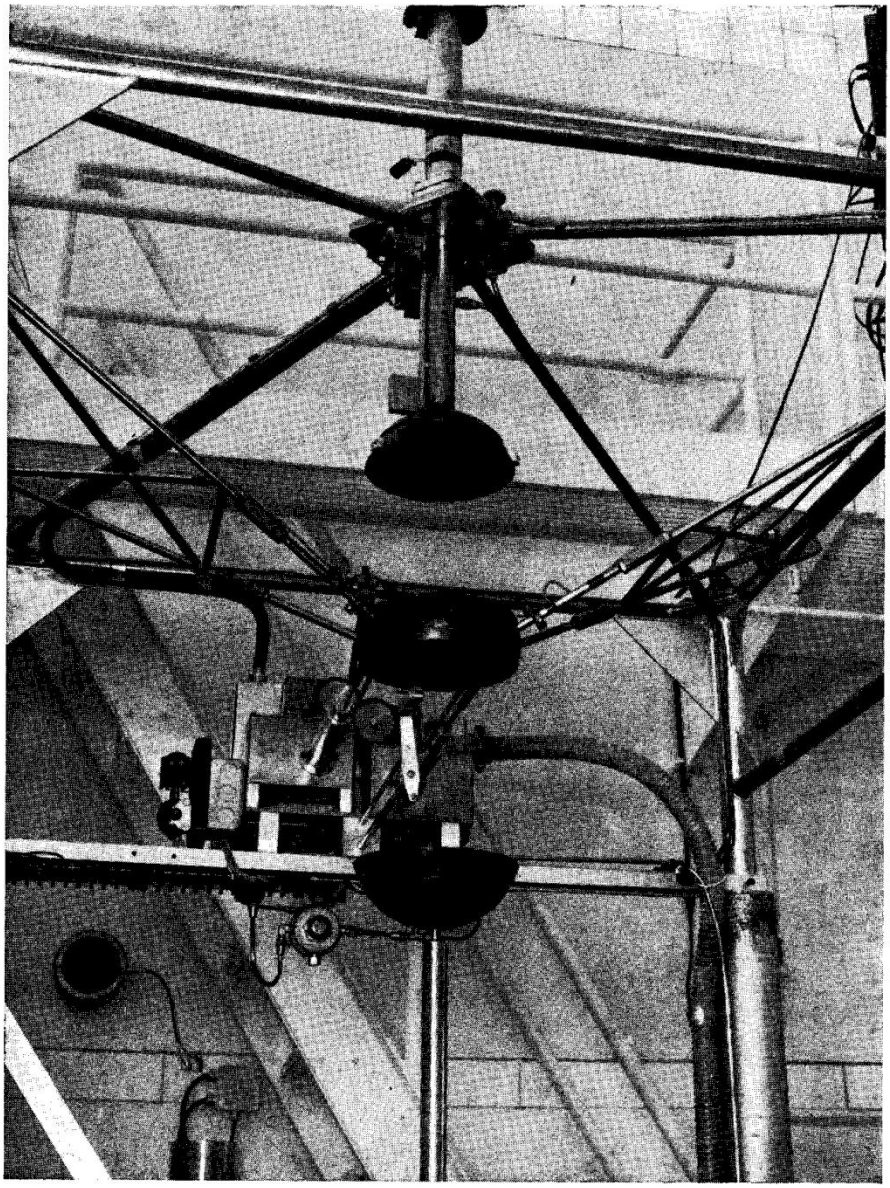

Figure 3. *LASL LADY GODIVA assembly (unreflected enriched uranium sphere) in the scrammed configuration.*

The first accidental excursion occurred during preparations for a scheduled prompt burst during a programme designed to measure the parameters associated with excursions. Generally, a burst was initiated by (*a*) establishing delayed critical by adjusting control rods, (*b*) lifting the top section to reduce reactivity to allow decay of the neutron population, and (*c*) lowering

the top section into position and rapidly inserting a burst rod worth slightly more than 1 dollar. A power excursion normally creating about 10^{16} fissions in 100μ s followed and in 40 ms the system was scrammed safely. As the only source of neutrons was that from spontaneous fission, it was customary to assemble to an excess reactivity of about 70 cents to accelerate the rise to sufficient power for establishing the settings for delayed critical in a reasonable time. This excursion was caused, apparently, because additional reactivity was inserted by error after assembly to 70 cents but before a fission chain started. The excursion yield was 5.6×10^{16} fissions, about six times the yield of the average burst. There was no radiation hazard, spreading of contamination, irradiation of personnel or significant damage to the major uranium parts. One piece was slightly warped and required remachining and several light steel supporting members were bent or broken.

The second accidental excursion occurred during preparations for an experiment in which GODIVA was to provide a pulse of fast neutrons. Again, the burst occurred during assembly to establish, in this case, a fiducial at about 80 cents excess reactivity. Control rods were to be adjusted on the basis of this period. The extra reactivity is thought to have been contributed by a large mass, which included graphite and polyethylene, that was to be irradiated. This mass had been moved closer to GODIVA, and either the change in reflection was underestimated or the material slumped further toward the reactor. The burst yield was 1.2×10^{17} fissions, about twelve times the standard excursion. The metal was severely oxidized, warped, and apparently had been plastic near the centre. The central burst rod was nearly ruptured and at its centre must have been within 100° C of the uranium melting temperature. Figure 4 is a photograph of several of the pieces. External damage was limited to the supporting structure; radioactive contamination consisted of oxide scale, and clean-up proceeded rapidly. Repair of GODIVA was not practical and construction of GODIVA-II, specifically designed for burst operation, was accelerated. Despite the severity of the excursion, operating personnel were not irradiated.

The behaviour of the GODIVA system during super prompt critical power excursions is well understood, both experimentally and theoretically. GODIVA-I experienced well over 1,000 safe, controlled bursts and a coupled hydrodynamics-neutronics code describes the behaviour of the system quite adequately. The first excursion (5.6×10^{16} fissions) must have been on an e-folding period of 6.4 μ s, equivalent to a reactivity excess over prompt critical of 0.15 dollar. The excess reactivity of the larger excursion (1.2×10^{17} fissions) was 0.21 dollar above prompt critical, corresponding to a period of 4.7 μ s. The fission yield of 1.2×10^{17} in the second incident is equivalent to the energy contained in 1.7 lb of high explosive but the damage (Figure 4) was much less than would have been caused by this quantity of high explosive. The above-mentioned code can predict the fraction of fission energy that is converted to kinetic energy, and, in this case, only about 1.4 % of the energy, or 0.024 lb high explosive, is available as kinetic energy to do damage. The damage was consistent with this latter quantity and it is evident that most of the fission energy was deposited as heat content.

Water-Moderated Metal Systems

Los Alamos, New Mexico — 6th June, 1945 (pseudosphere of uranium cubes, water-reflected; local control). The experiment underway was designed

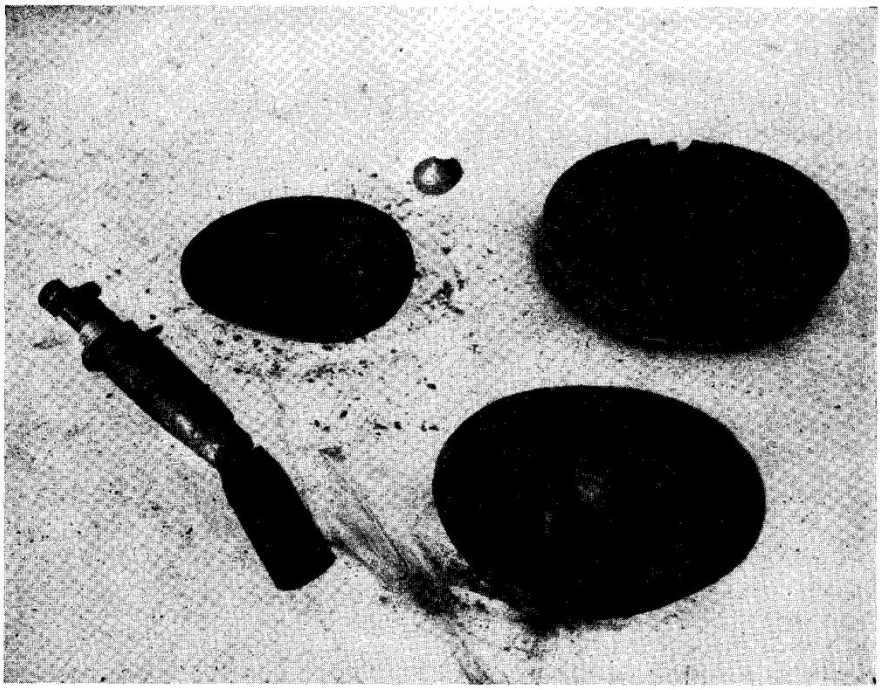

Figure 4. *Burst rod and several sections of LADY GODIVA showing the oxidation and warpage which accompanied the second accidental excursion.*

before the days of remote control and was intended to establish the critical mass of enriched uranium metal when it was surrounded by hydrogenous material. The uranium mass of 35.4 kg was stacked in the form of a pseudosphere constructed of 0.5-in. cubes and blocks 0.5 in. $\times$ 0.5 in. $\times$ 1.0 in. The core was in a 6-in. cubical polyethylene box, with the void space filled with polyethylene blocks. The whole assembly was placed in a large tank which was then partially filled with water.

The assembly became critical (unexpectedly) before water completely covered the polyethylene box. The situation was aggravated because no scram device was built into the system and the inlet and drain valves were 15 ft apart. Before the system was reduced to a safe subcritical state 5 or 10 s later, a total of 3 to 4 $\times$ 10^{16} fissions were created, an energy release sufficient to raise the average temperature of the metal more than 200° C. As subsequent examination of the polyethylene box showed that it was not watertight, it is probable that water seeped slowly into the assembly as the level was being raised above the bottom of the box. The additional moderation then caused the prompt critical situation which was quenched by boiling of the water within the box and next to the metal cubes.

The characteristics of excursions of large masses of fissile metal in water are at best poorly known. A calculation by Hensen has shown that for a U 235 sphere in water, 6.85 cm in radius, 15 % of the fissions occur in the outer 0.05 cm, and that the fission density in this region is six times that at

the centre. A burst of 3×10^{15} fissions would then raise the surface temperature 130° C while the central regions would remain relatively cool with a temperature rise of only 19° C. Apparently the first spike must have been of this order of magnitude with the majority of the fissions occurring at a much lower average power.

In this excursion three people received radiation exposures in the amounts of 66, 66, and 7.4 rep. There was no contamination and the active material was used again in three days.

The Los Alamos Scientific Laboratory — 1st February, 1951 (critical separation experiment; two large U 235 metal masses in water, control one-quarter mile away). A water-reflected system was set up in 1949 to obtain the neutron multiplication of a single piece or arrangement of fissile metal in water. The system had two scram devices. The first, with a quick response, consisted of a pneumatic cylinder which raised the unit out of the water, while the second and slower action drained the tank. A travelling support was added later so that critical separation distances between two units could be determined; a dropping cadmium screen provided an additional scram.

The experiment which precipitated the excursion consisted of measuring the critical separation distance of two enriched uranium masses in water, one a solid cylinder of 24.4 kg and the other a hollow cylinder of mass 38.5 kg, each of 93.5 % U 235 content. Sheet cadmium 10 mils thick was fastened to the outer surface of the solid cylinder and to the inside surface of the hollow cylinder, and a paraffin slug filled the cavity in the hollow cylinder. The general geometry of this experiment is shown in Figure 5.

At the completion of the critical separation experiment (at a multiplication of 65.5) the assembly was scrammed as a final flourish. Simultaneously, the water started draining, the cadmium screen dropped, the solid cylinder (left body in Figure 5) was lifting and an excursion (later determined to be 10^{17} fissions) was observed as jamming of counters and evidence on television of a vapour cloud above the water. Later reconstruction of the incident showed that the pneumatic tangential scram was the first to be effective and led directly to two types of difficulty. First, the centre of reactivity of the left-hand cylinder (Figure 5) proved to be below that of the stationary cylinder, and second, the rapid lift through the water created Bernoulli forces which swung the cylinders closer together. The combination of the two effects was enough to drive the assembly prompt critical and maintain this or a greater reactivity for 0.2 s, if the power excursion had not occurred. In a manner analogous to the earlier metal-water system, the first power spike is estimated to contain 6×10^{15} fissions. It is possible that one or more excursions into the prompt region followed as boiling was the primary quenching mechanism.

In this excursion of 10^{17} fissions no person was irradiated and no contamination was found in the experimental area. Damage to the uranium consisted of a very small amount of oxide flaking and blistering. The quarter-mile distant experimental area was in use two days later.

The Argonne National Laboratory — 6th July, 1952 (reactor mock-up; UO_2 particles in plastic, water-moderated). This incident occurred in a light water-moderated core in which 6.8 kg U 235 oxide were embedded in strips of polystrene plastic. The oxide particles were mostly (all but 0.5 %) 10 microns in diameter or less, the remainder of sizes up to 40 microns in diameter. Seven strips of the plastic fastened to six zirconium strips 0.91 in. × 0.110 in. × 43 in.,

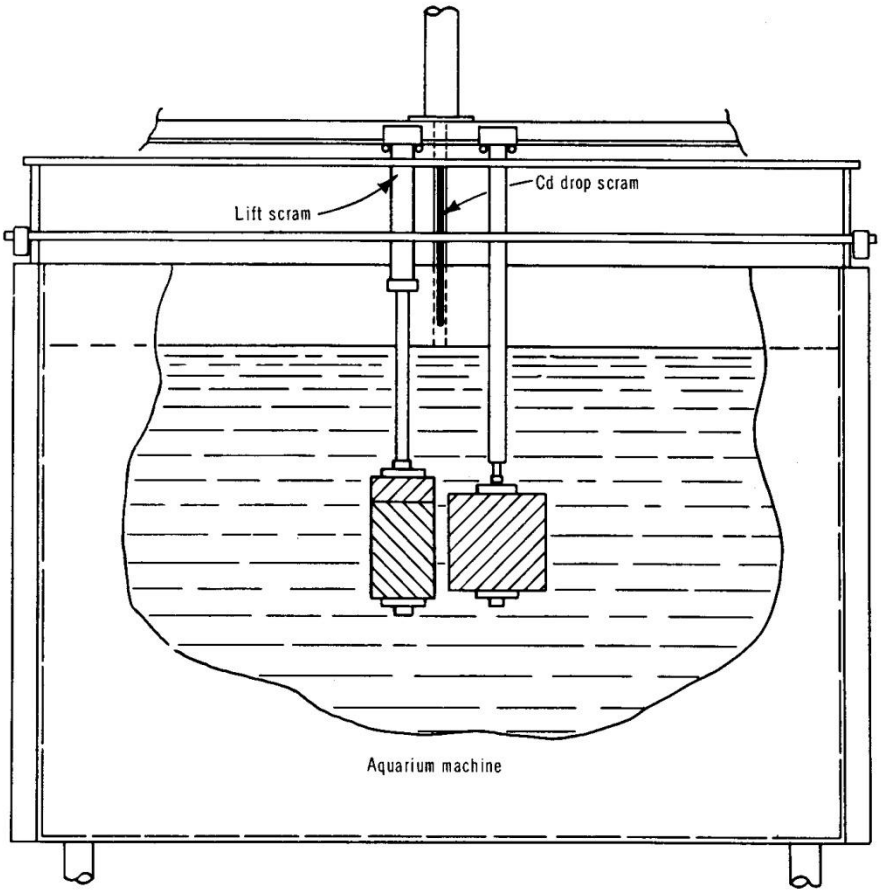

Figure 5. *Schematic of the LASL aquarium assembly machine which was employed for measurements of critical separation distances.*

formed one standard fuel element. The core was roughly cylindrical and contained 324 fuel elements. The zirconium, fuel strips and water occupied 60 %, 7.71 % and 32.2 %, respectively, of the core volume.

The experiment in progress at the time of the incident consisted of making comparisons of the worth of central control rods of different design. The system became super-prompt critical following an attempt (contrary to operating regulations) to replace the central control rod when the normal amount of water was in the core. Peripheral poison rods were in position but these were inadequate to maintain a subcritical reactivity. The quenching mechanism for this excursion of 1.22×10^{17} fissions was the near-uniform expansion of the plastic as the 10-micron size particles became hot, and bubble formation in the neighbourhood of the 40-micron size particles. This plastic expansion forced most of the water out of the core and the entire excursion was over about 0.6 s after the operator started raising the control rod. The maximum α (reciprocal period) was nearly 100/s, the maximum power was 1.7×10^8 watts and the half width of the power spike was 18.5 ms.

In this excursion, the core fuel elements were ruined but no significant amount of fissile material was lost. The activity in the reactor room was above tolerance for about a day, the core elements were removed after five days and decontamination was completed by a single application of detergent and warm water. Four persons were irradiated in the amounts 136, 127, 60 and 9 rep.

Atomic Energy of Canada, Limited, Chalk River, Ontario — 12th December, 1952 [3, 4, 5] *(NRX reactor; normal uranium rods, heavy water-moderated, graphite-reflected).* The NRX reactor is a natural uranium, heavy water-moderated system with the uranium rods cooled by a thin sheath of light water flowing between the aluminium clad rod and a slightly larger concentric aluminium cylinder. Space for 198 fuel or control rods is available in a graphite-shielded calandria of 8.75 ft dia. and 10.5 ft high.

Through a very complicated series of operator errors and electrical and mechanical safety circuit failures, the reactor was forced to be supercritical by about 0.6 dollar. Initially the power diverged rapidly but because of a slowly-moving control rod the reactor gave every indication of levelling off at a power of about 20 MW. Normally this would have been a high but tolerable power, and very likely the situation would have been controllable if the planned experiments had not required reduced light water cooling flow for several of the fuel rods. At a power of $\sim$ 17 MW the cooling water commenced to boil in those channels with reduced flow. This autocatalytic action (the light water was effectively a poison) increased the reactivity by about 0.2 dollar and the power again commenced to climb for a period estimated to be between 10 and 15 s. When the power reached 60-90 MW, the heavy water moderator was dumped and the reaction stopped.

The reactor power was greater than a MW for no more than 70 s and the total energy release is estimated to have been 4,000 MWs or about 1.2×10^{20} fissions. The core and calandria were damaged beyond repair and some 10^4 curies of long-lived fission products were carried to the basement by a flood of 10^6 gallons of cooling water. Irradiation of personnel was apparently low and the reactor was restored to operation in slightly over one year.

The Argonne National Laboratory, Idaho Reactor Testing Area — 22nd July, 1954 (BORAX reactor; aluminium-uranium alloy, water-moderated, remote control). This excursion was an incident only in the sense that it was larger than expected. The BORAX-I reactor had been built as a temporary affair, steady state and transient studies were regarded as complete and it was decided that the reactor should be forced onto a short transient period to obtain the maximum amount of experimental information before it was dismantled. The excess reactivity was chosen to produce a fission yield such that about 4 % of the fuel plates would melt.

The reactor consisted of 30 MTR-type fuel elements moderated by light water. Each element contained 18 fuel plates 2.845 in. $\times$ 0.060 in. $\times$ 24.6 in., consisting of aluminium-uranium alloy clad with about 0.020 in. aluminium. The whole core was in a semi-buried tank of 4 ft dia. and 13 ft high. It had been estimated from earlier controlled prompt excursions that about 4 % excess k would put the reactor on a period between 2.0 and 2.5 ms and that the resulting excursion would release about 80 MWs of fission energy. To perform this experiment, a larger than usual fuel loading was needed and a stronger central control rod required.

The excursion, following rapid ejection of the control rod, completely disassembled the reactor core and ruptured the reactor tank. Very extensive melting of the fuel plates had occurred, some elements remained in the tank while small pieces were found up to 200 ft away. " An example of the force of the explosion was the carrying away of the control rod mechanism. This mechanism, which weighed 2,200 lb, sat on a base plate, about 8 ft above the top of the reactor tank. Except for the base plate, about 4 ft square, the top of the 10-ft shield tank was essentially unobstructed. The force of the explosion, plus the impingement of water and debris on the base plate, tore the plate loose from its coverage, and, as revealed by high-speed movies, tossed the mechanism about 30 ft into the air. "

The total energy release was 135 MWs or, assuming 180 Mev deposited per fission, 4.68×10^{18} fissions. This energy is equivalent to that contained in about 70 lb high explosive but it has been estimated that between 6 and 17 lb of high explosive would produce comparable damage. The minimum period was 2.6 ms and the maximum power was about 1.9×10^{10} watts. It is apparent that the nuclear excursion was completed before the steam explosion destroyed the system.

In this excursion the reactor was destroyed, but because of the remote site, physical damage was limited to the reactor and personnel were not irradiated.

Vinca, Yugoslavia — 5th October, 1958 [6,7] *(unreflected, D_2O-moderated, natural uranium assembly ; unshielded).* The critical facility at the Boris Kidrich Institute in Vinca, Yugoslavia, is composed of an unreflected matrix of natural uranium rods moderated by heavy water. The aluminium-clad rods are of 2.5 cm dia. and 210 cm long; the total core uranium mass is 3,995 kg in a volume of 6.36×10^6 cm^3. Two cadmium safety rods were installed but were not interlocked with the flux recorder, and normally the liquid level was used to control the system reactivity (the critical level was 178 cm).

At the time of the incident none of the dosimetry, alarm and automatic safety control instruments was operating. The assembly became critical because of an unsuspected and uncontrolled rise of the heavy water moderator. The six persons in the assembly room (4 to 6 metres from the tank) first became aware of the reacting system by the smell of ozone, and the cadmium safety rods were used to stop the reaction after an energy release of 80 MWs and a time interval estimated to be between 3 and 7 min.

The critical level of D_2O was about 178 cm and near this level the D_2O was reported to be worth about $1.5\,\% \left.\frac{\Delta k}{k}\right/$ cm. The pumps operate at one of two speeds, either 2.5 cm/min or 0.8 cm/min. However, even the lower speed is incompatible with the energy release, as 0.8 cm/min for three minutes (the lower time-limit) would increase the reactivity by $3.6\,\% \frac{\Delta k}{k}$ (~ 4.5 dollars) and the energy release would have been far greater than 80 MWs. Since the observed energy release would raise the average temperature only 3.5° C, one must conclude that the pump was operating only intermittently or at a much lower rate than that reported, or the change in reactivity was much less than $1.5\,\% \left.\frac{\Delta k}{k}\right/$ cm.

Irradiations were very intense, being estimated at 400, 700, 850, 850, 850, and 1,100 rem. Of the six, one person died while the other five recovered after severe cases of radiation disease. The critical assembly absorbed the energy release of 80 MWs ($\sim 2.6 \times 10^{18}$ fissions) with no reported damage.

Stationary Low Power Reactor No. 1, Idaho Reactor Testing Area — 3rd January, 1961 [8, 9, 10] *(SL-1 reactor, aluminium-uranium alloy, water-moderated).* The SL-1 reactor (originally known as the Argonne Low Power Reactor) is a direct-cycle, boiling-water reactor of 3 MW gross heat capacity, using enriched uranium fuel plates clad in aluminium, water-moderated and cooled. Because the reactor was designed to operate for three years with little attention, the core was loaded with excess U 235. To counterbalance this excess of U 235, a burnable poison (B^{10}) was added to some core elements as aluminium-Boron 10-nickel alloy. Because these boron plates had a tendency to bow (and apparently to corrode, thereby increasing reactivity), some of them were replaced in November 1960 with cadmium strips welded between thin aluminium plates. At this time the shut-down margin was estimated to be 3 %, or about 4 dollars, compared to the initial value of 3.5 % to 4 %. The control rods were large cruciform cadmium sheets sandwiched between aluminium plates.

After having been in operation for about two years, the SL-1 was shut down on 23rd December 1960 for routine maintenance; on 4th January 1961 it was again to be brought to power. The three-man crew on duty the night of January 3rd were assigned the task of reassembling the control rod drives and preparing the reactor for start-up. Apparently they were engaged in this task when the excursion occurred. The fission yield, estimated to be 50 MW, may have created a pressure within the reactor of several hundred psi which was vented to the atmosphere through a number of partially-closed nozzles. Nevertheless, several shield plugs, portions of control rods, and some fuel were blown out of the reactor. Two men were killed instantly while the third died within two hours as the result of an injury to his head.

The cause of the incident and the sequence of events are not yet known. Investigations are hindered by the very high radiation field (500-1,000 r/hr) in the reactor room.

Miscellaneous Assemblies

Los Alamos, New Mexico — 11th February, 1945 (the DRAGON reactor, UH_3; pressed in styrex; shielded operation). The DRAGON reactor was the first fissile system designed for the purpose of generating prompt power excursions, and to my knowledge it was the first reactor of any kind whose reactivity exceeded prompt critical*. This was accomplished (by intent) on 8th January 1945; the temperature rise is quoted as 0.001° C and the yield (which is not quoted) can be estimated to have been about 2×10^{11} fissions.

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* Because of the similarity of the procedures used in these experiments with that of tickling the tail of a dragon (pointed out by R. Feynman) the experiment has been sometimes called the ' Dragon Experiment '. The name has stuck and is often applied to the class of prompt burst experiments where reactivity is added and subtracted mechanically, and where quenching mechanisms dependent upon the fission energy released do not contribute significantly to the shut-down process.

The DRAGON was made of enriched UH_3 pressed with a special plastic, styrex, into small cubes of average chemocal composition UC_4H_{10}. The stacking for the final experiments contained only 5.4 kg of this material, was diluted with polyethylene and reflected by graphite and polyethylene.

The reactor was made prompt critical for about 1/100 s by dropping a slug of this active material through a vertical hole in the remaining portion, which was stacked on a heavy (3/8 in.) steel table. The falling slug of active material was contained in a steel box and its path was closely defined by four guides. Generally, the fission energy did not contribute to the quench of the excursion; the burst size was determined by the background fission rate and the stacking on the table. Thus, the burst size could be controlled by moving a reflector nearer the assembly, or increasing the background fission rate. Both techniques were often employed and this may have been the case in the final experiment as the bursts were being made steadily larger. During the final excursion of about 6×10^{15} fissions, the UH_{10} cubes became so hot that blistering and swelling occurred. The whole system had expanded about 1/8 in.

In the final excursion the core material was damaged, but no active material was lost, there was no contamination and no one was irradiated.

The U.S.S.R. — 1953 or 1954 [11]. This incident apparently took place during 1953 or early in 1954. The location, date, facility, burst yield and causes are not mentioned. Two persons described as being " close to the reactor " and " in close proximity to the reactor " were subjected to " a short, general external gamma and neutron irradiation " equivalent to 300 and 450 r respectively. Their exposures could have been caused by a power excursion of any one of a large number of possible systems but one suspects a small critical assembly, possibly metallic, whose stacking was unintentionally increased to a reactivity well above delayed critical and possibly above prompt critical. It is unlikely that the burst yield exceeded 10^{16}-10^{17} fissions. It may be noted that the radiation dose at one metre from a GODIVA burst of 10^{16} fissions is 400 r (30 r gamma and 370 r neutron).

The Argonne National Laboratory, Idaho Reactor Testing Area — 29th November, 1955 [12,13] *(EBR-1, enriched uranium fast breeder reactor; shielded operation).* Design of the EBR-1 fast neutron reactor was started in 1948 with the objectives of establishing possible breeding values and demonstrating the feasibility of cooling a metallic reactor with liquid metals. These objectives were met and indeed in early 1952 the plant furnished more than enough electrical power for the reactor and the reactor building; excess steam was blown to the condenser.

The reactor core consisted of cylindrical, highly-enriched uranium rods of slightly less than 0.5 in. dia. canned in stainless steel with a bonding of NaK between rod and can. The total core mass of about 52 kg was bathed in a stream of NaK which served as a coolant.

The final experiment was designed to investigate coefficients of reactivity and in particular to study a prompt positive power coefficient for the case of no coolant flow. To do this, the system was placed on a period of 60 s at a power of 50 W. About 300 s later the power was 1 MW, the period had decreased to 0.9 s and core temperatures were rising significantly. The signal to scram the system was given, but by error, the slow-moving motor-driven control rods were activated instead of the fast-acting scram dropping

TABLE I. SOLUTION SYSTEMS

Date	Location	Active Material	Geometry	Total Fissions	Cause	Physical Damage
December, 1949	The Los Alamos Scientific Laboratory, New Mexico	~1 kg U 235 $UO_2(NO_3)_2$ in 13.6 litres water	Sphere graphite-reflected	$3\text{-}4 \times 10^{16}$	Manual withdrawal of two poison control rods	None
16th November, 1951	The Hanford Works, Richland, Washington	1.15 kg Pu $PuO_2(NO_3)_2$ in 63.8 litres water	Sphere 93 % full unreflected	8×10^{16}	Poison control rod run out too fast	None
26th May, 1954	The Oak Ridge National Laboratory, Tennessee	18.3 kg U 235 UO_2F_2 in 55.4 litres water	Cylindrical annulus unreflected	1×10^{17}	Tilting of inner poison cylinder	None
1st February, 1956	The Oak Ridge National Laboratory, Tennessee	27.7 kg U 235 UO_2F_2 in 58.9 litres water	Cylinder unreflected	1.6×10^{17}	Falling scram set up waves creating a critical geometry	Warping of bottom of cylinder
16th June, 1958	Y-12 Processing Plant, Oak Ridge, Tennessee	2.5 kg U 235 $UO_2(NO_3)_2$ in 56 litres water	Cylinder concrete reflected below	1.3×10^{18}	Wash water added to $UO_2(NO_3)_2$ solution	None
30th December, 1958	The Los Alamos Scientific Laboratory, New Mexico Pu Processing Plant	3.27 kg Pu $PuO_2(NO_3)_2$ in ~168 litres water	Cylinder water-reflected below	1.5×10^{17}	Agitator created a critical geometry	None
16th October, 1959	Chemical Processing Plant Idaho Reactor Testing Area	34.5 kg U 235 ~800 litres $UO_2(NO_3)_2$ water	Cylinder concrete reflected below	$\sim 4 \times 10^{19}$	Solution siphoned from safe to unsafe geometry	None
25th January, 1961	Chemical Processing Plant, Idaho Reactor Testing Area	8 kg U 235 $UO_2(NO_3)_2$ in 40 litres water	Cylinder	6×10^{17}	Solution pumped from safe to unsafe geometry	None

TABLE II. METAL SYSTEMS IN AIR

Date	Location	Active Material	Geometry	Total Fissions	Cause	Physical Damage
21st August, 1945	Los Alamos, New Mexico	6.2 kg δ-Phase Pu	Spherical core, tungsten-carbide reflected	$\sim 10^{16}$	Hand stacking reflector	None
21st May, 1946	Los Alamos, New Mexico	6.2 kg δ-Phase Pu	Spherical core, Be-reflected	$\sim 3 \times 10^{15}$	Hand stacking reflector	None
18th April, 1952	The Los Alamos Scientific Laboratory, New Mexico	92.4 kg uranium metal 93 % U 235	Cylinder, unreflected	1.5×10^{16}	Computation error	None
3rd February, 1954	The Los Alamos Scientific Laboratory, New Mexico	53 kg uranium metal 93 % U 235	Sphere, unreflected	5.6×10^{16}	Incorrect operation	Slight warping of pieces
12th February, 1957	The Los Alamos Scientific Laboratory, New Mexico	54 kg uranium metal 93 % U 235	Sphere, unreflected except for experiment	1.2×10^{17}	Shift of experiment	Warping, oxidation near melting close to centre

TABLE III. INHOMOGENEOUS WATER-MODERATED SYSTEMS

Date	Location	Active Material	Geometry	Total Fissions	Cause	Physical Damage
4th June, 1945	Los Alamos, New Mexico	35.4 kg uranium ~ 83 % U 235 0.5-in. cubes	Pseudosphere, water-reflected	$\sim 3 \times 10^{16}$	Water seeping between blocks	None
1st February, 1951	The Los Alamos Scientific Laboratory, New Mexico	2 cylinders uranium 24.4 and 38.5 kg 93 % U 235	2 cylinders, water-reflected	10^{17}	Scram increased reactivity	Slight oxidation
6th July, 1952	The Argonne National Laboratory	6.8 kg U 235 oxide particles in plastic	Inhomogeneous cylinder, water-reflected	1.22×10^{17}	Manual withdrawal of central safety rod	Plastic destroyed
12th December, 1952	Chalk River, Canada	Normal uranium	Rods, D_2O-moderated, graphite-reflected	1.2×10^{20}	Safety circuits failed, control rod maloperation	Core ruined
22nd July, 1954	The Reactor Testing Area, Idaho Falls, Idaho	U-Al plates clad with Al	Inhomogeneous cylinder, water-moderated	4.68×10^{18}	Estimate of expected excursion too low	Reactor destroyed
15th October, 1958	Vinca, Yugoslavia	3,996 kg normal uranium	Rods, D_2O-moderated, unreflected	2.5×10^{18}	Uncontrolled pumping of D_2O	None
3rd January, 1961	Idaho Reactor Testing Area	U-Al plates clad with Al	Inhomogeneous cylinder, water-moderated	1.5×10^{18}	Not yet determined	Not yet determined
		MISCELLANEOUS SYSTEMS				
11th February, 1945	Los Alamos, New Mexico	UH_3 pressed in styrex	Cylinder	$\sim 6 \times 10^{15}$	Reflector added and/or source too large	UH_3-styrex cubes swollen and blistered
1953	U.S.S.R.	Unknown	Unknown	Unknown	Unknown	Unknown
29th November, 1955	Idaho Reactor Testing Area	0.5-in. U 235 rods	Cylinder, rods cooled by NaK	4.7×10^{17}	Incorrect scram used	Core molten
3rd July, 1956	The Los Alamos Scientific Laboratory, New Mexico	58 kg uranium 93 % U 235, 2 and 5 mil foils	Cylinder	3.2×10^{16}	Too rapid assembly	None

under gravity of part of the natural uranium blanket as had been done to conclude similar experiments. This change in reactivity caused a momentary drop in power, but was inadequate to overcome the natural processes (very slight bowing inward of the fuel elements) adding reactivity to the system. After a delay of not more than 2 s, the fast scram was actuated both manually and by instruments and the experiment was completed. It was not immediately evident that the core had been damaged, but later examination disclosed that nearly one-half the core had melted and vaporized NaK had forced some of this molten alloy into the reflector. The theoretical analysis showed that the excursion was stopped by the falling reflector, after the power reached a maximum of 9-10 MW. The total energy release was close to 14 MWs or about 4.6×10^{17} fissions. The theoretical analysis was carried further in an attempt to determine if the core would have shut itself off in a non-catastrophic manner. The conclusion was that the energy release could have been nearly two-and-a-half times the observed yield, but would have been obtained in a non-explosive manner.

During this incident no one received more than trivial irradiation from airborne fission products and the direct exposure was essentially zero.

The Los Alamos Scientific Laboratory — 3rd July 1956 (the HONEYCOMB critical facility; U 235 foils moderated with graphite, control one-quarter mile away). The machine in which this excursion occurred is typical of several that are extant. The Los Alamos machine consists of a large matrix of 576 square aluminium tubes, 3 in. × 3 in. × 6 ft, split down the middle with one-half moveable on tracks. A photograph of the HONEYCOMB in the disassembled state is shown in Figure 6. Generally the facility has been used to simulate design features of complicated reactors, as there is considerable versatility in arrangements of uranium foil and various moderating materials. Inhomogeneous stackings in this and other machines of the same type are potentially the most dangerous critical assemblies in existence today. This conclusion stems from the apparent lack of any quenching mechanism, short of vaporization of the uranium foils or a sufficiently fast acting scram mechanism.

The stacking on 3rd July 1956 consisted of 58 kg enriched (93 % U 235) uranium in the form of 2 and 5 mil foils arranged between slabs of graphite, with some additional Be reflector surrounding the core. The total mass of graphite was 1,139 kg. At the time, some changes had been made in the reflector and graphite moderator, and critical was being approached for routine measurements. While the cart was in motion at a speed of about 0.2 in./s, the system became prompt critical, a burst occurred, and the scram system ejected Be control rods (reducing reactivity) and reversed the motion of the cart. The burst yield was 3.2×10^{16} fissions.

Apparently, this was a DRAGON-type excursion in that the excess reactivity was added and subtracted mechanically. There was no damage and no contamination. Since it was a remotely-controlled device at a distance of one-quarter mile, no person was irradiated.

Characteristics of Prompt Excursions and Quenching Mechanisms

Power excursions are regarded as dangerous because, for extreme cases, the exponential rise of power is so rapid that safety devices cannot react in time to prevent damage to the system and irradiation to personnel. Indeed,

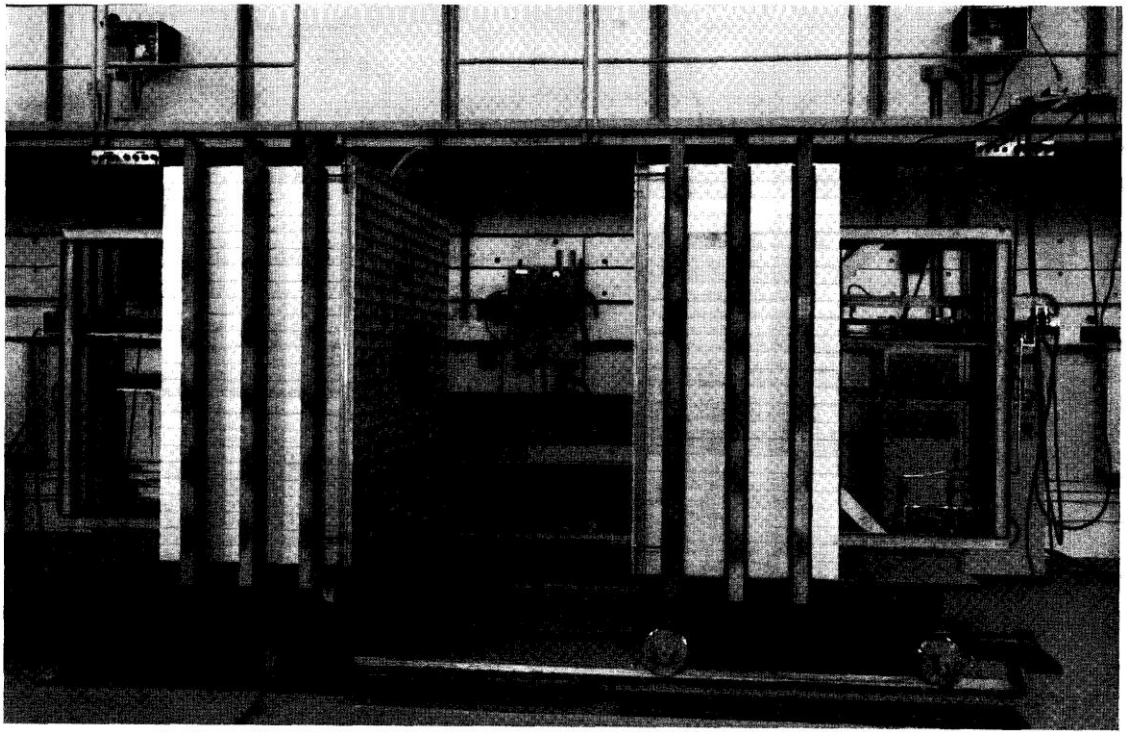

Figure 6. *LASL HONEYCOMB assembly machine. The movable section (right) is in the withdrawn position and the aluminium matrix is only partially loaded.*

this is often the case, but is not necessarily so. Some excursions have been terminated by safety devices while others have been terminated by natural quenching mechanisms that are roughly proportional to the fission energy that has been generated. This self-terminating characteristic has been demonstrated experimentally by the eight reactor systems that have been used in this field: the DRAGON, the BORAX systems, GODIVA-I, the SPERT reactors, the KEWB reactors, GODIVA-II, TRIGA, and TREAT. Of these the first three are no longer in existence, but the last five are working reactors. Several of these are no longer concerned with providing information about prompt excursions *per se*, but are used as devices to provide quick pulses of neutrons for other experimental purposes.

