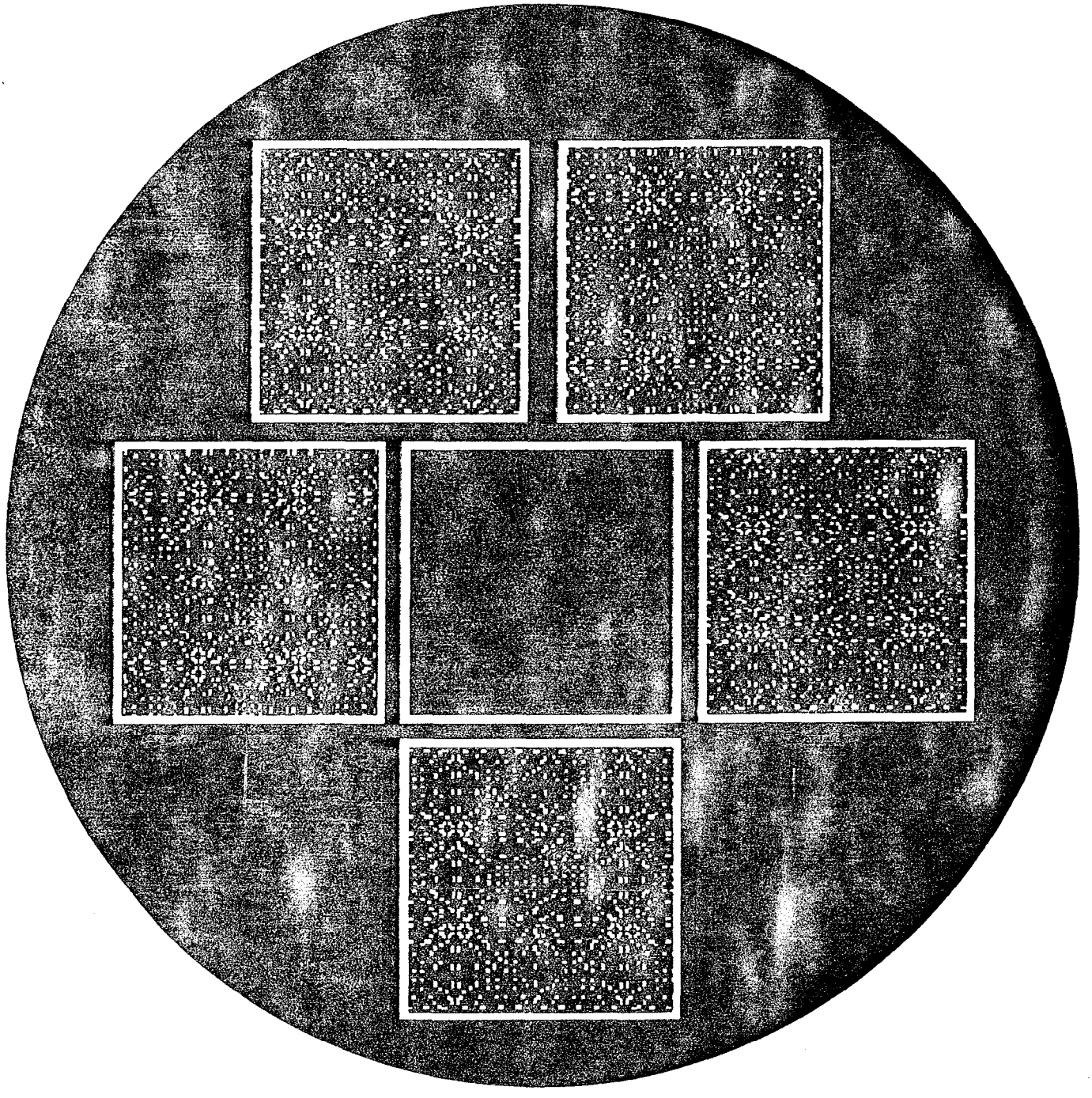
Nuclear Criticality Safety

Theory and Practice

Ronald Allen Knief



Prepared under the direction of the
American Nuclear Society with support from the
U.S. Nuclear Regulatory Commission

NORM PRUVOST
JUNE 3, 87

NUCLEAR CRITICALITY SAFETY

Theory and Practice

Norm—
With appreciation for
a fine presentation at the
'87 Short Course. May our
friendship continue to
grow.
Ron Knief
6-3-87

As new reference materials on nuclear criticality safety are identified by the author, an updated list is prepared. Distribution is announced in the newsletter of the ANS Nuclear Criticality Safety Division and on the computer bulletin board of Lawrence Livermore National Laboratory's Nuclear Criticality Information System (NCIS).

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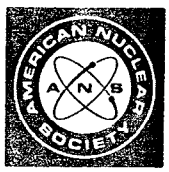

AMERICAN NUCLEAR SOCIETY
La Grange Park, Illinois USA

Dedicated to

Hugh Paxton and Dixon Callihan,

pioneers in the field of

nuclear criticality safety.

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Publisher's Foreword

The American Nuclear Society is pleased to publish this book on nuclear criticality safety. While there are a variety of existing proceedings, reports, and manuals on the subject, this is the first book. It deals with not only the theoretical bases for criticality safety, but also details practical applications in use today at facilities across the United States.

In addition to those individuals and organizations mentioned in the author's Preface, special thanks go to the U.S. Nuclear Regulatory Commission for its financial support.

W. Michael Diekman
Manager, ANS Publications

Preface

This book is intended to provide a comprehensive description of the nuclear criticality safety discipline. It addresses both the underlying bases in nuclear theory and a variety of innovative and practical in-plant applications. Consistent with its origins, the book is particularly well suited as a text for a basic short course on the subject or to provide a needed overview for a newly hired criticality safety specialist. It has also shown itself to provide a useful review and update for long-time practitioners. Although it is intended primarily for engineers, portions are also directly applicable to other responsible management and administrative personnel.

The history of this effort is closely tied to that of the University of New Mexico (UNM) Nuclear Criticality Safety Short Courses. The 1973 course, developed and its proceedings edited by Doug O'Dell, demonstrated significant interest in and support for the subject area by the nuclear community. This first course, however, turned out to be more of a multi-level seminar or conference than a true short course. Thus, in fall of 1974 when I took over the courses as a faculty member of UNM's Department of Chemical and Nuclear Engineering, my goal was to develop a program that would be responsive to the needs of those who are entering or have recently entered the discipline. The May 1975 Nuclear Criticality Safety Short Course and five successive offerings through 1983 have honed this concept well. The need for a comprehensive text was apparent from the outset.

Initial concepts and a brief topic outline for a book developed from discussions with B&W's Fran Alcorn and Fred Welfare, although actual writing was not pursued at that time. The impetus to begin the project was provided by Prof. Dean Eckhoff from Kansas State University when he spearheaded a project to develop nuclear fuel cycle education modules. With the help of project funding from National Science Foundation contract SED 75-04822 and U.S. Department of Energy (DOE) contract EY-76-5-09-095Z, I completed a draft of "Module 6: Criticality Safety" in summer 1980 and it was edited and finally issued by Eckhoff in December 1981.

Using the module as a starting point, roughly a year-and-a-half's worth of effort produced a draft book manuscript. Module or book chapters were audience-tested in a 1980 UNM graduate course on safety methods, the 1981 and 1983 Nuclear Criticality Safety Short Courses, a 1982 training session for operators at the Three Mile Island Unit 2 (TMI-2) reactor, and several 1983 DOE training courses on the prevention of significant nuclear events. This experience, input from a pair of comprehensive reviews by each of two reviewers, and a number of subject-matter-specialist critiques, helped to produce an expanded and thoroughly revised manuscript.

The book consists of three major portions. The first two chapters provide a statement of objectives, an overview, and a review of theoretical concepts. The next seven establish the principle methods of criticality safety—accident experience, Standards, experiments, computer calculations, limits, hand calculations, and regulation. The final two chapters examine integration of the methods into individual practices and overall facility operations. Appendix content focuses on specific computer and hand calculation examples, criticality alarm development, regulatory documents, and TMI-2 recovery applications.

Exercises at the end of each chapter have been designed to guide study by focusing attention on key concepts, exercising calculational methods, and occasionally introducing a new concept or otherwise drawing out an additional insight. A recommended approach for a novice to master the subject matter includes review of the exercises, reading the chapter and taking related notes, formulating answers "open book," writing answers "closed book," and comparison of the latter two. (Since the book

emphasizes criticality safety rather than general reactor theory and the nature of the fuel cycle operations themselves, reference to my *Nuclear Energy Technology* or other books may be appropriate from time to time.)

This book would not have been possible without the help of many individuals. Some of the more significant contributions are acknowledged here. First of all, Dean Eckhoff deserves great thanks for his initiative, perseverance, and patience with the project that got this all started. The ANS Nuclear Criticality Safety Division through its officers, executive committee, and membership provided much needed encouragement.

The participants in the short course programs (and other courses that used draft material) shared both their technical insights and, perhaps more importantly, their enthusiasm for the book. One particularly valuable perspective was provided by Bob Lidstone in a paper detailing a Canadian view of U.S. regulatory practices. Former UNM graduate students Bob Busch, Mark Hoover, David Price, and Don Cutchins helped work out hand calculation examples or reviewed early drafts of the text.

The faculty members from the short courses are largely responsible for my own maturation in the criticality safety discipline; Dave Smith (LANL) provided significant guidance in this regard. Others who have participated in several courses and have reviewed selected chapters include George Bidinger (NRC), Elliott Whitesides (ORNL), Sid Bierman (Battelle Northwest), Joe Thomas (ORNL), and Bob Wilson (ENICO). Additional subject-area support was provided by Jack McLendon (Y-12), Roger Carter (Rockwell Hanford), John McCarthy (Rockwell Rocky Flats), Bud Crocker and Jerry Roth (NRC), Bill Waltz (AGNS), Ken Elliott and Bill Jensen (DOE), and Marilyn Weber (ANS).

Special thanks goes to those who helped to write appendix sections—Ivon Fergus (Bechtel), Fred Welfare (GE, formerly B&W), and Martyn Evans (BNFL). By having each reviewed two entire drafts of the project, Hugh Paxton (LASL, retired) and Prof. Jack Courtney (LSU) have made invaluable contributions. Professor Bob Seale (U. Arizona) has been involved from the earliest days of the nuclear fuel cycle education modules through his role as continuing technical contact for publication by ANS.

The actual preparation of the various drafts and the final manuscript would not have been so readily accomplished without the skill, energy, dedication, and patience of Karen Reist and Norma Hummel. Graphic support from Ken Winters, Mike Boncoddo, and Marcia Moore and photographic assistance from Alan Brinser have added greatly to the visual qualities of the book. Mike Diekman and Lorretta Palagi from ANS have made the publication process surprisingly smooth. Mike guided the early manuscript over the hurdles required for ANS to accept it for publication and then saw it through to completion. Lorretta has combined excellent editorial skills and instincts with great patience and a talent akin to mind-reading to convert the manuscript into this well-produced book.

Finally and most importantly, I thank my wife Pam for her love for me and for her understanding support of this project (which admittedly did not always come without some effort). Her contribution has been monumental considering that the final sustained writing effort began a few months after we met, took up the better part of three years' worth of weekends, and did not conclude until almost a year-and-a-half after our marriage.

Ronald Allen Knief
April 1985 Middletown, Pennsylvania

Introduction 1

Goals and Objectives

The goal of this book is to provide the reader with a basic understanding of the need for and implementation of nuclear criticality safety in fuel cycle facilities. Specific objectives include the ability to:

1. Define nuclear criticality safety and identify basic safety principles that are applicable to it.
2. Explain the features of and lessons learned from the criticality accidents that have been reported in process facilities.
3. Define a Standard and describe the content of ANSI/ANS-8.1/N16.1 and several others that address nuclear criticality safety.
4. Describe the roles of benchmark experiments and of the transport theory and Monte Carlo calculational methods in assessing criticality.
5. Define "subcritical limit" and explain its relationship to process limits for fuel facility operations.
6. Use hand calculation methods to assess the criticality safety of single units and arrays of fissionable materials.
7. Describe the role of licenses issued by the U.S. Nuclear Regulatory Commission (NRC) and orders issued by the U.S. Department of Energy (DOE) in the practice of criticality safety for U.S. fuel cycle facilities.
8. Identify unique devices and practices designed specifically for implementation of nuclear criticality safety in fuel cycle facilities.

Overview

The primary purpose of nuclear criticality safety (or simply criticality safety) is to prevent nuclear chain reactions in operations with fissile materials outside of reactors. As with other safety-related activities, it must balance risks with costs and benefits.

This book examines the wide range of activities that constitute nuclear criticality safety. In many respects, criticality safety may be thought of as a microcosm of the nuclear industry since its theory and applications draw from almost every individual discipline of the conglomerate known as nuclear engineering. The following paragraphs highlight some of the more important topics of the chapters that follow.

A phrase like "production without undue risk to the public or the environment" is commonly used in describing an important requirement for all operations within the nuclear industry. Although production is emphasized, minimization of the associated risk is recognized as a necessity. Nuclear safety in fuel cycle facilities encompasses both nuclear criticality safety and radiation protection activities. General industrial safety and fire protection are also important adjuncts to overall facility safety. Each safety discipline tends to focus initially on the plant operations themselves, but then must interact effectively with the other safety activities. Good safety programs are an integral part of facility operations and generally encompass process and equipment design, administrative practices, and personnel training.

It is well recognized that past safety experience is an important guide to current and future operations. Analyses of both successful practice and accident experience are important contributors to overall safety. In criticality safety, a number of very important lessons have been learned from each of the several accidents that have occurred. Many of these and other experiences have been or are being included in the recommendations of consensus Standards.

Criticality safety depends heavily on experimental measurements for determining the compositions and con-

figurations that produce criticality. These results, in turn, are used to establish requirements for subcriticality. Experimental validation and verification of simple "hand calculation" methods and of extensive computer-based methods have added further flexibility to process design and utilization.

The NRC and DOE are charged with overseeing U.S. nuclear fuel cycle operations in the private and government sectors, respectively. The licenses issued by NRC, the orders of DOE, and similar activities by comparable regulatory bodies elsewhere in the world set requirements and otherwise provide guidance for nuclear criticality safety. Regulations and practices for radiation safety, fire protection, and material safeguards and security also affect criticality safety.

Complete fuel facility operations depend on integration of design-based safety features and wide-ranging administrative controls. Basic requirements are determined by the identity, mass, composition, and concentration of the fissile species and the presence of other materials. The features of a fabrication facility that handles only slightly enriched uranium, for example, are very different from those associated with fabrication of mixed-oxide fuels. The spent fuel reprocessing facility, which contains uranium and plutonium in a variety of forms and which must also accommodate highly radioactive fission products, is altogether different from either fabrication plant.

General References

Several of the important general references on nuclear criticality safety are listed below.

1. H. C. Paxton, "Criticality Control in Operations with Fissile Material," LA-3366 (revision), Los Alamos Scientific Laboratory (1972).
2. R. D. O'Dell, Ed., *Proc. Short Course Nuclear Criticality Safety*, Albuquerque, New Mexico, May 1973, TID-26286, U.S. Atomic Energy Commission (1974).
3. J. T. Thomas, Ed., "Nuclear Safety Guide/TID-7016/Revision 2," NUREG/CR-0095 and ORNL/NUREG/CSD-6, U.S. Nuclear Regulatory Commission (1978).
4. *Proc. ANS Topl. Mtg. Nuclear Criticality Safety*, El Paso, Texas, April 8–10, 1980, SAND80-1675, Sandia National Laboratories.
5. *Proc. ANS Topl. Mtg. Criticality Safety in the Storage of Fissile Material*, Jackson, Wyoming, September 9–12, 1985, American Nuclear Society, La Grange Park, Illinois (1985).

Appropriate sections of this book have benefited greatly from these documents and discussions with their authors and editors.

Sources of current information related to criticality safety include the following publications from the American Nuclear Society (the abbreviations following the titles are commonly used when referencing these publications):

1. *Nuclear Science and Engineering—Nucl. Sci. Eng.*
2. *Nuclear Technology—Nucl. Technol.*
3. *Transactions of the American Nuclear Society—Trans. Am. Nucl. Soc.*

The first two are journals published monthly. The third consists of summaries of papers presented at the Society's two annual national meetings and selected other meetings. At each national meeting, the Nuclear Criticality Safety Division typically conducts one general session for contributed papers entitled "Data and Analysis for Criticality Safety" and from one to four sessions for invited or contributed papers concerning specialized topics. *Transactions* summaries through early 1982 that are specifically related to criticality safety and critical measurements have been compiled in:

> B. L. Koponen, V. E. Hampel, "Nuclear Criticality Safety Experiments, Calculations, and Analyses—1958 to 1982: Compilation of Papers from the Transactions of the American Nuclear Society," UCRL-53369, Lawrence Livermore National Laboratory (1982).

A number of these papers and sessions, as well as more recent ones, are referenced in the remaining chapters of this book. (Citations showing a title but no author refer to entire sessions.)

A variety of U.S. government reports related to criticality safety are published by NRC, DOE, national laboratories, and other contract organizations. Those referenced earlier in this chapter and elsewhere in the book may be purchased from the National Technical Information Service (NTIS), U.S. Department of Commerce, 5285 Port Royal Road, Springhill, Virginia 22161. Selected publications from Canadian, European, and other international sources are also handled by NTIS.

Exercises

1-1. Considering summaries from the *Transactions of the American Nuclear Society*, identify examples of both an individual paper and a paper session from each of at least six chapter reference sections in this book.

1-2. The following acronyms are used as designators for technical reports that apply to criticality safety: AEC, AHSB, ARH, BNL, CEA, DOE, DP, ERDA, HW, K, LA, NUREG, ORNL, PNL, SAND, UCRL, WASH, Y. Use the chapter reference sections to identify the organization of origin for at least 12 of these.