The quenching actions that the above experimental reactors utilize and that have terminated many of the accidental excursions, are thermal expansion, rise in neutron temperature, boiling, and bubble formation from fission fragments. The order here is of no importance and not all are independent. In addition, in some reactors, more than one mechanism contributes to quench or shut-down of the excursion, and in many cases additional quenching actions set in when energy densities or temperatures reach some threshold value. The ramifications of this subject are varied and numerous, but the simplest and most generally applicable case is that of the energy model in which the change of reactivity is proportional to the release of fission energy.

For the special case of a step increase in reactivity, ΔK_o, we can write:

$$\Delta K(t) = \Delta K_o - bE(t) \tag{1}$$

where $E(t)$ is the fission energy released to time t, and b is a constant characteristic of the system. With this assumption the reactor kinetic equations have been coded for numerical solution by use of digital computers. Such codes exist in many laboratories; the results quoted here are from the Los Alamos RTS Code. Figure 7 illustrates a series of computations for hypothetical systems in which the step increase of ΔK is 1.20 dollars, the value of b is constant and the neutron lifetime, l, is varied from 10^{-8} to 10^{-4} s. The power and reactivity traces for the short neutron lifetime cases (simulating a fast reactor) are characteristic of prompt excursions in such systems. The very sharp rise and fall in power is called the spike, and the relatively constant power following the spike is the plateau. During the time interval of the spike, the reactivity changes by $2\Delta K_o$ — it reflects about prompt critical. The characteristics of these spikes are established almost entirely by the prompt neutrons. The traces for $l = 10^{-4}$ (simulating a moderated reactor) do not show the reflection about prompt critical and there is no well-defined plateau following the spike. The time scale is of the order of decay times of the delayed neutron precursors and the effects of these neutrons cannot be ignored in this case.

Figure 8 illustrates comparable data for a step increase in reactivity of one dollar. The time history of the reactivity and power in this case is quite different and, indeed, is typical of excursions in the delayed critical region. The time scale is much extended, allowing the possibility of mechanical devices to shut off the transient, the power peaks are broader, and the reactivity now attempts to reflect about delayed critical. It should be noted that the implicit assumption of no heat loss from the system cannot be realized in practice. Any such loss of energy could only hold the power to values greater than those plotted.

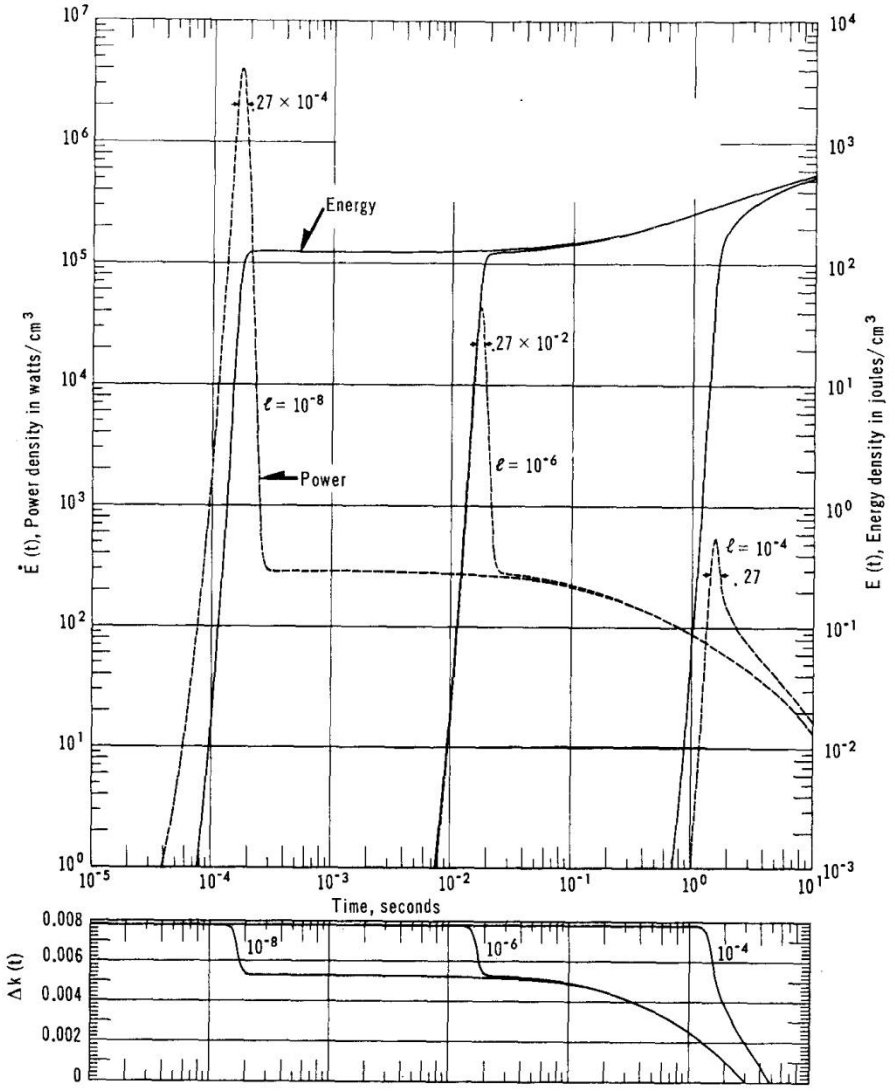

Figure 7. *Energy model computation of power versus time and energy release versus time, for an initial reactivity of 1.2 dollars above delayed critical for three different neutron lifetime values, $l = 10^{-8}$ sec, $l = 10^{-6}$ sec, and $l = 10^{-4}$ sec. Below are the corresponding curves of reactivity versus time.*

Some of the results illustrated in Figures 7 and 8 can be obtained analytically. For sufficiently large step increases in reactivity above prompt critical, the delayed neutrons may be ignored and the kinetic equations integrated to give the total excursion yield*.

* A similar result can be obtained for the delayed critical region, but the implicit adiabatic assumption vitiates the result.

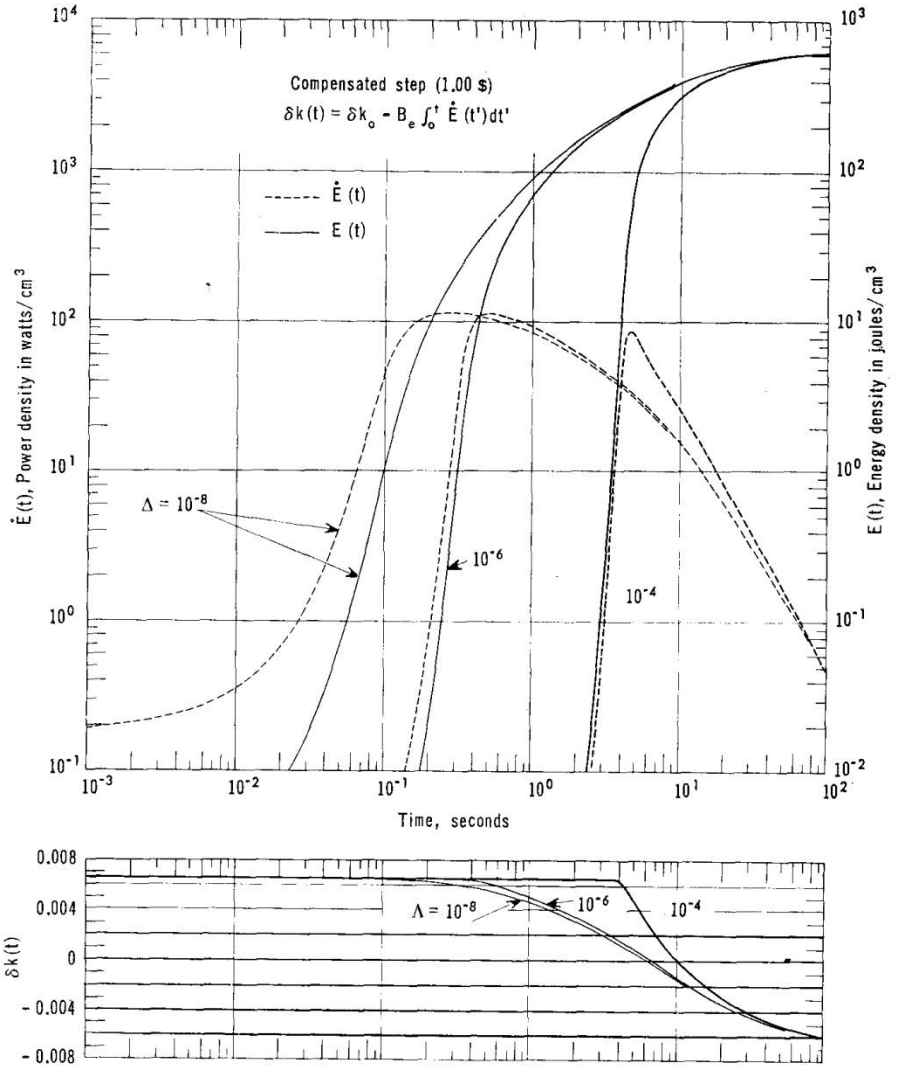

Figure 8. *Energy model computation of power versus time, for an initial reactivity of 1.0 dollar above delayed critical for three different neutron lifetime values, $l = 10^{-8}$, 10^{-6} and 10^{-4} sec. Below are the corresponding curves of reactivity versus time.*

$$E = \frac{2\Delta K_p}{b} \tag{2}$$

where ΔK_p is the step increase relative to prompt critical. The half-width of the spike is given by:

$$\Delta t_{1/2} = \frac{3.52l}{\Delta K_p} \tag{3}$$

where l = the neutron lifetime and the maximum power is:

$$\left(\frac{dE}{dt}\right)_{max} \approx \frac{2}{3.5}\frac{\Delta K_p^2}{bl} \qquad (4)$$

The experimental systems which have been most intensively studied and which exemplify best the data in Figures 7 and 8 and many of the possible hazards in the nuclear energy industry are the GODIVA, KEWB, and SPERT reactors. Each of these will be discussed briefly.

GODIVA-I (partly discussed earlier) and GODIVA-II are near-solid uranium (93 % U 235) metal critical assemblies, which were pressed into service as irradiation facilities. At a few cents above prompt critical, controlled prompt excursions provide an excellent experimental picture to complement the curves of Figure 7. The prompt negative temperature coefficient of reactivity of 4.3×10^{-3} dollar/°C arises from thermal expansion and is directly related to the deposition of fission energy. The transient proceeds so rapidly that no heat is lost from the system. When the step change of reactivity is increased to 8 or 9 cents above prompt critical, a new effect sets in. The power level rises to such high values that the thermal expansion lags the energy deposition and the simple ratio of E and ΔK_p in equation (2) is no longer true. At still higher step changes the energy release becomes proportional to the square, and eventually to the cube, of the initial excess reactivity.

The transient behaviour of solution systems has been studied with the two KEWB reactors. The KEWB-A core was a 13.6-litre stainless steel sphere containing 11.5 litres of enriched uranyl sulphate solution. The reflector was thick graphite. This reactor provided a means to study transients in solution systems during which the period was as short as 2 ms. The KEWB-B core was designed for the specific purpose of extending these measurements to a period of 1 ms. It was cylindrical in shape and during the transient experiments (up to about 5.2 dollars over prompt critical) contained 18 litres uranyl sulphate solution.

In the KEWB system, two quenching mechanisms seem to be dominant over a wide range of excursions. The first of these is the rise in neutron temperature and thermal expansion as the core temperature rises, which results in a prompt temperature coefficient equal to -0.02 dollar/°C at 30°C. This effect is sufficient to account for the observed yield of excursions starting near prompt critical, but is inadequate for more violent transient experiments. The second is that of bubble formation. The available evidence supports the contention that during the spike, void space consisting of many very small bubbles (micro-bubbles) with internal pressures in the range of 10 to 1,000 atm is created by the fission process. These small bubbles later coalesce and leave the system, giving the observed gas production coefficient of about 4.4 litres/MWs. The growth of these micro-bubbles seems to involve the repeated interaction between fission fragments and existing micro-bubbles from earlier fissions. Thus a quenching mechanism proportional to the square of the energy release can be invoked. This model is successful in describing the solution transients, notwithstanding imprecise knowledge of the manner in which the bubbles form and grow.

The family of SPERT-I reactor cores are composed of enriched uranium-aluminium alloy plates, clad with aluminium and arranged to be moderated and reflected by water. Between 12 and 24 of the roughly 3 in. × 24 in. × 0.060 in. plates are grouped in a single assembly in which the plates average 0.117 in. apart. The number of such assemblies required to complete a critical core is

between 28 and 64, depending upon the plate loading and the problem to be investigated. Cores have been designed to include the range from under-moderation to the hazardous region of slight over-moderation. Transients on these reactors have been intensively studied since 1957 to solve core design problems and find the limitations of such plate type reactors. In particular, the period and energy release which can cause damage has been carefully determined.

The shut-down of a power transient in the SPERT systems is more complicated than in the simpler reactors. The model developed includes (*a*) heating and density change of the water, (*b*) heating of the core structure including its own geometry changes and moderator expulsion from such changes, and finally, (*c*) the boiling of water next to the plates and loss of moderator when water is expelled from the core. The model is well understood except for the boiling process. The formation of steam voids under such transient conditions is not sufficiently well understood to permit the development of an exact theory. In spite of such difficulties, the empirical model permits a description of the transients and reasonable extrapolations to reactors of similar design.

Summary

Some of the conclusions of this paper are summarized in Tables IV to VI. Where possible and appropriate, the excursion fission energy is divided into that created in the spike and in the plateau, and some time-intervals of interest are listed: the spike width at half maximum, the time from prompt critical to peak power and the duration of the plateau. Also given are the maximum alpha (reciprocal period), the peak power during the spike, the initial plateau power, and the probable quenching mechanism. Many of these numbers are at best approximate, so that no estimate of probable error is attempted. As can be seen, the time scales vary from microseconds to seconds and for some excursions practically all fissions were in the plateau while others consisted of a single spike.

Though the accidental excursions are too few in number to permit reliable statistical conclusions, there are some observations that may be of general interest and importance. Of the twenty-four excursions studied, four have occurred in production plants (and therefore were a complete surprise), five in what must be called working reactors (the water boiler, the second GODIVA accident, the DRAGON, the SL-1 and the NRX) and the remainder in critical facilities where the properties of the assemblies themselves were being investigated. One suspects that the frequency of incidents may be related to the frequency of producing an unfamiliar critical configuration, though the statistics to prove this hypothesis are not available.

The causes of excursions can be generalized to some extent. Four resulted from failure of some part of the experimental equipment during operation near critical; two were induced by scram devices; the four production plant incidents must be related to inadequate control over large and complex systems; and two are unknown. The responsibility for the others invariably returns to the personnel operating the facility and these incidents must be attributed to impatience, lack of planning or understanding of the experiment or to simple carelessness.

The thought often has been expressed that, for reasons unknown, prompt critical incidents are limited in yield to a range roughly between 10^{16} and a

few times 10^{17} fissions. Recent excursions have demolished this myth rather effectively, for now the ratio of yields of the largest and smallest excursions is over 10^4. The ratio of fission densities, though less, is still a rather large number. It is quite true that, as the fission density increases, there appear more effective (and destructive) quenching mechanisms, such as boiling of water or melting and vaporization of metals. For the special case of slow insertion of reactivity, the fission densities corresponding to such phase changes can be regarded as the upper limits of order of magnitude for excursions of some systems, but these limits do not apply in general.

A potentially most dangerous manufacturing plant incident is possible in the handling and storage of large masses of fissile metal. The model to which one goes for information in this area is the GODIVA reactor for which experiment and theory are well enough established to permit reliable extrapolations to catastrophic conditions. One can imagine, for example, the consequences of inserting 3 dollars excess reactivity (relative to delayed critical) into GODIVA; the resulting excursion would produce 10^{19} fissions, vaporize the entire mass and probably destroy the building in which the experiment was being performed. Under some circumstances, the consequences of this type of incident can be estimated with reasonable accuracy. If the reactivity insertion rate is known, and if a source of neutrons is present and sufficiently strong to guarantee that a continuing fission chain starts at or before prompt critical, the burst yield can be calculated. A most extraordinary rate is then required to produce a catastrophic explosion. The case for a quick step increase of reactivity can also be calculated quite exactly. If a neutron source is not present, an explosion could be created.

This situation, in which no external source is present and a high reactivity insertion rate is imposed on the system, is currently a matter of experimental and theoretical investigation at the Los Alamos Scientific Laboratory, using the GODIVA-II reactor, and at the Idaho Reactor Testing Area using the SPERT-I reactor. The background fission rate of GODIVA-II is only 40/s, primarily from the spontaneous fission of the 6.5 % U 238, and quick (about 50 ms) changes of reactivity to a few cents above prompt critical can be made before a continuing fission chain is born. Generally, after such a step increase of reactivity, there is a waiting time of the order of seconds before a continuing fission chain develops. The statistics associated with these experiments are illustrated in Figure 9 where the fraction of the bursts occurring in one-second intervals is plotted against the waiting time in seconds. The most probable delay is about two seconds and observed waiting times have been as long as eleven seconds. These experiments are continuing for various steps and ramp increases of reactivity.

The application of these results to some U 235 metal safety problems is a two-edged sword. The very low neutron background associated with U 235 could allow (for example) sizable masses to fall together before a fission chain started. An external neutron source to guarantee initiation then would very substantially reduce the yield of such an accidental power excursion. On the other hand, in some cases the very real and possible waiting times could allow mechanical dispersion of the active material before a chain started. An external neutron source would then be the trigger for a power transient which otherwise might not have occurred. This dilemma does not exist for plutonium metal, for which the spontaneous fission rate guarantees a strong and reliable source.

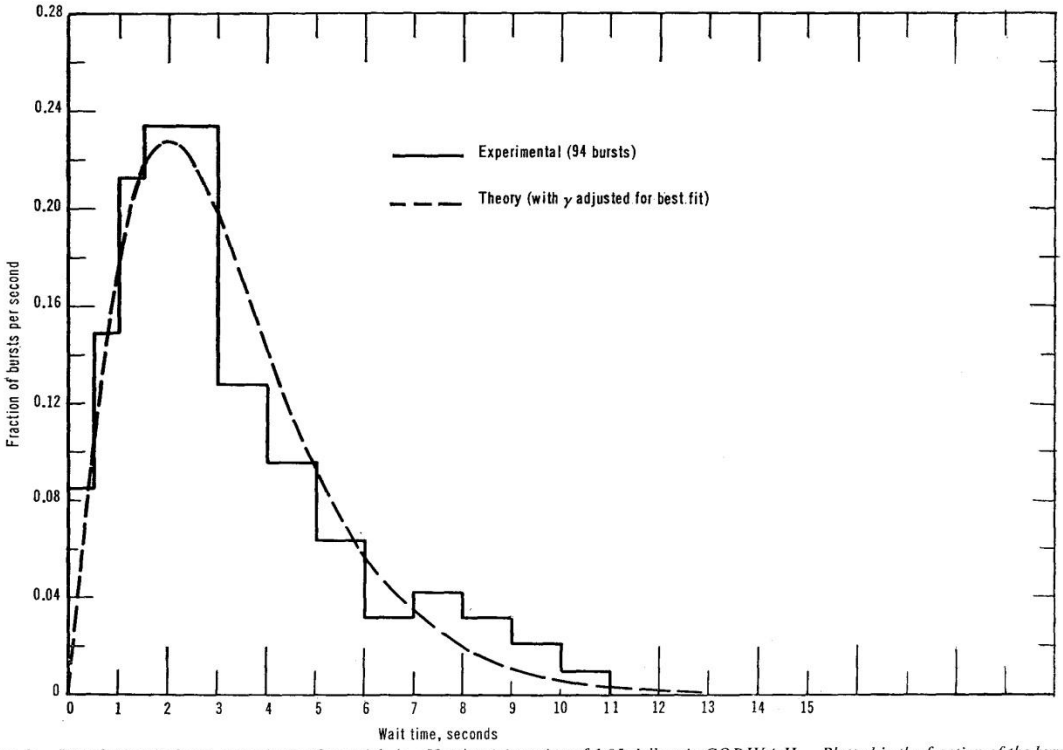

Figure 9. *Distribution in burst wait times after quick (~ 50 m/sec) insertion of 1.05 dollars in GODIVA-II. Plotted is the fraction of the bursts in each second versus the wait time in seconds.*

TABLE IV. SOLUTION SYSTEMS

Incident	Observed Fission Yield	Spike Yield (*Fissions*)	Plateau Yield (*Fissions*)	Maximum α (s^{-1})	Time Interval from Prompt Critical to Peak Power (*s*)	Width of Spike at One-Half Maximum Value (*s*)	Time Duration of Plateau (*s*)	Peak Power During Spike (*Fissions/s*)	Initial Plateau Power (*Fissions/s*)	Probable Quenching Mechanism
Water boiler reactor control rods removed by hand, December, 1949	$3\text{-}4 \times 10^{18}$	$\sim 3 \times 10^{18}$	2.7 3.7×10^{18}	6.25	~ 3.5	—	~ 1.5	—	$2\text{-}3 \times 10^{18}$	Rise of neutron temperature
Pu solution assembly cadmium rod removed too rapidly, 16th November, 1951	8×10^{16}	8×10^{16}	0.0	No estimate	No estimate	No estimate	0.0	No estimate	0.0	Micro-bubbles, neutron temperature and poison rod
Uranium solution assembly, central poison cylinder tilted to less favourable position, 26th May, 1954	10^{17}	5×10^{16}	5×10^{16}	4.8×10^{2}	7.8×10^{-2}	5.4×10^{-3}	~ 0.5	8.2×10^{18}	4.8×10^{16} to 1.4×10^{17}	Micro-bubbles and neutron temperature
Uranium solution assembly, wave motion created by falling cadmium sheet, 1st February 1956	1.6×10^{17}	1.6×10^{17}	0.0	6.1×10^{3}	7.4×10^{-3}	3.7×10^{-4}	0.0	$\sim 3.5 \times 10^{20}$	0.0	Micro - bubbles and neutron temperature
Uranium process solution combined with wash water in 55-gallon drum, 16th June, 1958	1.3×10^{18}	$\sim 10^{16}$	1.29×10^{18}	No estimate	No estimate	No estimate	170.0	No estimate	No estimate	Micro - bubbles, neutron temperature and boiling
Separated phases in plutonium process tank, 30th Dec., 1958	1.5×10^{17}	1.5×10^{17}	0.0	No estimate	No estimate	No estimate	0.0	No estimate	0.0	Micro - bubbles, neutron temperature and stirring action
Uranium process solution siphoned from safe to unsafe geometry, 16th October, 1959	4×10^{19}	$\sim 10^{17}$	$\sim 4 \times 10^{19}$	No estimate	No estimate	No estimate	No estimate	No estimate	No estimate	Micro - bubbles and neutron temperature
Uranium process solution pumped from safe tou nsafe geometry, 25th January, 1961	6×10^{17}	No estimate	No estimate	No estimate	No estimate	No estimate	No estimate	No estimate	No estimate	Micro - bubbles and neutron temperature

TABLE V. METAL SYSTEMS IN AIR

Incident	Observed Fission Yield	Spike Yield (*Fissions*)	Plateau Yield (*Fissions*)	Maximum α (s^{-1})	Time Interval from Prompt Critical to Peak Power (*s*)	Width of Spike at One-Half Maximum Value (*s*)	Time Duration of Plateau (*s*)	Peak Power During Spike (*Fissions/s*)	Initial Plateau Power (*Fissions/s*)	Probable Quenching Mechanism
Pu core reflected with tungsten carbide, 21st August, 1945	10^{16}	$\sim 1.8 \times 10^{15}$	$\sim 8.2 \times 10^{15}$	$\sim 10^{4}$	$\sim 2 \times 10^{-3}$	$\sim 3.5 \times 10^{-4}$	0.8	$\sim 5 \times 10^{18}$	$\sim 1.2 \times 10^{16}$	Thermal expansion
Pu core reflected with beryllium, 21st May, 1946	3×10^{15}	$\sim 1.8 \times 10^{14}$	$\sim 2.86 \times 10^{15}$	$\sim 10^{3}$	1.5×10^{-2}	$\sim 3.5 \times 10^{-3}$	0.28	$\sim 5 \times 10^{16}$	$\sim 1.2 \times 10^{16}$	Thermal expansion
JEMIMA, a cylindrical unreflected U 235 assembly, 18th April, 1952	1.5×10^{16}	$\sim 10^{15}$	$\sim 1.4 \times 10^{16}$	8×10^{2}	4.3×10^{-2}	4.4×10^{-4}	≤ 0.3	2.3×10^{18}	$\leq 10^{17}$	Thermal expansion
GODIVA, a bare U 235 sphere, 3rd February, 1954	5.6×10^{16}	5.6×10^{16}	0.0	1.6×10^{5}	2.4×10^{-4}	1.8×10^{-5}	0.0	3.5×10^{21}	0.0	Thermal expansion
GODIVA, a bare U 235 sphere, 12th February, 1957	1.2×10^{17}	1.2×10^{17}	0.0	2.1×10^{5}	1.9×10^{-4}	1.2×10^{-5}	0.0	10^{22}	0.0	Thermal expansion

TABLE VI. INHOMOGENEOUS METAL SYSTEMS IN WATER

Incident	Observed Fission Yield	Spike Yield (*Fissions*)	Plateau Yield (*Fissions*)	Maximum α (s^{-1})	Time Interval from Prompt Critical to Peak Power (*s*)	Width of Spike at One-Half Maximum Value (*s*)	Time Duration of Plateau (*s*)	Peak Power During Spike (*Fissions/s*)	Initial Plateau Power (*Fissions/s*)	Probable Quenching Mechanism
Pseudosphere of uranium cubes, water-reflected, 6th June, 1945	$3\text{-}4\times10^{18}$	$\sim3\times10^{15}$	$2.7\text{-}3.7\times10^{16}$	No estimate	No estimate	No estimate	~ 5.0	No estimate	No estimate	Boiling
Critical separation experiment, two large U 235 masses in water, 1st February, 1951	10^{17}	$\sim6\times10^{15}$	$\sim9.5\times10^{16}$	No estimate	No estimate	No estimate	~ 0.1	No estimate	$\sim 10^{17}$	Boiling
UO_2 particles in plastic, water-moderated, 6th July, 1952	1.22×10^{17}	1.22×10^{17}	0.0	100	~ 0.6	1.9×10^{-2}	0.0	5.4×10^{18}	0.0	Plastic bubbles
NRX reactor, normal uranium in D_2O, graphite-reflected, 12th Dec., 1952	1.2×10^{20}	Not prompt critical	Not prompt critical	$\sim$0.33	Not prompt critical	~ 30	0.0	$\sim 3\times10^{18}$	0.0	D_2O drained from core
Borax reactor AL-U alloy, water-moderated, 22nd July, 1954	4.68×10^{18}	4.68×10^{18}	0.0	385	7.9×10^{-2}	6.5×10^{-3}	0.0	6.6×10^{20}	0.0	Boiling
Critical assembly of normal uranium rods in D_2O, unreflected, 15th October, 1958	2.5×10^{18}	Not prompt critical	Not prompt critical	No estimate	Not prompt critical	No estimate	0.0	No estimate	0.0	Cadmium rods inserted in core
SL-1 reactor AL-U alloy, water-moderated, 3rd January, 1961	1.5×10^{18}	1.5×10^{18}	0.0	No estimate	No estimate	No estimate	0.0	No estimate	0.0	Boiling
MISCELLANEOUS SYSTEMS										
The DRAGON reactor UH_3, pressed in styrex, 11th February, 1945	$\sim6\times10^{15}$	$\sim6\times10^{15}$	0.0	No estimate	$\sim 10^{-2}$	No estimate	0.0	No estimate	0.0	Thermal expansion
Experimental reactor, U.S.S.R. 1953	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown
EBR-1 fast breeder reactor, U 235 rods in NaK, 29th November, 1955	4.7×10^{17}	Not prompt critical	Not prompt critical	$\sim$3.6	Not prompt critical	~ 1.0	0.0	$\sim 3\times10^{17}$	0.0	Reflector dropped from core
The HONEYCMOB, U235 foils moderated with graphite, 3rd July, 1956	3.2×10^{16}	3.2×10^{16}	0.0	$\sim$40	~ 1.1	0.1	0.0	9.4×10^{17}	0.0	Motion of safety rods

A possible application of these results can be found in the conflicting economic and safety requirements associated with complicated or multiple fuel element castings. Some such operations, now forbidden for safety reasons, might be reconsidered if heavy shielding were available to protect operators and if strong neutron sources were present to guarantee initiation. Under these conditions, the risk of a non-explosive power excursion might be acceptable.

The two most recent chemical plant incidents along with the KEWB reactor experiments (see page 514) bring into focus the possibility that less stringent criticality rules may also be applicable to some U 235 solution operations where the operators are protected by heavy shielding. Neither of these two incidents caused injury to personnel, nor did any of the four plant incidents cause damage to equipment. It is possible that significant economic savings could be realized with no increased radiation hazard if certain plants were designed with the expectation that such incidents do occasionally occur. The KEWB experiments provide much information in regard to transient pressures, temperatures, fissions yields, neutron and gamma ray fluxes that are associated with excursions of solutions. The results also suggest the conclusion that only with difficulty can a solution system be forced to explode violently if it fills only a small fraction of the available volume. As was discussed above, enriched U 235 systems probably require an external neutron source.

These possibilities must be viewed with more caution when plutonium solutions are considered. It is well known that plutonium-rich, silica-like sludges tend to accumulate in significant quantities in chemical plants that have been in operation over a period of years. The second plant incident was caused by such an accumulation. Furthermore, these deposits have been discovered in several plants, sometimes in more than modest amounts. Fortunately, dissolution of the sediments has generally been accomplished without incident. These deposits can be created with a relatively low H/Pu atomic ratio (of the order of 20) thus raising the possibility of an autocatalytic nuclear excursion. One can imagine a mass of this material forced to critical or above by immersion in water, and when boiling commences, mixing caused by bubble action could raise the H/Pu ratio. As mixing with water will (in this case) reduce the critical mass, the net effect is to increase the reactivity of the solution and cause the power to diverge even more. However, even for such an apparently dangerous situation it is difficult to imagine an explosive reaction. Again, it is quite possible that thick shielding could nullify radiation hazards.

These chemical plant incidents also point up the problem of the large and complicated interconnected network of piping and tanks required for the dissolution of fuel elements and the separation of uranium, plutonium, fission fragments and alloying materials. All four plant incidents have been caused by solutions flowing from safe to unsafe geometries, and three of these transfers were caused by failures in the network or were in some way associated with peripheral operations. The possibility of making large auxiliary containers nuclearly safe and the insertion of positive blocks in certain lines has been considered, but it would be presumptuous to suggest detailed design features in this review.

ACKNOWLEDGMENTS

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REFERENCES

1. W.R. STRATTON, " A Review of Criticality Accidents ," *Progr. Nucl. Energy, Series IV, Technology, Engineering and Safety*. V. 3, Pergamon Press, London, 1960.
2. W.L. GINKEL, C.W. BILLS, A.O. DODD, K.K. KENNEDY and F.H. TINGEY, " Nuclear Incident at the Idaho Chemical Processing Plant ", *IDO-10035* (1960).
3. W.B. LEWIS, " The Accident to the NRX Reactor on December 12, 1952 ", *DR-32*, Chalk River (1953).
4. W.J. HENDERSON, A.C. JOHNSON and P.R. TUNNICLIFFE, " An Investigation of some of the Circumstances Pertinent to the Accident to the NRX Reactor of December 12, 1952 ", *NEI-26*, Chalk River (1953).
5. D.G. HURST and A.G. WARD, " Canadian Research Reactors ", *Progr. Nucl. Energy, Series II, Reactors*, V. I, Pergamon Press, London, 1956.
6. " Yugoslavian Criticality Accidents ", *Nucleonics, 17*, No 4, 106 (April 1959).
7. H. JAMMET, G. MATHE, B. PENDIC, J. DUPLAN, B. MAUPIN, R. LATARJET, D. KOLIC, L. SCHWARTZENBERG, Z. DJUKIC and J. VIGNE ," Study of Six Cases of Accidental Acute Whole-Body Irradiation ", *Revue Française d'Etudes Cliniques et Biologiques, 4*, 210 (1959).
8. A. SMAARDYK (Ed.), " Hazard Summary Report on the Argonne Low Power Reactor ", *ANL-5744* (Oct. 1957).
9. " Army Reactor Program ", *Nucleonics, 17*, No 2, 54 (Feb. 1959).
10. " Special Report on SL-1 Accident ", *Nuclear News* (An American Nuclear Society Publication), *4*, No. 2, 4 (Feb. 1961).
11. A.K. GUSKOVA and G.D. BAISOGOLOV, " Two Cases of Acute Radiation Disease in Man " in *Proc. First U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1955*, V. *11*, 35.
12. R.O. BRITTAN, " Analysis of the EBR-1 Core Meltdown " in *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 12*, 267.
13. J. KITTEL, M. NOVICK and R. BUCHANAN, " The EBR-1 Meltdown — Physical and Metallurgical Changes in the Core ", *ANL-5731* (1957).

DISCUSSION

A.F. THOMAS, United Kingdom:

I agree with Dr. Stratton in general about the danger being fairly similar when the system is stopped just short of critical and when it is being run critical. It all depends on the method of control. In approach machines it depends on the time required to get the system back to where it started from as a function of machine speed, electronic delays and so forth. The time scales might be slightly different with control rod mechanisms but the principle is the same. But I disagree with his idea of letting systems go critical or allowing them to be potentially super-critical behind shielding. The main difficulty is not in letting them go critical; it is in knowing just where they're going to go critical. One decides what the critical parameter is for the system

with a certain measure of uncertainty. The permissible increase allowing the system to go, say, up to one dollar above critical would be very small and the cost of shielding would almost certainly outweigh any gain in process time, or batch production.

W.R. STRATTON, United States:

Certainly, if the cost of shielding is higher than the saving in using larger batches, there is no point in changing the operation. If the shielding is available, it could be made use of, or if the operation is something we can do no other way, then rather than *not* do it and give something up, we could risk it.

E.D. CLAYTON, United States:

I should like to comment that Plant Managers in general have been rather reluctant to assume the risk of a criticality incident even behind heavy shielding. In the case of plutonium, I think the problem may be rather different and should be considered in a slightly different light, because although the physical damage might be insignificant in the event of a criticality incident — if even a few drops of plutonium solution were forced out of a line somewhere into an operating area, then the entire plant might be closed down for a period of weeks or perhaps months while personnel were decontaminating the facility. In the case of plutonium it is primarily the contamination problem from the plutonium itself that would be the most serious consequence of a criticality incident. This is assuming there was shielding for personnel during the incident.

W.R. STRATTON:

Dr. Clayton is certainly right. Plutonium is more hazardous than U 235 and one has to take more care.

F.R. CHARLESWORTH, United Kingdom:

I would like to add to Dr. Clayton's remarks, that quite frequently in dealing with non-active material one has quite a clean-up problem to face in re-purifying the material that has gone critical.

R. GALLEY, France:

J'ajouterai aux observations formulées par M. Clayton que le fait d'avoir une protection épaisse n'assure les opérateurs que pendant une période d'exploitation normale. Ainsi, dans le cas de la purification du plutonium, le fait d'avoir une protection épaisse qui ne serait pas nécessaire en raison de la faible activité gamma, mais aurait été installée pour des raisons de criticalité, ne protégerait absolument pas les ouvriers chargés de l'entretien qui seraient appelés à pénétrer en cas d'accidents graves ou d'importantes réparations à l'appareillage ; de ce fait, il n'est pas certain que dans tous les cas la protection soit non seulement nécessaire, mais souhaitable.

W.R. STRATTON:

One would hope that by taking these extra risks which I have suggested, one would perhaps lower the probability of severe accidents, even though

a milder accident might occur more often. Certainly one has to worry about the people who clean up. In the event of an accident there has to be some means of dispersal or saving of the material, but one would hope that by such a method we could possibly decrease the chance of a severe accident.

K.M. HILL, United Kingdom:

When processing very considerable quantities of fissile materials through unsafe geometry, I think this question of building plants which will normally operate with a higher probability of criticality is a very useful concept as an outside case. In other words one incorporates all one's normal protections against the system reaching criticality. In the case of solvent extraction equipment, there may be various concentration monitors, feed controls, etc., but in the event of these failing in a large plant of unsafe geometry, I think that this approach is well worth carrying out. The magnitude of the physical effects does appear to be small and within the limits of normal engineering equipment used for this sort of processing. The extra shielding cost is significant but is in any case not of a different order of cost to the instrumentation costs against criticality excursions. However, I think that with this approach, two things need sorting out. First of all, if one considers the possibility of an occasional criticality incident — I am assuming that the operator doesn't actually want to run a boiling water reactor — I think a better method of assessing the probability of an accident will be needed. While this is not in any way my field, I feel that much of the work which has been done on the probability of accidents occurring in civil engineering structures, or aircraft, for example, has generally been based on the failure rate, which works back to the reliability of the original data. I think that much of this sort of approach could be applied by the criticality expert who is used to working in similar mathematical terms. But the further thing which I think must be sorted out before one could confidently go ahead with this type of design, is that if one looks at the energy requirements for turning a given release into a physically damaging effect, in the many typical process volumes involved the energy difference is very small compared with the energy which is going to be liberated anyway. The factor of increase of say 50 % on the amount of energy is small when one normally talks in factors of 10, and in some systems, particularly small ones, an increase of 50 % in the energy liberation could mean the difference between the thing blowing up or just warming up very gently. Before one could confidently go ahead and design on this basis, I think there must be more certain ways of assessing what the top level of energy would be in, say, the first spike.

W.R. STRATTON:

I find your comments very interesting. You were referring to solution systems when you said a change of 50 % could mean the difference between safety and something unsafe? Are these closed volumes, whereas the solution fills the entire system?