Fundamentals 2

Much of the overall character of the field of nuclear criticality safety derives from the unique features of nuclear fission as an energy source. Criticality safety is an interactive part of the nuclear fuel cycle and, as such, employs the theoretical concepts and methods applicable to all fission systems. Of comparable importance, however, are a variety of sound principles drawn from a wide range of safety activities.

Overview

Every nuclear engineer and reactor operator is intimately familiar with each of the three individual words in the term "nuclear criticality safety." Surprisingly few, however, have been aware of the overall meaning. Even involvement with such common activities as reactor fueling and defueling may not have led to recognition that the procedures and equipment benefited from the practice of criticality safety. For this reason it is appropriate to explore the definition for and scope of the field and to examine briefly its history.

Definitions

The present nuclear energy industry is based on the nuclear fission process. When a neutron fissions a nucleus of uranium or plutonium, the reaction produces energy, fission fragments, neutrons, and various radiations.

The very large amount of energy per reaction and the neutrons that can cause a self-sustaining chain reaction are the major advantages of the fission process. The neutrons and other radiations pose a potential hazard to personnel and equipment. Since the fission fragments are radioactive, they can present a long-term radiation hazard.

Nuclear reactors are specifically designed to produce energy from fission by controlling the neutron chain reaction while shielding against the radiation and containing the radioactive products. Fuel cycle operations, however, generally do not provide for control, shielding, and containment. Thus, an inadvertent fission chain reaction may produce harmful radiation-related effects on nearby personnel. It is also possible that the energy generated could result in harm to personnel or equipment. The characterization[1] in Fig. 2-1 was used to emphasize these hazards during training and education following the accident at the Oak Ridge Y-12 Plant (described in Chapter 3).

Nuclear criticality safety has been defined as "the prevention or termination of inadvertent nuclear chain reactions in nonreactor environments." [2] In practice, the first concept—prevention—is by far the primary goal. An equivalent goal, emphasized later in this chapter, is to maintain a state of subcriticality.

Nuclear criticality safety is also defined as the "art

Fig. 2-1. A criticality accident.[1]

of avoiding a nuclear excursion." [3] The use of the word "art" emphasizes that it, as well as all other types of safety, is not subject to strict scientific development. The high degree of dependence on the actions of individual operators and supervisors can lead to a substantial amount of variability in accomplishing the same overall safety goals. In line with an earlier observation, "the art and science of not building a nuclear reactor without shielding, coolant, and control" [4] is also a good definition.

Two other definitions state that criticality safety may be considered "protection against the consequences of a nuclear excursion" [3] or "protection against the consequences of an inadvertent nuclear chain reaction, preferably by prevention of the chain reaction." [5] Here it is recognized that design can provide near-complete mitigation of the consequences of an accident. A properly shielded and ventilated facility, for example, may experience no unacceptable personnel radiation exposure or equipment damage from a nuclear excursion. This latter approach is likely to be costly, however, especially under current regulatory policies, since any accident will initiate an investigation and require corrective action before operations resume. A major impact of a nuclear excursion in any facility is likely to be the loss of production while the facility is shut down for study, decontamination, repair, or modification. The statement of the second of the two definitions recognizes these concerns.

An additional definition combines a number of the features described above. It states that nuclear criticality safety is "prevention or termination of inadvertent nuclear criticality, mitigation of consequences, and protection against injury or damage due to an accidental criticality." [6]

The terms "nuclear criticality safety," "criticality safety," and "criticality control" may be considered equivalent from a practical standpoint. Although it is also called "nuclear safety" at some plants, this term does not provide adequate distinction from radiation safety, or even reactor safety, for most general applications.

Scope

Nuclear criticality safety, according to Alcorn,[4] is a multifaceted discipline with three major components—neutron physics, engineering, and administration. Only with proper interaction among the three can the ultimate safety goals be expected to be met.

Although the underlying principles of neutron chain-reacting systems have been well established in the field known as reactor physics or reactor theory, criticality safety applications are somewhat different. The generally well-defined environment of reactor systems allows criticality, power shapes, control rod worths, and depletion or burnup effects to be computed in relatively straightforward manners. The nuclear criticality safety evaluations by contrast must consider a wide variety of fuel-bearing equipment under diverse conditions of quantity, shape, spacing, chemical and physical form, and presence of other materials. Such concerns have led to development of some different techniques and approaches within the general framework of neutron physics.

The engineering design process determines the quantities and configurations of nuclear fuel constituents, process equipment, and other in-plant materials. Integrated design practices can provide needed processing capacity and quality while reducing the likelihood of criticality for normal activities and credible abnormal events.

Since nuclear fuel processing operations include people, sound administrative practices are of the utmost importance. Appropriate engineering that makes proper operation easier than maloperation can ease the administrative burden greatly. Clear operating rules and procedures, personnel training, and evaluation and feedback mechanisms are among the vital interrelated components of nuclear criticality safety programs.

Integration of the neutron physics, engineering, and administrative facets must be facilitated by a nuclear criticality safety specialist. The designated individual generally supplements personal knowledge and experience with input from other subject-area experts to coordinate development of an overall program that is consistent with production needs. Since the operating organizations have ultimate responsibility for the design and conduct of facility operations, the specialist's role should be more that of a consultant (and, perhaps, catalyst) than a dictator. It is, however, also necessary that the criticality safety specialist have sufficient independence and authority to assure that production considerations do not overwhelm valid safety concerns.

A statement by Brown[7] provides further perspective on the overall scope of nuclear criticality safety:

> Of the many technological and safety disciplines that overlay the nuclear energy industry, criticality safety stands out as unique—unique because it is the discipline that most constrains productivity and is thus viewed as economically counterproductive. The objective of the criticality safety engineer is to impose the limitations necessary to avoid the accumulation of a critical mass of nuclear fuel. His professional inclinations are toward small unit masses of fuel, small-diameter vessels, well-spaced equipment, and predictable process chemistry. The objective of the design engineer, on the other hand, is to meet product quality expectations as safely, efficiently, and economically as possible. His professional inclinations are toward large unit masses, large conventional vessels, close arrangements to conserve plant floor space, freedom from concern about unusual chemistry, or where in his process nuclear fuel may be intentionally or unintentionally diverted.
>
> So over the years, a necessary and mutually profitable partnership has developed between the criticality safety engineer, the nuclear engineer responsible for design, and the engineer responsible for operational activities. Through ingenious, creative, and innovative efforts, most problems brought about by criticality safety

restraints have been solved. No doubt it is because of this close partnership that the nuclear industry has safely and successfully reached its present level of technology. Today we have a reasonably sound criticality safety technology and a methodology that very well complements the needs of the nuclear industry.

History

The history of nuclear criticality safety has been traced by Paxton[8,9] and others.[7,10–13] Its conception coincided with the design and operation of the Oak Ridge Gaseous Diffusion Plant and the Hanford Chemical Processing Plant as elements of the "Manhattan Project" and the development of the first nuclear explosives. Since, by necessity, both facilities had to proceed without reliable criticality information, substantial conservatism was incorporated. Recognition of increased demands on these and other soon-to-be-built processing plants, however, led to the conduct of critical experiments (as soon as enriched uranium and plutonium became available). The need to correlate experimental data to theory and, ultimately, to translate both into plant practices provided the impetus for evolution of the first nuclear criticality safety specialists.

Originally, batch processes with enriched uranium and plutonium employed administrative controls on batch size as the primary means of avoiding criticality. As criticality data were developed and specialists contributed to process design, the use of geometrically favorable equipment, for example, allowed reduced dependence on administrative controls. A lag in application of available technology during a time of expanding production, however, was at least partially responsible for the series of six criticality accidents that occurred in the seven-year period from 1958–1964. As detailed in Chapter 3, the accidents led the industry to recognize the need for more effective criticality controls and, in the process, elevate criticality safety as a discipline to its current level. The impressive, ongoing development of Standards (considered further in Chapter 4) is, perhaps, the clearest indication of the field's maturity.

Relatively broad interaction among nuclear criticality safety specialists began in about 1955 when representatives from U.S. Atomic Energy Commission facilities began meeting on an informal basis to exchange information, discuss common safety problems, and coordinate experimental activities. From an initial participation of 15 to 20, interest grew to the point where the informal sessions would no longer provide an adequate forum. At about this time, major symposia on criticality safety at Karlsruhe, Federal Republic of Germany,[14] in 1961 and at Stockholm, Sweden,[15] in 1965 provided a major international perspective. Then the focus shifted back to the United States and to the American Nuclear Society (ANS) where the success of a 1966 national topical meeting[16] encouraged formation of the Nuclear Criticality Safety Technical Group in 1967. The formation of a full-fledged Nuclear Criticality Safety Division in 1970 completed the rapid establishment of the discipline within ANS. The division, with a sustained membership of slightly over 300, has been very active in providing the major international arena for formal exchange of information through technical program sessions, development of standards, promotion of training and educational activities, presentation of awards in recognition of significant contributions, and encouragement of federal support for needed experimental and other research programs.

Principles of Safety

Nuclear criticality safety is but a single aspect of overall industrial safety in nuclear facilities. The basic principles of safety are just as applicable to criticality prevention as to any of the other concerns.

Roy Reider, former safety director of the Los Alamos Scientific Laboratory, now Los Alamos National Laboratory, was an especially intuitive and articulate spokesman for sound safety practice. Because, in part, of extensive interaction with the nuclear criticality safety community, his observations as he wrote them[17,18] and as reported by others[19–21] provide some valuable insight, which is the basis for the following paragraphs.

Social concern for safety is traced from biblical times, through the industrial revolution, to the present. One notable historical milestone was boiler inspection legislation passed in 1882, which over the next decade cut steamboat explosions significantly. It was the forerunner of the American Society of Mechanical Engineers' boiler codes.

Perhaps most significant to safety were the first workmen's compensation acts in 1911–1912, which imposed automatic payment for employment-related injuries. The resulting tie to economic considerations (direct and through insurance ratings) began to justify safety on a cost reduction basis alone. After World War II, an increasing social awareness of risk developed along with a more general concern for the impact of all technology on life and the environment.

Reider observed that "any new risk tends to be reduced as the technology becomes more widely used, or, conversely, new technologies may have to prove their safety before full acceptance." [17] Applicability was noted for the nuclear industry in general and for the evolution of radiation protection standards in particular. (Specific criticality risks are considered in Chapter 3.)

Reider also delineated seven "fundamentals of safety." [18] They state generally that sound safety practice includes:

1. management leadership in the declaration of policy and assumption of responsibility for control of accidents
2. assignment of responsibilities to operating offi-

cials, safety and health personnel, supervisors, and technical committees

3. establishment of requirements for formal written procedures and their frequent review
4. maintenance of safe working conditions, including inspections by specialists (of cranes, elevators, high-pressure equipment, fire protection devices, etc.), committee inspection, proper purchasing and acquisition, supervisory interest, and other elements
5. safety training for supervisors and employees, which could include first aid, emergency response, review of accidents, technical information, protective clothing, safety fundamentals, and a variety of other specific subjects
6. medical and first aid planning, including preplacement and periodic examination of equipment, treatment of injuries, and health counseling
7. a system for reporting and recording accidents, including near misses or potential mishaps, which can alert personnel concerned to needed protective measures or procedural changes.

Although most of these safety fundamentals are self-explanatory, some of particular importance in criticality safety are expanded below. Specific applications and examples are considered later in the book.

The most important principle in accident prevention is the assignment and acceptance of responsibility at every level of supervision. Upper management must not only establish safety policy but must also provide leadership and resources for its implementation. The supervision closest to the operation must have assigned responsibilities plus the authority to carry them out. Safety "professionals" can provide valuable assistance only if supported by all levels of management and supervision.

Many unsatisfactory accident records can be traced directly to ineffective procedures. To be useful, procedures should be developed at the working level and reviewed by safety professionals and management. Since safety should always be an integral part of an operation rather than something externally imposed, "natural and workable" procedures are of the utmost value. Continuing review by operators, supervisors, and others can assure that a procedure satisfies its intent.

Descriptive reviews of accidents and near-accidents can be very valuable as warning signals of potential deficiencies in processes, procedures, equipment, maintenance, training, or supervision. Accident experience for both a given facility and a whole industry should be a powerful tool in accident prevention.

The training of personnel is important at all levels. Management needs to understand the risks inherent in the various facility operations. Safety officials should be fully aware of state-of-the-art practices for the industry. Supervisors and operators must be trained in general policies and the specific safety-related aspects of their assignments. Training programs for new employees may include formal activities conducted by a personnel department, but will be most effective when safety representatives and supervisors facilitate both classroom and on-the-job interactions.

An alternative formulation of "practical nuclear safety fundamentals" delineated by Paxton,[3] former head of the critical facilities at Los Alamos Scientific Laboratory, recognizes that:

> 1. Safety is an acceptable balance of risk against benefit, it is meaningless as a concept isolated from other goals. It follows that safety should be considered one of the goals of design and operation instead of something superposed.
>
> Although experience has shown that criticality hazards are no more serious than other industrial hazards, controls for balancing criticality risk against benefit are somewhat more stringent than is usual in nonnuclear industry. It is reasonable that there be some allowance for the uneasiness naturally associated with this new type of hazard. But the extreme concept of risk elimination (as implied by any claim that certain controls "assure" safety or "ensure" safety) is dangerously misleading. Dismissing risk as nonexistent can detract from the continuing job of maintaining an acceptable low risk level.
>
> 2. Accident prevention depends upon responsibility for safety implementation (and commensurate authority) at the supervisory level closest to operation, under the general direction and policies set by higher management. Attempts to control detail at a remote level are misguided.
>
> Because of the requirement for governmental regulation, great care is required to preserve this precept in criticality safety. Remotely administered detail discourages the on-the-job alertness required for effective control, because it encourages the attitude, "Someone else is taking care of us."
>
> 3. Safety regulation should be based upon professionally generated standards and should preserve alternative routes to safety objectives. The arbitrary selection of a single route (as by rule) may eliminate the best economic balance or the most convenient scheme.
>
> Inflexible rules hamstring the designer in his traditional search for the most satisfactory way to fulfill his many objectives. The result is to set safety apart from other objectives, and increase the chance of an awkward operation that invites improvisation. Flexibility frees the designer to apply to integrated process design the considerable experience that has accumulated in nuclear industry.
>
> 4. Other things being equal, simple, convenient safety provisions are more effective than complex or awkward arrangements. Similarly, "free" (no cost) contributions to safety should be nurtured.
>
> As an example of this principle, criticality safety is enhanced by arrangements of material and equipment that tend to make proper operations convenient and maloperation inconvenient.

Review of Reactor Theory

A number of simplified methods of reactor theory[a] are useful for calculating criticality in the regular geometries typical of reactors. Although, as noted previously, the varied geometries of fuel facilities limit most direct criticality safety applications, the criticality indices can serve to sharpen perceptions of neutron processes and to suggest forms for empirical correlations of experimental data.

Criticality Indices and Correlations

The state of a neutron chain-reacting system can be described conveniently in terms of its effective multiplication factor. Denoted as k_{eff}, or simply k, it is defined as

$$k_{eff} = k$$

$$k_{eff} = \frac{\left(\begin{array}{c}\text{number of neutrons}\\ \text{in one generation}\end{array}\right)}{\left(\begin{array}{c}\text{number of neutrons in}\\ \text{the just-previous generation}\end{array}\right)}$$

or, alternatively,

$$k_{eff} = k = \frac{\text{neutron production rate}}{\text{neutron loss rate}} \quad .$$

When production and losses exactly balance, k is unity and the system is said to be critical. The three situations of most interest with respect to the multiplication factor are

$k < 1$ subcritical

$k = 1$ critical

$k > 1$ supercritical .

Subcritical systems have zero power or a decreasing power level depending on the initial state of the system. The critical condition is a very delicate balance characterized by a steady power level. Supercritical systems have a growing neutron population and increasing power.

Criticality safety is concerned with preventing both criticality and supercriticality. It seeks to assure that operations with fissionable materials outside of nuclear reactors are always subcritical.

Nuclear reactor design has led to the development of a number of formulations for criticality. The four-factor formula is

$$k_\infty = \varepsilon p \eta f \qquad (2.1)$$

where

k_∞ = infinite multiplication factor
ε = fast fission factor
p = resonance escape probability
η = number of neutrons produced per neutron absorbed in fuel
f = thermal utilization.

Since the infinite multiplication factor k_∞ is the value that k would have in an infinite system (i.e., one from which neutrons do not leak), it is related to k by

$$k = k_\infty P_{fnl} P_{tnl} \quad , \qquad (2.2)$$

where

P_{fnl} = fast neutron nonleakage probability
P_{tnl} = thermal neutron nonleakage probability.

Combining Eqs. (2.1) and (2.2) results in the expression known as the "six-factor formula":

$$k = \varepsilon p \eta f P_{fnl} P_{tnl} \quad . \qquad (2.3)$$

Diffusion theory relates a quantity known as the buckling, B^2, directly to neutron leakage and to the dimensions and composition of a critical system. The material buckling, B_m^2, is a function of the composition while the geometrical buckling, B_g^2, depends on system dimensions. When the two terms are equal, the system they describe is just critical. If $B_m^2 > B_g^2$, supercriticality results because the geometry provides less leakage than the material requires for criticality. The system is subcritical when $B_m^2 < B_g^2$. Expressions for B_g^2 for the most common regular geometries are provided in Table 2-I.

Since diffusion theory tends to overpredict actual critical dimensions, values used in the buckling calculation must be adjusted. An extrapolation length, d, defined such that

$$X_a = X_{dt} - d \qquad (2.4)$$

is generally used to relate the actual critical dimension X_a to the dimension X_{dt} predicted by diffusion theory calculations.