K.M. HILL:

I think that one can take it that nearly any plant handling, particularly plutonium, but even uranium 235, is closed. It may have a vent system but there will always be a lid on the vessel.

W.R. STRATTON:

I must confess I speak in some part from innocence of real knowledge of processing plants. The two most recent processing plant accidents in the U.S. were both in vessels in which the solution filled only a small fraction of the available volume. This is a relatively ' safe ' situation. If it's a closed system it's quite different, the vessel could rupture the vessel.

H.C. PAXTON, United States:

There is a very important situation in which one must have shielding but in which men do not go behind shielding for maintenance purposes: this is the processing of irradiated fuel. Rather than talk about the case in which we really expect an accident once a month or so, one might ask whether one puts the same requirements on processing equipment, that is behind shielding for other purposes, as one would on processing equipment for unirradiated fuel for material out in the open. For example, sometimes the double contingency requirement strains the nuclear safety specialist, and I wonder whether there might be some relaxation of this requirement behind shielding? To be more specific, frequently there is, let us say, at least an uneasiness about the use of soluble poison as one contingency. Might it not be possible that soluble poison could be considered an acceptable contingency behind shielding, whereas one might prefer not to use it where there are people near the equipment?

J. BOURGEOIS, France:

En France, nous avions envisagé à un moment donné de protéger de façon permanente certaines parties d'une usine qui nous semblaient moins sûres que les autres. Nous entendions par protection le principe selon lequel l'ouvrier le plus exposé ne devait subir qu'une irradiation de 2 rems lors d'une excursion de 10^{18} fission. M. Stratton envisage-t-il un mode de protection analogue et les hypothèses que nous avions retenues lui semblent-elles raisonnables ?

W.R. STRATTON:

I don't know what the accepted levels are in the U.S., but there are definite values. I think that as a general rule, people who do work in criticality laboratories and in production plants suffer more risk than the general public.

D. CALLIHAN, United States:

The Oak Ridge National Laboratory recommends for normal operations, a maximum total-body exposure of 1/10 rem per week, 12 rems annual, with an age-dependant employment-lifetime accumulation. However, in an emergency, some health physicists believe that single exposures as high as 25 rems or even 100 rems might be taken once in a lifetime. This sort of thing one does in order to save a life or in an action of equal importance.

A.F. THOMAS:

I would like to comment on Dr. Stratton's answer to Mr. Ford's question about the provisions of criticality alarms in fast systems. I took the implication to be whether it is worth providing detector heads in systems which

will be essentially fast, because the operator may not have time to run before it's all over. I think that the fast incidents in the U.S. have all had mechanical withdrawal systems which have operated before the system has had time to cool down again. If the system — GODIVA for instance — had been kept in position and cooled down, then it would have settled down at a pretty high power level and would stay expanded. Perhaps Dr. Stratton would like to comment on a system such as this or, for instance, large lumps of uranium in graphite moulds which would be essentially fast, where the operator would be receiving a fairly heavy dose though not the burst dose.

W.R. STRATTON:

In the GODIVA excursions the yield in the first spike was in the order of 10^{16} fissions or perhaps 1/3 MWs, the power level in the plateau following the spike was of the order of 1 MW. This is sufficient power to melt GODIVA, or start melting the pieces, in four or five seconds, I believe. As soon as it starts to melt, it would turn itself off by dripping on the floor, so that the total would be perhaps 4 or 5 MW following the initial power spike compared to 1/3 MW, or perhaps 1/2, in the initial power spike. The yield would be larger in the part following the spike if there were no mechanical separation.

A.F. THOMAS:

I wasn't thinking so much of GODIVA itself, but more of a system such as a melt in a graphite crucible where the thing would go critical and, unless there were sufficient mechanical damage to break the crucible, molten metal would already be there and it would continue for a long while; or perhaps the graphite mould might break and fall into the bottom of a badly-designed system where it could still retain a supercritical configuration.

W.R. STRATTON:

In this hypothetical case, I suggest that one would have to have means to break the mould in some sense to allow dispersion of the liquid to safe geometry. I think that safe geometry could be found which would be quite adequate.

K.M. HILL:

I think we should keep a sense of proportion. Even if the thing were nuclearly safe I don't feel that any plant operator would be happy at the thought of molten uranium dripping on to the floor or crucibles breaking. We consider it a serious incident if uranium catches fire; it it drips on to the floor it will not only catch fire, but set a lot of other things on fire as well.

A.F. THOMAS:

The point at issue is not whether it's going to be an awful mess afterwards but whether it's worth providing a criticality incident detector.

W.R. STRATTON:

A detector of some kind would be necessary.

F.R. CHARLESWORTH:

One could certainly think of a case where it would be of economic advantage to operate somewhat nearer to the estimated critical condition, namely in the processing of low-enriched fuel which had been irradiated, where one was uncertain of the precise reactivity of the system when in nitric acid. If one could control the rate of addition of metal and the rate of take-off of solution, with perhaps a modicum of analytical control across the system, one could fairly confidently operate with say 90 % of the minimum critical mass estimate, or perhaps even a little above the minimum critical mass estimate, since the rods would, in fact, be in random array rather than in the appropriate array for a minimum critical.

H.J. HEIN, Germany:

In his paper, Dr. Stratton describes an incident which happened to an uranium oxide plastic core. Can anyone here tell me more about the quenching mechanism of the uranium oxide plastic dispersions, in particular about bubble formation which must be similar to the well-known trouble with the AGN reactor ?

W.R. STRATTON:

The incident you referred to is one that occurred in the Argonne National Laboratory in 1952. As I understand it, people in this operation were guilty of a gross violation of their own safety regulations, which was the cause of the accident as they were replacing a central poison rod with water remaining in the system. The water should have been drained before they attempted to replace the rod. The plastic apparently swelled and boiled and finally blackened near the ten to twenty micron-size uranium oxyde particles that were embedded in the plastic. The system was thoroughly quenched by this swelling which forced water out of the system, decreasing the moderation. This stopped the reaction.

J.G. WALFORD, United Kingdom:

I would refer to the emphasis placed by some earlier speakers on the possibility of severe public relations or political consequences following a radiation incident. I rather formed the impression from Dr. Stratton's remarks, and also from the apparently quite minor consequences of the latest processing plant accident at Idaho, that the stage of serious political consequences has now been passed as far as criticality incidents are concerned, provided no personal injury or very severe damage is caused. I would like to ask Dr. Stratton whether this really is so in the United States, bearing in mind that the fact that an incident has occurred cannot fail to become generally known. (The Idaho accident illustrates this latter point since plant evacuation was necessary even though it was only for a short time.) I would hesitate to believe one could now assume a similar situation to apply in European countries, including the United Kingdom.

W.R. STRATTON:

I do not think this is the situation in the United States. I believe there is still a large amount of public apprehension about nuclear incidents. The

tragedy with this SL-1 reactor was very bad and very widely publicised. For minor plant incidents I think it is nearly as it was a few years ago; the public has not yet been educated to accept these risks.

J.G. WALFORD:

Perhaps I should have emphasized that when I referred to the Idaho incident as one which did not cause, shall we say, any great public apprehension, I was not speaking of the SL-1 incident but of the more recent one in the chemical plant.

W.R. STRATTON:

It is quite true that the second plant incident caused only an evacuation of the processing plant and received much less publicity than had many previous incidents, but I'm not sure that this one may just have been swamped publicity-wise by the tragic incident only a few weeks earlier.

F.R. CHARLESWORTH:

I would like to add that a recent plant designed in the U.K. is concentration controlled and it is recognised that there is always a possibility of a critical assembly with such a plant and the limiting dose to operators has been set at 5 rems in the case of an emergency.

R. CAIZERGUES, France:

M. Stratton a mentionné tout à l'heure l'arrêt du premier pic de puissance par ébullition. Je pensais que cet arrêt était également provoqué par la création de bulles de radiolyse dans les réacteurs homogènes. D'autre part, comme la durée du premier pic de puissance est très faible, les bulles ne doivent pas avoir le temps de se dégager et se dégagent donc après le premier pic de puissance. J'aimerais savoir s'il a été procédé à une étude de ces bulles de radiolyse et à une mesure de leur diamètre moyen, par exemple.

W.R. STRATTON:

There is some information on this question of the size of bubbles and how fast they escape in a report written by a group headed by Mr. J. Flora of Atomics International.

H.K. SAUER, Germany:

There has been a rumour about a revised edition of the " Nuclear Safety Guide ". Could someone give us any information on this?

D. CALLIHAN:

There has been in existence for some years in the United States an A.E.C.-published document entitled " Nuclear Safety Guide " (TID 7016) which has received some usage in the United States and perhaps some abroad also. This document has been worked upon for the past year or so by a group in the United States and has been re-issued with better and more readily interpretable information. This revision is in the process of publication and will be sent to all those taking part in this Symposium.

Session VIII

ORGANISATION OF EXPERIMENTAL CRITICALITY RESEARCH

ORGANISATION DE LA RECHERCHE EXPÉRIMENTALE EN MATIÈRE DE CRITICALITÉ

Chairman - Président :

M. R. GALLEY
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ORGANISATION DE LA RECHERCHE EXPÉRIMENTALE EN MATIÈRE DE CRITICALITÉ*

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INTRODUCTION

L'étude des problèmes de criticalité fait apparaître deux possibilités :

a) La définition directe, partout où cela est possible, de paramètres de sécurité pour les installations industrielles en s'appuyant soit sur des résultats généraux provenant d'études antérieures, soit sur des calculs théoriques limités ou non à un cas particulier.

b) L'expérimentation à caractère fondamental (définition des paramètres cités ci-dessus) ou à caractère particulier, en vue d'une vérification bien précise.

En France, les principaux problèmes de criticalité posés jusqu'à présent ont pu trouver des réponses, en général provisoires, dans les connaissances de base fournies par la littérature ou dans les calculs. Il était urgent de définir, pour les installations existantes ou à démarrer, les mesures indispensables pour assurer la sécurité du personnel.

On peut citer, parmi les problèmes déjà abordés :

— L'étude de la criticalité de la première usine de plutonium de Marcoule, comportant des problèmes relatifs aux solutions, aux dispositifs de transport et de stockage des produits finis (sel, métal).

— L'examen des chaînes de fabrication et de récupération des éléments combustibles d'uranium légèrement enrichi (sels, oxydes) et l'étude du stockage et du transport de ces mêmes produits.

Mais il apparut rapidement que les projets futurs d'usines traitant l'uranium ou le plutonium ne pouvaient pas être résolus de façon suffisamment rationnelle en s'appuyant uniquement sur les données de base utilisées jusqu'alors.

Il faut pourtant reconnaître que les calculs devenaient plus nombreux, et mieux adaptés grâce à l'utilisation des machines électroniques, et que les connaissances générales devenaient de plus en plus étendues, fournissant des bons ordres de grandeur.

Cette amélioration des moyens d'étude n'a pas semblé suffisante. Pour éviter d'arriver à des coûts d'installations prohibitifs, provenant soit de l'appli-

* Conférence présentée par M. C. Clouet d'Orval.

cation de coefficients de sécurités incertains donnés par le calcul, soit de l'utilisation directe des normes étrangères, il fut décidé de procéder, dans certains cas, à des expériences.

Le but de ces expériences était de résoudre des problèmes parfaitement définis, aux limites assez précises. Il fallait pourtant que, tout en fournissant des résultats généraux, elles permissent de serrer d'assez près la réalité. Un autre impératif était d'apprendre à réaliser des dispositifs expérimentaux présentant toutes les conditions de sécurité requises et permettant d'entreprendre quelques études simples de base.

Deux étapes caractérisent les possibilités actuelles de nos recherches expérimentales :

a) Utilisation d'un bâtiment existant à Saclay (Seine-et-Oise) abritant la pile homogène PROSERPINE, où une autre expérience simple pouvait être montée dans un temps assez bref pour mettre au point les techniques.

b) Construction d'un nouveau laboratoire, dit « Station de Criticalité », situé aux environs de Dijon (Côte-d'Or), où seront réalisées des expériences plus importantes sur des milieux solides et liquides.

Le programme expérimental immédiat lui-même peut se résumer ainsi :

— Etude des stockages spéciaux, c'est-à-dire sortant du domaine des normes connues pour le plutonium, en milieu correctement modéré.

— Examen de certains problèmes posés par l'uranium 235, dans des configurations particulières, et à des taux de modération variables.

Ce programme trouve des applications très directes dans les installations pour lesquelles il a été élaboré, mais on s'attache à lui garder un caractère général qui rende utilisables les résultats dans un domaine plus vaste.

Nous nous attacherons à décrire ici les laboratoires où sera réalisé ce programme et aussi les principaux dispositifs expérimentaux utilisés ou prévus.

Le Laboratoire de Saclay (Proserpine)

Le bâtiment (figures 1 et 2)

Il est constitué d'un hall de dimensions 30 m × 15 m × 10 m entièrement fermé et étanche, permettant de réaliser deux expériences en même temps.

Les premières expériences ayant eu pour objet le plutonium, ce hall est équipé d'une ventilation climatisée, renouvelant l'air cinq fois par heure, et maintenant une dépression de 10 mm d'eau. Des appareils de détection suivent l'activité alpha de l'air et, sur incident, la ventilation peut être stoppée, et le hall isolé par des vannes. L'air est alors filtré. Le revêtement du sol et des murs est aisément lavable.

Une salle de décontamination permet un éventuel nettoyage des surfaces internes de boîtes à gants. On y entre en scaphandre autonome.

Un mur de protection en béton ordinaire, d'épaisseur au moins égale à 1,30 m, de 5 m de hauteur, entoure les expériences de tous côtés. Jusqu'à 10 m, on trouve des épaisseurs de 60 ou 30 cm de béton.

En cas d'excursion critique, on a calculé ou vérifié, à puissance faible pour nos expériences, que la dose totale restait, pour 10^{18} fissions, inférieure à 1 rem. Le rayonnement diffusé compose la majeure partie de cette dose,

Figure 1

Le bâtiment PROSERPINE du Centre d'Études Nucléaires de Saclay (plan).

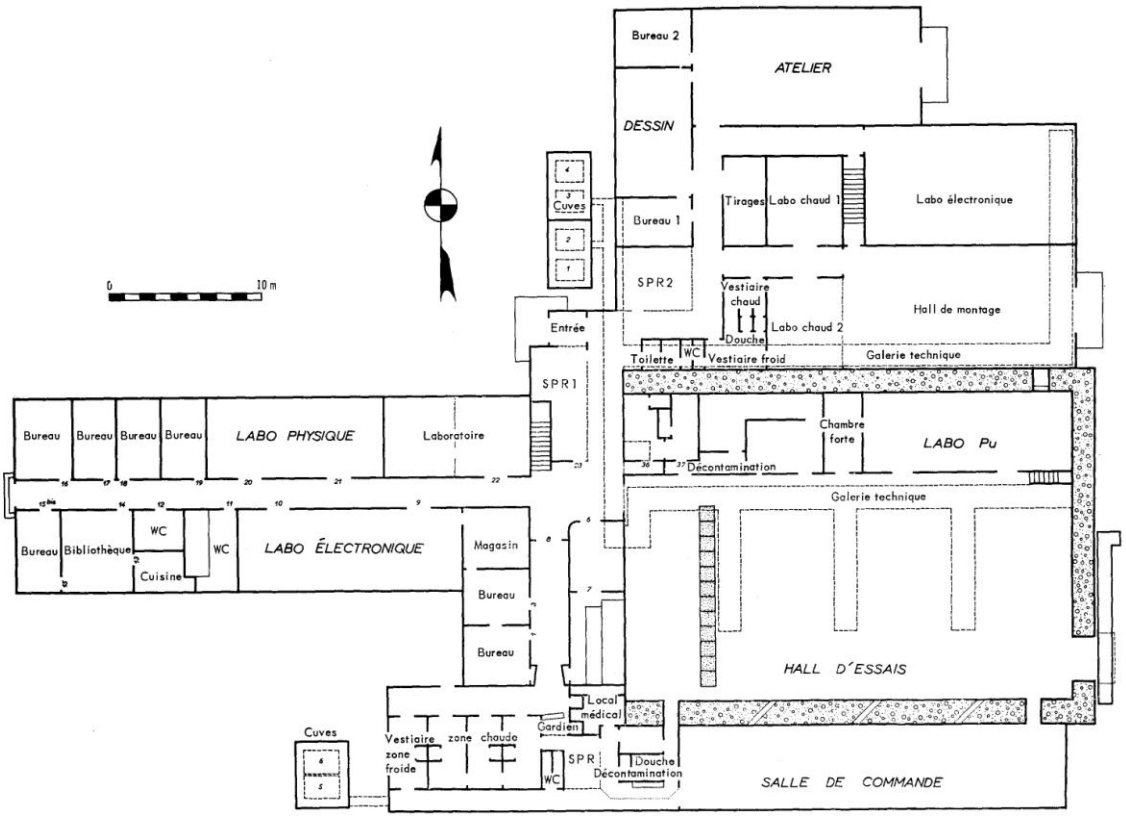

Figure 1. *Le bâtiment PROSERPINE du Centre d'Études nucléaires de Saclay (plan).*

malgré la présence d'un écran (réflecteur lui-même, ou toit de paraffine borée) situé à la partie supérieure des expériences.

Un laboratoire de chimie, contigu au hall d'essais, permet la préparation des matières fissiles. Seules des expériences sur des solutions sont entreprises dans le bâtiment.

Le produit concentré, sorti du laboratoire de chimie, est introduit dans la partie stockage ou préparation de la solution des dispositifs expérimentaux, où il est amené à la concentration désirée.

Une aile située dans le prolongement du hall contient les annexes, principalement un laboratoire d'électronique, un bureau d'études mécaniques, un laboratoire de construction d'appareils de physique.

Dans ce bâtiment, deux dispositifs expérimentaux, de longue durée, existent : la pile PROSERPINE, en fonctionnement, et l'expérience ALECTO, en démarrage.

PROSERPINE [1] [2] [3]

Conçue à l'origine (1958) pour étudier la physique d'un milieu homogène concentré, composé de plutonium et d'eau, une nouvelle orientation de son utilisation lui a été donnée, grâce à la possibilité d'une expérience (1961) employant l'uranium enrichi à 90 %. Le programme sera probablement poursuivi dans l'avenir en utilisant l'uranium 233.

La comparaison des masses critiques et des grandeurs neutroniques caractéristiques des deux milieux permet une connaissance meilleure des constantes à utiliser pour les calculs, ces constantes pouvant être employées ensuite dans d'autres cas. Elle entre, à ce titre, dans la catégorie des expériences dites de criticalité.

Description (figure 3)

La pile elle-même est constituée d'une cuve qui, d'abord en acier inoxydable, a ensuite été remplacée par une cuve en zirconium, de diamètre 25 cm, de hauteur 30 cm, remplie aux deux tiers environ d'une solution de plutonium ou d'uranium enrichi. Cette cuve est entourée d'un réflecteur d'oxyde de béryllium d'épaisseur moyenne, 27,5 cm, puis d'un réflecteur de graphite, épais de 50 cm, ayant une forme extérieure cubique.

La composition de ce réflecteur était justifiée par le désir d'obtenir une masse critique aussi faible que possible.

Un tube fin, traversant l'empilement à la base, réunit la cuve à un réservoir de stockage, identique à la cuve, situé dans une boîte à gants proche où l'on prépare la solution, à partir de quantités amenées d'une autre boîte à gants de transfert manuel.

A la partie supérieure de la cuve, fermée par un couvercle, partent des tubes traversant l'empilement et débouchant dans une boîte à gants par laquelle on peut introduire différents appareils expérimentaux.

Le circuit de pile constitue une enceinte étanche, entourée soit par des boîtes à gants, soit par des enveloppes de vinyle ou de métal destinées à recueillir une fuite éventuelle.

Un dispositif, composé de deux barres horizontales de cadmium pénétrant dans l'oxyde de béryllium, assure la sécurité, doublée par la chute par gravité du liquide de la cuve. Une cabine thermostatée entoure complètement la pile.

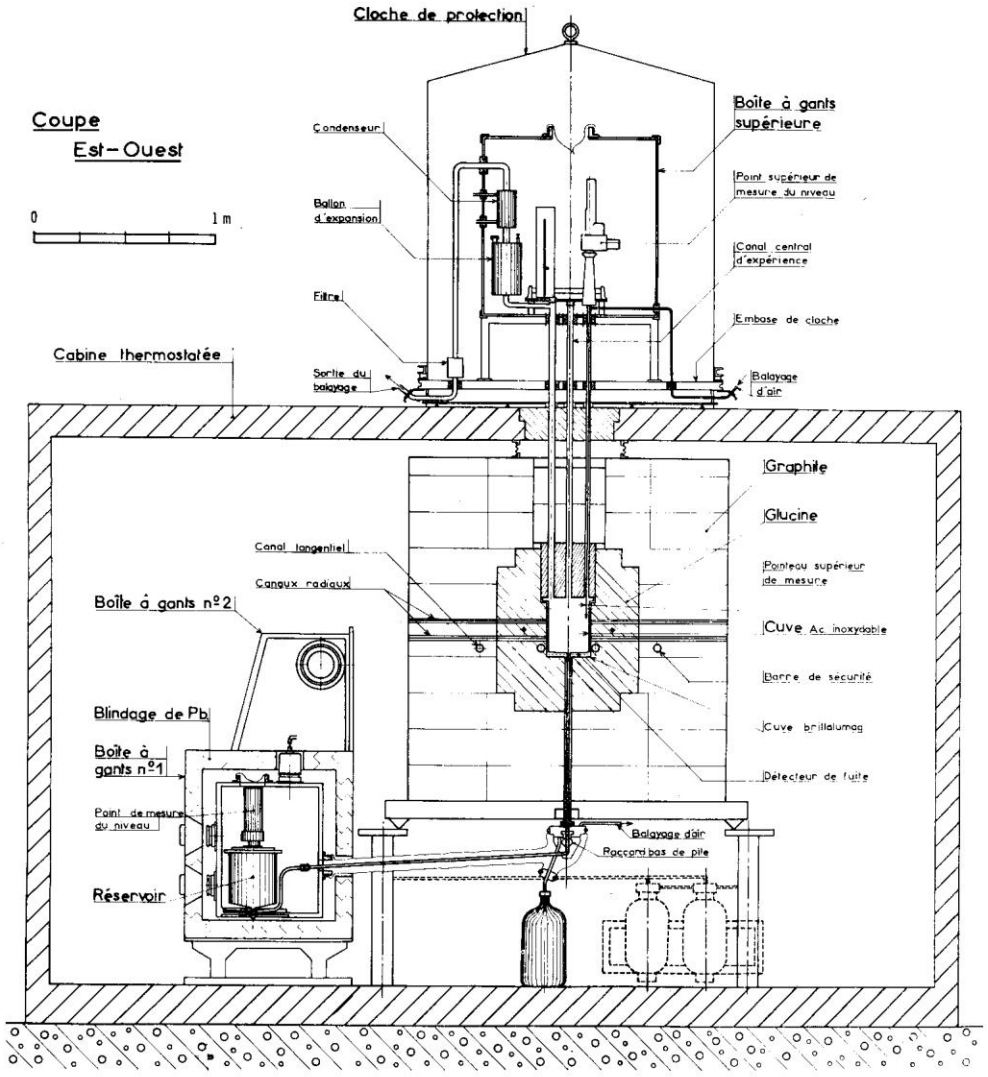

Figure 3. *PROSERPINE (coupe verticale).*

Il est intéressant de mentionner ici les principales techniques mises au point à l'occasion de la construction de PROSERPINE et dont l'utilisation peut être assez générale [4] :

— La stabilité du niveau dans la cuve est obtenue grâce à une régulation à air comprimé qui maintient le liquide dans un puits ($\Phi = 6$ mm) du réservoir inférieur. Ainsi, lorsque la cuve est remplie, le réservoir du bas est presque vide, excepté ce puits. Des variations de 1 cm dans ce puits correspondent à des variations de 5.10^{-3} mm dans la cuve.

— La mesure des niveaux cuve et réservoir de stockage est réalisée grâce à des pointes à contact électrique pouvant lire des variations de niveau de 1/100 de mm.

Expériences

Les expériences réalisées sur PROSERPINE sont de deux sortes :

a) *Etude de masses critiques*

Elle a été effectuée en faisant varier les paramètres hauteur et concentration (figure 4). Cette étude nous a amenés à conclure que, d'une part l'importance énorme du réflecteur, d'autre part le rôle joué par le vide situé au-dessus de la solution, ne permettent pas, avec les méthodes de calcul simples, d'approcher la masse critique exacte à mieux de 10 à 20 %. Une méthode, utilisant la théorie du transport en deux dimensions, permettra peut-être de rapprocher les calculs des résultats expérimentaux.

L'expérimentation a montré par ailleurs que, pour obtenir une masse critique par extrapolation sous-critique à mieux de 2 %, il fallait approcher à moins de $200\ 10^{-5}\frac{\Delta k_{\text{eff}}}{k_{\text{eff}}}$ de l'état critique.

b) *Étalonnages particuliers* [5] [6] [7]

Nous avons été amenés, pour mieux connaître le comportement neutronique de la pile, à effectuer les mesures suivantes :

— Détermination d'un étalon de réactivité par empoisonnement homogène au bore ;
— Réactivité, en fonction de leur enfoncement, des barres de contrôle ;
— Etude des courbes de flux thermiques et rapides et étalonnage en puissance ;
— Etude du spectre de neutrons.

c) *Mesures diverses*

D'autres mesures, sortant de l'étude propre des masses critiques, ont été entreprises et intéressent surtout le temps de vie des neutrons prompts [5] et la proportion effective des neutrons retardés.

Par ailleurs, le phénomène de fluctuation de la population neutronique au voisinage de l'état critique a fait l'objet d'une recherche systématique axée sur les mesures de variance.

*ALECTO** [8]

Cette nouvelle expérience a plus spécialement pour objet l'étude systématique de certaines masses critiques. Elle se présente comme une installation pilote, se prêtant à la mise au point de méthodes de mesures et de techniques expérimentales. Elle est cependant susceptible, bien qu'elle soit la première

* ALECTO est l'une des trois Furies, filles de PROSERPINE.

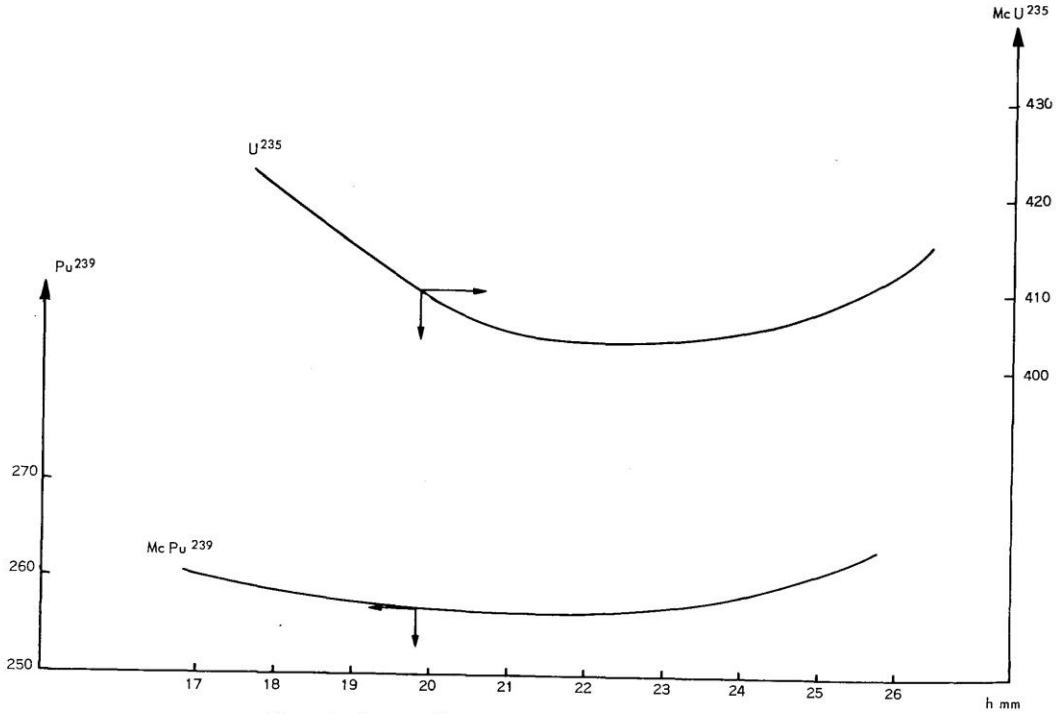

Figure 4. *Masses critiques Pu 239 et U 235 de PROSERPINE.*

d'une série dont on parlera plus loin, de fournir des résultats intéressant directement certaines de nos installations industrielles.

Il a été décidé, pour commencer, de réaliser à l'aide d'ALECTO certaines expériences systématiques, partiellement effectuées à l'étranger et présentant les deux caractères suivants :

— Pouvoir se monter simplement dans le hall existant, en donnant l'occasion de mettre au point des montages et des méthodes de mesure.
— Répondre rapidement à diverses questions sur la validité de certains coefficients de sécurité d'usines.

Description (figure 5)

Le but d'ALECTO est de pouvoir tester les conditions critiques de récipients cylindriques, contenant une solution de nitrate de plutonium, avec ou sans réflecteur. L'eau a été choisie comme premier réflecteur, mais l'installation peut être transformée pour en étudier d'autres. On peut considérer : le bloc pile, les stockages.

a) *Le bloc pile*

Il est constitué d'une grande cuve cylindrique fixe ($\Phi = 110$ cm) pouvant contenir intérieurement des cuves plus petites (diamètre de 20 à 50 cm, hauteur 100 cm). La cuve intérieure contient la solution de plutonium amenée par le fond. Cette cuve est surmontée d'une boîte à gants formant couvercle, où viennent prendre place les dispositifs de sécurité (deux barres de cadmium plongeant dans la solution fissile) et les appareils de mesure.

L'ensemble cuve intérieure-boîte à gants sécurité est démontable, pour le changement de cuve, et se retire par une ouverture pratiquée dans la cabine qui entoure le montage.

Cette cabine est destinée à prévenir la rupture du dispositif décrit ci-dessus et réalise une enceinte étanche et ventilée à travers filtre. Elle est posée dans le hall PROSERPINE.

Entourée de murs épais de béton et surmontée d'un toit de paraffine, elle laisse le passage à des communications, d'une part avec un ballon d'eau par un gros tube ($\Phi = 125$ mm) de vidange rapide, d'autre part avec la cabine des stockages plutonium, par un tube de vidange de la solution de plutonium ($\Phi = 16$ mm).

b) *Les stockages*

La quantité de plutonium à stocker étant de 2 kg, des précautions ont été prises pour réaliser un stockage sûr par sa géométrie. Il comporte deux parties :

— Le stockage primaire, ou d'arrivée, formé de bouteilles de diamètre 10 cm, hautes de 100 cm et espacées de 50 cm d'axe à axe. Elles communiquent par l'intermédiaire d'une bouteille « balance » avec un stockage secondaire.
— Le stockage secondaire est formé de deux réservoirs annulaires identiques, dont l'un peut contenir toute la solution nécessaire pour une expérience (au maximum 80 litres). Leur épaisseur de 4 cm et leurs écrans internes et externes cadmium-paraffine les rendent sûrs à

Figure 5

ALECTO (coupe verticale et plans).

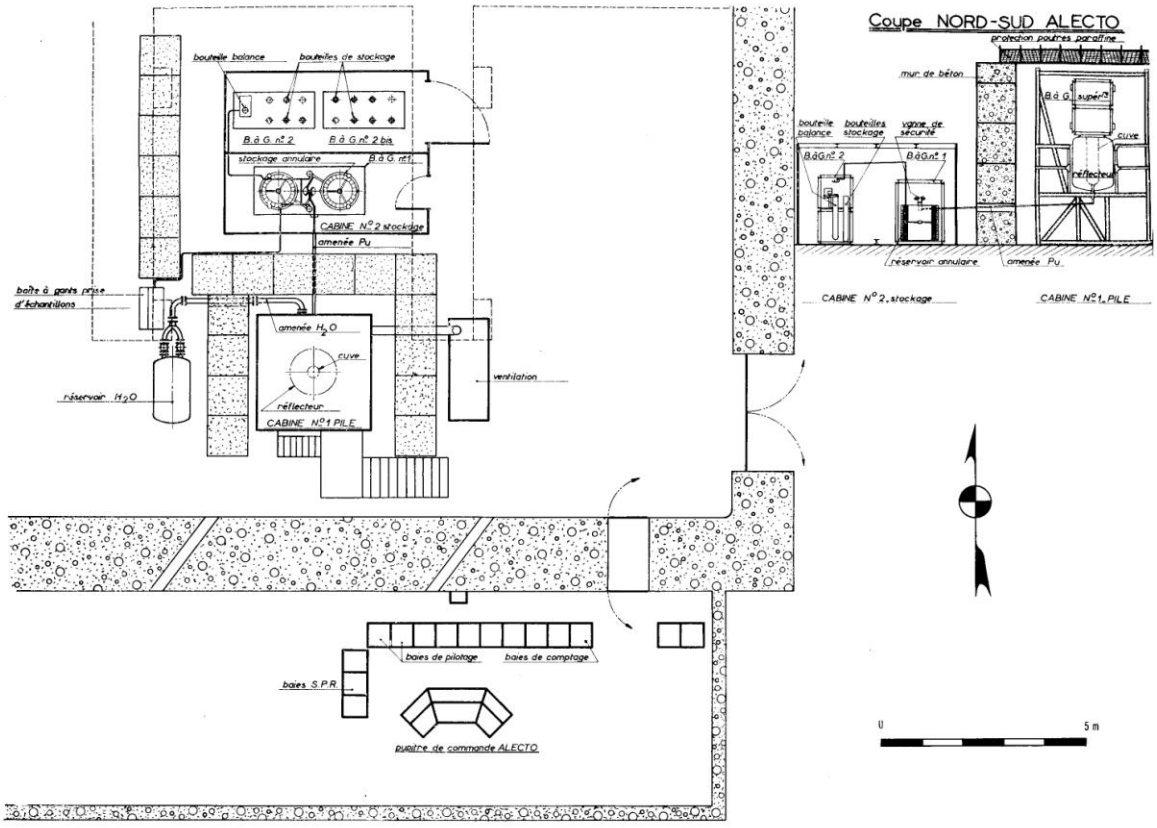

Figure 5. *ALECTO (coupe verticale et plans).*

toute concentration. Ils permettent une homogénéisation rapide et simple de la solution. L'un d'entre eux est sélectionné, après cette opération, pour communiquer avec la cuve à essayer.

Ces stockages sont disposés dans une seconde cabine adjacente à la première.

Programme expérimental

Il comprend l'essai de cuves de diamètre variable, à hauteur variable. Les cuves sont réfléchies soit latéralement (cuves exactement cylindriques), soit également sur la partie inférieure (cuves particulières).

Outre les mesures de taille critique avec diverses concentrations de solutions, et en utilisant des cuves de dimensions variées, il est envisagé d'associer à ALECTO une petite source de neutrons pulsés. Cette technique, qui a déjà été utilisée sur d'autres assemblages critiques et sous-critiques, doit permettre des études particulièrement intéressantes pour les questions de criticalité. La mesure de l'antiréactivité de forts absorbeurs disposés dans des cuves de stockage peut être, par exemple, effectuée par cette technique, avec une précision sans doute meilleure que celle obtenue en utilisant d'autres techniques plus classiques. On sait qu'après l'injection d'une bouffée de neutrons dans un milieu multiplicateur une phase de transition s'établit, au cours de laquelle la population neutronique se met en équilibre, puis décroît exponentiellement en fonction du temps ; après la décroissance des neutrons prompts, il subsiste un groupe de neutrons décroissant lentement et dû aux neutrons retardés. La mesure du coefficient α caractérisant la décroissance exponentielle des neutrons thermiques dans l'assemblage peut, moyennant quelques simplifications, être reliée au temps de vie des neutrons l et au coefficient de multiplication effectif k_{eff} par la relation :

$$\alpha = \frac{1}{l}\,[1 - (1 - \beta)\,k_{\text{eff}}]$$

α étant la fraction des neutrons retardés pour le système considéré. En posant :

$$\rho = \frac{k_{\text{eff}} - 1}{\beta}$$

on a au second ordre près :

$$\alpha = \frac{\beta}{l}(1 - \rho)$$

ρ étant la réactivité du système. L'appareillage mis en œuvre se compose essentiellement d'une petite source de neutrons pulsés, qui est maintenant construite par différentes sociétés, et d'un sélecteur en temps muni d'une centaine de canaux.

Techniques particulières

Il faut citer, parmi les études propres à cette expérience, la réalisation des appareils suivants :

— Vannes de sécurité doubles ; sur tube de diamètre $\Phi = 16$ mm, d'encombrement : $35 \times 15 \times 30$ cm ;

— Pompe à plutonium à membranes, de très petites dimensions d'encombrement total 15 × 15 × 24 cm. Débit max. 2,5 litres/min ;
— Pointes de mesure de niveau à câbles précises au 1/10 de mm.

Tous ces appareils sont situés en boîte à gants.

Matériel expérimental

La nécessité d'effectuer des mesures dans des milieux très ramassés, approchant de près la criticalité et fortement contaminés, a conduit à la réalisation d'appareils divers.

a) *Chambres à fission miniatures*

Ces chambres dont les diamètres s'échelonnent de 3 mm (efficacité 10^{-4} Hz par n/cm^2 s en υ_5) à 10 mm (efficacité 2 10^{-2} Hz par n/cm^2 s en υ_5) sont construites en nickel ou en zirconium. Remplies à l'argon, sous plusieurs atmosphères, elles donnent des impulsions facilement détectables après plusieurs mètres de câble porteur.

Elles ont été relayées, pour les faibles flux, par des compteurs BF 3 suédois, type BB, dont les faibles impulsions ont nécessité la réalisation d'un préamplificateur à faible souffle [9] [10].

b) *Détecteurs*

Des courbes de flux ont été établies dans la solution fissile elle-même, grâce à des détecteurs d'activation placés sous plusieurs enveloppes de polyvinyle. Une fois ces enveloppes soigneusement enlevées, il est possible d'effectuer un comptage dans des installations « inactives ».

c) *Sécurités diverses*

Un aspect intéressant des divers appareils de détection utilisés pour la sécurité est leur vitesse de réponse.

L'emploi de photomultiplicateurs a permis de ramener les temps de réponse à des valeurs négligeables devant les vitesses des barres, ceci même à des niveaux neutroniques bas.

— Sécurité à échelle rapide :

Une échelle rapide (temps mort 0,1 μs) compte les impulsions reçues d'un photomultiplicateur sensible aux photons. Elle est remise à zéro à intervalles réguliers, ces intervalles étant fixés par un oscillateur. Si, pendant l'intervalle fixé, pouvant être par exemple de 1/50 de seconde, l'échelle n'est pas remplie, elle est automatiquement remise à zéro. Dans le cas contraire, une sécurité de fin d'échelle fonctionne. En faisant varier la vitesse de remise à zéro pendant la montée normale de la puissance, on peut suivre parfaitement le niveau neutronique, tout en gardant une sécurité très proche.