TABLE 2-I
Geometric Buckling For Regular Geometries

Geometry	Buckling B_g^2
Spheres of radius r	$\left(\frac{\pi}{r}\right)^2$
Cylinder of radius r and height h	$\left(\frac{2.405}{r}\right)^2 + \left(\frac{\pi}{h}\right)^2$
Cuboid of dimensions a, b, and c	$\left(\frac{\pi}{a}\right)^2 + \left(\frac{\pi}{b}\right)^2 + \left(\frac{\pi}{c}\right)^2$

[a]Reactor theory, or the theory of neutron chain-reacting systems, is assumed to be familiar to the reader. This section serves to emphasize aspects of special interest in criticality safety. Those not familiar may wish to consult a general nuclear engineering textbook.[22-26] A review of nuclear physics and reactor theory at a level especially appropriate to the following discussion is contained in Chapters 2, 4, and 5 of Ref. 26.

The effect on critical dimensions of a uniform reflector surrounding a unit may often be approximated in terms of reflector savings, δ_r, according to

$$X_r = X_a - \delta_r = X_{dt} - d - \delta_r \qquad (2.5)$$

for actual reflected critical dimension, X_r; unreflected actual and diffusion theory predicted dimensions, X_a and X_{dt}, respectively; and extrapolation length, d.

Examination of Eq. (2.5) shows that the reflector savings augments the general diffusion theory correction (i.e., extrapolation length) to provide the net adjustment for the presence of the reflector. As a practical matter, the extrapolation length d itself may be considered to be the reflector savings associated with a surrounding vacuum. (The use of the symbols d and δ_r for the extrapolation length and reflector savings, respectively, is common in nuclear reactor theory.[22] However, in the "buckling conversion" hand calculation method for nuclear criticality safety, it is customary to label an extrapolation distance δ, which is equivalent to the extrapolation length for a bare system or to the sum of the extrapolation length and the reflector savings for a reflected system; values are assigned according to material and reflection as described in Chapter 8.)

The neutron nonleakage probabilities in the six-factor formula may be developed through age-diffusion theory as

$$P_{fnl} = \exp(-B^2\tau) \quad , \qquad (2.6)$$

where τ is the neutron age and

$$P_{tnl} = \frac{1}{1 + L^2B^2} \quad , \qquad (2.7)$$

where L is the thermal diffusion length.

When these and the previous parameters are developed from general correlations, they can be quite useful for describing basic features of reactor systems. The detailed computer calculations required for design purposes often generate the equivalent of the parameters to aid in the understanding of basic system behavior.

The total nonleakage probability P_{nl} according to the age-diffusion method, is the product of the fast and thermal terms. For a large system, an often valid approximation is

$$P_{nl} = P_{fnl}\,P_{tnl} = \frac{\exp(-B^2\tau)}{1 + L^2B^2} \sim \frac{1}{1 + M^2B^2} \quad , \qquad (2.8)$$

where $M^2 = \tau + L^2$ is the migration area. Thus, the multiplication factor k may be represented as

$$k = \frac{k_\infty}{1 + M^2B^2} \quad , \qquad (2.9)$$

where k_∞ and M^2 are properties of the material and B^2 depends only on the geometrical configuration.

Multigroup diffusion theory models a neutron population as if its overall behavior could be described by the average behavior of neutrons within predetermined energy groups, e.g., one-group theory treats them as a single population, while two-group theory considers the effects of fast and thermal components, respectively. More general formulations are described in Chapter 6. Since diffusion theory does not consider neutron directions explicitly, it is most applicable to large, moderately absorbing, homogeneous systems. Although the appearance of reactors is that of physical heterogeneity, their regular arrangements of fuel, cladding, and moderator provide a certain homogeneity from the standpoint of neutron behavior. Diffusion theory, thus, is often applicable, as long as "effective" region-wise nuclear parameters are computed by more sophisticated means.

By contrast, diffusion theory is seldom applicable to nuclear criticality safety, which usually deals with geometries that are irregular in both a physical and a neutronics sense. Some of the diffusion theory concepts, however, are quite useful after some redefinition or modification. The buckling B^2, for example, is employed as a measure of neutron leakage in a powerful hand calculation procedure for judging the criticality of material in different geometrical shapes, as considered in Chapter 8.

Experiments and calculations have shown it possible to determine a value for M^2 that will make the migration area formulation in Eq. (2.9) relatively exact for a wide range of sizes and shapes of a given material. Thus, it is a common practice in criticality safety to plot or tabulate k_∞ and M^2 as a function of material composition.[27] Then, by calculating B^2 for a desired geometry, it is possible to use Eq. (2.9) to obtain a very good estimate of the multiplication factor and, thus, the state of criticality of the system.

Neutron Balance Controls

Criticality depends on the relative magnitudes of the neutron production and loss mechanisms. In a critical system,

production rate = loss rate

or

production rate = absorption rate + leakage rate .

In typical power reactors, control rods made of neutron-absorbing or "poison" material perform routine adjustments of the balance by changing the absorption term. Critical, supercritical, and subcritical configurations are established for steady power production, power escalation, and power reduction (including shutdown), respectively.

Since the goal of criticality safety is to assure that all operations are subcritical, the ability to adjust the balance

routinely is, of course, not necessary. Any methods that favor some combination of low production and high absorption and leakage may be employed. It is important to recognize that the amount of neutron moderation may have a significant impact on all three terms.

Although none of the terms in the neutron balance is ever completely independent of the others, each is discussed separately in this section. An example of the interactive nature is provided after these preliminary discussions.

The production rate for neutrons depends on the amount and type of each fissionable material present in a system. The fissile nuclides (i.e., those that can be fissioned by neutrons of any energy, such as ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu) are much more important than the nonfissile nuclides (i.e., those like ^{232}Th, ^{238}U, and ^{240}Pu that are fissionable but have fission threshold energies) in terms of neutron production. Thus, removal of fissile material is generally most significant in achieving subcriticality. Limits on fissile mass and enrichment are related to the control of the production term in the neutron balance.

Absorption processes remove neutrons that would otherwise participate in the fission chain reaction. Nonfissionable constituents usually absorb some neutrons and thus may act as neutron poisons. Specific additives (e.g., boron, cadmium, or gadolinium) in solid or solution form may be used to raise absorption significantly. The increased presence of nonfissile constituents can also have a net poisoning effect. Absorption, then, may be controlled by limiting enrichment, controlling chemical composition, and by adding solid or soluble poisons.

Neutron leakage is dependent on system geometry and density. For a given composition and quantity of material, a geometry of increased surface area (e.g., long, thin cylinder or long, wide, and thin slab) or decreased density enhances leakage. Neutron reflectors decrease the net leakage by scattering back neutrons that would otherwise have been lost. Limits on dimensions, densities, and reflection may be employed to control leakage.

Controlling leakage by geometry is especially important to nuclear criticality safety. When a container or piece of equipment cannot hold enough fissionable material to produce criticality regardless of enrichment, concentration, or external reflection, it is referred to as being "geometrically favorable" (e.g., see Fig. 10-5 in Chapter 10). Equivalent terms are "geometrically subcritical" and "subcritical by geometry." The term "geometrically safe" has also been used synonymously but is discouraged since such containers are not necessarily "safe" from criticality if more than one is present.

In facilities or portions thereof that handle only certain types of fissionable material, a geometrically favorable container may be defined with stated restrictions. A light water reactor (LWR) fuel fabrication facility, for example, may employ equipment and containers that would be "favorable" for its slightly enriched uranium product, but would not be favorable for highly enriched uranium or plutonium. However, if neither of the latter species could be expected to be introduced by accident, the designation geometrically favorable is quite appropriate. A container that would be subcritical by geometry only for a given range of concentrations, on the other hand, cannot be considered to be geometrically favorable.

If the density of a given homogeneous sample changes, so does the relationship among the neutron production, absorption, and leakage rates. The leakage effect, however, is dominant in determining the state of criticality of the system. For example, the critical mass of a bare (i.e., unreflected) sphere decreases with density according to the expression[3]:

$$\frac{m}{m_0} = \left(\frac{\rho_0}{\rho}\right)^2 , \qquad (2.10)$$

where m_0 is the initial critical mass, ρ_0 is the initial density, m is the final critical mass, and ρ is the final density. This expression contains the underlying basis of an implosion-type nuclear explosive. Since an increase in density reduces the critical mass, an already critical system would become increasingly supercritical with compression or, conversely, increasingly subcritical with expansion.

Nuclear reaction probabilities (or equivalently neutron cross sections) are highly dependent on neutron energy. Scattering reactions between high-energy neutrons and low-mass nuclei are likely to result in sizable energy changes. Thus, these light nuclei are effective at slowing down (moderating or thermalizing) the initially high-energy fission neutrons. The slower neutrons in turn are often more likely to cause fission in fissile nuclides (i.e., the fission cross sections of such nuclides are large for very low energy neutrons).

When a moderating material is added to a fissionable composition, the neutron multiplication may increase. Reflectors of moderating material can increase multiplication in two ways—by returning neutrons that would otherwise leak and by reducing their energies to levels where the probability of fission is increased. Moderation control of criticality is based on limiting the concentration of solutions, the moisture content of solids, and the presence of reflectors.

The extent of interaction between units that are subcritical when considered individually presents an additional serious concern. The potential for neutrons to leak from one unit only to become a source for another is the crux of the problem. Control may be maintained by limiting the numbers, types, and relative spacings of fissile-bearing process elements such as those in solution vessels, machining operations, storage racks and vaults, and transport carts. The actual or potential presence of interspersed

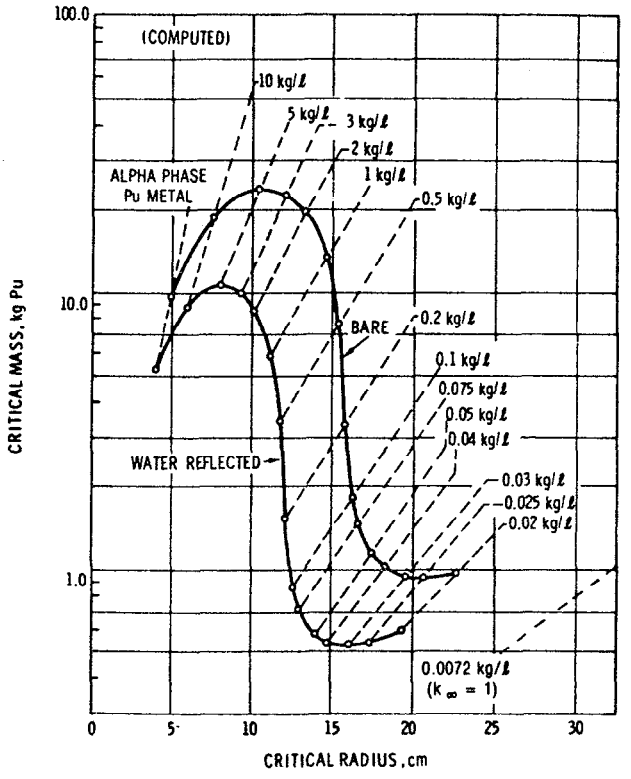

Fig. 2-2. Calculated critical mass and radius for plutonium-water spheres in bare and reflected configurations.[29]

poison and moderator must also be recognized as affecting the system neutron balance.

Criticality Example

The chemical hazard of a water solution can usually be reduced by dilution with more water. With a fissile solution, however, such a dilution may move the system either toward or away from criticality. A given mass of fissile material might produce criticality at as many as three different concentrations as illustrated by the following example from Clayton.[28,29] The results of a series of computer calculations on plutonium critical mass as a function of concentration with water and required size (in this case, radius of a sphere) are shown in Fig. 2-2 for an idealized system of pure ^{239}Pu mixed with water. [Although reality dictates multi-isotope plutonium, dissolution in nitric acid rather than water, and some kind of outer container(s), such differences do not affect the general trends and conclusions that can be drawn from the data on the figure.]

The upper curve on Fig. 2-2 shows mass versus critical radius for a bare system. The corresponding concentrations are shown parametrically. The lower curve contains similar information with a close-fitting water reflector of an "optimum thickness," i.e., thick enough to reflect approximately as many neutrons as would an infinite thickness of water.

The bare critical mass of alpha-phase plutonium metal is 9.8 kg. At a density of 19.6 g/cm^3, this corresponds to a critical radius of 5.0 cm as shown by Fig. 2-2. Initially, dilution increases both the critical mass and radius. The slight amount of moderation reduces some fission-neutron energies to the extent that resonance absorption is increasingly favored over fission; stated another way, the average number of fission neutrons produced per neutron absorbed in fuel decreases. An additional effect of the water is to reduce the density and increase the leakage.

Increasingly effective moderation, however, allows more and more neutrons to escape capture at resonance energies. The effect, as shown in Fig. 2-2, is that the upper curve levels off (near a mass of 23 kg and a concentration of 5 kg/ℓ and then drops sharply. At a density near 25 g/ℓ, an "optimum moderation" condition exists with an overall minimum bare critical mass of ~0.9 kg. This latter value is more than an order of magnitude *less* than that for the pure metal, but the associated radius and volume are ~4 and 64 times *greater*, respectively.

The minimum critical mass is associated with the fuel and moderator configuration that most effectively balances the results of hydrogen's opposing roles in thermalizing neutrons (increasing the probability of plutonium fission) and absorbing them. Dilution by adding more moderator causes the absorption process to become dominant, leading to an increase in critical mass.

The critical mass and radius will both continue to increase with dilution beyond the amount shown in Fig. 2-2. However, the concentration will approach that for an infinite (i.e., zero leakage) critical system. The dashed line in Fig. 2-2 represents a concentration of 0.0072 kg/ℓ, the asymptote corresponding to an infinite critical system. The plot of critical radius versus critical concentration in Fig. 2-3 provides another perspective on the behavior of the system at low fissile concentrations.

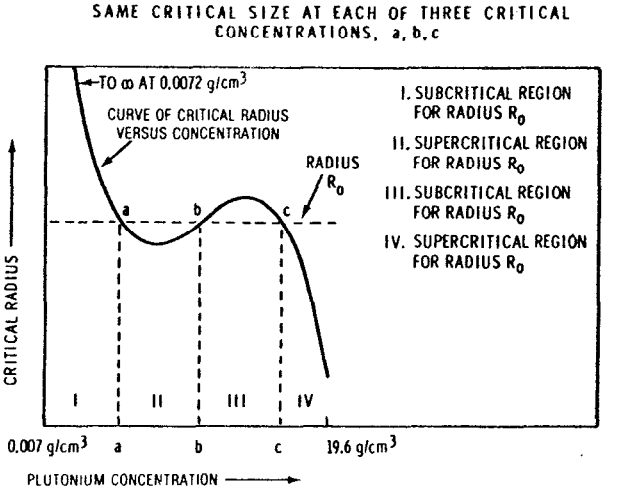

Fig. 2-3. Triple point of criticality for idealized plutonium-water sphere showing the same critical size at each of three concentrations.[29]

The lower curve on Fig. 2-2 shows that the addition of the reflector decreases the metallic critical mass to ~4.1 kg. Since the metal itself accomplishes essentially no slowing down of the fission neutrons, the water reflector is doubly effective by both returning and thermalizing the neutrons. The presence of the reflector may be noted to produce an effect somewhat like that of added dilution to a bare mass, except that it is more effective (i.e., results in lower critical masses at each radius). As the system grows larger and the reflector affects a smaller and smaller fraction of the neutrons, the critical mass approaches that of the bare system.

This idealized example in Fig. 2-2 shows that:

1. the maximum and minimum bare critical masses differ by a factor of >20
2. certain masses can be critical at three different concentrations (or, as shown by Fig. 2-3, certain sizes can be critical at up to three different concentrations)
3. full-water reflection decreases the critical mass substantially for each concentration and each radius shown in Fig. 2-2
4. at high dilutions, the effect of the reflector diminishes as both the bare and reflected systems approach the critical concentration of an infinite critical system.

The complex behaviors in the plutonium-water system are typical of those for other fissile nuclides whether or not triple-criticality points exist.

The interaction of production, absorption, leakage, moderation, and reflection effects complicates the identification and implementation of criticality safety controls. The potential for interactions of bare and fully and partially reflected units with each other is an additional concern.

Reactor Kinetics

Precise criticality depends on both prompt neutrons and delayed neutrons. As noted for reactor systems, control is sufficiently delicate that criticality is not likely to be achieved by accident. Thus, in a fuel cycle facility, an inadvertent "critical excursion" is actually the result of a supercritical configuration.

A supercritical system is characterized by a power level that increases exponentially. The time constant for the increase depends on the amount by which the multiplication factor k exceeds unity or equivalently on the magnitude of the reactivity ρ defined by

$$\rho = \frac{k - 1}{k} \quad . \tag{2.11}$$

When ρ is much less than the delayed neutron fraction β, the time scale of the delayed neutrons tends to dominate and result in relatively slow (seconds to minutes) changes. As ρ approaches more closely to the prompt critical condition of $\rho = \beta$ (often quantified as one dollar, 1$, of reactivity), fewer delayed neutrons are required for criticality and power increases occur with a time constant more comparable to the prompt neutron lifetime (10^{-8} to 10^{-4} s). Increasingly prompt supercritical ($\rho > \beta$) systems respond on time scales that approach the prompt neutron lifetime more closely.

As a supercritical excursion raises the power level and temperature, feedback effects tend to reduce the multiplication. Density decrease, with the greater leakage that results, is the major feedback mechanism for metal systems. If there is a sizable amount of nonfissile material, increased absorption through Doppler broadening of resonance cross sections may also be important to the reduction of neutron multiplication. Expansion (with or without voiding or boiling) is the major feedback mechanism for solution systems.

If supercriticality continues despite these feedback effects, disassembly will ultimately terminate the excursion. Metal systems may break apart or melt. Solution systems are likely to disassemble by splashing or boil to subcriticality.