— Utilisation d'un photomultiplicateur en courant :

L'utilisation directe du courant fourni par un photomultiplicateur permet de faire marcher des relais très rapides (1 ms) en conservant un temps de réponse faible.

D'autre part, la dérivée de ce courant peut donner un signal de réactivité tout à fait utilisable pour un dispositif de sécurité.

Pour le moment, seule la détection des photons est employée, mais on peut, si les niveaux neutroniques le permettent, se servir de cristaux sensibles aux neutrons, ce qui serait intéressant dans le cas d'un système à réflecteur métallique.

La station de criticalité de Dijon

Présentation

Le laboratoire a été conçu pour permettre une expérimentation poussée. C'est-à-dire :

— Il présente une protection fixe homogène, tenant particulièrement compte du rayonnement diffusé ;

— Il permet l'utilisation de quantités de matières fissiles importantes. Pour les solutions, des stockages fixes sont prévus, qui rendront les opérations de transfert plus rapides et plus sûres ;

— Il est situé dans une région de densité de population très faible.

Il est destiné à répondre le plus directement et le plus rapidement possible à des problèmes de stockages dans les usines, sans que l'on y néglige pour autant l'aspect très général de certaines études.

Sa conception est particulièrement étudiée pour reproduire des appareils et des installations industrielles, dans leur ensemble ou en partie.

Description du bâtiment (figure 6)

Le bâtiment est composé principalement de deux cellules indépendantes, dont l'une est encore divisée en deux.

Leurs dimensions sont :

— Grande cellule (1) : $19 \times 12 \times 10$ m

— Petites cellules (2) : $9 \times 12 \times 10$ m

La protection de ces cellules consiste en un mur de béton ordinaire, d'épaisseur 146 cm, et d'un plafond, d'épaisseur 70 cm.

La dose calculée pour une excursion de puissance, sur les expériences prévues, de 10^{18} fissions est inférieure à 0,5 rem.

Ces cellules, deux laboratoires de chimie et des stockages en sous-sol constituent la zone active, que complètent des salles de décontamination et des aires de montage, et un atelier actif hors des cellules.

L'ensemble de cette zone active est reliée, par un bâtiment comprenant les servitudes (vestiaires, ventilation) à une zone inactive qui comporte les ateliers, laboratoires et bureaux.

Les dispositifs expérimentaux

Les expériences présentent des caractères communs que nous donnerons ici et nous nous limiterons à certaines d'entre elles, en ce qui concerne les détails.

a) Elles sont contenues dans des caissons étanches démontables, dont les panneaux se fixent sur un quadrillage du sol. Pour les solutions, les stockages primaires (produit concentré) et secondaires (solution préparée pour une manipulation) se trouvent sous les laboratoires de chimie; les stockages tertiaires contenant les quantités juste nécessaires pour une manipulation, se trouvent sous les cellules.

Une boîte à gants transfert permet les différents mouvements.

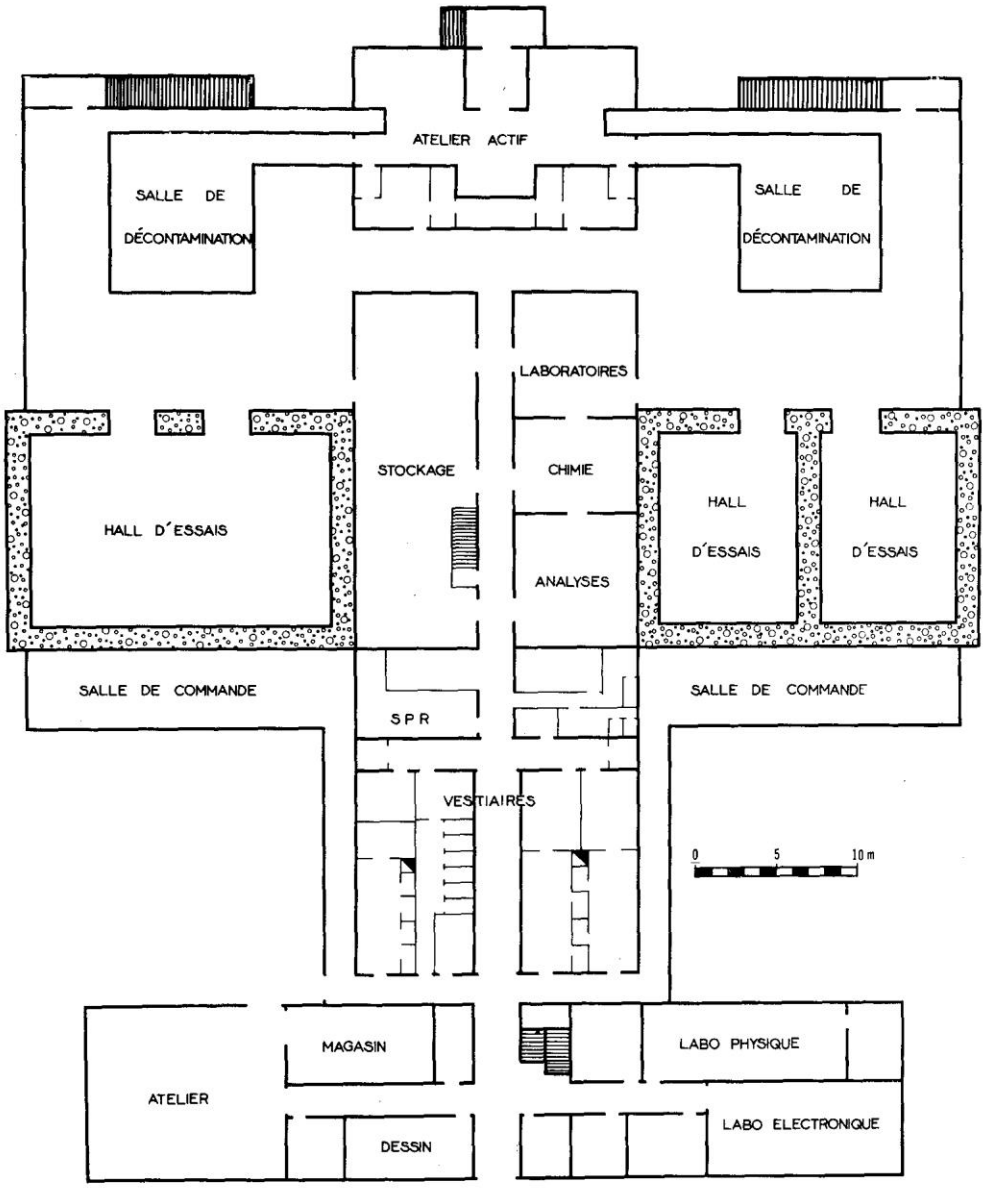

Figure 6. *La station de criticalité de Dijon.*

b) Le but des expériences initiales est de définir des paramètres de sécurité pour les stockages de solutions de nitrate de plutonium. Mais, en même temps, ces premières expériences serviront à définir les stockages définitifs eux-mêmes de la station.

c) Le programme expérimental comprend l'étude des conditions critiques d'un cylindre annulaire isolé, avec et sans réflecteur, le creux central

étant vide ou plein. Ensuite sera entreprise l'étude de l'interaction de deux réservoirs annulaires.

Les calculs d'interaction de réservoirs entre eux sont, en général, fort complexes si l'on veut les traiter complètement. Pour étudier expérimentalement cette interaction, une solution envisagée consiste à rendre l'ensemble critique et à moduler légèrement la réactivité de l'un des assemblages. La détection de la perturbation ainsi réalisée est une fonction des coefficients de couplages thermiques et rapides des divers assemblages. La théorie de ces assemblages couplés a été étudiée par plusieurs auteurs, en particulier R. Avery, et des expériences ont été réalisées aux États-Unis et en France. Les résultats mettent en évidence l'intérêt d'une telle étude pour les problèmes de criticalité. On doit pouvoir atteindre ainsi une mesure directe des coefficients de couplage des divers assemblages.

d) Le dispositif lui-même comprend une grande cuve à murs démontables contenant l'eau du réflecteur et suffisamment vaste ($320 \times 210 \times 150$ cm) pour y faire les expériences d'interaction.

Cette cuve supporte une armature métallique où viennent se poser les cuves à essayer.

La solution de plutonium arrive par la partie inférieure du pot au moyen de tuyaux souples branchés sur un bloc de répartition à plusieurs ouvertures.

Un mécanisme permet le déplacement des récipients vides, dans les problèmes d'interaction.

La sécurité du système repose d'une part sur la vidange rapide des solutions par des vannes de sécurité, d'autre part sur l'introduction de barres absorbantes dans la solution fissile.

Conclusion

Il semble que, même avec les résultats publiés, il soit difficile de traiter tous les problèmes de sécurité relatifs aux installations nouvelles.

— L'application directe des normes à des systèmes de forme et de composition complexes est délicate.

— Le coût des installations devient souvent prohibitif.

Les calculs eux-mêmes ne serrent pas d'assez près la réalité pour que l'on puisse accorder aux facteurs de sécurité estimés une confiance totale.

C'est pour ces raisons que l'on est amené, dès que les installations industrielles prennent quelque ampleur, à créer des laboratoires de criticalité destinés à donner des réponses aux problèmes les plus délicats.

Malgré l'utilisation très directe des résultats attendus, il faut garder aux programmes un caractère assez général. Ceci peut être réalisé en coordonnant les recherches des différents laboratoires et en s'efforçant de créer, lorsque cela est possible, des échanges fréquents portant sur les techniques mises en œuvre et sur les résultats.

REFERENCES

1. J. Bertrand, P. Bonnaure, C. Clouet d'Orval, J. Corpel, J. de Lamare, P. Lecoustey, I. Prevot, R. Roche, M. Sauve, J. Tachon et G. Vendryes, " Proserpine : expérience critique homogène avec le plutonium ", *Actes Deuxième Conf. Int. Nations Unies sur l'utilisation de l'énergie atomique à des fins pacifiques, Genève, 1958, 6*, 802.

2. C. CLOUET D'ORVAL, " Rapport de Sécurité de Proserpine ", *EC/S 76.*
3. J. TACHON, " Étude neutronique d'une pile à neutrons thermiques au plutonium : Proserpine ", Thèse, *C.E.A. 1547* (1960).
4. F. BERGER et J. BERTRAND, " Mesure et régulation du niveau d'un réacteur homogène au plutonium ", *C.E.A. 1054* (Déc. 1958).
5. J. BERTRAND, C. CLOUET D'ORVAL et J. TACHON, " Détermination de l'étalon de réactivité et mesure de l'efficacité des neutrons retardés dans Proserpine ", à paraître dans *J. Nucl. Energy.*
6. C. CLOUET D'ORVAL, E. DELIGAT, M. GUERY et J. TACHON, " Etalonnage en puissance de Proserpine ", *EC/S 24.*
7. C. CLOUET D'ORVAL, J. BERTRAND, P. LECOUSTEY et J. TACHON, " Mesure du temps de vie dans Proserpine par modulation de la réactivité ", *C.E.A. 1595* (Févr. 1960).
8. R. CAIZERGUES, C. CLOUET D'ORVAL, R. KREMSER et P. VERRIERE, " Rapport de Sûreté Alecto ", *EC/S 86.*
9. R. AGAISSE et C. CLOUET D'ORVAL, " Fonctionnement d'un compteur de neutrons type BB ", *EC/S 34.*
10. R. AGAISSE, " Étude d'un préamplificateur pour la détection d'impulsions faibles de neutrons issus d'un milieu critique (Proserpine) ", *C.E.A. 1596* (Jan. 1960).
11. R. AVERY, C.E. BRANYAN, G.S. BRUNSON, C.E. COHN, G.S. FISCHER, H.H. HUMMEL, W.Y. KATO, F.S. KIRN, D. MENNECHETTI, F.W. THALGOTT et B.S. TOPPEL, " Réacteur de puissance surrégénérateur à couplage rapide-thermique : expérience critique ", *Actes Deuxième Conf. Int. Nations Unies sur l'utilisation de l'énergie atomique à des fins pacifiques, Genève, 1958, 12,* 478.
12. A.V. ANTONOV, A.I. ISSAKOV, I.D. MOURINE, B.A. NEOUPOKOEV, I.M. FRANCK, F.L. CHAPIRO et I.V. CHTRANIKH, " Étude de la diffusion des neutrons dans le béryllium, le graphite et l'eau par la méthode des impulsions ", *Actes Première Conf. Int. Nations Unies sur l'utilisation de l'énergie atomique à des fins pacifiques, Genève, 1955, 5,* 3.

THE EQUIPMENT AND METHODS USED IN BRITISH CRITICALITY LABORATORIES

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INTRODUCTION

Experimental criticality studies occupy a somewhat specialized position, shown diagrammatically in Figure 1, within the much broader field of reactor physics research. They have, as their ultimate purpose, the prevention of criticality in processing plants, laboratories, fissile material stores and other places where it must at all costs be avoided, in contrast to other reactor physics studies which are aimed at the efficient attainment of a nuclear reaction where criticality is an essential.

This difference in purpose is reflected in the design of facilities and equipment, in the experimental techniques and in the organisation of the two types of reactor studies; at the same time, each field can contribute very usefully to the needs of the other. On the one hand, every measurement made in exponential experiments and zero-energy reactors can be said to be of some potential value in the safety assessment of processing plant. On the other hand, well-designed critical assembly experiments can and should be used to yield data of fundamental value in reactor analysis; indeed, it is extremely desirable from an economic standpoint that they should be so used.

Critical facilities can also contribute directly in a wider sphere since they can be used for setting up simplified reactor mock-ups in the earliest stages of a reactor project, for the operation of pulsed or ' fast burst ' reactors, and for certain types of irradiation and dosimetry experiments. At the other end of the power scale, the facilities are used for subcritical neutronic experiments such as interaction parameter studies and accurate multiplication measurements, which are often adequate in themselves for nuclear safety assessments. These several extensions of the scope of critical assembly laboratories are shown within the broken line in Figure 1.

CRITICAL ASSEMBLY LABORATORIES

Critical experiments on behalf of plant safety have been in progress in the United Kingdom for almost ten years and have been carried out at three different A.E.A. Establishments — Harwell, Aldermaston and Dounreay — of which only the last two are in use at present. The division of work between these laboratories has depended primarily on the physical form of the fissile materials, experiments with metals being carried out almost entirely at Aldermaston and those with solutions at Harwell (up to 1957) and Doun-

Figure 1

The position of critical assembly laboratories within the field of reactor studies.

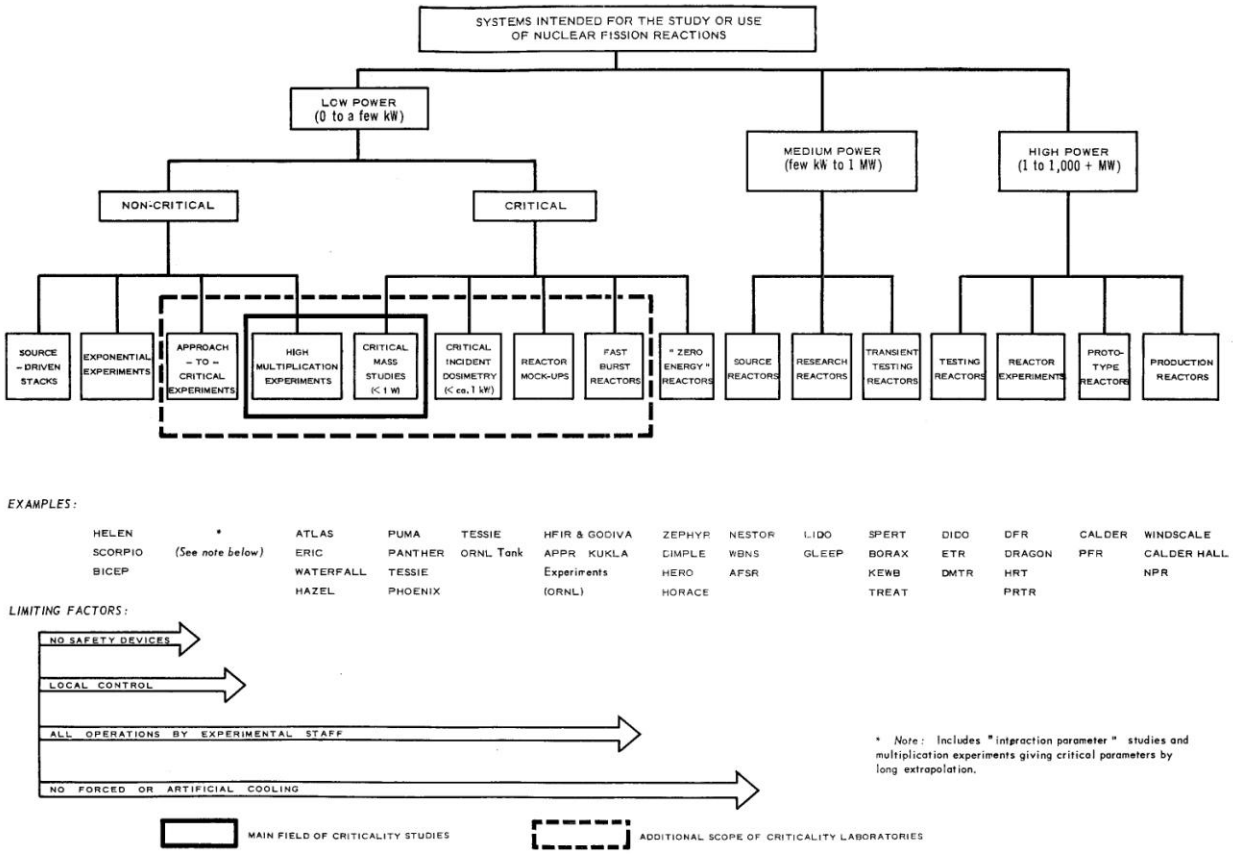

Figure 1. *The position of critical assembly laboratories within the field of reactor studies.*

TABLE I

COMPARATIVE FEATURES OF UNITED KINGDOM CRITICAL ASSEMBLY LABORATORIES AND CELLS

TABLE I. COMPARATIVE FEATURES OF UNITED KINGDOM CRITICAL ASSEMBLY LABORATORIES AND CELLS

Location	A.E.R.E. (Harwell) (Typical Cell)	A.W.R.E. (Aldermaston)	D.E.R.E. (Dounreay)			
			Cell 1	Cell 2	Cell 3	Cell 4
Date of operation	1952-1957	From 1952	From 1957	1959-61	from Nov. 1959	from Nov. 1960
Main types of experiments	Light and heavy water solutions of Pu, U 235 and U 233 in simple geometry	Metals and moderated solids (U 235 and Pu); interaction studies	Solutions of U 235; including large systems and *ad hoc* tests on process vessels	Solutions of U 235 in simple geometry (up to 3 ft overall dia.)	Moderated solids (U 235), especially large and heavy systems; interaction studies on solutions and solids	Plutonium solutions and moderated solids, usually in simple geometry
Number of rigs accommodated	One	Three	One or two	One	One	Two
Control room position	Local	Remote (100 yds)	Remote (100 yds)	Local	Local	Local
Exclusion area	On rear side of cell; also on upper floors of building	100 yds radius	100 yd radius; also earth embankment on open side of cell	None	None	None
Shielding material and thickness	Temporary concrete cairn (3 walls: 3 ft roof and usually none on rear wall)	Brick (1 ft) (normal structure, with unshielded doors and windows)	Concrete (1.5 ft) on three walls; *none* on fourth wall and roof	Below ground (rock) (concrete roof: 2 ft minimum)	Concrete (walls, 5 ft,, roof, 2 ft)	Concrete (walls, 5 ft,, roof, 2 ft)
Containment	None	None	None	None	Not designed, but all cell entrances and ducts can be closed	Steel pressure vessel (leakage $< 0.3\%/24$ hr at 10 lb/in.^{2} pressure)
Dimensions (ft): length	12	30	25	10	27	27 dia.
breadth	12	30	15	10	17	—
height	30	22 (also 8 ft deep well in floor)	18.5 (also 7 ft high, 7 ft square extension above roof)	10	18	22.5 (also 10 ft deep, 12 ft square pit in floor)
Ventilation	None, apart from glove-box extract and general active area ventilation	Unfiltered extract; filtered and heated input	None (can be open to atmosphere)	Recirculation and low capacity extract through filter	Filtered extract; filtered and heated input (both ducts sealed on emergency trip)	*i*) Cell *open* - filtered extract to stack; heated and filtered input. *ii*) Cell *closed* - recirculation through filter and heater
Temperature control	Conventional	Conventional	None	Conventional	$\pm 2^\circ$ C (no cooling)	$\pm 2^\circ$ C (no cooling)
Access to cell: personnel	Removable labyrinth of concrete blocks	Normal doors	Steel door	Labyrinth and stairs (removable blocks)	Labyrinth (fixed)	Removable labyrinth in concrete and wicket door in pressure vessel
equipment	Rear side of glove-box (open)	Double doors	Double doors in thin front wall	By removal of concrete roof beams	By removal of concrete blocks in 12 × 8 ft hole in end wall	Steel plant door, 8 ft square, in pressure vessel wall
Ancillary services: solution storage	e.g. 600 litres (2 tanks)	None	600 litres (8 tanks)	150 litres (2 tanks)	Up to 100 litres (temporary only)	550 litres (7 tanks)
solution processing	Available on site	None	Two batch evaporators (4 litres/hr total)		None	Continuous evaporator (5 litres/hr)
Metal processing	—	Available on site	—	—	—	—
Moderated solid processing	—	Available on site	Recovery line for U/H mixtures		Storage only	Fabrication line for Pu/H tablets
Lifting capacity in cell	e.g. 5-ton gantry for cell erection	2.5-ton gantry; mobile hand hoist	2-ton monorail; 2 × 0.25 ton monorails	None	3-ton gantry	2-ton monorail (internal) 2-ton monorail (external)

reay. Work on solids other than metals including, in particular, mixtures of fissile material and moderator, is currently in progress at both Aldermaston and Dounreay.

The design of the facilities and the experimental procedures at the three sites have been somewhat different and they are described separately. The main features of the several facilities are summarized in Table I.

Harwell

No laboratory intended specifically for criticality studies was set up at Harwell. The experiments all employed aqueous solutions of the three fissile isotopes (Table II) and were carried out at intervals between 1952 and 1957 in a succession of temporary concrete cairns erected inside buildings used for chemical engineering research [1,3]. Each cell was relatively small in size (approx. 12 ft square) and contained only the critical assembly itself, the storage and dispensing equipment for the fuel solutions and all control and nuclear instrumentation being located in separate cubicles just outside the 3 ft thick cell walls. The cells were not contained in any way, though all were situated in active areas having filtered extract ventilation. It was a feature of the operational procedure at Harwell that no assembly was taken nearer to critical than 98 % or 99 % of the critical mass.

TABLE II. CRITICAL EXPERIMENTS CARRIED OUT AT HARWELL
(Systems of interest in the nuclear safety field)

Fissile isotope (X)	I	II	III	IV
	Pu 239	U 233	U 234 (45 %)	U 235 (45 %)
Moderator	Water	Water	Water	D_2O
Chemical form	Nitrate solution (acidity 1.7 N)	UO_2F_2	UO_2F_2	UO_2F_2
Core shape	Cylinder	Cylinder	Cylinder	Cylinder
Core diameter *(cm)*	30.5	30.5	30.5	71
Reflector: radial	Water	Water	Water	Graphite (46 cm)
axial	None	None	None	None
H/X ratio: min.	400	250	260	1940
max.	890	850	680	6720
Apparatus	ZETR. 0	ZETR. 1*a*	Zetr. 1*a*	HAZEL
Date	1952	1956	1955	1957

Aldermaston

At Aldermaston, various critical assembly tests and critical mass determinations have been made in a facility comprising a single large experimental cell operated from a control room 100 yards away [4]. The cell is of conventional brick construction and is surrounded by an exclusion area of 100 yards radius, from which all persons must be evacuated when a critical experiment is in progress.

The experimental cell is 30 ft square and 22 ft high and is thus large enough to house three critical assembly machines (ERIC, WATERFALL and ATLAS, Tables III and IV). It is a disavantage that only one of these can be used at one time, and also that setting-up work cannot be carried out on one machine while another is being used in an approach-to-critical experi-

ment. It is possible, however, to change over the control equipment and radiation detectors from one machine to another in a few minutes.

The cell is equipped with an air-conditioning system providing a heated and filtered air input. There is no extract filter. It is the practice at Aldermaston, as it was at Harwell, to stop each approach experiment slightly short of the critical condition. The final subcritical reactivity depends on the nature of the experiment and may be as little as a few cents with a well understood metal assembly of 'clean' geometry, but is more usually 2% or 3% of the critical mass.

The equipment of the laboratory is of a kind appropriate to solid criticality work. The cell itself contains a 2.5-ton gantry crane with electric hoist, a mobile hand crane, and a fume hood for building up small fuel sub-assemblies and for cleaning and monitoring fissile components. Adjacent to the cell are a change room, a small workshop and a plant room; a store for machine accessories, solid reflector materials and portable neutron sources lies a short distance from the cell. There are no facilities for solution handling nor for the storage of fissile solutions.

Dounreay

Construction of the Dounreay facility started in 1956 and the first of four experimental cells became operational in 1957 [5]. Two further cells, completing the part of the laboratory intended for uranium work, came into use during 1959. A fourth cell, designed primarily for experiments with plutonium-bearing materials, has recently been brought into use. A site plan of the area is shown in Figure 2 and a photograph of the facility in Figure 3.

The first cell resembles that at Aldermaston in being operated from a control room 100 yards away. There is, however, provision for some additional attenuation of neutrons by 18-in. thick concrete walls on three sides of the cell and by an earth bank on the fourth. The cell has no installed ventilation system and is uncontained. It is considerably smaller than that at Aldermaston but can accommodate two critical assembly rigs without undue difficulty. It was intended for studies on the criticality of uranium solutions and has been used for this work almost exclusively. A semi-permanent solution handling system is installed, with storage capacity for 600 litres of fuel solution in eight separate vessels; part of the latter is located in an annex which also serves as a change room. This cell shares with that at Aldermaston the disadvantage that only one assembly rig can be operated at one time, with the further restriction that fuel solution for a later experiment cannot be prepared while an earlier one is in progress.

Cell 2 consists of an underground pit in the form of a 10-ft cube, covered by a roof of removable concrete shielding blocks. The control room is at ground level and to the side of the cell roof, entrance to the pit being by way of a labyrinth in the concrete cairn and a ladder. The working volume is sufficient for only a single rig of limited size; the cell has therefore been used entirely for studies on uranium solution systems up to 16 in. in core diameter. The cell is uncontained, but is provided with filtered extract and recirculatory ventilation of low capacity. All solution handling and storage equipment is outside the pit and can be used whatever the state of the critical experiment. This feature, together with the proximity of the control room, have made for considerably greater speed of working than is possible in Cell 1. The proximity of the cell walls to the core of an assembly render this cell

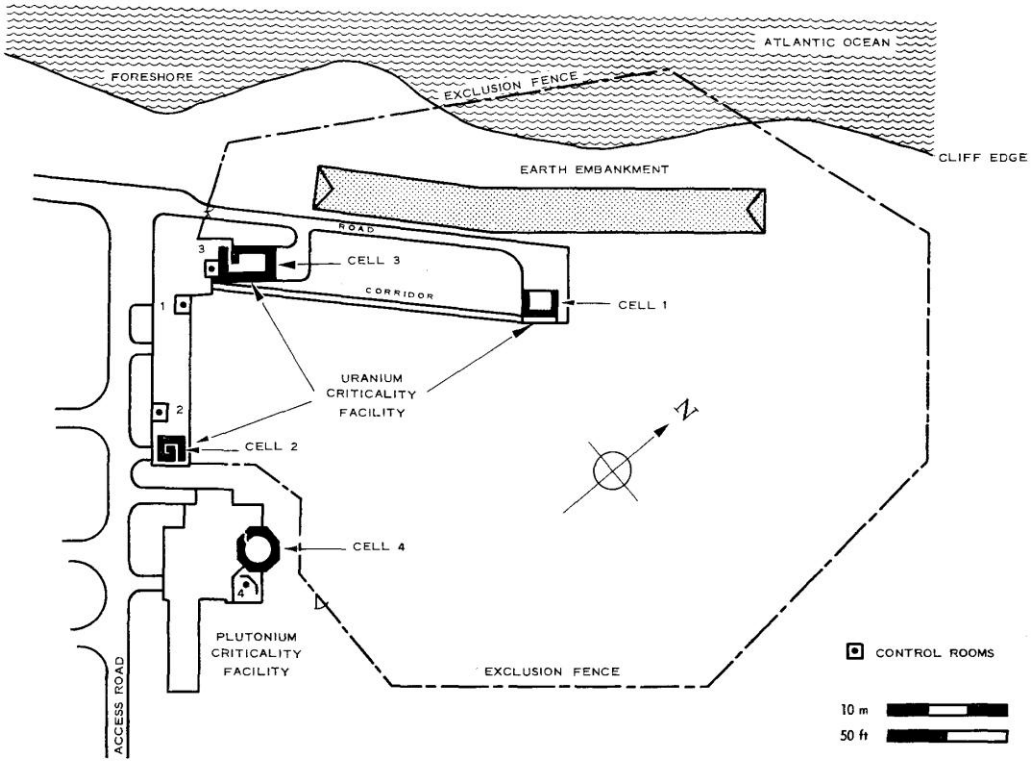

Figure 2. *Site plan of the Dounreay criticality laboratory.*

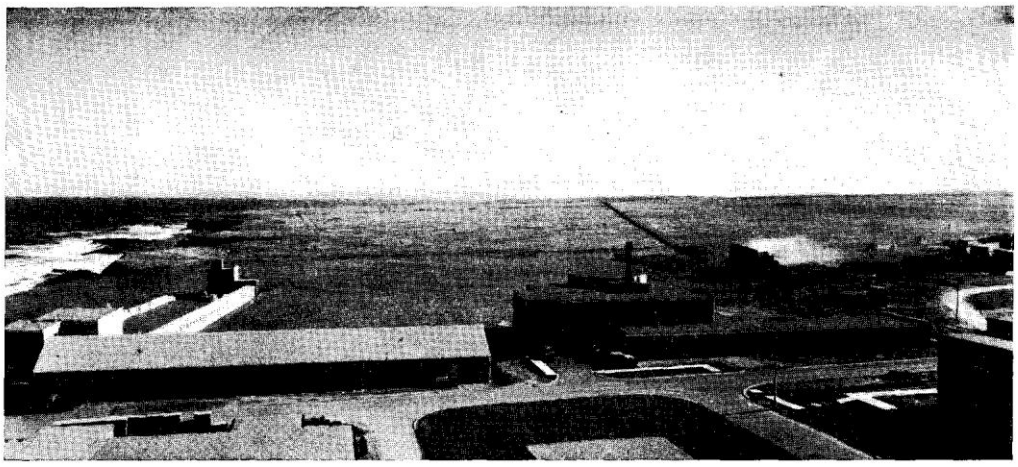

Figure 3. *The Dounreay criticality laboratory seen from the south-west.*

unsuitable for valid critical mass measurements on unreflected systems. Its small dimensions and the difficulty of access for large equipment have been a considerable inconvenience and have detracted from the safety of the work. For these reasons this cell, which was a temporary addition to the Dounreay resources, has recently been dismantled.

The third Dounreay cell is in the form of a concrete blockhouse, 27 × 17 ft in area and 18 ft high, with walls 5 ft thick and a roof 2 ft thick. The control room is situated alongside the cell, and access to the latter is by way of a labyrinth built into one of the concrete walls. In conformity with accepted United Kingdom philosophy, the cell shielding is designed to protect persons in the control room against the radiation from a critical incident yielding at least 10^{18} fissions and no exclusion area is required. The cell has its own separate input and extract ventilation systems, the former being heated; both are filtered and can be closed automatically by a trip signal. Although the cell cannot be sealed, its structure provides quite a high degree of containment of released activity.

Cell 3 was designed to house a single large assembly machine (TESSIE) for experiments on solid uranium systems up to twenty tons in weight. The equipment includes a three-ton gantry crane and a store for solid fuel units. Future experiments will employ uranium solutions contained in two or more interacting vessels (SIRIUS, see Table IV) and to provision these, two independent solution-handling systems with simple storage and glove box facilities have been installed.

The fourth Dounreay cell, intended specifically for work on plutonium systems, is considerably more elaborate than its predecessors and is housed in a separate building (Figure 4). It consists of a cylindrical steel shell of 27 ft dia. and 22.5 ft high, surrounded by a 5 ft thick concrete shield with a 2 ft thick roof. There is a steel-lined pit, 10 ft deep and 12 ft square, in the cell floor for housing dump tanks, control mechanisms, etc. An 8 ft square door in the steel shell, in line with a removable portion of the concrete shield, allows heavy plant to be brought into the cell; a smaller steel door and a labyrinth in the concrete are available for personnel access.

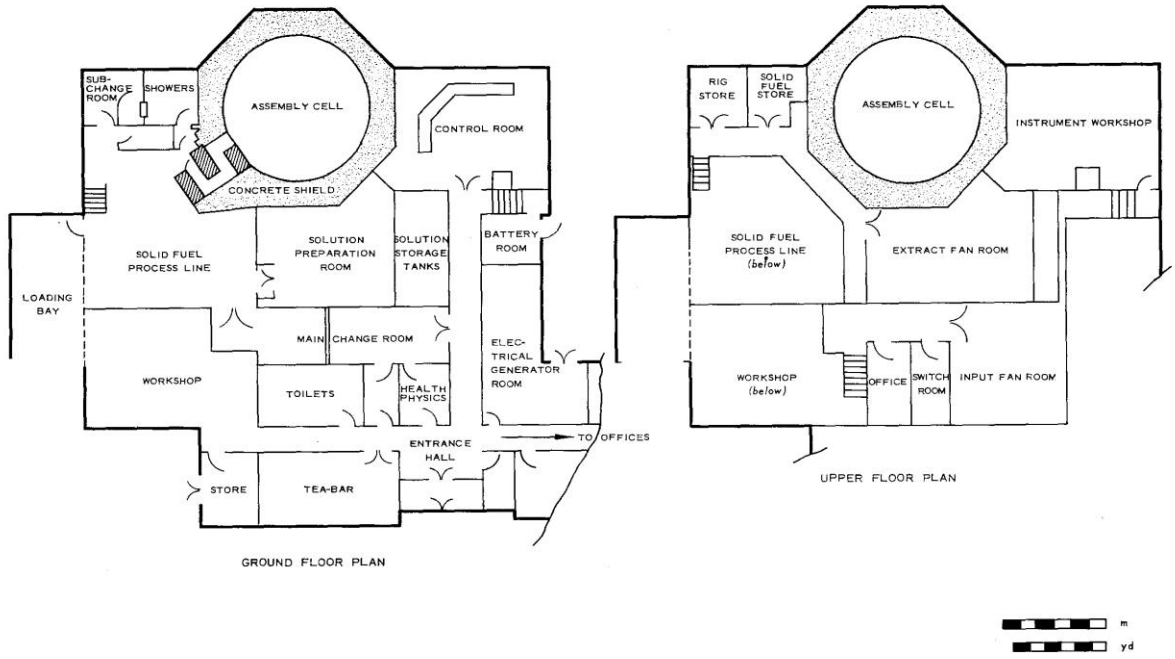

Figure 4. *The Dounreay plutonium criticality facility (Cell 4).*

The steel shell provides a high degree of containment of radioactivity (shown on test to be much better than the specified maximum permitted leakage of 0.3 % per 24 hours at 10 lb/in.^{2} excess pressure). To preserve this containment, all cables and pipes enter the cell through sealed service plugs; the larger and smaller doors are sealed and must be closed whenever reactivity is added to an assembly. No air lock is provided, the proposed procedure in the event of a critical incident being to keep the cell sealed until clean-up action can be taken by frog-suited workers. A sub-change room and control point, equipped with radio and television links to the cell, are provided for this purpose close to the cell entrance.

The cell is able to accommodate two critical assembly rigs, each housed within separate containment boxes having their own recirculatory ventilation systems. These are intended to minimise the spread of activity and to assist clean-up in the event of a critical incident. It is thought probable that the boxes prevent the general contamination of the cell unless the incident were unusually severe. The cell itself is equipped with a recirculatory ventilation system, comprising a filter and air heater, which serves both for temperature control (to within 2° C) and for clean-up. The main extract from the cell discharges through a filter to a stack and is operative only when the cell doors are open, the extract duct being sealed by a motor-driven valve during critical experiments.

Support facilities for the critical experiments are disposed as far as possible around the outside of the cell at two floor levels, with offices and general building services at a greater distance (Figure 4). The active facilities are on the side of the cell nearest its entrance and include two fuel processing laboratories, stores for solid and liquid fuels, the emergency change room already described and the extract fan room. These are separated by a fire-break wall from the inactive rooms, comprising the control room, an electronics laboratory which can be used to house the additional instruments needed in certain experiments, and a motor-generator plant giving stand-by electrical supplies from batteries. A main change room, health physics room, input fan room, small engineering workshop and staff accommodation complete the facility.

The uranium and plutonium criticality laboratories at Dounreay include limited provision for the preparation of fuels for use in the experiments, though not for their purification and recovery. Plutonium nitrate solutions are stored in seven slab-shaped tanks having a total capacity of 550 litres. These are independently valved, permitting solutions of several concentrations to be stored, but are all connected to a common mixing tank in which solution for a critical experiment is prepared. The latter is also connected to a continuous steam-heated evaporator in the form of a titanium cylinder, having a throughput of 5 litres/hr, and to two diluent tanks holding dilute nitric acid and condensate water. A solution of any desired concentration up to about 500 g Pu/litre can thus be prepared in the mixing tank, after which it is transferred to a holding tank which supplies the critical assembly apparatus, leaving the mixing tank and other vessels available to carry out further processing. Solution returned from an experiment is passed directly to the mixing tank and thence to one of the stock tanks. Mixing can be effected in all tanks by compressed air sparge and all tanks can be independently sampled for analysis. Level indication is by sight-glass. All transfers other than to and from the critical assembly are by the application of vacuum; transfer to the assembly

apparatus takes place in two stages, first through a service plug in the cell shield to a small intermediate tank, and thence to the core tank itself; both these transfers are by air-operated diaphragm pumps.

Equipment for the preparation of solid fuel tablets of plutonium and moderator is set up in a glove-box line close to the cell entrance. This includes balances and rotary mixers for preparing homogeneous dispersions of plutonium oxide and polyethylene powder, small hydraulic presses, and provisions for packing the pressed tablets in unsealed boxes and for the external decontamination of these boxes. Steam-heated and water-cooled dies of up to 2 in. square section have been used in the presses. Refabrication of the tablets by diluting with additional polythene is carried out in a further glove-box containing a heated extrusion press, granulator and grinder, for reducing the tablets successively to thin rods, chips and powder.