Criticality accidents such as those considered in Chapter 3 have common features. An initial supercritical excursion usually drives one or more of the feedback mechanisms to return the system to a subcritical state. The result is a power pulse or spike. Figure 2-4 shows a typical

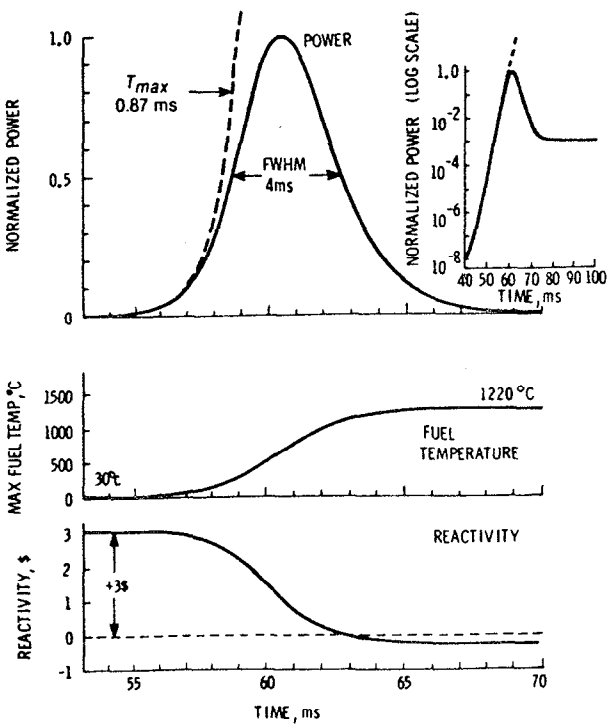

Fig. 2-4. Typical power pulse for the ACRR pulse reactor following a large reactivity insertion. The qualitative behavior of power, temperature, and reactivity is similar to that which would occur in a criticality accident.[26]

relationship among power, temperature, and reactivity as exhibited for a pulse from Sandia National Laboratories' Annular Core Research Reactor (ACRR). The pulse is characterized by a maximum power; a time, usually the full-width in time of the pulse at one-half the maximum power, or the FWHM; and an energy release equal to the integral of the power—in the case of criticality excursions, reported in terms of total number of fissions.

The size and composition of a system at the time of an excursion determine its characteristics. Metallic systems tend to produce pulses of very short duration because the expansion and heat transfer mechanisms are extremely rapid. Solutions tend to have broader pulses. If disassembly does not occur, additional pulses or sustained criticality are also possible.

If the initial excess multiplication is not too great, it is possible for a solution to reach an equilibrium power level. This can occur in a manner similar to the way a boiling water reactor uses variations in coolant density and void content for automatic adjustment of operating power. Under such conditions, a system may fluctuate around the delayed critical condition for an extended period of time until enough boiling occurs to produce a subcritical concentration or until corrective actions are taken.

In the event of a solution excursion, the state of moderation of the system (e.g., as described earlier using Fig. 2-2) affects the strategy for termination. Dilution of an overmoderated system would be expected to produce the desired move toward subcriticality. On the other hand, it could be counterproductive to do the same for an undermoderated system since supercriticality would actually be increased. Solution boiling or splashing is likely to be the more effective shutdown mechanism for the latter situation. Since the cause of an excursion is not likely to be recognized immediately (otherwise, the accident would probably have been prevented in the first place), the appropriate termination strategy may be difficult to identify.

Nuclear Fuel Cycle Concerns

Nuclear criticality safety plays an important role in all nuclear fuel cycle steps where accidental criticality is credible.[30] A generic fuel cycle diagram including uranium and thorium material flows and recycle of ^{233}U, ^{235}U, and plutonium is provided in Fig. 2-5. Steps where criticality safety is required are indicated.

The following paragraphs provide an overview of criticality safety considerations for each of the fuel cycle steps.

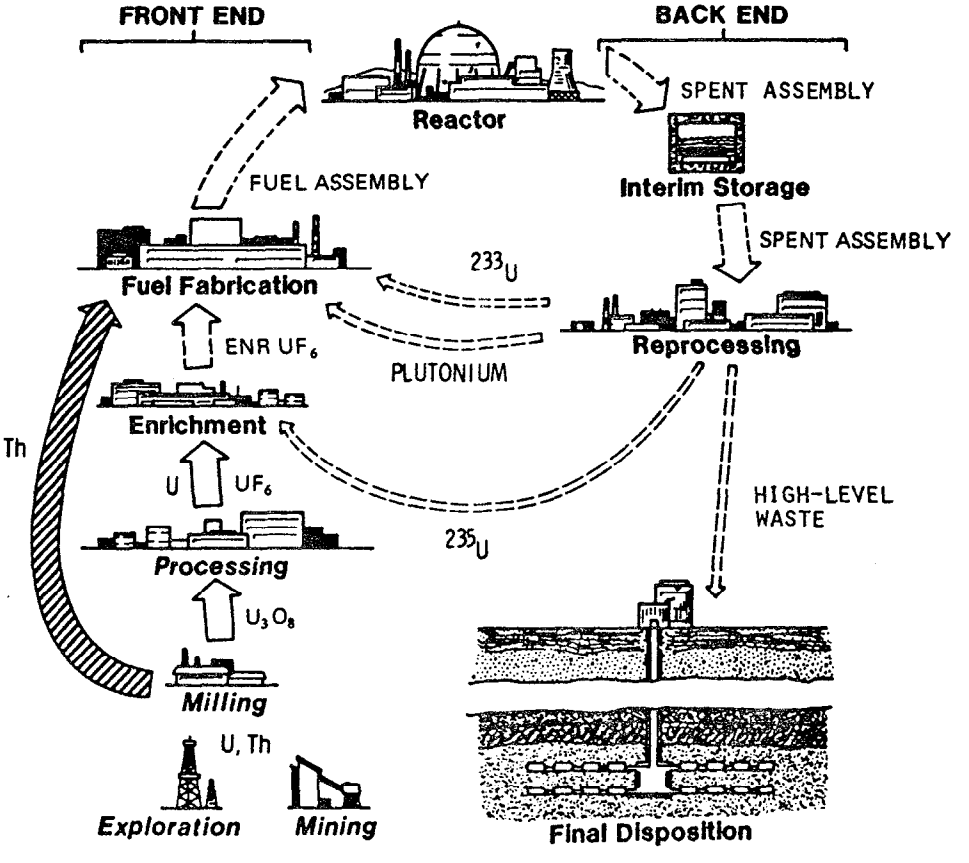

Fig. 2-5. Nuclear fuel cycle operation and transportation steps where accidental criticality is credible are indicated by bold labels and dashed arrows, respectively.

The last chapter addresses some significant features in more detail for LWRs, CANDU, gas-cooled, liquid-metal fast-breeder (LMFBR), and research reactors. (Background information is available from the references on the fuel cycle processes themselves [26,31,32] and the reactor types.[26])

The mining, milling, and processing steps of the uranium fuel cycle have no credible criticality safety concerns. This also applies to fabrication and fuel storage for the CANDU heavy-water reactors and the gas-cooled reactors that use natural uranium fuel. Natural uranium at 0.711 wt% ^{235}U can support a self-sustaining chain reaction only under very special conditions of moderation and geometric arrangement. Although it is theoretically possible for a natural uranium and water system to achieve criticality, the prerequisites of extremely high purity, precise heterogeneous arrangements, and large size could not be met by accident. The other usual moderators—heavy water, beryllium, and graphite—are not present in fuel cycle facilities in sufficient purity and quantity to result in a critical configuration with natural uranium. The thorium stream, being essentially all nonfissile isotope ^{232}Th, by itself poses no criticality concern.

Criticality in the slightly enriched uranium of the enrichment and fabrication steps of the LWR fuel cycle requires water moderation. With highly enriched ^{235}U or ^{233}U for the high-temperature gas-cooled reactor (HTGR), plutonium for the LMFBR, or one or more of these fissile species in other commercial or military fuel cycles, dry criticality is an additional possibility.

Fuel in the reactor itself is generally excluded from the purview of nuclear criticality safety. However, a significant exception exists at Three Mile Island Unit 2 where accident-damaged fuel is to be removed. (Criticality safety aspects of the process are described in Appendix G.)

Fresh LWR fuel assemblies at reactor sites may be stored and handled in air or water in initial full-core or reload batches. Spent fuel is generally stored and handled underwater to provide both cooling and shielding as required by the radiation from its fission product inventory. Maintaining adequate spacing between fuel assemblies is an effective criticality control measure.

The current absence of commercial reprocessing in the United States has resulted in large inventories of spent fuel at some sites with a resulting emphasis on design and use of high-density, poisoned storage structures. Irradiated fuel from other commercial or military sources must also be handled and stored in a manner consistent with providing adequate criticality control, cooling, and shielding.

Reprocessing operations start with spent fuel assemblies, separate fuel material from fission product and transuranic wastes, and separate fuel constituents from each other. While the products of the LWR fuel cycle are slightly enriched uranium and plutonium, cycles that contain thorium produce ^{233}U. The resulting diversity of forms leads to interesting criticality control strategies. However, the shielding and containment required for the fission products already in the fuel could be expected to mitigate the radiological consequences of most potential criticality accidents.

Criticality concerns for high-level reprocessing wastes are relatively minor since the content of fissile materials is intentionally very low. However, long-term concentration effects in disposal media must be assessed. The situation is more extreme if spent fuel elements are the waste form. The latter require controls similar to those for spent fuel storage in the short term as well as evaluations for long-term effects.