A uranium processing laboratory is available to serve all three uranium assembly cells, though it has no direct pipeline connexions to any of them. This laboratory contains two batch evaporators having a total throughput of 4 litres/hr and which are suitable for uranyl fluoride solutions of all enrichments. Facilities for the recovery of some residues, tissues, etc., are available and two glove-boxes are equipped for the conversion of solid mixtures of uranium tetrafluoride and paraffin wax (used as homogeneous briquettes in critical experiments at low H/U ratios) to uranyl nitrate solutions.

Equipment for Criticality Experiments

The critical assembly rigs in use in the United Kingdom can be divided into two main classes, according to whether the fuel and reflector materials are both solid or whether one at least is in liquid form. The two classes differ markedly in their design, experimental use and safety features and are described separately. Attempts have been made, however, to standardize as much of the equipment as possible, particularly that used for measurement and control, and this common equipment is considered first. The main characteristics and uses of the various rigs of both types are summarized in Tables III and IV.

General Design of Critical Assemblies

A critical assembly rig consists essentially of the following basic features:

a) A means of supporting or containing the fuel material and its reflector (if any);
b) Some means, closely controlled and readily reversible at all times, of increasing the reactivity of the system;
c) A means of measuring the ' dependent variable ' by which the reactivity is controlled;
d) Equipment for monitoring the reactivity of the system;
e) One or more shut-down devices, of the highest possible reliability, capable of reducing the reactivity into the subcritical regions in all eventualities.

Of these, items (*a*), (*b*), (*c*) and (*e*) are peculiar to each particular rig, though much can be done to standardize the control and measuring equipment and the system for actuating the shut-down devices. Item (*d*), the reactivity monitoring equipment, can be standardized to a very large extent and will be considered first.

The basic method, used with all the assemblies, of monitoring the reactivity during an approach to criticality is by the response of boron trifluoride-filled proportional counters to a neutron source placed within or close to the core. The BF_3 counter tubes are used either as Hanson and McKibben ' long ' counters or are inserted into the reflector of the assembly; counters are not normally used inside the core owing to the need to preserve the clean geometry of the latter. Pulses from these counters are fed via amplifiers to linear ratemeters equipped with adjustable trip relays (which actuate the shut-down devices) and also to scalers for the accurate measurement of the relative neutron multiplication during an approach to criticality.

A minimum of three BF_3 counting channels is provided in all experiments, partly to increase the reliability of trip actuation but more to avoid the need to interrupt an experiment should a channel become ' noisy ' or prove insensitive to the reactivity of the system. The use of several channels also improves the accuracy of the estimate of the critical size of a system which cannot be taken as far as the critical point. It has been found expedient at Dounreay to employ at least five BF_3 pulse channels, particularly when working with vessels of complex shape or when a high flux level is to be reached. In the latter circumstance it is often necessary to cut out one or more of the channels in the course of an experiment, though it is not the practice to use fewer than two BF_3 channels and an automatic shut-down is brought about if any attempt is made to cut out all of them.

Unless a system has a high internal production of neutrons, as from spontaneous fission or an (α, n) reaction, the presence of a neutron source is essential to the safety of an experiment. When the critical point is to be found by extrapolation from subcritical multiplications, a ' mock fission ' source is used in a fixed location as near as possible to the centre of the fully assembled system. The correction to be applied to the measured critical mass to take account of the source cavity must then be found by a subsidiary ' material replacement ' experiment. In experiments intended to attain criticality, the source must be removable (by remote control), though its position and emission spectrum are less important. At Dounreay, Po — Be sources of 10^7 n/s yield are used, the normal working position being about 5 cm outside the core surface.

Several methods have been used for withdrawing the source when near to the critical point, the two most convenient being a pneumatic tube and a system of cable control. Both methods are flexible and can be used to bring the source quite close to the core without impairing its geometry. The pneumatic tube is the more rapid and can easily be arranged to transport the source far from the assembly; the cable system is slower but more positive in operation and a special container, shielded by cadmium and wax, for example, must be provided to screen the source when it is withdrawn from the assembly. Whatever method is used, there must be a positive indication in the control room whenever the source is remote from the core. A continuous indication of source position is an advantage and can be provided by a synchro geared to the cable drive.

Additional radiation detectors are used at Dounreay with systems which are to be made critical [5]. These normally include two argon-filled ionization chambers (Type TPA) and one or more BF_3-filled chambers (Type RC-1). The former feed linear amplifiers provided with high and low current trips, and at least one of these channels must be operative at all times. The BF_3

chambers feed logarithmic amplifiers and period meters and three such channels are used in all experiments on plutonium assemblies; these are arranged to give an excess reactivity trip on a two-out-of-three basis when the doubling time falls below five seconds. Period meters are also occasionally used in conjunction with BF_3 pulse counters but have proved unsuitable for trip initiation. Fission counters, coated with U 235, U 238 and Pu 239, are used for special purposes, notably spectral index and fast fission factor measurements, but are not used for control of the assemblies.

In addition to the nuclear instrumentation, the control circuitry of the assembly machines has been standardized to a useful extent in the two laboratories. At Aldermaston a single control desk serves all three machines, a particular area of the desk being used for each one (Figure 5). Each machine also has its local control panel in the cell which can be used for setting-up purposes and which is rendered inoperative by a 'remote/local' key during active runs. At Dounreay, indentical relay boxes and 'jumper panels' of modular construction are installed as permanent features of each control room. The rig control panels on the control desk consist of nothing more than switches and indicator lights, all the required control operations, interlocks and sequencing being provided by cross-connections at the jumper panel. The system has proved remarkably versatile and trouble-free and all

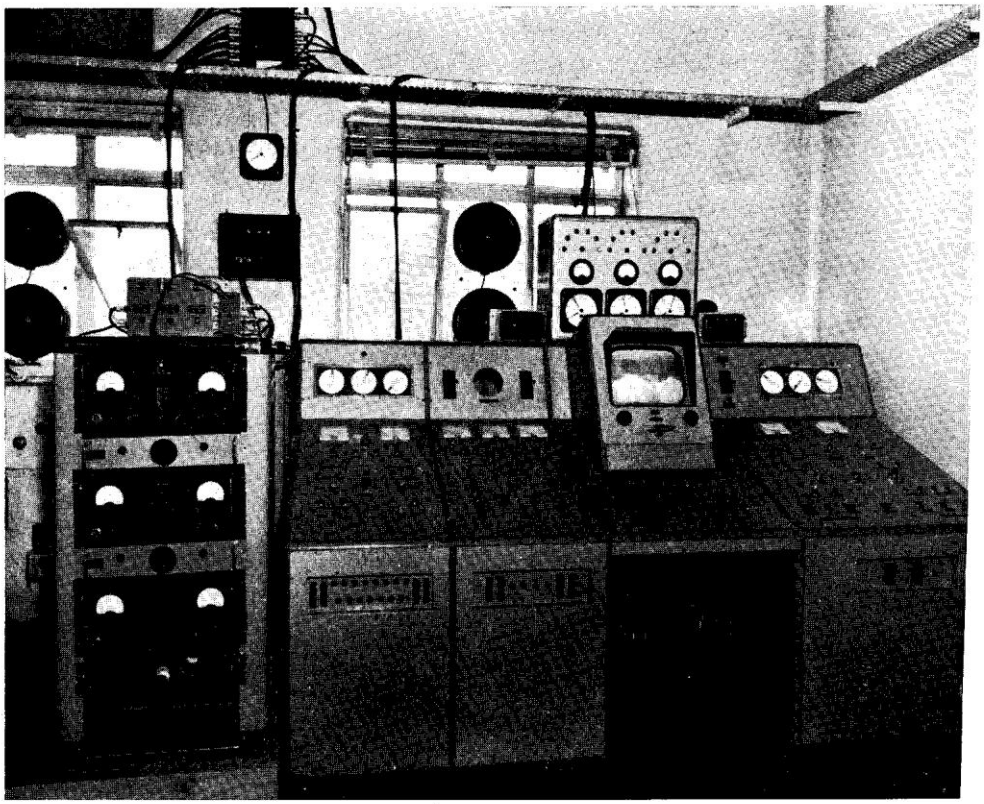

Figure 5. *Main control of the Aldermaston critical assembly laboratory.*

the control facilities for an entirely new experiment can be set up and tested in a few hours, while the diagnosis and cure of electrical faults has been much accelerated.

Machines for Experiments on Solid Systems

Four assembly machines are in current use for experiments on entirely solid systems; their principal features are given in Table III. Two of these, ERIC (Figure 6) and ATLAS (Figure 7), are at Aldermaston and have been in use in one form or another since 1952; the others, PUMA (Figure 8 and TESSIE (Figure 9), are at Dounreay and were installed in 1959 and 1960 respectively. All four machines are essentially similar in that, on each, an

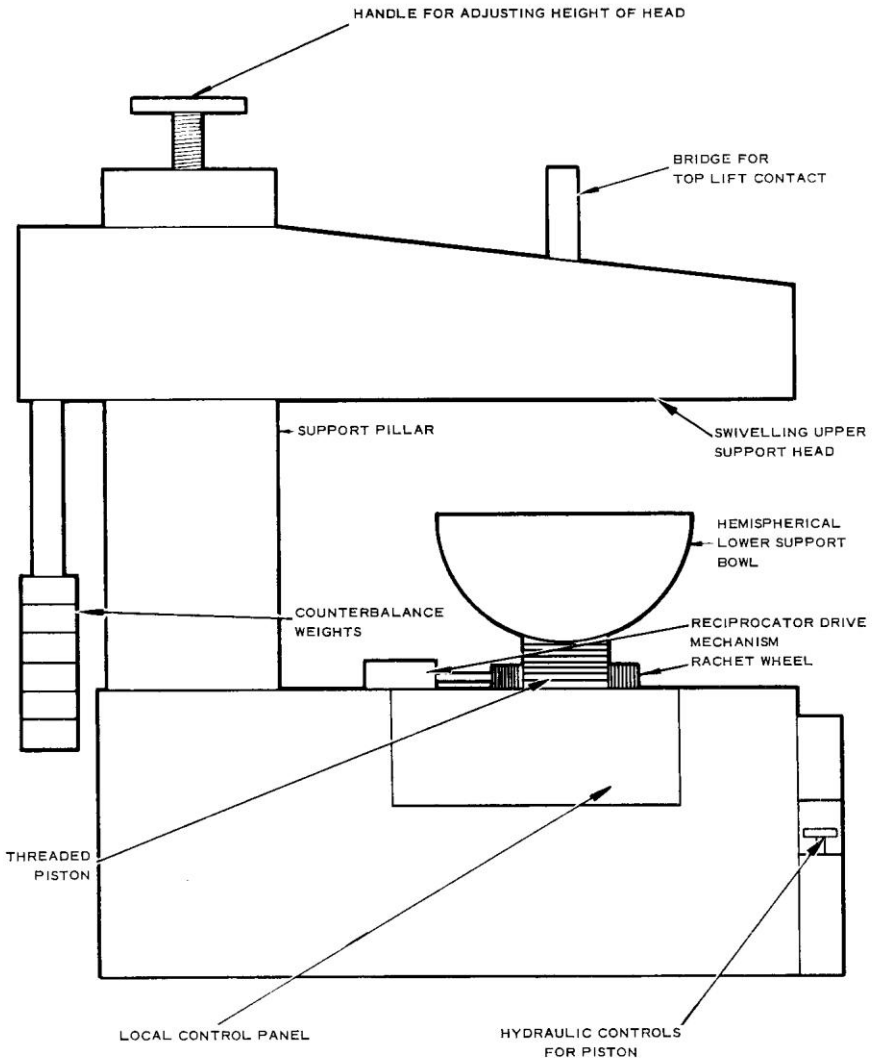

Figure 6*a*. *The vertical critical assembly machine ERIC (not to scale).*

TABLE III

Characteristics and uses of United Kingdom critical assembly machines:
MACHINES FOR ENTIRELY SOLID SYSTEMS

TABLE III. CHARACTERISTICS AND USES OF UNITED KINGDOM CRITICAL ASSEMBLY MACHINES
MACHINES FOR ENTIRELY SOLID SYSTEMS

Designation	ERIC	ATLAS	PUMA	TESSIE
Location	Aldermaston	Aldermaston	Dounreay (Cell 4)	Dounreay (Cell 3)
Date of first operation	1952	1952	Nov. 1960	Nov. 1959
Illustrations	Figure 6	Figure 7	Figure 8	Figures 9 and 15
Principal Characteristics:				
Basis of design	Vertical	Vertical	Horizontal	Horizontal
Basis of control	Movement of *lower* table	Movement of *lower* table	Movement of *both* tables	Movement of *both* tables
Basis of shut-down	Physical disassembly	Physical disassembly	Physical disassembly	Physical disassembly
Capacity: load (total)	0.25 ton	5 tons	1 ton	20 tons
size (total)	18 in. dia.	5 ft dia.	3 × 3 × 6 ft	6 × 6 × 10 ft
Max. separation of platforms	15 in.	68 in.	40 in.	74 in.
Control Mechanisms (moving table):				
Type of drive	Screw jack, with ratchet nut rotated by hydraulic reciprocator	Hydraulic ram, with forward motion limited by 3 lead screws driven by 'Varimag' motor	Lead screw, rotated by 1 of 3 constant speed electric motors	Lead-screw, rotated by 1 of 4 constant speed electric motors
Range of travel	15 in.	68 in.	30 in.	60 in.
Forward speeds (continuous): fast	0.4 in./min	4.5 in./min	3.0 in./min	17 in./min
intermediate (if any)	Continuously variable	Continuously variable	—	3.15 in./min
slow	Zero	0.075 in./min	*a*) 0.125 in./min *b*) 0.095 *c*) 0.040 *d*) 0.028	*a*) 0.63 in./min *b*) 0.38 *c*) 0.20 *d*) 0.12
Availability of forward speeds: fast			30 to 2 in.	60 to 20 in.
(for table separations) intermediate (if any)	All platform separations	All platform separations	—	60 to 4 in.
(adjustable settings) slow			All separations	All separations
Forward motion - fine adjustment	0.001 in./stroke	—	0.001 in./stroke	0.005/.0025 in./stroke
Reverse speed (continuous)	No steady speed	420 in./min	3.0 in./min	17 in./min
Reverse motion - fine adjustment	None	None	0.001 in./stroke	0.005/.0025 in./stroke
Control Mechanisms (fixed table):				
Type of drive	None	None	Pneumatic ram (oil-damped), hydraulic return buffer	Hydraulic ram, with hydraulic return buffer
Range of travel	Manual adjustment	Fixed platform	12 in.	14 in.
	—	—		
Shut-down Mechanisms:				
Number	One	One	Two	Two
Primary: type	(1) Release of lower platform, with initial free fall under gravity		Release of 'fixed' platform with initial acceleration of 0.1 g down 5° slope	
delay time after trip	135 ms	90 ms	150 ms	
free travel distance	3 in. (vertical)		2 in. (inclined)	4 in. (inclined)
retardation	Hydraulic damping over final 12 in. of travel	Hydraulic damping over final 15 in. of travel	Hydraulic damping over final 10 in. of travel	Gradual closure of oil return valve
Secondary: type	None	None	Reversal of motion of 'moving' platform, driven by *either* a.c. mains *or* d.c. battery supplies	
Measurement of Table Separation:				
Method of indication	Synchros (3) geared to screw-jack	Synchros (3) geared to lead screws	Metal scale and vernier, viewed by TV (12 in. lens)	Metal scale and vernier, viewed by TV (12 in. lens)
Error at small separations	0.003 in. max.		0.001 in.	0.002 in.
Reproducibility	0.001 in.	0.002 in.		
Typical Experimental Systems:				
Material and nature of core	Small assemblies of Pu or U 235 metal parts	Large U 235 metal assemblies, solid mixtures of enriched UO_2 and wax moderator	Solid mixtures of plutonium oxide and plastic moderator	Solid mixtures of low enrichment UF_4 and paraffin wax moderator (see also SIRIUS - Table IV)
Peak neutron flux $(n/cm^2/s)$	10^4	10^5	10^7	10^8
Max. thermal power	~ 0.1 W	~ 0.1 W	10 W	5 kW

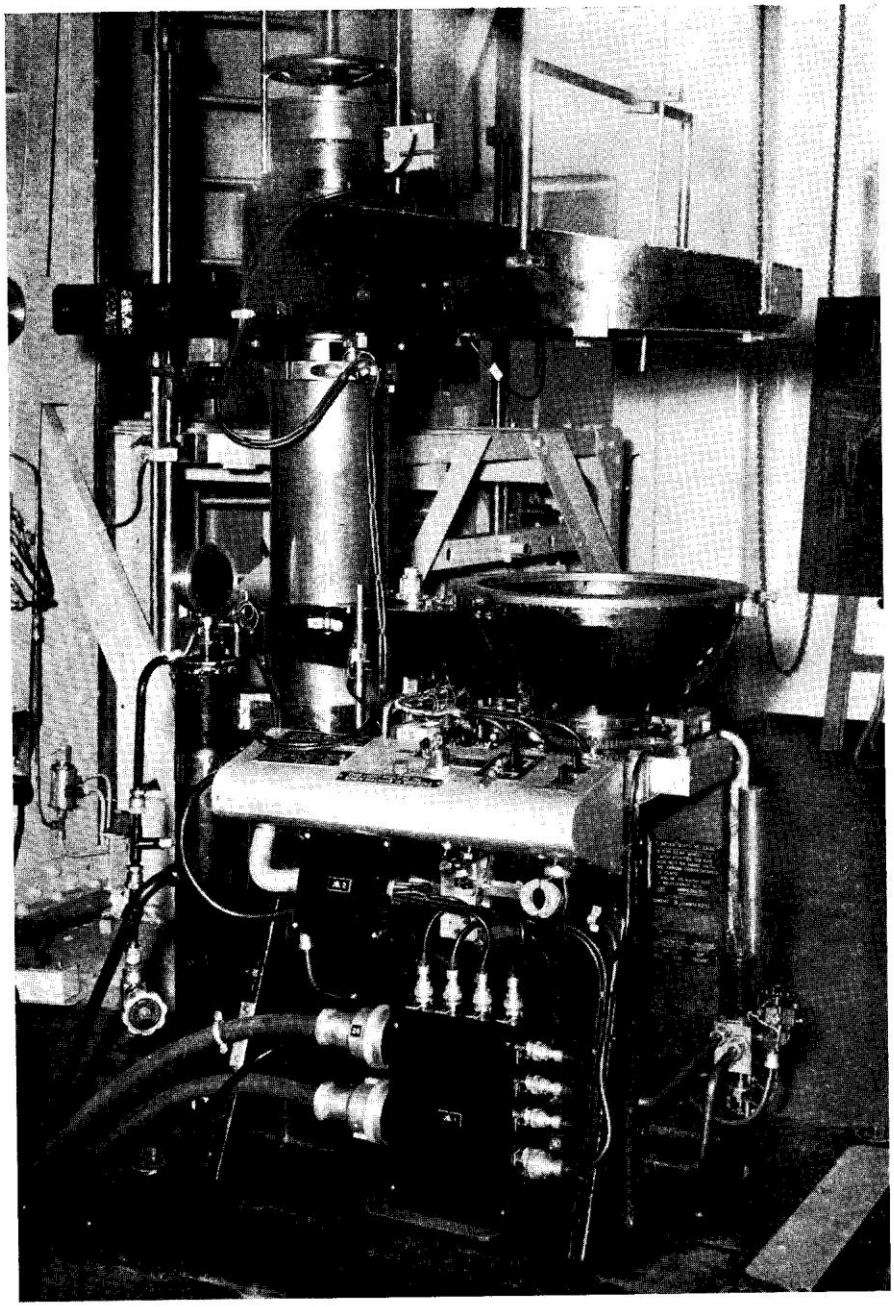

Figure 6*b*. *The vertical critical assembly machine ERIC.*

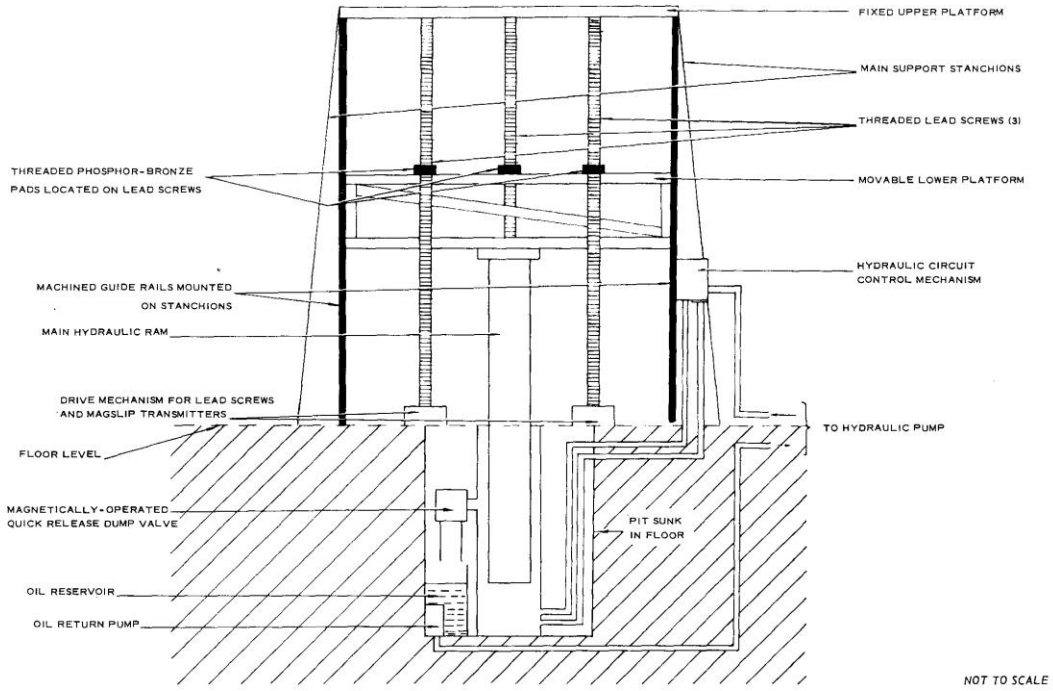

Figure 7*a*. *The ATLAS high capacity vertical assembly machine. (side elevation).*

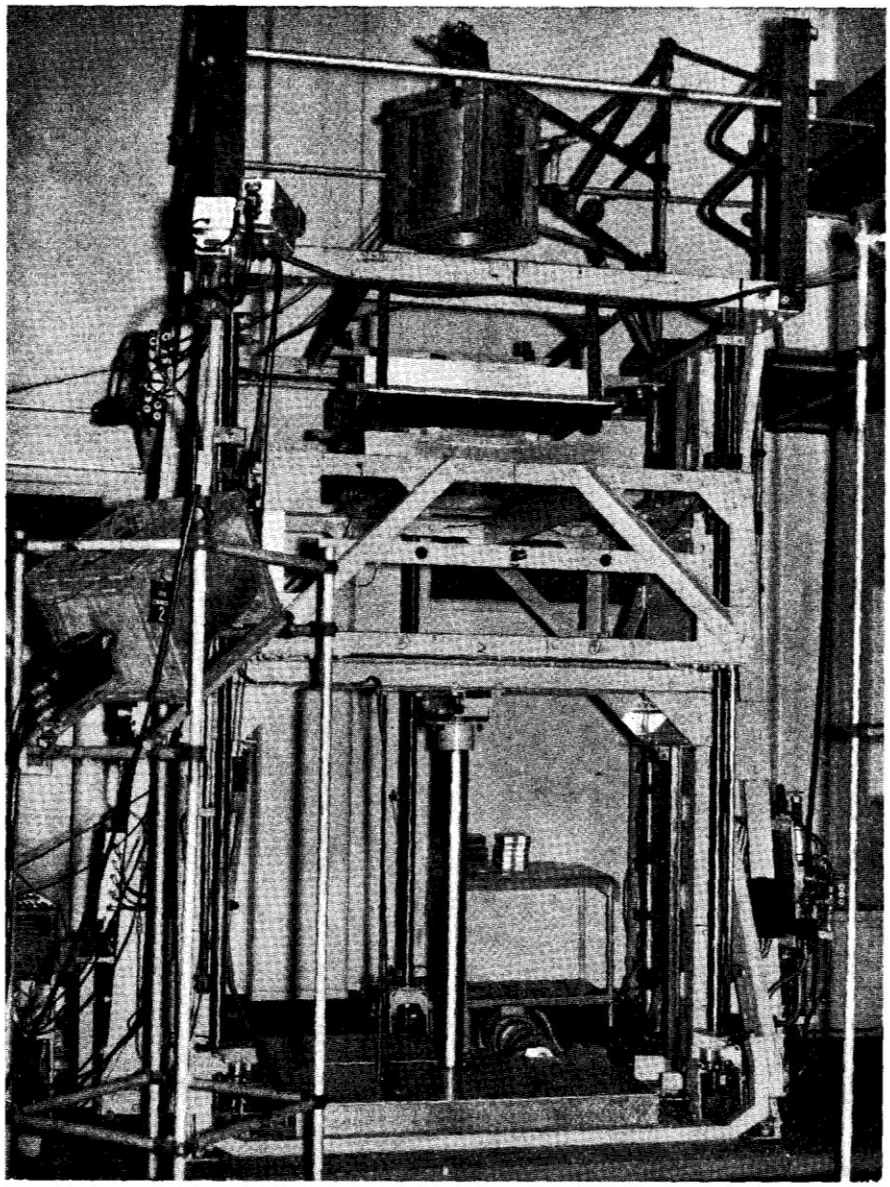

Figure 7*b*. *The ATLAS high capacity vertical assembly machine.*

assembly is first built by hand in two parts, one of which is then driven towards the other under remote control. No control or safety rods are employed with any of them, safety being dependent primarily on the limited speed of approach of one part of the assembly to the other and on the rapid separation of the two parts in the event of a trip. Only a single shut-down mechanism is provided on each: the rapid separation of the two parts under gravity.

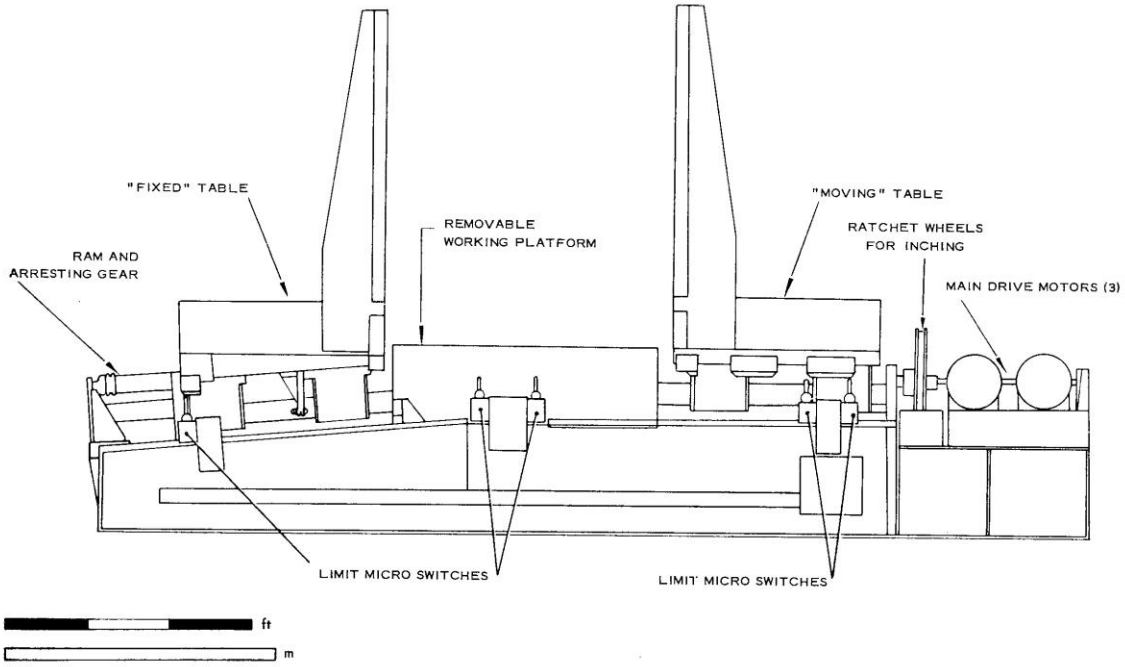

Figure 8. *Elevation of PUMA assembly machine, showing tables in retracted positions.*

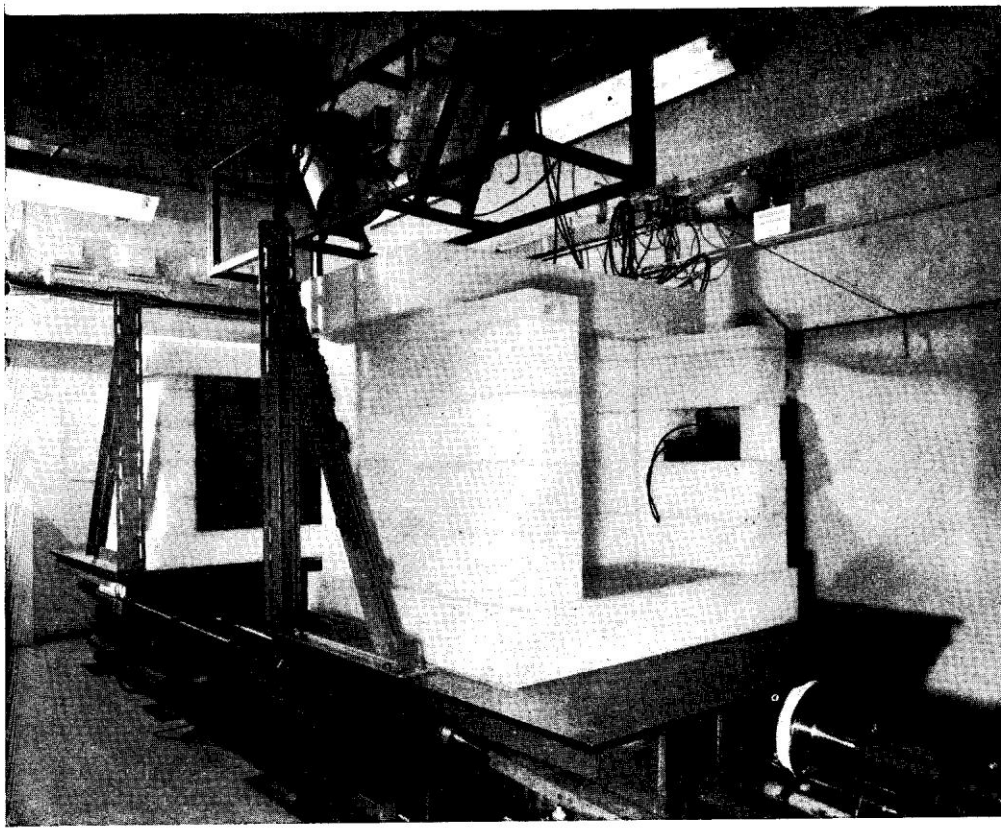

Figure 9. *The TESSIE horizontal assembly machine, showing a cubical core of low enrichment uranium tetrafluoride and paraffin wax mixture with thick polyethylene reflector.*

The two Aldermaston machines are characterized by the movement of a lower platform vertically upwards towards a fixed upper one, the lower platform being able to fall away freely under gravity in the event of a trip. In the Dounreay machines assembly takes place in a horizontal direction and both platforms can move, though the movement of one serves only to disassemble the system on trip. The two types of machine are largely complementary in function and it is an advantage to be able to select whichever is most appropriate for a particular experiment.

The vertical machines are especially suitable for high density systems where external clamping arrangements are undesirable or are difficult to provide; they have the merit of the very effective shut-down action which the free fall of the lower platform makes possible. It is fairly easy to ensure accurate mating and alignment of the two halves. The symmetry about a vertical axis is convenient in work on heterogeneous systems and it is somewhat easier than the horizontal arrangement for demonstrating the safety of a subsequent stage in an experiment. It is also of great value in some interaction studies on mixed systems where a liquid reflector or a tank of fissile solution is to be brought towards a solid core or reflector. The vertical arrangement

has the disadvantage that the upper part of all but the smallest assemblies requires to be supported by a membrane of foreign material, the effect of which must be found in a tedious and sometimes unreliable subsidiary experiment. Care must be taken whenever additions or alterations are made to the upper part of the assembly, owing to the risk of fuel or reflector material falling onto the lower part.

The horizontal design of the machine eliminates the need for any supporting plate inside the core, making it possible to measure the critical sizes of cores in the absence of internal perturbations. Its main disadvantage is the difficulty of providing a rapid and fail-safe disassembly mechanism. On both the Dounreay machines, one of the tables is mounted on a sloping track, down which the table slides with an acceleration of about 0.1 g in the event of a trip; although reliable, this arrangement does not provide as rapid a reduction in reactivity as might be desired. More rapid separation can be achieved by some form of energy storage such as springs, suspended weights or compressed air cylinders, but only at the cost of securely clamping the assembly to the table. Even in the sloping track design, external clamps are often needed to maintain the stability of unreflected assemblies and allowance has to be made for unwanted neutron reflection from the clamping structure. Provision has to be made, also, to absorb the kinetic energy of the moving platform after disassembly. The horizontal design has proved convenient for experiments requiring frequent changes to the core geometry and where ready access to the core centre is needed for flux distribution and spectrum measurements, material replacement tests, etc; it is less satisfactory for work with liquid reflectors or where the entire reflector has to be varied in nature or thickness, owing to the difficulty of access to the underside of the core. A high load capacity is more easily achieved with the horizontal design.

Summarizing, the horizontal type of machine is thought to be somewhat more versatile and convenient in operation than the vertical arrangement. It would probably be slower in regaining a subcritical condition in the event of a reactivity burst, but does not demand such close administrative control of loading or core changes during normal use. The critical parameters of cores unperturbed by the presence of foreign materials or voids can be measured directly on a horizontal machine; experiments on bare cores free from unwanted neutron reflection are more readily performed in a vertical arrangement.

The smaller vertical machine, ERIC (Figure 6), is used almost exclusively for experiments on small metal assemblies, usually of high density and simple shape. It is suitable for systems, either bare or reflected, up to 18 in. dia. and 500 lb weight. The upper part of a proposed assembly is attached to a light alloy frame, adjustable in height but fixed during a critical approach. This frame has a limited amount of side float and is equipped with levelling screws to ensure correct mating, without risk of jamming, of components during the final stages of assembly. A counterbalance weight is used to reduce distortion of the machine structure when carrying heavy loads on the upper platform. The lower section is carried on a screw-jack capable of very slow and precise upward movement, the screw being raised or lowered by a ratchet nut, itself turned by a hydraulic reciprocator which can be operated either in single strokes (giving a movement of the load of 0.001 in.) or continuously at speeds up to 400 strokes/min (0.4 in./min). The precision of the screw and nut is such that the actual position of the platform lies within 0.0008 in. of the calculated position over the whole 15 in. range of travel and is repro-

ducible to within 0.0001 in. The separation of the tables is displayed, both on the machine and in the control room, by synchros geared to the screw-jack and having a maximum following error of 0.001 in. Transmitter-receiver synchronization is checked by electrical contacts on the screw-jack at 0.1-in. intervals. Rapid fall of the lower platform on trip is achieved by disengaging the nut and screw from the driving mechanism; the release action is fail-safe and is assisted by the weight of the load; the total release time is about 135 ms, after which the platform falls freely for 3 in. and is subsequently brought gradually to rest over the remaining 12 in. by a hydraulic damper.

The larger vertical machine, ATLAS (Figure 7), has a load capacity of 2.5 tons per platform and can accept assemblies up to about 5 ft dia. The main structure consists of two substantial 10 ft high stanchions carrying the fixed upper platform. The lower platform travels vertically within guides on the main stanchions and is located by roller bearings over its full travel of 68 in., its weight and that of the load being supported by a 4 in. dia. hydraulic ram in a well below the machine. Precise control of the movement of the lower platform is provided by three vertical lead screws, driven simultaneously by a 'Varimag' electric motor and carrying threaded pads to which limit switches are fixed. The platform can rise freely until it reaches the pads, after which contact between them is maintained automatically throughout the upward movement. The speed of vertical movement can be varied continuously from 0.075 to 4.5 in./min. The position of the three pads, and hence of the platform, is displayed by synchros geared to the lead screws and readable to 0.002 in. ; the position of the platform when not in contact with the pads is adjustable, by control of the oil flow to the ram, to better than 0.1 in. On trip, a quick-release dump valve in the hydraulic circuit allows the platform to fall at about 7 in./s, until it is brought smoothly to rest over the last 15 in. of travel by the gradual closure of the oil dump valve.

The two horizontal machines, TESSIE and PUMA, are similar in design, differing mainly in size and hence in their load capacity and fineness of control. The smaller of the two, PUMA, is shown in Figure 8 and is intended primarily for experiments on solid moderated systems of plutonium or highly-enriched uranium. It has a load-bearing capacity, without more than 0.005 in. distortion of the structure, of 500 lb per table and can accept assemblies having dimensions up to $3 \times 3 \times 6$ ft overall, including reflector. The two tables are each carried on four recirculating ball bushings running on two parallel round bars, an arrangement giving extremely free movement. The bars supporting one table are inclined at an angle of 5° to the horizontal. This table is pushed by a pneumatic piston through a distance of 12 in. to its normal operating position at the top of the 5° slope, where it is held by an electromagnet. The other table is driven horizontally towards the other from its maximum distance away of 30 in., first at a 'fast' speed of 3 in./min and later at one of four pre-selected 'slow' speeds within the range 0.028 — 0.12 in./min; a pre-set electrical cut-out, adjustable in position, prevents use of the fast speed at separations smaller than a few inches. Fine adjustment of the table position can also be made by an inching mechanism, consisting of a pneumatically driven pawl which engages a ratchet wheel on the lead screw; two such mechanisms are provided, allowing the table to be either advanced or retracted by 0.001 in./stroke. The separation of the two tables is indicated directly by a metal scale and vernier, visible on the machine itself and also in the control room over a closed circuit television link. The separa-

tion can be read, using a 12-in. camera lens, to within 0.001 in. and is reproducible to this accuracy up to separations of several inches.

Rapid disassembly of PUMA, on receipt of a trip signal, is brought about by the movement of both tables. The normally fixed table is released by its magnet after about 150 ms from trip initiation, accelerates freely under a 10 % component of gravity up to a separation of 2 in. and is subsequently brought smoothly to rest by a hydraulic buffer after travelling its full distance within 6 s. The reverse movement of the other table (at 3 in./min) is scarcely less reliable, although dependent on electrical supplies; it occurs provided that *either* the 240 V a.c. mains *or* a 24 V d.c. battery supply to an additional motor is maintained.

The TESSIE horizontal assembly machine (Figure 9) is suitable for systems up to 20 tons in total weight and several cubic yards in volume. Its general structure and control mechanisms resemble those of PUMA very closely. The fixed table is raised by a hydraulic, rather than a pneumatic, piston and is arrested after a trip by the gradual closure of a throttling valve in the oil circuit. The travel of the moving table is 5 ft, and three forward speeds are provided for its control during an approach to criticality. Typical ranges of availability of the speeds are: 17 in./min from 60 to 20 in.; 3.15 in./min from 60 to ; 4 in. and 0.63 in./min (or 0.12 in./min) from 60 in. to zero separation. Forward and reverse inching in steps of 0.005 in. is also provided. The TESSIE machine is used for measurements on homogeneous stacks of low enrichment uranium and moderator mixtures or other large dilute systems, for *ad hoc* experiments on processing plant vessels and for carrying subsidiary experimental rigs used in interaction studies, such as the SIRIUS apparatus described below.

Equipment for Liquid Critical Assembly Experiments

Critical assemblies in which either or both of the core and reflector are in liquid form have the advantage that the liquid level can be used to provide very sensitive control of reactivity. By the same token, dumping the liquid under gravity — especially when this is the core material — can be made to serve as a convenient and highly effective shut-down device. On the other hand, systems using liquid cores are in two respects considerably more troublesome to operate than solid systems: a fairly elaborate solution storage and handling system, preferably including facilities for dilution and evaporation, is needed to supply the fissile core solutions and, where plutonium solutions are concerned, must be provided as a permanent installation; and the need to break fuel lines when changing core vessels retards working ow ng to the need to follow active handling procedures.

It is a relatively simple matter to vary the moderation of a solution-cored system, at least within the range of chemical solubility; it is very much less easy to change the size of core vessel. In solid systems the reverse obtains, the re-assembly of core components into different core shapes being straightforward, whereas a homogeneous change of moderation necessitates the reprocessing of the entire core. In solution systems, the fuel density is related to the moderation in a definite manner and only elementary precautions need be taken to eliminate voids and maintain homogeneity; in contrast the achievement of densities close to theoretical in solid homogeneous mixtures of fuel and moderator is often difficult. A supercritical excursion in a solution system is demonstrably self-limiting by virtue of the void coefficient of reactivity

for radiolytic gas formation; this is not necessarily so in a moderated solid assembly unless it is initially free from built-in voids.

A number of rigs has been in use at Harwell and Dounreay since 1952 for studies on entirely liquid systems, details of the majority of which are given in Table IV. Three such rigs — PANTHER, PHOENIX and TOAD — are in current operation at Dounreay and are described in this report. Two additional machines (WATERFALL and SIRIUS) are in use for experiments with part-liquid, part-solid systems, and a design study has been completed for a third (SORCERER).

The WATERFALL machine (Figures 10 and 11) installed at Aldermaston, is used to surround small solid cores, usually of metal, by liquid reflectors up to 24 in. dia. The core, previously shown to be subcritical and safe to handle by an experiment on an approach machine, is supported on a light aluminium tube at the centre of a spherical copper vessel. Reflector liquid is pumped from a reservoir into this sphere by a piston pump driven by a second, double-acting piston actuated by oil pressure; the stroke and speed of the latter can be varied so as to insert ' shots ' of reflector liquid between 1.6 and 60 cm^3 in volume either singly or continuously at speeds from 30 to 200/min. An additional valve allows liquid to be withdrawn from the reflector vessel at the same rates. A trip signal interrupts the power supply to the pump and to a magnetic valve in a large diameter dump pipe at the base of the

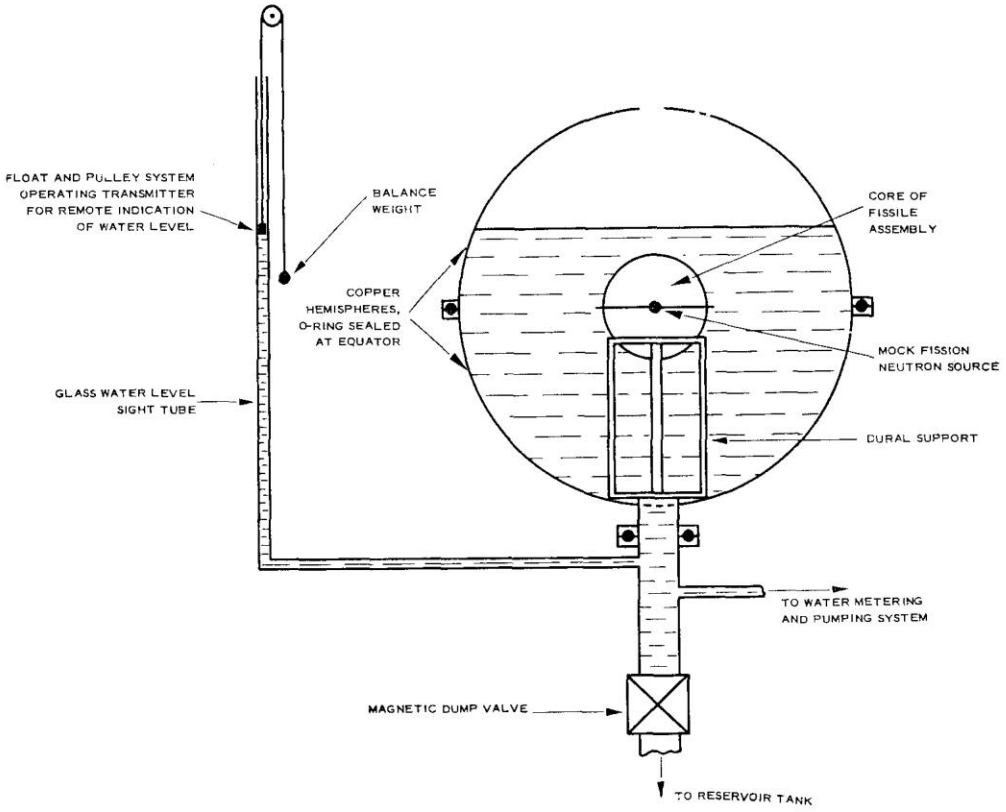

Figure 10. *The WATERFALL critical assembly equipment (vertical section).*

TABLE IV

Characteristics and uses of United Kingdom critical assembly machines: EQUIPMENT FOR PARTLY OR WHOLLY LIQUID SYSTEMS

TABLE IV. CHARACTERISTICS AND USES OF UNITED KINGDOM CRITICAL ASSEMBLY MACHINES
EQUIPMENT FOR PARTLY OR WHOLLY LIQUID SYSTEMS

Designation (with earlier designation)	WATERFALL	ZETR-0 (Pu ZETR)	ZETR-*1a*	ZETR-*1b/c*	HAZEL (ZETR-II)	PANTHER	TOAD (ZETR-III)	PHŒNIX (HAZEL-II)	SIRIUS	SORCERER
Location	Aldermaston	Harwell	Harwell	Dounreay (Cells 1 and 2)	Harwell	Dounreay (Cell 4)	Dounreay (Cell 1)	Dounreay (Cell 1)	Dounreay (Cell 3)	(for Dounreay Cell 1)
Date of first operation	ca. 1954	1952	1955	Aug. 1957	April 1958	1961	1961	July 1960	May 1961	(Design study: 1961)
Date dismantled	—	ca. 1953	1956	Aug. 1960	Sept. 1958	—	—	—	—	—
Illustrations	Figures 10 and 11	Not shown	Not shown	Not shown	Not shown	Figure 12	Not shown	Figure 13	Figure 14	Not shown
Principal Characteristics:										
Core: isotope	U 235 or Pu	Pu	U 233/U 235	U 235	U 235	Pu	U 235	U 235	U 235	U 235
enrichment (%)	Various	—	45	30 or 93	45	—	30	30 or 93	30 or 93	30 or 93
moderator	None	H_2O	H_2O	H_2O or TEP,OK	H_2O	H_2O	H_2O	H_2O	H_2O	H_2O
phys. form	Metal	Solution	Solution	Solution or complex	Solution	Solution	Solution	Solution	Solution	Solution
chem. form	Metal	Nitrate	UO_2F_2	Nitrate or UO_2F_2	UO_2F_2	Nitrate	UO_2F_2	UO_2F_2	UO_2F_2	UO_2F_2
shape	Sphere (eg)	Cylinder	Cylinder	Cylinder (1 or 2-zone)	Cylinder	Cyl./sphere	Cyl./sphere (1,2 or 3-zone)	Cyl./sphere	2 × Slabs (interacting)	Slab
Reflector: material	Water/oil	Water	Water	Water/bare	Graphite	Water/bare	Water/bare	Water/bare	Polythene	Various
coverage	Complete	Radial	Radial	Radial	Radial	Complete/side	Complete/side	Complete/side	All faces	1 or 2 faces
thickness	Inf.	Inf.	Inf.	Inf.	47 cm	Inf.	Inf./various	Inf.	Inf.	Various
Core vessels: material	—	S/steel	S/steel	S/steel	S/steel	Al	Al	S/steel or Al	S/steel	Al
Peak flux (n/cm²/s)	10^4	10^4	10^5	10^6	10^5	10^8	10^8	10^7	10^7	10^7
Max. thermal power (W)	—	—	—	0.1	1	10	10	100	100	100
Control Mechanisms:										
Core	(Fixed)	Suction	Suction	Suction/Pump	Pump	Pump	Pump	Pump	Pump	Pump
Reflector	Pump	Pump	Pump	Pump	(Fixed)	Pump	Pump	Pump	(Fixed)	(Fixed)
Other	—	—	—	Reflector shield	Reflector shield	—	—	—	Table separation	—
Shut-down Mechanisms:										
Number	One	Two	Two	1 or 3	Two	2 or 3	2, 3 or 4	2 or 3	2 per tank	1 or 2
Primary	Reflector dump	Reflector shield	Reflector shield	Core dump (Reflector shield)	Core rods (2)	Core dump (Reflector dump)	Core dump (Reflector dump)	Core dump (Reflector dump)	Core dump (2)	Core dump
Secondary	—	Reflector dump	Reflector dump	(Reflector dump)	Reflector shield	(Core pump-out)	(Buffer dump; core pump-out)	(Core pump-out)	Table drop	Reflector drop
Other	—	—	—		—				Table drive	—
Range of Core Vessels:										
Close gaps: Spheres - Number	(Reflector:	—	—	—	—	10	6	1	—	—
Spheres - Diameters (in.)	24 in. dia.)					9 to 38	9 to 22	38		
Close gaps: Iso. Cyls. - Number		—	—	—	—	3	2	—	—	—
Iso. Cyls. - Diameters (in.)						8.5 to 12	10 and 12.5			
Close gaps: Tall Cyls. - Number		1	2	4	1	5	6	6	—	—
Tall Cyls. - Diameters (in.)		12	5 and 12	5, 8, 12, 16, 36	24	5.5 to 9	7 to 10	7.5 to 40		
Tall Cyls. - Max. Ht. (in.)		24	36	36	84	60	30	108		
Close gaps: Squat Cyls. (slabs) - Number		—	—	—	—	4	Plus 6 thin reflector tanks; plus 7 buffer and specimen cores	—	4 × 2 slabs	1 slab
Squat Cyls. (slabs) - Depths (in.)						2.5 to 5			1.5 to 4	Up to 18
Squat Cyls. (slabs) - Area (in.)						30 in. dia.			48 in. square	48 in. square

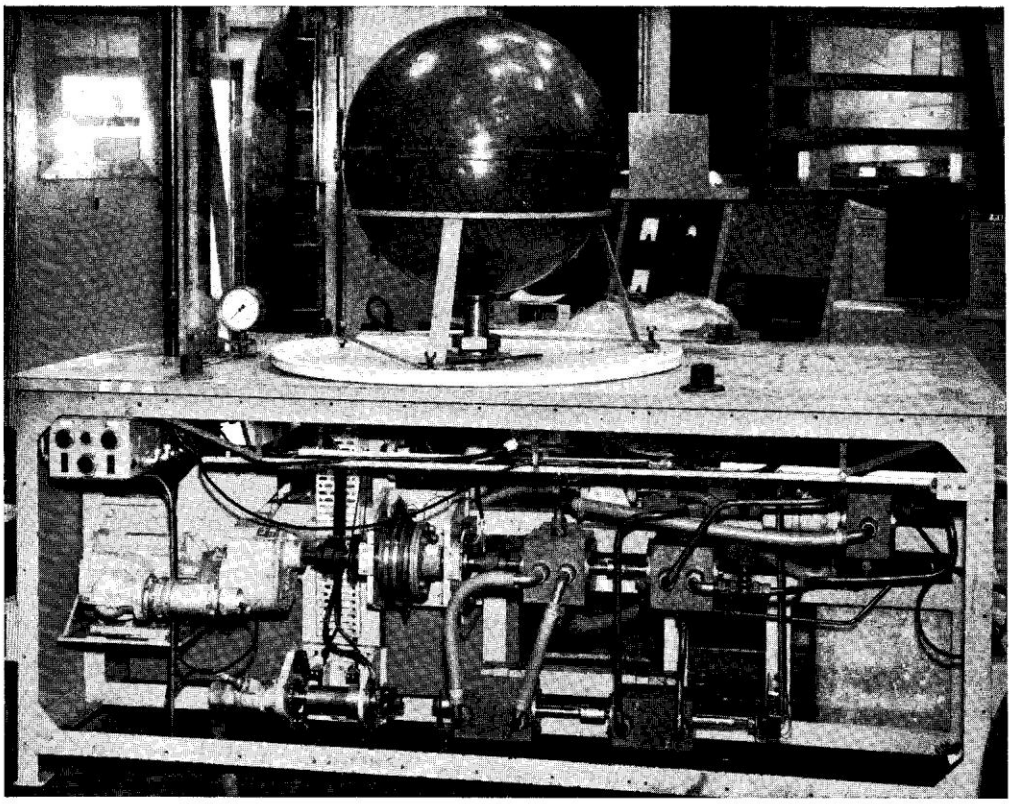

Figure 11. *The WATERFALL critical assembly equipment, viewed from the side.*

reflector tank. The reflector level is indicated locally by a vertical sight glass alongside the machine; a float in this tube, joined to a counterweight by a silk thread passing over the drive pulley of a synchro, transmits this level to the control room with a reproducibility better than 0.02 in. over the full range of 26 in.

The three Dounreay solution rigs are all basically similar, differing mainly in the size range of their core tanks and in the degree of containment. All three are more versatile and more rapid in operation than any of their predecessors. The most recently constructed of these, PANTHER (Figure 12), has been designed to measure the critical parameters of plutonium nitrate solutions, with or without a water reflector, over the whole accessible range of concentrations. It consists essentially of four units: a core tank, interchangeable with others in a comprehensive range of shapes and sizes; a removable cylindrical reflector tank surrounding the core tank; two dump tanks, one for fuel solutions from the core and one for reflector liquid. The whole apparatus, with the exception of the reflector dump tank, is contained in a transparent glove-box equipped with tent flanges to enable personnel to carry out core tank changes and general maintenance. The range of interchangeable core tanks includes ten spheres (9 to 38 in. dia.), three isometric cylinders

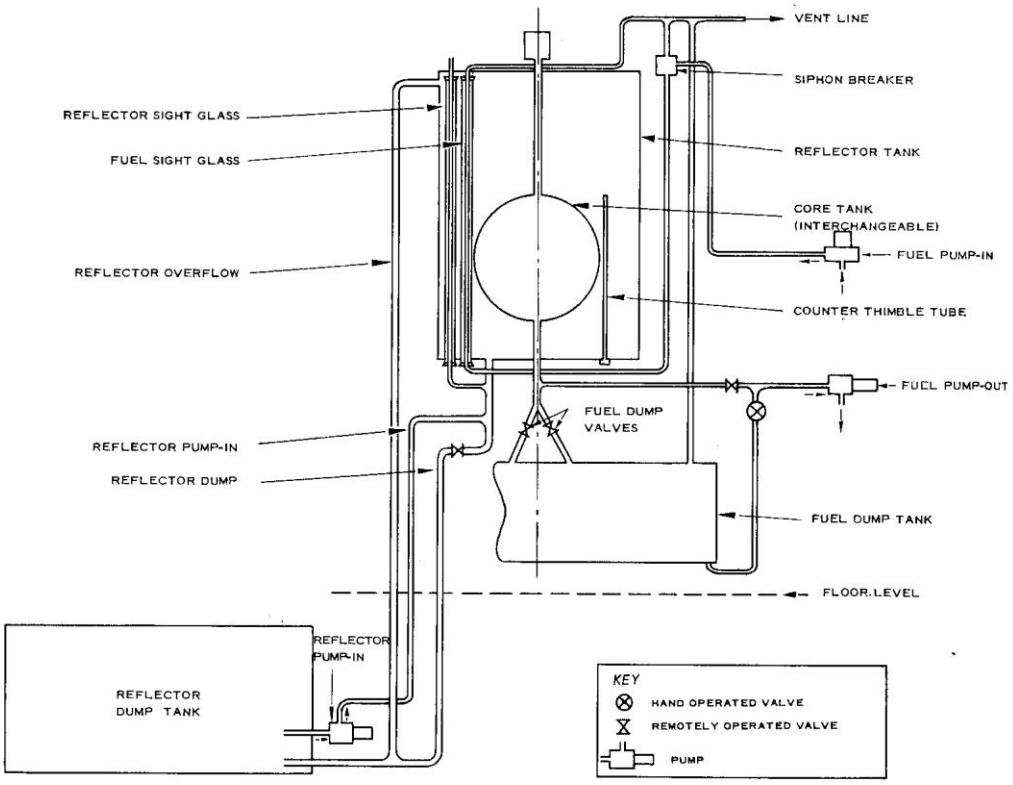

Figure 12. *PANTHER. A critical assembly apparatus for plutonium solutions.*

(8.5 to 12 in. dia.), and four shallow cylinders (30 in. dia. and 2.5 to 5 in. deep), all of which can be fully reflected by water if required. There are, in addition, five tall cylinders (60 in. high and 5.5 to 9 in. dia.) which can be reflected in a radial direction only. The core tanks are of 0.064 in. thick aluminium, protected against attack by the fuel solutions by an epoxy resin lacquer. A vertical pipe at the base of the core tank serves as the normal filling and emptying line and also as a fuel dump pipe through which a portion of the core solution can be rapidly discharged to a geometrically safe vessel in the event of a trip; a similar pipe at the top of the core tank serves as a pressure balance line.

Plutonium nitrate solution is supplied to the PANTHER rig from the holding tank of the fuel storage system previously described, in two stages: in the first, a predetermined volume of solution is pumped quite rapidly through the wall of the cell into a 20-litre intermediate vessel inside the cell; in the second stage, this solution is transferred at a predetermined rate into the core tank itself. The two-stage procedure eliminates the need for any direct connection between an ' unsafe ' vessel (the core tank) and a virtually unlimited source of fuel, thereby providing protection against over-running of the pump; it also enables the cell to be completely sealed whenever reactivity is added to the system. The return of solutions from the core tank to storage is effec-

ted in a single operation. Reflector water is pumped, when required, from a dump tank in a pit below the rig into a large cylindrical vessel surrounding the core. The levels of fuel and reflector liquids can be read locally by sight glasses and in the control room over a closed-circuit television link with an accuracy, after previous calibration, of $\pm$ 0.020 in.; complete filling of the spherical and closed cylindrical core tanks is also detected by electrical contact probes.

In the absence of a liquid reflector, rapid shut-down of PANTHER, as also of TOAD and PHOENIX, is entirely dependent on a single safety mechanism: the removal of fuel solution by dumping under gravity. To enhance the reliability of this action, two parallel dump lines, each closed by a large diameter, air-operated diaphragm valve controlled by its own trip-responding circuit, are provided. In addition, duplicate electrical contact probes ensure that the dump tank is empty whenever reactivity is added to the system. It is not always convenient, nor is it necessary, to provide dump tank capacity for the entire contents of the core vessel. The principle guiding the design of a dump tank is that it shall, by accepting at least 20 % of the volume of the largest core tank, enable reactivity to be reduced at a rate approaching several per cent k/s up to a total of a few per cent in k; further reduction in reactivity then continues at a slower rate by the pumped removal of solutions from the dump tank until the system is empty. The gravity flow of fuel by dumping is, as nearly as can be contrived, proof against failure. Its continued removal by pumping is somewhat less reliable, being dependent on electrical supplies to a pump, though these are duplicated. In experiments employing a liquid reflector, the latter is also dumped under gravity in the event of a trip. For convenience, the level of the reflector trip is usually set below that of the core trip to permit a quicker resumption of an approach to criticality should the trip have some trivial origin such as counter noise. The reflector dump system is not duplicated, though, to give a comparable rate of reactivity reduction, it must be of larger flow capacity than that serving the core. The dump tank for the reflector is also the storage tank and can thus accept the whole volume of reflector liquid.

The TOAD liquid critical assembly equipment closely resembles PANTHER in its design and method of operation. It is intended primarily for basic criticality studies on fairly small vessels containing uranyl fluoride solutions of medium or high U 235 isotopic content; separate containment of the rig has not therefore been considered necessary. The range of fourteen interchangeable core tanks includes spheres, isometric cylinders and tall cylinders up to a maximum core diameter of 22 in. (see Table IV). In addition, seven special cylindrical tanks are available for studies on two- and three-region cores, including replacement experiments in which the reactivity of a specially prepared test sample can be compared with that of a void. This latter facility has been included as a convenient if somewhat approximate means of assessing the criticality of ' awkward ' materials such as wet powders, curds, sludges, and solutions containing dispersed or lumped neutron poisons. The provision of three core regions allows the neutron spectrum of the test specimen to be roughly matched with that of the main or ' driver ' part of the core by a ' buffer ' region of similar composition to the specimen itself. Six further tanks are available for experiments on tall cylinders having thin or nominal water reflectors of 0.5, 1 and 2 in. radial thickness; these are intended to provide critical data applicable to process plant situations where

the neutron reflection, although not negligible, is demonstrably less than that in a fully-reflected environment. All the TOAD core tanks are of aluminium alloy, usually 0.128 in. thick. The fuel and reflector dump systems are similar to those of PANTHER, with the addition of a third dump system to serve the buffer region of the core when this is in use. The levels of liquids in the three core regions and the reflector are reproduced in parallel sight glasses within the field of a single television camera and can be read in the control room to an accuracy of $\pm$ 0.030 in.

The PHOENIX equipment (Figure 13) is used for critical mass studies on enriched uranium solutions in vessels of considerable diameter or height; it is thus complementary in scope to TOAD and resembles it in design and operation. The range of core tanks includes tall or ' near-infinite ' cylinders (9 ft high and 7 to 10 in. dia.), large cylinders (up to at least 40 in. dia.) for studies on shallow slabs and on low concentration systems, and a 38 in. dia. sphere. The reflector tank can be readily removed, making the equipment suitable for examining the effects of *ad hoc* plant conditions, such as reflection by persons or by walls or other building structures. The basic equipment can also be used to operate a specimen of a proposed process vessel as a solution-filled critical assembly and thus to give direct information on the nuclear safety of such vessels.

An apparatus has recently been constructed at Dounreay for measuring the degree of nuclear interaction between vessels containing solutions of U 235. In its initial form the equipment, designated SIRIUS (Figure 14), consists of two parallel slab-shaped tanks, each mounted vertically on one of the platforms of the TESSIE assembly machine (Figure 9). Pairs of tanks of various internal thicknesses between 1.5 and 4 in.) are available; they are 4 ft wide and are normally filled with solution to a depth of 4 ft. The primary purpose of the experiment is to measure the critical separation of the two tanks as a function of the tank thickness, while keeping the facing area of the tanks and the solution concentration constant. By comparing the critical tank-separation/tank-thickness curves for vessels separated by air, water and constructional materials such as concrete, beechwood and compressed timber (with or without neutron poisons such as cadmium sheet), it is hoped to obtain the basic data needed for the safe and economical design of high capacity solution stores. Each slab tank is treated as a potentially critical assembly in its own right and is filled by remote control from its own solution storage and supply system. Each tank also has its own primary shut-down mechanism — the dumping of 10 litres of fuel solution into a safe cylinder carried on the approach machine table, the separation of the two tables on trip serving as a secondary shut-down device. To economise in fuel, the outward-facing sides of both slabs are reflected by a permanent solid reflector such as polyethylene, while the solid ' decoupling ' material under examination is clamped in contact with the inward-facing side of one slab tank. The liquid levels in both tanks are measured by sight glasses and are transmitted by television to the control room, together with the table separation.

Finally, a design study has been carried out for a further solution-cored assembly machine, SORCERER, proposed for use at Dounreay to compare the effectiveness of different materials as neutron reflectors. The significance of a structural material as a reflector is greatest, probably, when it lies in close proximity to a slab-shaped vessel containing solution of maximum reactivity since in such circumstances a normally trivial uncertainty in the reflector

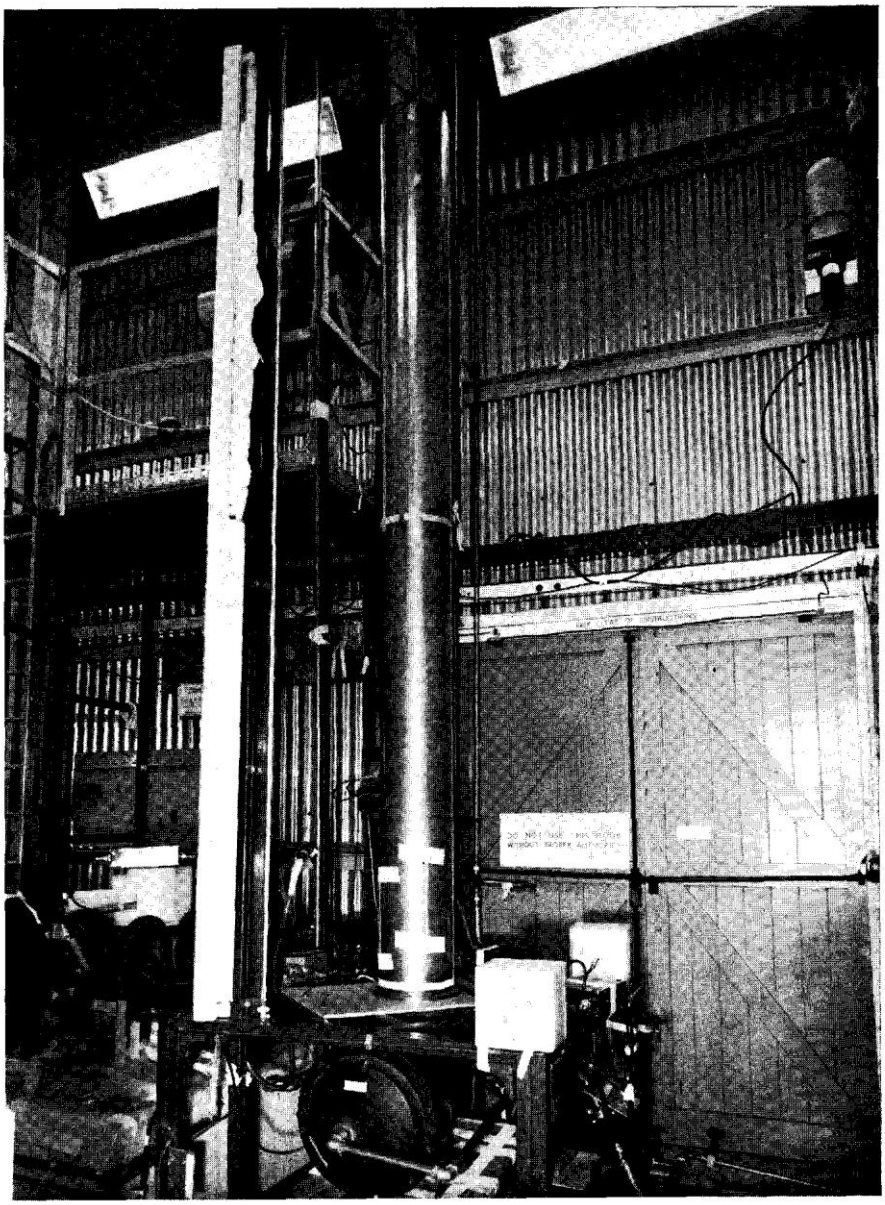

Figure 13. *The PHŒNIX uranium solution reactor, as used for critical height measurements in a tall unreflected cylinder.*

saving of the material can amount to a proportionately large uncertainty in the safe thickness of the slab-tank. The central feature of SORCERER is, therefore, a horizontal slab of uranium solution contained in a 4 ft square, open-topped tank having a thin aluminium base. The tank is supplied with uranyl fluoride solution from the Cell 1 storage system and the fuel depth

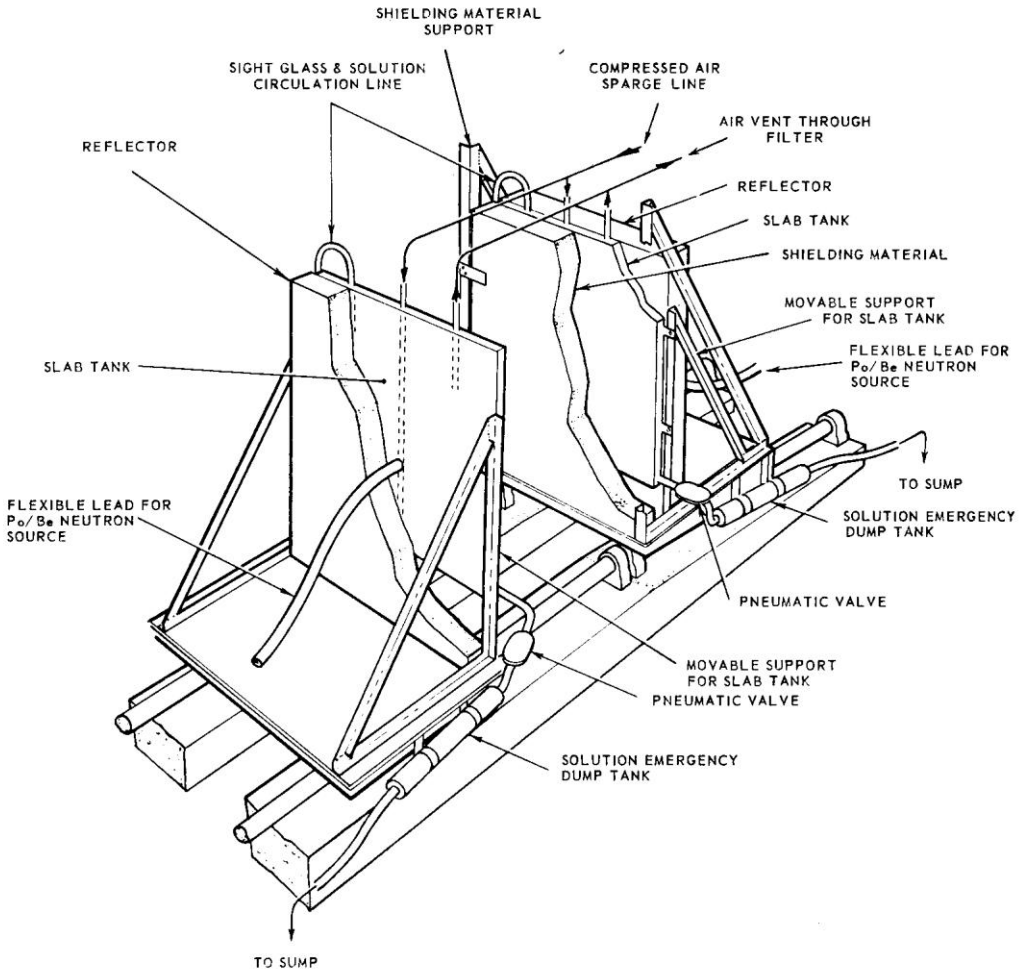

Figure 14. *SIRIUS interacting slab assembly rig (schematic).*

measured by televised observation of a sight glass; a duplicated solution dump system serves as a shut-down device. Arrangements are provided for levelling the tank and also for maintaining its base flat to within 0.050 in. by a removable honeycomb support which contributes a minimum of unwanted neutron reflection. In this, its simplest form, the equipment is suitable for measuring the critical thickness of an isolated and unreflected 4-ft square slab of solution over a range of concentrations and enrichments. The saving afforded by a solid reflector can then be measured directly, as the reduction in this critical thickness, by raising a slab of the test material into contact with the underside of the core tank. A platform, mounted on horizontal rails and raised hydraulically, is used to support the test reflector and permits it to be assembled and accurately aligned at a distance from the core tank. Tests on ' non-infinite ' reflectors are carried out using an aluminium honeycomb as a lower support. The effect of water reflectors of various thicknesses

can also be studied, using one of several flooded honeycombs inside a second tank carried on the lower platform. A further addition to the equipment, an upper platform supported inside the solution tank and manually adjustable in height, enables the saving afforded by placing solid reflectors on the two faces of the fuel slab to be measured. This latter facility is intended to confirm the validity of measurements using a one-sided reflector rather than for routine use.

The Operation of a Criticality Laboratory

The operator of a critical experiment laboratory is faced with two not always compatible objectives: the absolute requirement to ensure the safety of all personnel, whether they are concerned in the critical mass work or not, together with the only slightly less imperative need to ensure the safety of site installations and of the laboratory equipment and materials; and the need, which derives from the high capital value of the equipment and fuel materials and from the operational difficulties of the work, to obtain the greatest possible yield of critical mass, reactor physics or nuclear data from each programme of experiments. Some indication has been given in the preceding section of the ways these two objectives are met in the United Kingdom laboratories [5]. It is appropriate, however, to summarize the main features of safety philosophy and experimental procedure adopted in the two laboratories currently in operation.

Safety Philosophy and Procedures

At Aldermaston, all experiments are terminated slightly short of the delayed critical point by an interval dependent chiefly on the familiarity of the system being studied. Each approach to criticality is carried out in a stepwise manner and extreme care is taken to obtain an accurate estimate of the critical point from an early stage in the approach. Whenever possible, therefore, a 'normalizing' experiment using a non-fissile replica in place of the intended fissile core is carried out prior to the 'live' run. The ratio of the response of each neutron detector in the live run to that in the normalizing run, for similar arrangements of the neutron source and detectors and of scattering and absorbing material in the system, then provides a satisfactory measure of the relative neutron multiplication (M) at each stage of the approach. The reciprocal of this relative multiplication (1/M), which tends to zero as the system approaches criticality, is plotted as a function of the control variable: table separation, liquid level, etc. Extrapolation of the 1/M curve provides an increasingly accurate estimate of the critical point as the experiment proceeds; in addition, it enables the size of the next step to be decided and the increase in multiplication, and hence in counting-rate, corresponding to the chosen step to be predicted. It is the practice at Aldermaston to proceed by stages such that the relative multiplication is approximately doubled in each, the trip level being normally set at 50 % above the predicted counting rate for a proposed step, while the 1/M curve is re-examined if the predicted level is exceeded by more than 10 % in any counter.

At Dounreay, account is taken of the fact that reactivity excursions have occurred on quite a number of occasions in critical assembly laboratories and the facilities and experimental procedures have been designed accordingly. The philosophy has been adopted that, if an accident capable of creating a prompt critical condition does happen, there is little that can be done to pre-

vent it by trip-actuated shut-down devices; on the other hand, such devices can do much to limit the total yield of the excursion by suppressing bursts after the first as well as any subsequent fission plateau. It is considered, therefore, that the likelihood of an incident yielding more than about 10^{18} fissions is small. In fact, the shielding of the cells, by distance or concrete or both, has been made such that an incident yielding 10^{19} fissions would not result in a direct radiation dose to personnel exceeding the emergency tolerance level. In a similar manner, the isolation of the uranium cells and the containment afforded by the steel pressure vessel of the plutonium cell provide adequate safeguards against the release of fission products and the natural activity of plutonium from incidents having yields up to 3×10^{18} fissions.

The acceptance of a slight element of risk in the Dounreay experiments leads to a considerably greater speed of working than is possible at Aldermaston, as well as to greater flexibility in the physical arrangement of the assemblies. It is not usually essential to obtain an accurate measure of the relative multiplication early in an approach, and a normalizing run is thus rarely required; a neutron source of adequate emission is essential, but it need not be positioned inside the core, nor need it emit neutrons in a fission spectrum. The steps in an approach can safely be of a size such that the multiplication is increased by a factor of three or four, while the final steps to the critical point can be made equivalent to a maximum reactivity increase of about 0.5 % in k. The actual critical point is located whenever possible by the extrapolation to zero of two slightly supercritical pile period measurements, obtained after removal of the neutron source.

In both United Kingdom criticality laboratories, as elsewhere, the safety of working depends primarily on the experience and vigilance of the personnel rather than on an excess of mechanical interlocks and administrative regulations. It is the procedure, however, before starting a new series of experiments, to prepare a Safety Principles Document or Experimental Plan describing the measurements to be made, the control and safety features of the equipment and the experimental techniques. This document must show that all locally-controlled operations, such as the hand-stacking of a sub-assembly, are demonstrably safe or will be shown to be so by a previous stage in the experiment; it must show that all reactivity additions are reversible; it must consider all foreseeable accidents and indicate the steps taken to avoid them; and it must show that the consequences of the maximum credible accident are acceptable. It is submitted to, and must be approved by, a local Safety Committee, composed of persons largely independent of the operation team, after which the responsibility for the safety of day-to-day operations rests with the leader of the experimental group. The conduct of a critical experiment itself is under the personal control of an authorized Team Leader, chosen for his experience of the type of system being studied; his contribution is particularly valuable in the interpretation of approach curves and in the disposition of the neutron source and counters so as to yield meaningful 1/M curves from the earliest possible stage in the approach. During an approach, independent calculations of the multiplication and graphical predictions of the critical point must be made independently by two members of the team, and they must agree on the safety of a further stage before the experiment can proceed. At least two, and preferably three, members of a team must be qualified scientific staff.

The function of shut-down devices in limiting the yield of a prompt

critical excursion has already been described. They serve also to terminate the less severe surge of power likely to remain within the delayed critical region, to be expected following the over-running of a fuel pump or machine drive motor. Whenever possible, two (or more) physically dissimilar shut-down mechanisms are provided on a machine. One of these is arranged to be fairly fast-acting and, in consequence, may control only a small amount of reactivity; the other is usually slower in operation but is capable of shutting the system down entirely in all circumstances. Examples of both are the separation into two halves of a solid assembly and the removal of core or reflector liquid. The insertion of solid neutron absorbers into the core is not favoured, owing to their somewhat unpredictable effects on reactivity in frequently-changed core configurations and to the difficulty in providing spaces within the core to contain the absorbers while preserving a ' clean ' geometry. Fast-acting ' safeties ' frequently invoke the direct fall under gravity of part of a solid or liquid core or — less usually — a reflector. Slow-acting devices may require an electrical supply to a motor, either to drive the two halves of a solid assembly far apart horizontally or to remove the entire contents of a liquid core by pumping; such mechanisms cannot be made entirely fail-safe, though the chances of failure can be made almost negligible by the provision of alternative mains and battery supplies, the latter preferably unfused. In systems, such as bare solution cores, where two different safety devices are difficult to provide, a single one (dumping of solution), duplicated in its entirety, is used. This insures against failure of the mechnical or electrical parts but not against the physical inadequacy of the method of reducing reactivity.

The rates of reactivity addition are limited during particular experiments by the motor or pump speeds and by limit or level switches restricting the ranges of availability of the several speeds. However, the gear ratios of mechanical drives, the displacement of pumps, and the positions of limit switches are all adjustable and their settings are considered as part of each Experimental Plan. Interlocks and sequencing circuits are used to ensure that safety devices are cocked and that a neutron source is inserted prior to starting a critical approach. Beyond this, the conduct of an experiment is very largely at the discretion of a team leader; in particular, many — though not all — of the radiation sensing instruments can be bypassed, and it is the experimenter's responsibility to ensure that an adequate number of effective detectors are available to initiate a trip at any stage in an approach.

Economic Operation of Facilities

The high cost of critical assembly experiments derives from two main causes: the relatively high investment of fissile materials, and the need to conduct the experiments without hazard to persons and equipment. The former is determined by the critical masses and volumes of the systems being studied and cannot be reduced materially as long as a direct measurement of the critical parameters is required. The latter — which is reflected in the costs of shielded cells, containment vessels, complex instrumentation, remotely-controlled operations and active-handling procedures — is part of the safety philosophy of the nuclear industry and cannot be compromised. It is therefore essential to obtain the highest possible utilization of the facilities, materials and staff if criticality studies are to be economically worthwhile. This need can be met partly by designing the buildings, assembly machines and other

installations to be of high flexibility and partly by extracting from each programme of experiments all the information readily obtainable regardless of its relevance to the immediate critical mass determinations. To conclude this paper, some observations will be made on how these objectives are being approached in the two laboratories in question.

It is a pre-requisite of flexibility that several assembly machines are available. For work on solid systems, it is not unduly inconvenient for these to be within the same cell, though each should be provided with its own control instrumentation. For work on solutions or on a mixture of solid and solution systems, it is almost essential that each apparatus shall be in a separate cell if work is not to be hindered by the impossibility of setting up one rig when an adjacent one is operating in a near-critical condition. Even where separate machines and cells are available it is often found that the major part of the time is occupied by engineering or other preparatory work and it is probable that a critical facility concerned with a variety of systems cannot be operated with reasonable efficiency if fewer than three cells are available; only one or two of these are then likely to be in use for actual measurements at any one time.

Several procedures have been adopted at Dounreay in an attempt to reduce the proportion of time spent on setting up an experiment. These include the use of a standard control wiring installation, incorporating a modular system of plug-in relays to provide interlock and sequencing facilities and including a telephone-type distribution or ' jumper ' panel at which all the cross-connections needed to change from an old rig to a new and entirely different one can be made and tested in a few hours. It is the practice to pre-test all new control circuits prior to installation, or even to delivery, of a new rig by using an electrical rig simulator. Likewise, the hydraulic behaviour of the liquid circuits of solution rigs is tested in a mock-up prior to being set up in a cell. Closed-circuit television is frequently used for transmitting to the control room physical measurements such as liquid levels, platform positions and even some meter dial readings in preference to more conventional electrical methods This is a particularly convenient procedure where the need is of short duration and where faulty instrument indications must make themselves immediately apparent.

A final requirement of a versatile criticality facility concerns the ancillary services. An adequate instrument department, integral with the laboratory and familiar with pneumatic and hydraulic as well as with the more conventional electronic and electro-mechanical techniques, is highly desirable. For work on solutions, and probably also on solid moderated fuels or ' mock-precipitates ', there must be a chemical technology department within the facility. Mechanical engineering services must be available for programmed work such as core tank changes and to provide day-by-day services in support of the less predictable needs of the experiments; the major design of new machines is normally outside the scope of the laboratory staff, though it remains their responsibility to specify their requirements. Ready access to computer facilities, if necessary over a telex data link, is desirable for numerous purposes, from the routine checking of pulse counting channels for statistical purity to the analysis of foil activation data, the computation of reactor parameters from distribution measurements and the treatment of reactor period data.

The principle of exploiting each experiment to the full can be followed

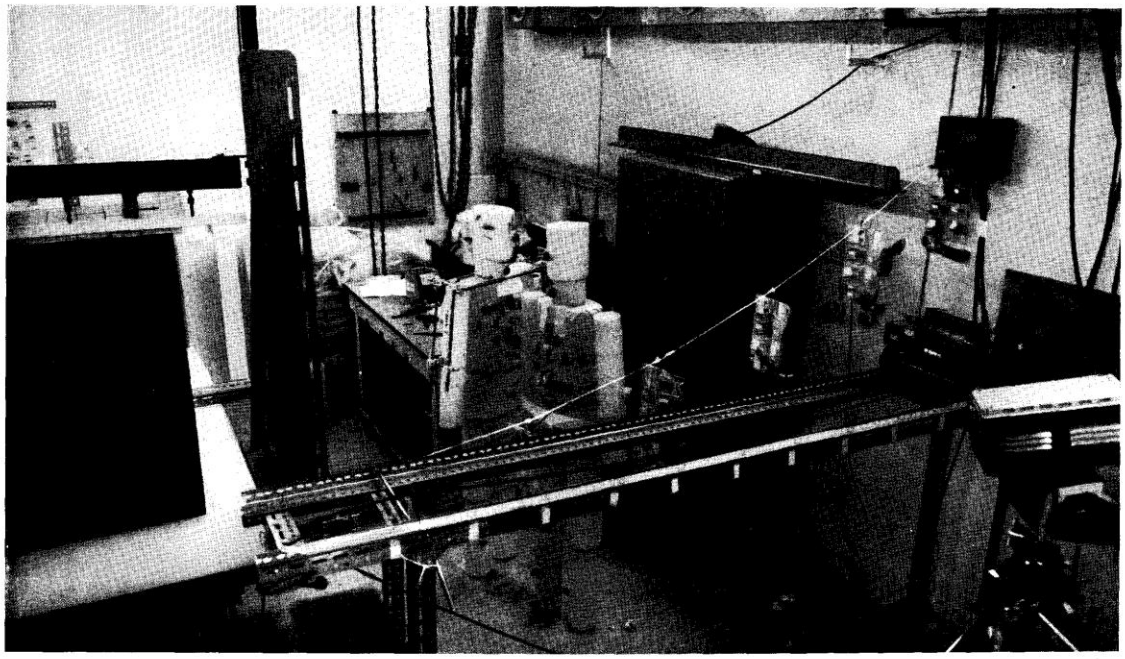

Figure 15. *The use of the TESSIE reactor for the irradiation of dosimetry lockets mounted on " phantoms " and at other fixed locations in the vicinity of the critical assembly.*

in a general way by carrying out neutron-distribution, integral reactor spectrum studies, importance or replacement experiments and fission rate measurements whenever opportunity permits, and in as comprehensive a manner as possible; some of these studies are only practicable in a critical system and may require operations up to a power of a few watts. In particular instances, other extensions of the scope of critical mass experiments are possible. One such extension, shown in Figure 15, has involved the use of the Dounreay TESSIE assembly machine for the irradiation of neutron dosimeters and ' phantoms ', as part of a programme to develop a critical incident ' locket ' for use in fissile material processing plants throughout the United Kingdom. In this, operation of the assembly at a power of over 1 kW was required, until the yield of the run exceeded 10^{17} fissions and the radiation dose to the nearest phantom was over 300 rad. In another experiment, special core tanks in the PANTHER equipment are being used, in conjunction with a neutron chopper, for time-of-flight measurements of the spectrum in plutonium solutions of various concentration; the differential spectra thus obtained are being compared with the integral measurements made in the critical spheres. The same vessels are also being used to obtain the $\eta(239/\eta(235)$ ratio.

In conclusion, it may be stressed that continuity of work, and hence the economic usage of a facility, can only be ensured by including in its programme one or two longer-term experiments able to absorb the surplus efforts of scientists, but which do not make very heavy demands on the chemical or engineering resources of a laboratory.

Examples of such work are the fundamental neutron interaction experiments carried out on multi-component metal systems at Aldermaston [4] and the basic solution critical parameter surveys in progress at Dounreay [4] in the PHOENIX and TOAD reactors [6]. The former have led to the steady development of a widely applicable theory, using the concept of an interaction parameter, for calculating the safe number of fissile units in an array or the safe spacing of such units. The latter have embraced a wide range of core shapes, sizes and concentrations and are gradually defining the bounds of uranium solution criticality in the medium enrichment range. Both types of experiment have served as a reservoir of effort, available for urgent plant safety work as occasion demands, and have acted as a training ground for new staff.

REFERENCES

1. C.C. HORTON and J.D. MCCULLEN, " Plutonium-Water Critical Assemblies ", *Proc. First U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1955, 5*, 156.
2. W.G. CLARKE, C.C. HORTON and M.F. SMITH, " Critical Assemblies of Aqueous Uranyl Fluoride Solution. I - Experimental Techniques and Results ", *AERE R/R. 2051* (Sept. 1956).
3. J.R. HARRISON, M.F. SMITH, W.G. CLARKE, A.M. MILLS and J.A. DYSON, " Critical Assemblies with Heavy Water Solutions of Uranyl Fluoride (HAZEL). II - Physics ", *AERE R/R 2703* (Nov. 1958).
4. A.F. THOMAS and R.A. SCRIVEN, " Neutron Interaction in Fissile Assemblies ", *Progr. Nucl. Energy, Series IV, Technology, Engineering and Safety*, Pergamon Press, London, *3*, 253, 1960.
5. G.W.K. FORD and J.G. WALFORD, " Critical Assembly Experiments at Dounreay ", *Proc. Second U. N. Int. Conf. Peaceful Uses Atomic Energy, Geneva, 1958, 17*, 545.
6. J.C. SMITH *et al.*, " Criticality of 30 % Enriched Uranium Solutions in Cylindrical Geometry ", *DEG-Memo-663* (1960).

EXPERIMENTS FOR CRITICALITY CONTROL

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INTRODUCTION

The prevention of unwanted and uncontrolled nuclear chain reactions is of the greatest importance to the advancement of nuclear energy because of both the personnel and property damage which may result and the stigma attached to incidents directly associated with this new and often misunderstood energy source. Public reaction to such occurrences is often far out of proportion to their consequences, tragic though they may be to individuals directly concerned, when measured by the scale used to assess industrial accidents in general and the day-to-day accident experience of individuals. It is particularly necessary that nuclear chain reactions be avoided in operations for processing fissionable materials where, in general, there is little protection of personnel from the attendant radiation and where the personnel themselves are not nuclear reactor specialists knowledgeable in the properties of such chain-reacting systems. On the other hand, these processes must be designed and operated with some appreciation of their costs. The specification of these non-reactor operations with fissionable materials must, therefore, be very carefully prepared in order to achieve both safety and reasonable economy. This requirement demands, *a priori*, sound technical information as design bases. Sources of such information are, at one extreme, direct measurement of equipment, piece by piece, to assure its safety and, at the other extreme, a nuclear-reactor-type calculation of tolerable dimensions and batch sizes.

Present practices in nuclear safety rely strongly upon experimental evidence of the conditions necessary for a particular material to be critical in a particular configuration since uncertainties inherent in the results of reactor calculations, due to deficiencies in both the method and the nuclear constants which must be inserted into the analyses, demand the application of inordinate safety factors. That the emphasis may shift from such a strong dependence upon experiment as the field matures is recognized.

Since a majority of the experiments have provided quite basic information, interpolation and short extrapolation of the results allow their application to many specific problems. The experimental results also serve as important guides in the development and evaluation of analytical methods.

This paper discusses the experimental methods which yield information useful in the solution of criticality problems. Reference is made only to activities within the United States. The discussion is very general, merely indicating the type of experiments and the laboratory requirements. There is no theoretical treatment of the experimental methods.

Experimental Methods

The experiments to be considered may be roughly grouped into two types. One includes those in which observations of the neutron multiplying properties of the test material are made directly and is illustrated by well-known critical experiments. In the other type, observation is made of the distribution, in space or in time, of neutrons within the test material and is characterized by exponential experiments.

Critical Experiments

The performance of a critical experiment yielding only a value of a critical quantity is very straightforward and the observations are direct. The fissile material is arranged, with due regard for potential hazards, in the amount required to support a chain reaction. The result defines a critical system under the conditions of the experiment. The establishment of this chain reaction is achieved in the following idealized manner. The requirements are the test material, a source of neutrons, and a detector of neutrons. Observations are made of the increasing neutron count rate as the material is assembled, preferably by remote operation, around the source. These observations provide guidance in the experiment and, finally, establish the system as critical. The increased count rate arises, of course, from fission-produced neutrons supplementing those from the source itself.

A property of a neutron multiplying medium, k, known as the neutron reproduction factor, is defined as the ratio of the neutron population in successive neutron generations. In anticipation of the discussion which follows, it is noted that if k is equal to unity, an assembly is critical by definition; if k is either greater or less than unity, the assembly is either supercritical or subcritical. The count rate observed during the construction of a critical assembly is the sum of source neutrons and those arising from fissions produced both by source neutrons and by progeny of neutrons born in earlier fissions. The count rate, C_t, is, in this simple consideration:

$$C_t = C_o + kC_o + k^2C_o + \ldots + k^nC_o$$

where C_o is the initial count rate in the absence of any neutron multiplying material and the exponents designate the neutron generation in which the fissions responsible for that term first occurred. This expression can be written as:

$$\frac{C_t}{C_o} = \frac{1}{1-k} \quad \text{for} \quad k < 1$$

and is the source neutron multiplication produced by the assembly as constructed at time t. It is observed that the state is steady as long as k is a constant less than unity. Increasing the size of the assembly increases k and increases the neutron multiplication. In the limit of $k = 1$, the source multiplication becomes infinite. During the course of the assembly the reciprocal of the source multiplication, C_o/C_t, is plotted as a function of the controlling variable. Extrapolation of this plot to zero intercept yields the critical value of the variable, and the approach to a critical condition is thereby guided.

It is interesting to observe that k is independent of the absolute value of the neutron flux and, since $k = 1$ is the necessary condition for criticality, critical systems can be operated at very low power. For this reason informa-

tion applicable to megawatt power reactors may be obtained from milliwatt critical experiments. It may also be noted that k of a finite volume is a function of both the material and the geometry since the latter determines the loss of neutrons by leakage. For an infinite volume, $k = k_\infty$, a property of the material alone.

To ascertain that a system is indeed critical requires removal of the neutron source, or the production of sufficient fission neutrons to mask its presence, to assure that the fission neutron population is constant in time and that the chain reaction is self-sustaining. Deviations in k from unity produce exponential changes in the neutron level at a rate depending upon the average neutron lifetime. Since about 0.7 % of the neutrons arising from fission are emitted by fission products which decay with finite half lives, the time behaviour of an assembly with k no greater than 1.007 is governed primarily by these delayed neutrons. The control of reactors is, indeed, possible only through these neutrons. If k exceeds 1.007 the system becomes prompt critical and the uncontrollable rate of increase in power is governed by the properties of the material. The margin between delayed critical, $k = 1.000$, and prompt critical, $k = 1.007$, is, perhaps, of the order of 1 % of the critical mass. It is for these reasons that critical experiments must be done with caution and in specially-designed laboratories.

To extract information other than dimensions from critical experiments requires much more effort. It is necessary to evaluate the conditions particular to the experiment, the effect on the critical volume of the container or the supporting structure, for example. It is also desirable to measure the spectral and spatial distribution of the neutron flux within and adjacent to the assembly. It is necessary to know in detail the properties of the experimental materials — both fissile and those needed for the experiment — requiring chemical and isotopic analyses. Additional knowledge of the properties of the critical assembly may be obtained from its kinetic behaviour, that is, the time rate of change of the neutron population as a function of the perturbation which caused it. From many data can come an analysis of the experiment and correlation with a definitive theoretical model.

Neutron Multiplication Experiments

The establishment of critical dimensions by neutron multiplication experiments, sometimes called 'critical approach' or 'subcritical' experiments, is a variation on the critical experiment described above. In these, as before, the test material, a source of neutrons, and a neutron detector are required. The same assembly procedure is followed, except that the assembly of material is stopped short of critical by an amount depending on the confidence in the measure of the multiplication. Many data obtained in laboratories from experiments of this kind have yielded valuable information on the critical dimensions through extrapolation of the reciprocal neutron-multiplication curve. The method has been used extensively in at least one process plant to determine the margin of safety in many operations.

In these experiments it is important to establish a truly representative reciprocal multiplication curve which can be extrapolated with confidence. This will require careful location of the components to ensure observation of a neutron count rate characteristic of the assembly and not one distorted by extraneous reflections from nearby structure. It is most desirable, for example, to place the source within the fissile unit.

Two applications of this type of experiment to process testing, made at the Rocky Flats Plant, are described here. The first problem, reported by Bidinger [1], was undertaken to ascertain the feasibility of increasing the U 235 capacity of a storage tank by first filling the tank with short sections of borosilicate glass tubing known as Raschig rings. The experimental arrangement is shown in the upper section of Figure 1. The glass rings (3.8 cm long, 3.8 cm outside dia., 0.2 cm wall thickness, 12.5 % B_2O_3 content) when packed in the tank occupied 17.8 % of the volume and established a boron density of 15.4 g/litre. The tank was surrounded by concrete to provide neutron reflection. The neutron source was placed in the lower part of the cylindrical tank and the neutron counters beneath it. Aqueous uranyl nitrate solution, containing uranium of about 90 % U 235 content, was used in these experiments at two chemical concentrations. The lower section of Figure 1 shows the results in a plot of the reciprocal neutron multiplication observed as solution was added to the tank. A balance between neutron leakage and neutron production after a particular height is exceeded is shown by the parallelism of the curves and the abscissa. This indicates that a tank infinitely tall will be subcritical.

The second example illustrates an examination of the safety of a storage array. The units to be stored were cylindrical containers of an enriched uranium solution mounted in a supporting frame. The neutron source was located inside the container which was to become the most central in the array and the boron-containing neutron detectors were placed at its base as shown in Figure 2. The result of assembling up to 25 conterminous units in a square pattern is shown in the graph. Figure 3 is a photograph of the completed array.

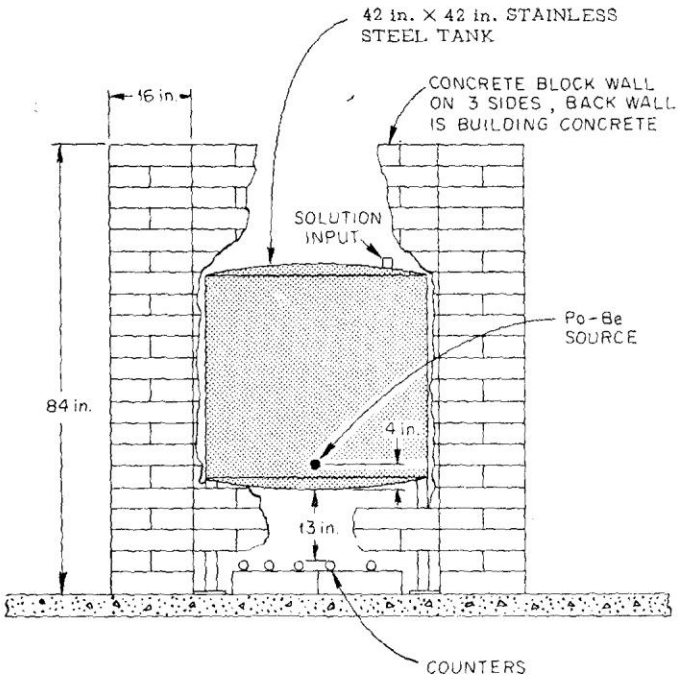
It is pointed out that this type of experiment does not yield basic information beyond the critical dimensions obtained from the extrapolation of the reciprocal multiplication curves. These remarks have been purposely detailed to emphasize the advantages of the method in a direct evaluation of process-plant safety.

Material Replacement Experiments

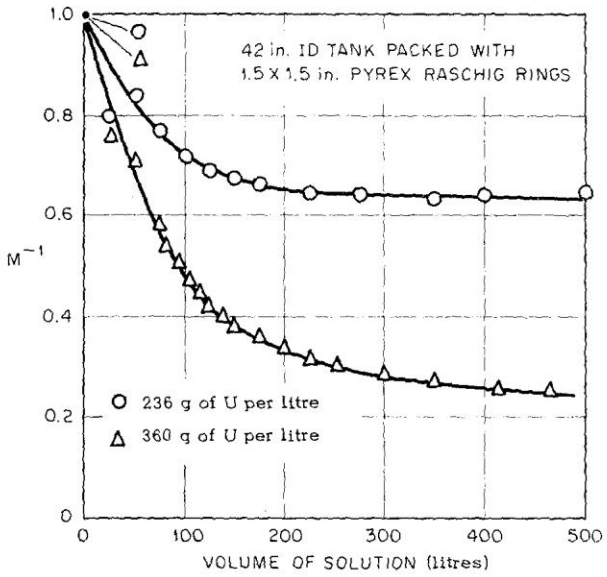
A low-power reactor or critical assembly is required for experiments of this type. The assembly is equipped with a device for precisely adjusting the reactivity and is provided with a means of introducing samples into a central region. It essentially compares the neutron multiplying properties of a test sample with those of a standard.

The properties of a number of materials have been studied extensively at the Hanford Laboratories by this method in the Physical Constants Test Reactor described by Donahue [2]. The PCTR may be briefly described as a graphite-moderated critical lattice of enriched uranium rods enclosing a central cavity having a capacity of a few hundred litres. The assembly is approximately a 2-m cube. It can be separated into two sections for operational convenience. Although the central cavity is filled with the test material, only a small portion at the centre is designated as the sample. The purpose of the remainder, called a buffer, is to establish at the sample a neutron spectrum characteristic of the test material itself in contrast to the spectrum in the critical uranium lattice. The property evaluated in the PCTR is the neutron multiplication factor of an infinite sample, k_∞.

The basis of the experiment is the condition that exchange of two samples

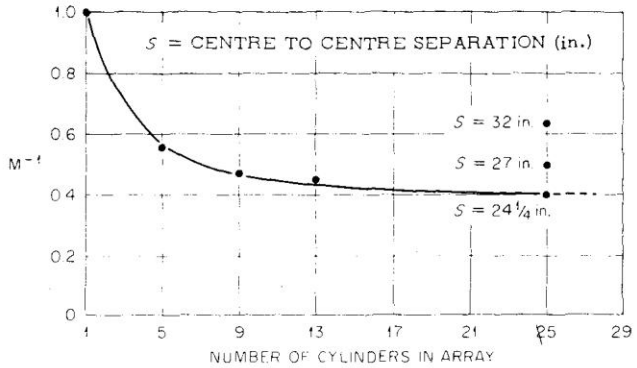


Experimental tank set-up.

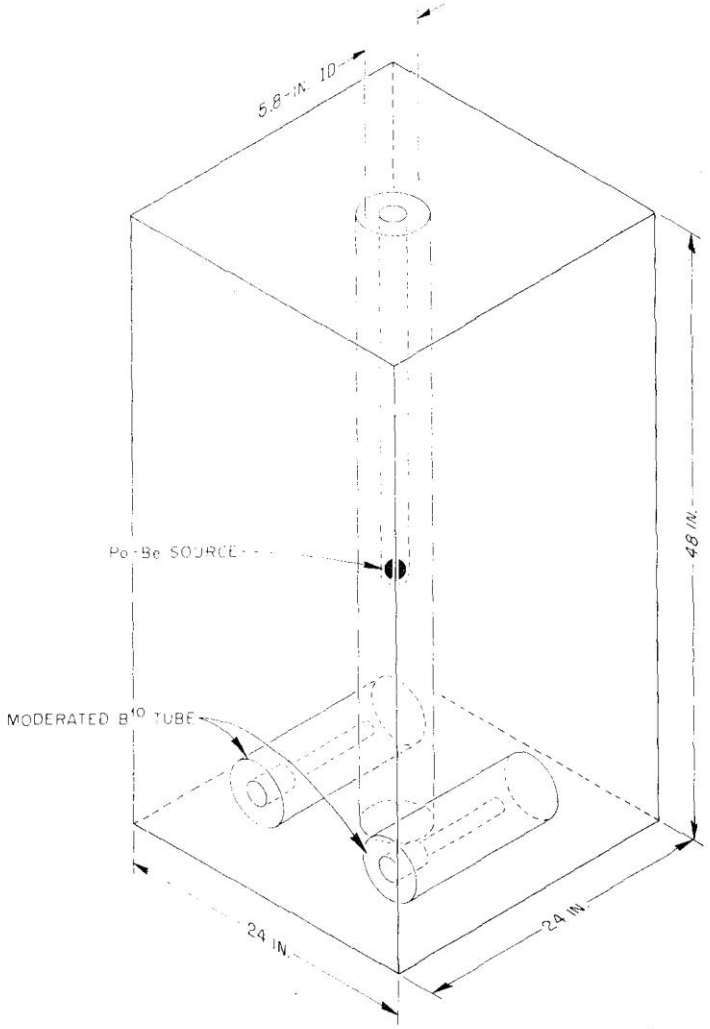


Reciprocal neutron multiplication (M^{-1}) as a function of $UO_2(NO_3)_2$ solution volume in litres.

Figure 1.



Reciprocal multiplication as a function of number of cylinders in array.



Central source container.

Figure 2.

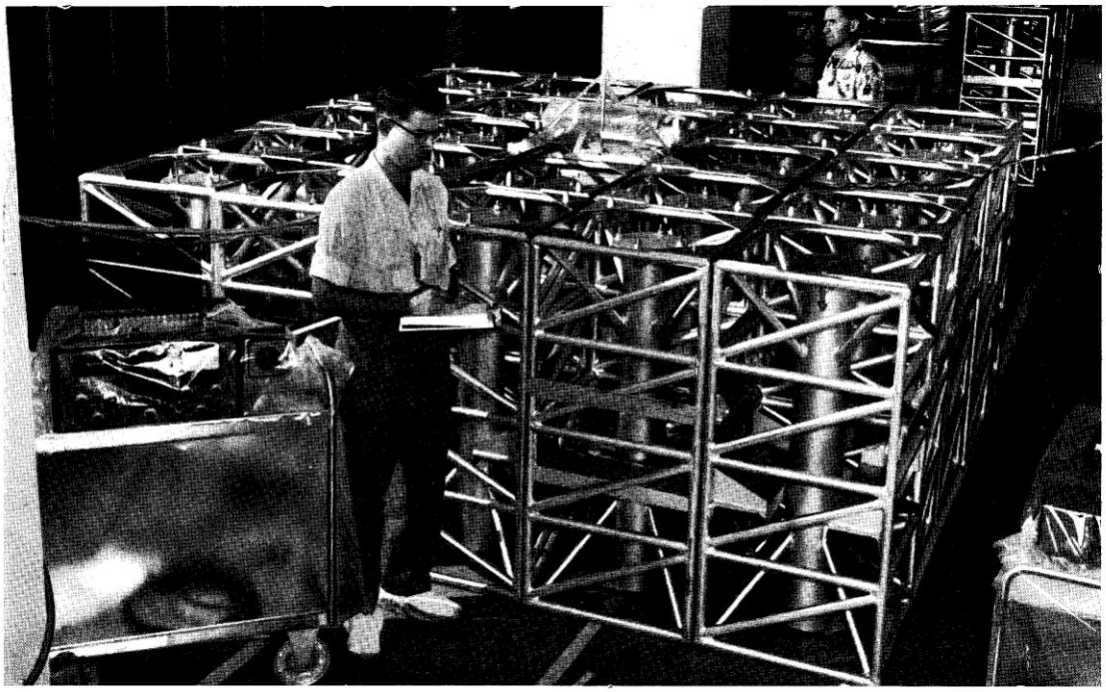


Figure 3. *Completed subcritical array of cylindrical containers of U 235 - enriched solution mounted in 61 cm (2 ft) square supports.*

of equal k_∞ within the assembly will not change the reactivity of the assembly. In the PCTR experiments one of the samples, the standard with k_∞ unity, is a vacuum or, for practical reasons, helium. The procedure, therefore, is to adjust the amount of a neutron absorber, of known properties, distributed within the material under study, including the buffer, until substitution of the standard sample for the central poisoned test sample causes no change in the overall reactivity of the assembly. This condition is achieved, of course, when the control devices remain fixed for both sample conditions.

The value of k_∞ can be inferred from the quantity of neutron absorber required to establish this identity of samples and its neutron absorption cross-section.

Figure 4 is a photograph of the PCTR and Figure 5 is a close view of the buffer section. The test sample measured 15 × 15 × 30 cm.

The application of the PCTR is illustrated by the recent experiments by Handler [3] establishing the limiting critical U 235 enrichment of uranium homogeneously moderated by hydrogen. The test material was uranium oxide mixed with an hydrogenous plastic. The value of k_∞ was determined for each of several mixtures of varying hydrogen content over a range of U 235 enrichment. Interpolation of the results defined a combination of these quantities ($\sim$ 1 %U 235 enrichment and an H/U ratio of about 5) which, without any extraneous neutron absorber, would effect no change in the reactivity of the assembly upon substitution for the standard.

Exponential Experiments

Information describing neutron multiplying properties may be obtained from exponential experiments, an example of the type depending primarily upon neutron distribution measurements. Exponential experiments can be performed with considerably smaller quantities of the material of interest than are required for critical or even for multiplication experiments. An idealized exponential experiment consists of a block of the test material placed with its base adjacent to an extended source of neutrons. The source may be a beam of neutrons from an operating reactor or it may be one or more capsules of a mixture of polonium and beryllium, or of plutonium and beryllium, embedded in a neutron diffusing material such as graphite.

The performance of the experiment consists in measuring the steady state spatial distribution of slow neutrons in the multiplying medium with appropriate neutron detectors. The material buckling, B^2_m, a property only of the material, can be obtained from this distribution in the following manner. Neutron diffusion theory [4] relates the buckling and the slow neutron distribution by the expression:

$$\left(\frac{\pi}{a}\right)^2 + \left(\frac{\pi}{b}\right)^2 - \gamma^2 = B^2_m$$

In this relation a and b are the effective dimensions of the block in the plane parallel to the base and $1/\gamma$ is the relaxation length of the neutrons in the direction perpendicular to the base. The relaxation length is the distance within which the neutron flux decreases by a factor e, the logarithmic base, as is seen from the expression $\Phi(z) = Ce^{-\gamma z}$, describing the exponential dependence of the flux on the distance from the source. The quantities measured in the experiment are a, b, and γ. Recognition must be made of the deficiencies of diffusion theory in the interpretation of these measurements.

Figure 4. *Physical Constant Test Reactor.*

The solution of the equation describing the flux distribution in a particular critical assembly yields a value of the geometric buckling, B_g^2, a property of the geometry. Since the values of the geometric and the material

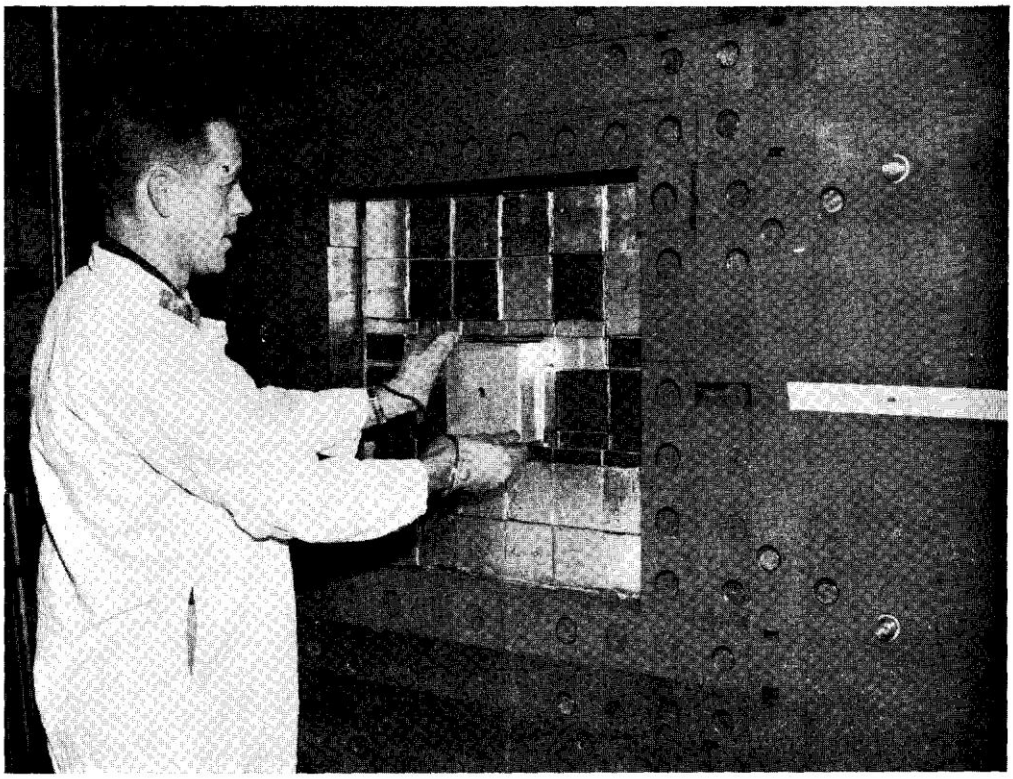


Figure 5. *Close view of PCTR near midplane.*

bucklings of a critical system are equal, determination of the latter from an exponential experiment enables the critical dimensions of the material in a particular geometry to be calculated by equating these two characteristic quantities. It is emphasized that dimensions appearing in the expressions for bucklings define a fictitious boundary where the neutron flux, not the physical boundary, effectively vanishes. These two dimensions differ by the extrapolation distance, a difficult quantity to evaluate since it depends in part upon the multiplying medium, the shape of the boundary and the reflector condition. Uncertainties in extrapolation distances can introduce significant errors in the critical dimensions evaluated in this manner.

Many exponential experiments have been described in the literature [5] and reference is made to them for details.

Pulsed Neutron Experiments

In the second type of experiment predicated on neutron distribution, short bursts of fast neutrons are injected into a sample of the multiplying material and the exponential decay of the prompt neutrons from fission is observed. Characteristic lifetimes in moderated multiplying media are of the order of milliseconds and are significantly less in fast neutron systems.

The prompt neutron decay constant governing the exponential decay is the quantity measured in pulsed neutron experiments.

Treatment of the time-dependent prompt neutron flux decay by reactor analysis relates the decay constant to the buckling of the medium. In principle the prompt critical buckling is the intercept obtained by extrapolation of the curve relating the measured decay constants of assemblies of a particular composition to their geometric bucklings. In some cases the delayed critical buckling can be inferred from the results of this type of experiment, supplemented by knowledge of the properties of the material.

The equipment required for these experiments includes a source of fast neutron pulses and a neutron detecting system capable of measuring the neutron intensity as a function of time within intervals in the O.01 μs to millisecond range. The source is usually the D-T nuclear reaction which yields 14 MeV neutrons. A beam of deuterons, accelerated by potentials of the order of 100 kV by a high voltage device, is caused to fall upon a tritium target only during short periodic intervals by a pulsing electrostatic field through which the beam passes. Equally short and periodic bursts of neutrons are thereby produced. Neutrons are detected by crystal scintillators. The time distribution of the neutrons following a burst is measured directly or is converted to a pulse-height distribution which is sorted into intervals by a pulse height analyzer. The output of the analyzer is stored until the number of pulses observed is sufficient for adequate statistics. The original distribution in time is then displayed.

A number of pulsed neutron experiments have been reported and reference is made to them for details of the analyses [6].

That quantitative measures can be made of process equipment sufficiently subcritical to meet safety requirements has not been clearly established. A proposal for effecting such measurements has been made by Silver [7] in which the decay constant is first determined with the equipment water-filled, and then when filled with a solution of known concentration of fissile isotope. A linear extrapolation of these two points yields the concentration which would be prompt critical and an estimate can be made of the delayed critical concentration. Values of the decay constant measured during subsequent operations indicate the degree of approach to a potentially hazardous condition. Favourable comparison of the values of k describing an assembly of water-moderated and reflected reactor fuel elements obtained both by a pulsed neutron experiment and by a more conventional method support the proposal. The minimum value of k in this experiment was 0.87.

This technique, in its present development, is applicable to homogeneously distributed materials in simple geometry.

The availability of compact portable sources of neutron pulses makes desirable an exploration of this method of safety evaluation.

Laboratory and Equipment Requirements

Any experiment with fissile material is potentially hazardous to a degree depending upon the nearness of the approach to criticality in its performance. The experiments considered above, therefore, require specialties in laboratory and equipment design commensurate with both the radiation hazard involved and the possible severity of radioactive contamination arising from the test materials. Brief comment on accepted practices will be made in this Section.

Critical Experiments

Of the several types discussed, critical experiments are more vulnerable to nuclear incidents. In fact the record in the United States shows a greater preponderance of incidents of this kind in critical experiments laboratories than in all other nuclear activities combined. This susceptibility is not unexpected, because these experiments are purposely exploratory and because they are short-ranged and do not warrant the detailed operational mechanisms prepared for long-term operating reactors. On the other hand, although incidents in critical experiments may be quite severe in their immediate environs, the hazard to an extended area is negligible since, in low-power operation, no inventory of radioactive fission products has accumulated. For these reasons the critical experiments laboratory is designed to protect primarily the operating staff. This protection against direct radiation is provided by local shielding or by adequately separating the potential accident site from the operating centre. Both of these methods must be supplemented by control of personnel to ensure their proper location at times of accident potential. This control may be accomplished by interlocking equipment with access barriers and by strict administrative practices. Potential accidents necessitate these protective measures, not the usual low-power operations.

The Los Alamos Laboratory is admirably located in a system of connecting canyons which allows the ' safety-by-distance ' solution of the radiation problem. A plan of the laboratory site is shown in Figure 6. The experiments, assembled in buildings designated as kivas, are made critical by remote operation from the control centre some 400 m distant. Natural shielding provides protection of one kiva from the other. Requirements of long cables for instrument and control signals and inconvenient access by personnel for equipment adjustment are obvious disadvantages of this laboratory plan.

The recently completed Plutonium Critical Mass Facility at Hanford described by Reardon and his colleagues [8] is an example of a laboratory with test and control areas relatively proximate and radiation protection provided by local shielding. The reactor room walls and the roof are of ordinary reinforced concrete 1.5 m and 0.6 m thick, respectively. The structure is designed to provide protection both from the radiation accompanying a nuclear excursion and any concomitant internal pressure. Figure 7 is the floor plan of the facility of which Figure 8 is an exterior photograph.

Special equipment is required in some laboratories to contain contaminating materials which may be used in their programmes. Figure 9 is a photograph of a compartment to house the vessels in which the plutonium-solution critical experiments will be performed.

The basic requirements of the equipment for critical experiments can assume many variations in conformity with the needs of particular assemblies. The requirements, succinctly enumerated, are: a neutron source, three or more neutron detectors, and a method of changing the reactivity of the assembly by remote control.

The source can be a capsule containing a mixture of polonium and beryllium, plutonium and beryllium, or radium and beryllium, although the gamma radiation from radium is undesirable. The neutron source may be an inherent property of the test material exemplified by spontaneous fission in U 238 and in Pu 240, a contaminant of Pu 239; or by the α-n reaction on the oxygen in aqueous solutions of plutonium, a strong alpha particle emitter.

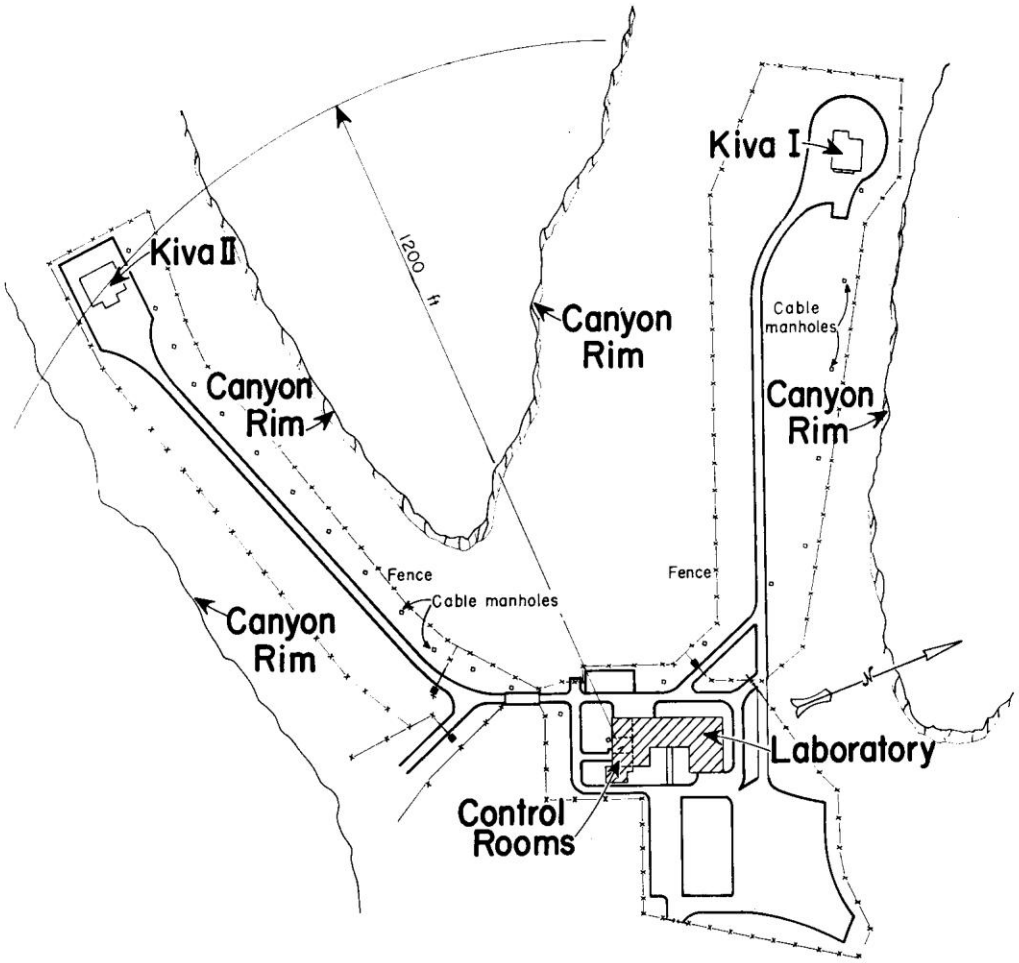


Figure 6. *Plan of Los Alamos critical experiments laboratory site.*

Although a proportional neutron counter, with associated amplifier and scaler, is required for observing the source-neutron multiplication in the approach to criticality, operation both at and near critical can be best guided by a continuous recording of the output of an integrating device such as an ionization chamber. Two or more of the sensory circuits, preferably of very rapid response, are required to signal an emergency and to actuate the shut-down mechanism if a predetermined power or neutron flux level is exceeded. A safety circuit responsive to the time rate of change of the flux level is a desirable supplement.

Auxiliary necessities are standard health physics-type instruments for the control of contamination which can arise from the materials under test, and for radiation surveys both during normal experimentation and in the event of an incident.

The details of the method of controlling the reactivity may vary widely,

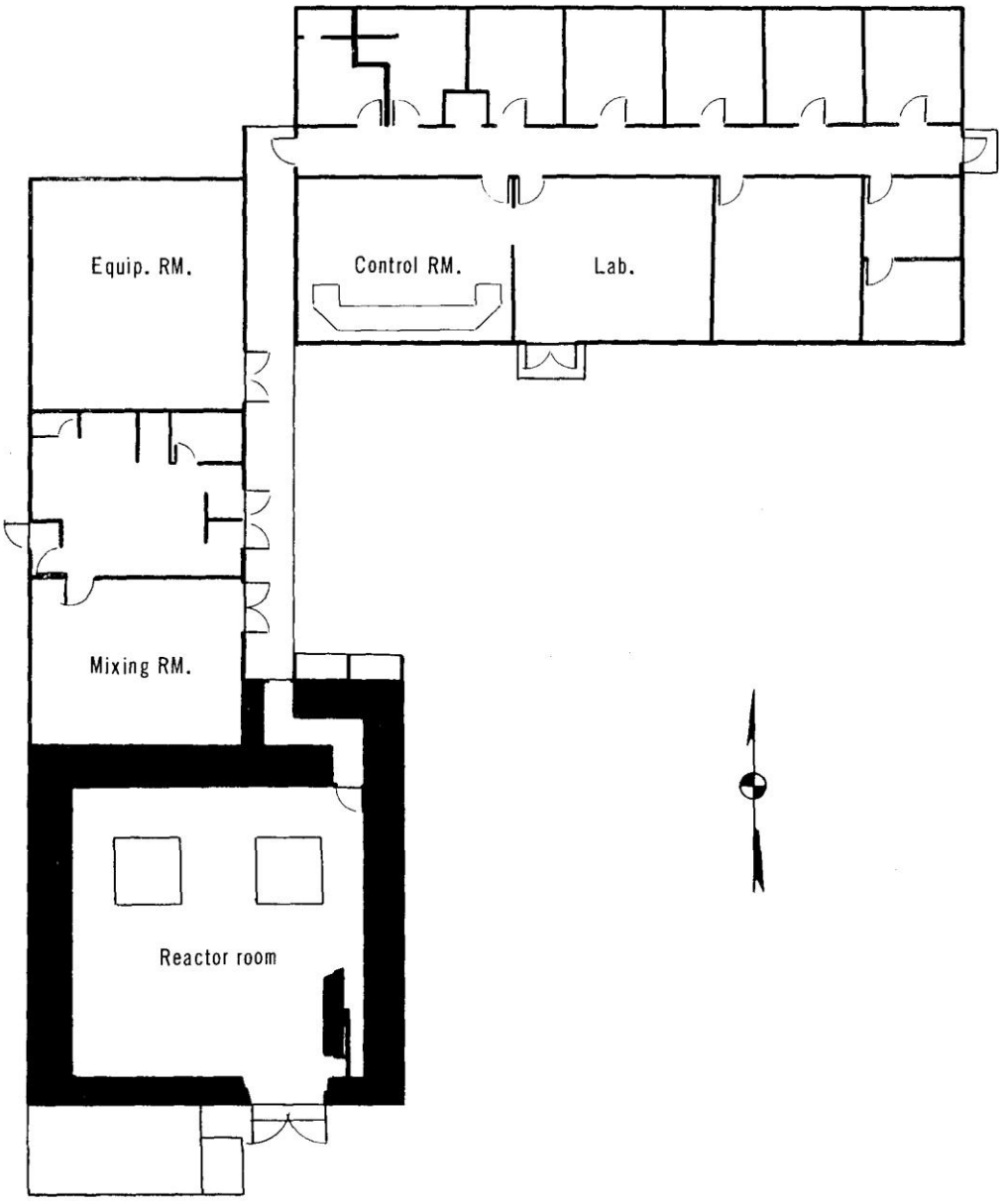


Figure 7. *Floor plan of Hanford plutonium critical mass facility.*

depending upon the nature and the materials of the experiment, provided two basic requirements are met. One of these requirements is remote operation and the other is, in effect, reversibility of the operation with the rate of reactivity removal greater than the rate of addition. At this point the discussion may diverge along many paths. Possibly the simplest example, and one often employed, is the stepwise construction of an array in the presence of a

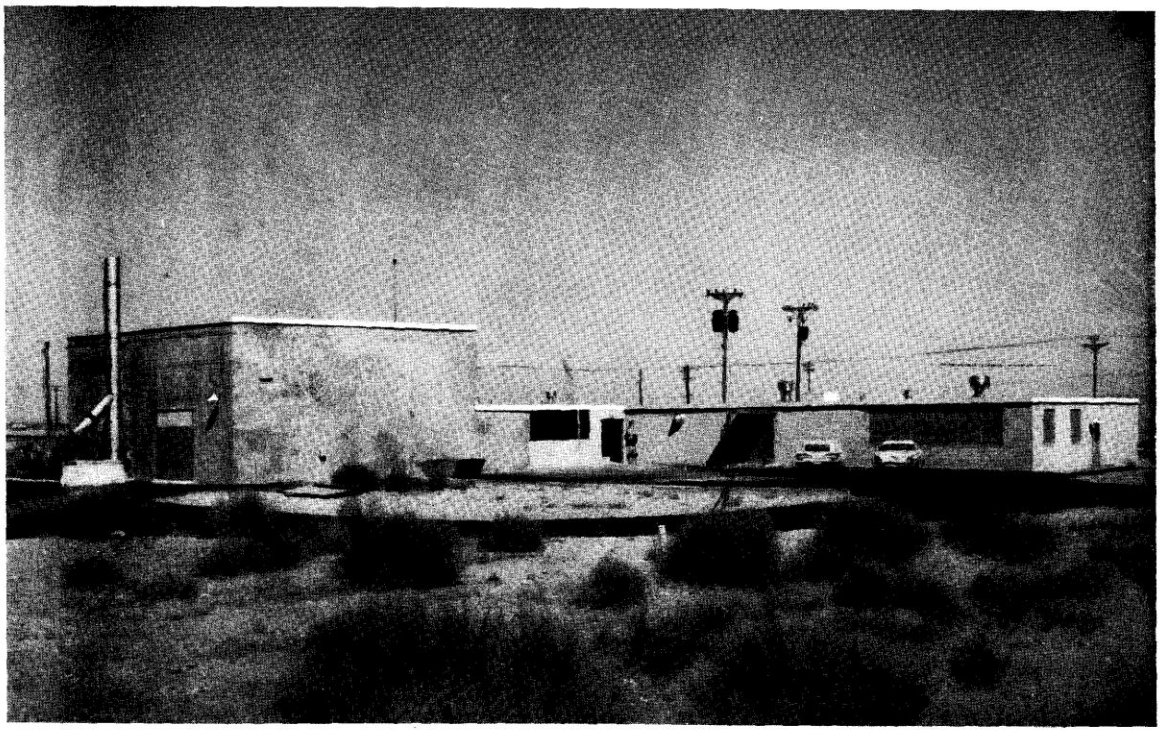


Figure 8. *Exterior of the Hanford facility.*

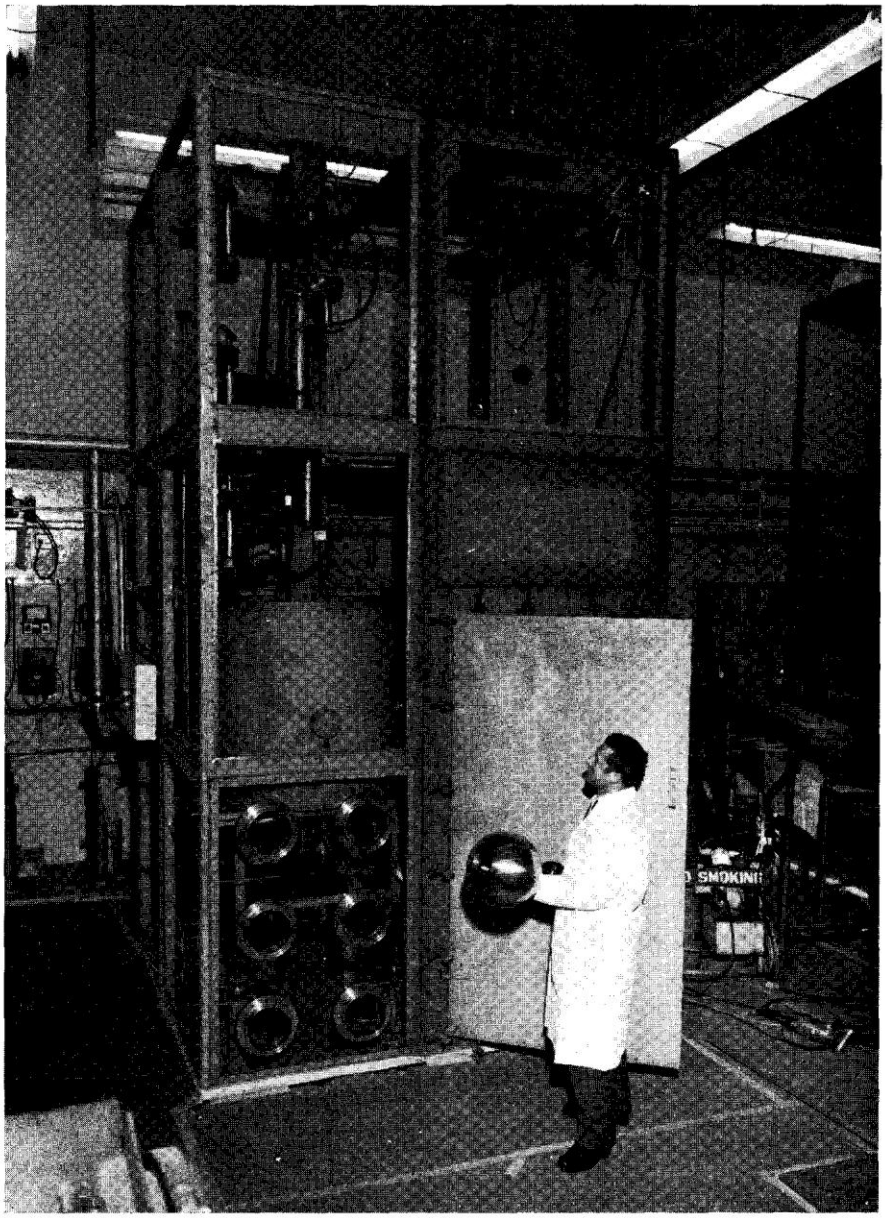


Figure 9. *Containment structure for plutonium critical experiment equipment.*

strong neutron-absorbing element which can be withdrawn by remote control. If, further, this absorber is supported against gravity by an electromagnet whose current is controlled by the radiation-sensitive detectors, its reinsertion serves the shut-down requirement.

A well known method of reactivity control in experiments with solid

materials is the construction of the assembly in two sections, one being movable by a remotely-operated motor. Upon emergency signal the sections are driven apart at a speed greater than their approach. A remotely-operated pump may transfer a liquid to the experiment — for example the fissile material in solution or an aqueous neutron moderator or reflector for a solid-fuel system. Removal of the liquid can be through a pipe, larger than the entry line, equipped with a 'normally-open' valve controlled by the radiation sensor. It is appropriate to provide a second and independent shut-down method to guard against the failure of the primary one. This may also be the insertion of an absorber, or the removal of moderator or fuel, by gravity or by a spring-loaded device. Many effective and acceptable combinations of these basic control methods can be devised.

It is difficult to illustrate the details of actual experimental arrangements. The assembly for each of two recent tests made in Oak Ridge are shown in Figures 10 and 11. In the first of these the material mounted on the platforms is a mixture of UF_4 and paraffin formed into compact blocks and wrapped in thin aluminium foil to facilitate handling. The platform on the left is movable by controls located in a well-shielded area. The approach to criticality is effected by observing the neutron multiplication as successive blocks are added. The base is a low density aluminium structure to minimize the return of neutrons in this nominally unreflected array.

A photograph of an array of 98 plastic bottles containing 93% U 235 enriched uranium in solution (Figure 11), is even less definitive of detail. The purpose of the experiment was to establish safety specifications for storage of these units, each of which contained about 6 kg U 235, by determining the critical number as a function of their spacing. Although the contents of most of the units remained fixed during the experiment, five centrally located units were joined to a common supply of solution and could be filled by a remotely-operated pump. They could be emptied through a large pipe fitted with an automatically controlled valve. It is, perhaps, obvious that this experiment followed many simpler ones which established the critical dimensions of smaller arrays.

Neutron Multiplication and Exponential Experiments

There are no unique laboratory requirements imposed by safety on neutron multiplication and exponential experiments. Fissile materials and neutron sources are used in both, so it is imperative that the quantities of the former be kept well below critical amounts and that the hazards of neutron sources, *per se*, be recognized. If the assemblies under study are nominally unreflected, it is necessary that they be located with reasonable separation from extraneous reflecting structures.

The instrumentation for the neutron multiplication experiments is the same as that required for the initial steps of a critical experiment.

Pulsed Neutron Experiments

The above comments on quantities of fissile material and neutron reflection are applicable to pulsed neutron experiments with perhaps even greater emphasis on minimizing spurious neutron reflection. Personnel shielding during these experiments is necessary because of the intensity of the neutron source.

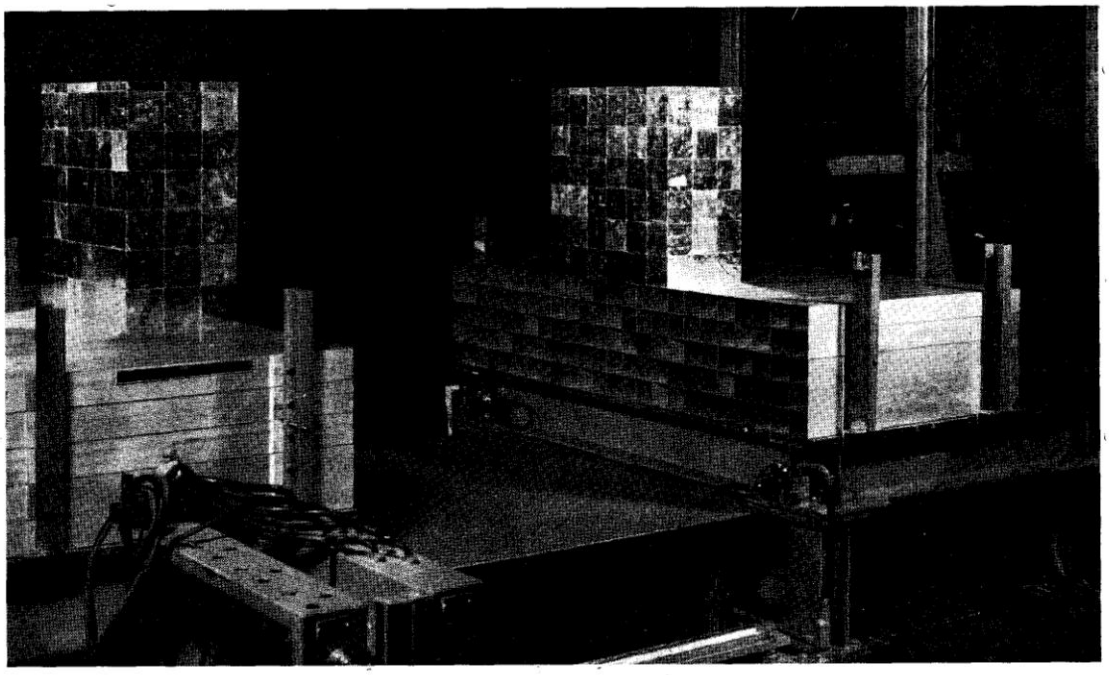


Figure 10. *Assembly of solids containing U 235-enriched uranium on separable table.*

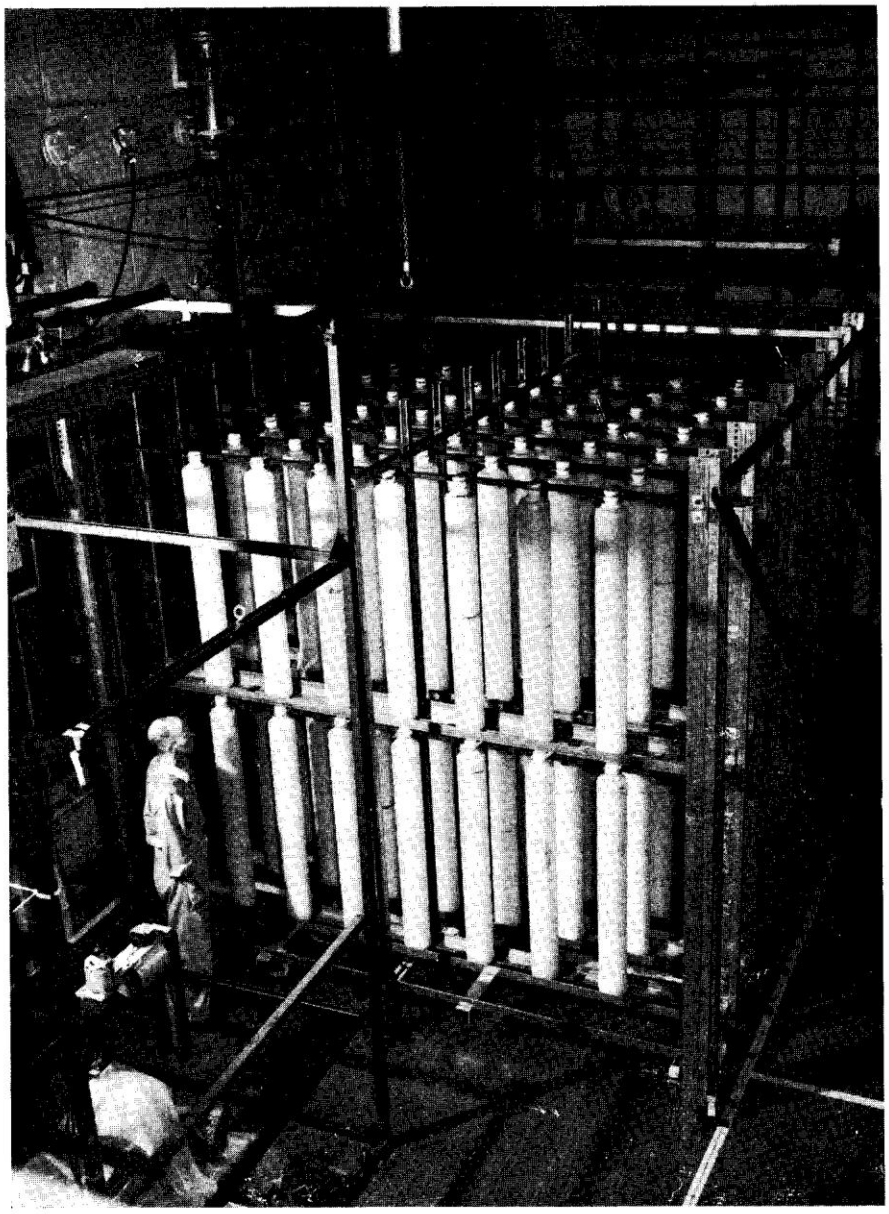


Figure 11. *Array of U 235-enriched uranium solution containers.*

Personnel Requirements

Little need be said about the qualifications of personnel for these experiments beyond pointing out that each is a research type project requiring qualified and knowledgeable staff. The performance of critical experiments is

not to be likened unto the operation of a power reactor. Most reactors are well-established machines incorporating thoroughly tested safety devices and automatic control mechanisms whose surveillance can be made a routine.

Future Demands on Experiments

There are deficiencies in the basic information applicable to nuclear safety problems. It is the intent of this Section to list some of those areas in which deficiencies are believed to exist; the selection is based on personal observations and is not purported to be complete.

Enriched Uranium

1. Perhaps the most interesting and demanding gap in the data at this time is the dependence of critical parameters on the density of U 235 in process materials. Although the mass of U 235 per unit volume of solution is established by solubility, the mass per unit volume of a slurry of a sintered oxide, whose density is nearly that of the crystal, will be significantly different at the same moderation. Experiments performed with high density compounds, highly enriched in U 235, compacted homogeneously with hydrogenous solids, will furnish information on this density effect.
2. The relation between critical dimensions of standard-density, highly-enriched uranium compounds as a function of moderation should be extended to zero moderation.
3. Another area, at present under investigation but requiring extension, is that of arrays of individually subcritical units. To generalize even a semi-empirical interpretation of data and to provide for development of a theory requires knowledge of the effect on a critical array of the following:

a) The moderation of a single unit;
b) The U 235 density and the U 235 enrichment of a single unit;
c) Neutron reflectors closely fitting single units;
d) A moderator distributed between units;
e) The reflection of the array.

4. The knowledge of uranium of intermediate enrichment should be broadened by repeating item 1 with 30 % U 235 material.
5. The field of low enrichment should be extended by repeating both items 1 and 2 with 5 % enriched uranium.
6. Very little is known of the properties of enriched uranium moderated by other than hydrogenous materials. Suggested for study as moderators are: D_2O, D_2O-H_2O mixtures, beryllium and carbon. The last two, in combination with water or organic substances, will yield information useful in some reactor fuel fabrication processes. Common organic materials encountered in industry may be included in this list.

U 233 and Plutonium

It is believed that only the more basic information describing U 233 and Pu 239 is required and that effects on attendant criticality problems of special moderators, etc., and the interaction between subcritical units can be extrapolated from U 235 data with support from a few representative measures. The topics suggested are, therefore:

a) The critical dimensions of water-reflected and unreflected spheres of aqueous solutions in an H/U 233 range of 70 to 1,000;
b) A study of H/U 233 values between 0 and 50;
c) Examination of U 233-Th mixtures;
d) An extensive programme yielding basic information describing Pu 239, in simple geometry, over a wide moderation range including H/Pu 239 of 0 to 50.

Mixed Fissile Isotopes

The critical dimensions of binary and ternary mixtures of U 233, U 235 and Pu 239 will be useful to some applications.

Neutron Absorbers

Investigations furthering the use of neutron-absorbing elements as either liquid or solid constituents of process streams will support this method of increasing unit capacity. Distinction should be made between the effect of a solid constituent as truly an absorber, and its effects as a diluent and in producing inhomogeneity.

Experimental Methods

Improvement can be made in existing experimental techniques and in developing practical in-process methods of evaluating nuclear safety. An example is the application of pulsed neutron experiments.

ACKNOWLEDGEMENTS

Appreciation is expressed to D. Clayton, H. Paxton and C. L. Schuske for descriptions of experimental facilities at Hanford, Los Alamos and the Rocky Flats Plant, respectively.

REFERENCES

1. G.H. BIDINGER, C.L. SCHUSKE and D.F. SMITH, " Nuclear Safety Experiments on Plutonium and Enriched Uranium Hydrogen-Moderated Assemblies Containing Boron ", *RFP-201* (1960).
2. D.J. DONAHUE, D.D. LANNING, R.A. BENNETT and R.E. HEINEMAN, " Determination of k_∞ from Critical Experiments with the PCTR ", *Nucl. Sci. Eng. 4*, 297 (1958).
3. H.E. HANDLER, " Nuclear Physics Research Quarterly Report ", *HW-54591*, p. 41 (Dec. 1957).
4. For details of exponential experiments the reader is referred to any treatise on reactor theory, for example: A.M. WEINBERG and E.P. WIGNER, *The Physical Theory of Neutron Chain Reactors*. p. 428-431, The University of Chicago Press, 1958:
S. GLASSTONE and M.C. EDLUND, *The Elements of Nuclear Reactor Theory*, p. 281. Van Nostrand, New York, 1952.
5. For a review of early experiments and descriptions of their use see: D.E. DAVENPORT, " Exponential Experiments on Graphite Systems ", in *Proc. First. U. N. Int. Conf. Peaceful Uses Atomic Energy. Geneva, 1955, 5*, 309.
H. KOUTS and R. SHER. " Experimental Studies of Slightly Enriched, Water-Moderated Lattices ", *BNL-486* (1957).

6. For example see: E.C. CAMPBELL and P.H. STELSON, " Physics Division Semi-annual Progress Report ", *ORNL-2204*, p. 34 (Sept. 1956).
F.A. KLOVERSTROM and T.T. KOMOTO, " Pulsed Neutron Source Studies of Graphite-Enriched Uranium Systems ", *UCRL-5477* (1959).
A more extensive discussion is given by: G.F. von DARDEL and N.G. SJOSTRAND, *Progress in Nuclear Energy, Series I, Physics and Mathematics*, V. 2, p. 183, Pergamon Press, London, 1958.
A survey of the use of pulsed neutrons in reactor studies was presented at the International Accelerator Conference in Amsterdam. See: K.H. BECKURTS. " *Nucl. Instr. Methods, 11*, 144 (1961).
7. E.G. SILVER, " A Proposal for Criticality Control Measurements Using Pulsed Neutron Methods ", *CF-60-5-130* (1961).
8. W.A. REARDON *et al.*, " Hazards Summary Report ", *HW-66266* (Aug. 1960).

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DISCUSSION

G. BLÄSSER, Euratom:

Je serais heureux de savoir si l'on peut lire les variations du niveau de la cuve du réacteur PROSERPINE à 0,01 mm près ou 0,1 mm, ainsi qu'il est dit ailleurs dans le rapport de M. Clouet d'Orval. Je me demande également si la quantité de solution qui reste toujours dans le réservoir pour des raisons de capillarité n'introduit pas une erreur dans la détermination de la hauteur critique de la cuve.

C. CLOUET D'ORVAL, France:

Nous pouvons effectivement apprécier des variations de hauteur de 0,01 mm. Le tube qui traverse l'empilement et reste rempli de solution contribue bien sûr à la masse critique de l'ensemble. Nous n'avons pas pu en déterminer correctement l'effet, mais nous pensons qu'il est faible.

W. SCHÜLLER, Eurochemic:

I was surprised to hear from Mr. Walford that concrete is a better reflector than water. Could he specify whether there are any results and to what cases this applies?

J.G. WALFORD, United Kingdom:

We have carried out only one main type of experiment on the effectiveness of concrete as a neutron reflector; this made use of a 1.4 % enriched uranium system of near-optimum moderation. We found that water gave a reflector saving to this system of about 3.5 cm whereas the reflector saving due to thick concrete was almost 5 cm. I would not like to suggest that these figures are directly applicable to any other system. I do believe, however, that our experiments place the various reflectors in the correct order of effectiveness, at least for thermal neutron systems, and the high neutron reflecting power of concrete should therefore be seriously considered when assessing the safety of process vessels.

C. SCHUSKE, United States:

What H/X ratio were these systems?

J.G. WALFORD:

The comparison of concrete and water as reflectors was mainly carried out on solid near-cubical assemblies of 1.4 % enriched uranium at an H/U (total) ratio of 8, corresponding to an H/U 235 ratio of about 570. This system is well moderated. The slide showing the interacting slab experiments depicts a system which will first be used with 30 % enriched uranyl fluoride solutions having an H/U 235 ratio of about 130, which we find is near the moderation giving minimum critical volume.

D. CALLIHAN, United States:

We obtained very provisional data on a 3 %-U 235 enriched uranium, hydrogen-moderated system at an H/U 235 of about 130. An 18-in. thickness of concrete seems to be a slightly better reflector than a similar thickness of paraffin, which in turn is a slightly better reflector than an equal thickness of water. With thinner layers of the individual materials, this relation might be somewhat different. I point out further that these results are strongly dependant upon the material being reflected.

P. BENOIST, France:

Que penserait M. Walford de l'explication suivante sur l'efficacité du béton, de l'eau et de la paraffine comme réflecteur : étant donné que les fuites de neutrons dans un système aqueux sont essentiellement des fuites de neutrons rapides, la présence dans le béton d'éléments lourds pourrait ralentir la thermalisation des neutrons dans le béton ; il en résulterait qu'un neutron rapide aurait une chance moindre de devenir thermique dans le béton et d'être capturé par la forte section efficace de l'hydrogène ; en d'autres termes, il aurait donc une chance plus grande d'être renvoyé dans le cœur.

J.G. WALFORD:

We in the United Kingdom, agree very much with what Mr. Benoist has just said. I did not wish to imply that we were completely without an explanation of the effect when I mentioned it as a side-issue from the experimental programme, but rather to suggest that it is one of those things that have perhaps been glossed over slightly in the early, more simplified treatment of criticality problems.

E.R. WOODCOCK, United Kingdom:

The fact that concrete is a better reflector than water can be roughly explained in this way. The concrete contains more atoms heavier than hydrogen than does water. These heavier atoms will turn a neutron back to the core much more easily than hydrogen will, because scattering with hydrogen in a laboratory system is always forward. We know that, in many cases, the thermal neutrons returning to the core are more effective than the fast neutrons returning to the core, but in concrete it seems that the actual number of neutrons returned is the overwhelming factor.

J.G. WALFORD:

I think this may be one of quite a number of instances where even a relatively crude experimental measurement can point the way to a more elegant theoretical treatment which can in its turn explain the phenomenon. Sometimes the more subtle phenomena tend to be overlooked in the rush of other computational work until some experimental evidence has been obtained.

R. GALLEY, France:

M. Walford a déclaré, au cours de son exposé qu'il était enclin, selon son opinion, qui est également celle de ses collègues, à recommander aux ingénieurs l'empoisonnement par différentes formules de poison, plutôt que des mesures pouvant porter sur la masse, la concentration, etc. Il a noté que ses suggestions relatives à cette forme d'action sur les masses critiques ne suscitaient que très peu d'enthousiasme. Ce manque d'intérêt ne serait-il pas partiellement imputable au fait que la plupart des gens ignorent la redoutable efficacité des poisons par suite de la rareté des connaissances diffusées à ce sujet ?

D. CALLIHAN:

I do believe that what Mr. Galley says is entirely true. In this new approach to safety, however, there is much to be learned about the effectiveness of the materials as neutron absorbers but more particularly much has to be learned about their integrity, their chemical and mechanical stability, and since this practice is new, I think it is being correctly applied with caution.

J. GUNTHER, France :

Je me demande également si le scepticisme qui règne vis-à-vis de l'emploi des poisons n'est pas dû au fait que leur efficacité est surtout portée vers l'augmentation de la masse critique et ne joue pas beaucoup, ou du moins à un degré qui n'a pas été précisé, sur l'élargissement de la géométrie critique. Or, la tendance actuelle est de contrôler le plus possible par la géométrie, de sorte que l'existence d'une masse critique plus grande n'apparaît peut-être pas, à de nombreuses personnes, comme un gain manifeste. Il est certain que l'effet d'un poison neutronique est beaucoup plus sensible dans des solutions très bien modérées qui donnent justement les masses critiques minima, alors que, pour les solutions peu modérées, l'effet des poisons dont l'absorption n'est sensible que dans le domaine thermique présente évidemment un intérêt beaucoup plus restreint.

E.C. WOODCOCK:

With regard to the graphs of experimentally measured multiplications shown by Dr. Callihan, I wonder if I may be forgiven for saying one general word about multiplication. Essentially a multiplication is measured by placing a neutron source at some point near or in the system and measuring a flux at some different point, and it is the ratio of this neutron flux to neutron source which gives us a measure of multiplication. This means that there are as many different multiplications as there are pairs of points at which one can put the source and counter. Some of these multiplications are very well behaved

and some are not; the surface multiplication of which I have spoken is one of the former. It is a measure of the skill of the experimenter, that he can place his source and counter in such a position that he gets a well-behaved multiplication which can extrapolate reliably to the critical size, but it is quite possible to place the source and counter in such a position that a most misleading multiplication is obtained. One may get very little effect until the system is very close to critical and then one will get a curve such as Mr. Callihan showed, which suddenly dropped down to the axis. This is perhaps a word of warning which should be borne in mind.

D. CALLIHAN:

As an experimentalist I showed that misbehaving multiplication curves can be obtained, and my remark was cautionary.

A.F. THOMAS, United Kingdom:

I agree thoroughly with both Dr. Callihan and Mr. Woodcock that one has to be very cautious about these things. At Aldermaston, we normally try to measure the central source multiplication of the system which is a very well-behaved quantity: but we feel that if one is going to measure multiplications other than the central source multiplication, or even central source multiplication itself, one must try to get the physics right. We believe that the multiplication to be used as an index of criticality is the ratio of the flux with the fissile material present to the flux with non-fissile replica. One is then measuring a ratio, and things like scattering, transmission of reflectors, etc., become a second-order effect — except with reflectors like water.

J.G. WALFORD:

I agree entirely with the points made by the last three speakers. The fact that the central source multiplication is one of the very few measures of multiplication which is well behaved is one of my reasons for having an objection, perhaps rather biased, to multiplication measurements as a class; this is that in order to use a central source you have of necessity lost the clean geometry you desire, which is a system without a space for a central source. Compensation for the source cavity is possible but is itself a potential source of error.

E.D. CLAYTON, United States:

In all these multiplication measurements people usually use at least three different counters or neutron detectors and these are placed at different positions with respect to the assembly, so that experimentally we are never in a position where we have to rely on one counter or one so-called multiplication, but have multiple curves which must all indicate the same critical mass, as it were; usually that curve is used which predicts the smallest critical mass during the approach.

H.C. PAXTON, United States:

I might mention that we are in the somewhat ridiculous situation of using central source multiplication as a safety index in our own regulations. In the older days when our work was confined almost entirely to fast neutron

systems, this was not so bad; central source multiplication of ten meant three-quarters of the critical mass, all within perhaps $\pm 5\,\%$ limits. This is no longer the situation, but we have not learned how to write simple regulations in any other terms and to us the multiplication of ten means 75 % of the critical mass regardless of how the latter is established. There are some real problems where one is given a fixed fissile object and asked how safe it is to handle. One can only judge on the basis of multiplication measurements and, as Mr. Thomas has mentioned, the required replica becomes a very awkward thing where there is any hydrogenous material present.

P. BENOIST:

Il me paraît intéressant de noter que la méthode de substitution progressive de Bacher et Naudet, qui est utilisée en France depuis plusieurs années pour l'expérimentation sur des réseaux au graphite et à eau lourde et qui a fourni des résultats très satisfaisants, pourrait être utilisée également pour la détermination du " buckling " dans des solutions aqueuses thermiques. Il me semble en effet que l'interprétation de cette méthode de substitution doit être plus facile en milieu homogène qu'en milieu hétérogène par suite de la disparition des effets d'anisotropie, d'une part, et de la disparition des effets dus à l'écart par rapport à la symétrie de révolution, d'autre part.

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