Transportation among all appropriate nuclear fuel cycle steps requires criticality controls conceptually similar to those applied to the same materials within the facility. Specially designed containers for each fuel form must also account for the possibilities of accidents, which, for example, could result in substantial geometry changes as well as water flooding with the inherent changes in neutron moderation.

Exercises

2-1. Define "nuclear criticality safety" as:

a. in Standard ANSI/ANS-8.1/N16.1-1975 and its successor ANSI/ANS-8.1-1983
b. an "art" according to Paxton
c. contrasted to "reactor safety"
d. in DOE Order 5480.1A.

2-2. Describe nuclear criticality safety's

a. three components and role of the specialist according to Alcorn
b. unique role in and three principal conflicts with production according to Brown.

2-3. Based on the history of criticality safety, identify:

a. the two initial facilities
b. the principal causes of the early accidents
c. the evolution of the American Nuclear Society's role.

2-4. Compare the observations on safety by Reider and Paxton relating to risk, responsibility, and procedures. Describe the statements of either or both on the subjects of training, accident reporting, standards, regulation, and proper operation.

2-5. List the three terms in the neutron balance equation and identify at least two methods by which each may be controlled for purposes of nuclear criticality safety.

2-6. Define "geometrically favorable" and explain its

importance in criticality safety. Explain why the term "geometrically safe" sometimes used in its place is generally considered misleading.

2-7. Referring to Fig. 2-2, describe the characteristics of the plutonium-water system at each of the three critical points where a line at 10 kg would intersect the curve. Describe how concentration or dilution would affect the critical state of the system at each of the points; relate this to the concepts of undermoderation and overmoderation.

2-8. Explain the physical phenomena that result in the typical nuclear excursion pulse shape, e.g., as shown in Fig. 2-4.

2-9. Considering a generic nuclear fuel cycle, e.g., as in Fig. 2-5, explain:

a. why the mining, milling, and processing steps do not have criticality safety concerns
b. at least one unique feature from the standpoint of criticality safety for the CANDU, LWR, HTGR, and LMFBR reactor designs
c. the range of criticality concerns that could be associated with various final waste management options.

See also Chapter 11.

2-10. Use the age-diffusion model to estimate:

a. k_{eff} of an individual pressurized water reactor (PWR) fuel assembly
b. k_{eff} of two PWR fuel assemblies (side-by-side)
c. the number of fuel assemblies required for criticality (if treated as a cylindrical arrangement of fuel pins with the properties of the individual assemblies).

Assume $k_\infty = 1.38$, migration area $M^2 = 60$ cm^2, reflected extrapolation distance $d + \delta_r = 7.5$ cm (see also Table 8-I), and the assembly is 20 × 20 cm in cross section and 5 m in length.

2-11. Consider a 20-cm-radius critical sphere. By equating bucklings, determine the equivalent dimensions (i.e., those that lead to the same leakage) for:

a. a cube
b. a cylinder with height-to-diameter (H/D) ratio of 1.0
c. a cylinder with H/D of 0.1
d. a cylinder with H/D of 10
e. a slab infinite in extent in two of three dimensions.

Neglect extrapolation distances.

2-12. Repeat exercise 2-11 applying extrapolation distance $d = 2$ cm to each dimension. (See Table 2-I and also Table 8-I.)

2-13. Repeat exercise 2-11 applying a reflected extrapolation distance $d + \delta_r = 6$ cm to each dimension. (See Table 2-I and also Table 8-I.)

2-14. Compare the results of exercises 2-11, 2-12, and 2-13 in terms of the volumes associated with each of the shapes.

2-15. Consider a 10-kg critical mass. Calculate the new critical masses associated with 10% density decrease and increase, respectively. If the initial density is 20 g/ml, calculate the new critical volumes.

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Criticality Accidents 3

The evolution of present-day nuclear criticality safety practice has been heavily influenced by previous experience. The several accidental criticality excursions described in this chapter are one important facet. The more positive experience of accident-free operations is the other input. Both have contributed to the development of the professional Standards that guide sound criticality safety practice as considered in Chapter 4.

Accident Experience

Stratton[1] has compiled a complete listing of pre-1967 criticality accidents at reactors, critical-measurement facilities, and processing plants. The processing events have also been described—and updated, as appropriate—in Refs. 2 through 9.

The following discussions consider two early critical-measurement events and the eight processing plant incidents. In all cases the impact on the practice of nuclear criticality safety is emphasized.

The first two fatal criticality accidents occurred at Los Alamos Scientific Laboratory (LASL) and involved a 6.2-kg sphere of plutonium being used for critical measurements. On August 8, 1945, an experimenter was hand stacking tungsten-carbide bricks for a reflector (a partial stack is shown in Fig. 3-1) when the last one slipped and

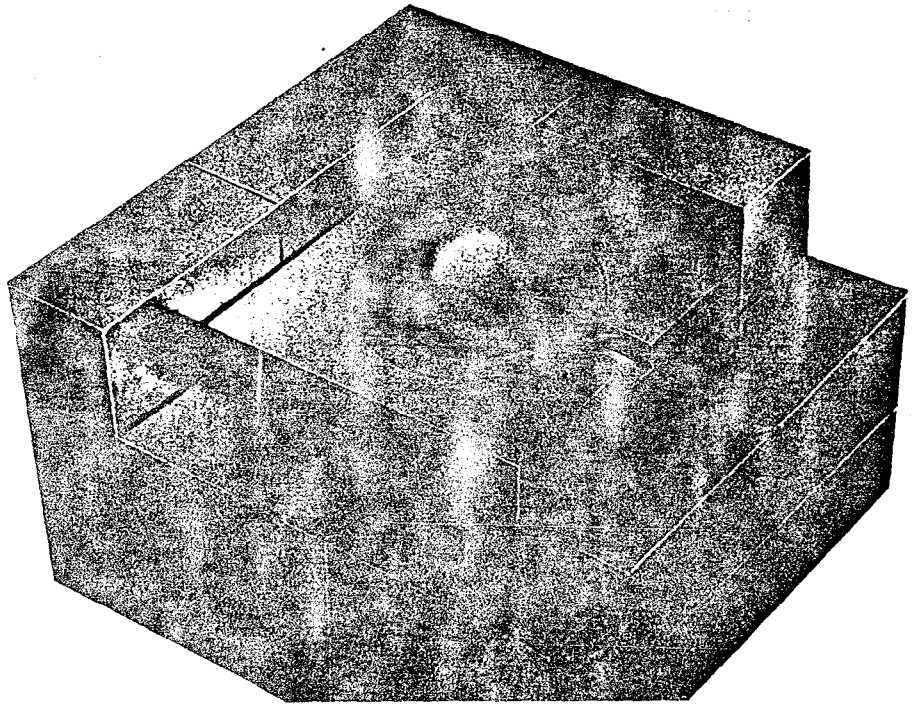


Fig. 3-1. Configuration for the critical-experiment accident at LASL involving a plutonium sphere reflected with tungsten-carbide blocks.

fell on the sphere. The resulting configuration was supercritical and produced a visible "blue flash" from the roughly 10^{16} fissions that accompanied the event. The experimenter unstacked the reflector, put the sphere away, and walked to the hospital. The combination of the excursion and the cleanup activities resulted in an exposure of ~800 rad and his death 28 days later. A guard standing ~12 ft away suffered no lasting injury. (Since these were the first large, acute radiation exposures, they were the subject of intense interest and extensive study.)

The second event occurred on May 21, 1946, when a senior scientist was demonstrating the method for performing a critical experiment to his replacement and six observers. This time the plutonium sphere was partially surrounded by two beryllium hemispheres as shown in Fig. 3-2. The proximity of the reflectors to the sphere was designed to produce criticality. Just before the accident, the scientist had removed the assembly's safety "spacers" and was holding the upper cap slightly open with a screwdriver in one hand and with the other thumb in the cap's center hole (a procedure that is now occasionally referred to as "tickling the dragon's tail," albeit this latter phrase originated with the production of superprompt pulses in the Dragon assembly). When the screwdriver slipped, a supercritical excursion occurred before the shell slid off. All present left the area promptly. The scientist received a dose that proved to be lethal nine days later. His colleague received a large, but nonlethal, dose while the observers received relatively lower doses.

These accidents demonstrated the acute need for remote handling operations, adequate shielding, and proper procedures. Their subsequent implementation in critical-measurement facilities (described further in Chapter 5) has forestalled additional deaths in U.S. facilities despite the fact that more accidental excursions have taken place.[10]

Process Criticality Accidents

There have been eight documented supercritical excursions in chemical process equipment, but none associated with mechanical processing, storage, or transportation. All occurred with aqueous solutions; five involved highly enriched uranium and three involved plutonium. Four of the excursions took place in shielded facilities designed for processing irradiated fuel so personnel were protected from direct radiation.

The consequences of the eight accidents have been two deaths, 19 significant overexposures to radiation, no equipment damage, and negligible loss of fissile material. The general public was not at danger from any of the excursions.

None of the incidents was attributable to faulty criticality information or to error in its interpretation. Instead the cause of each was related to difficulties with equipment, procedural inadequacies and violations, or a combination thereof.

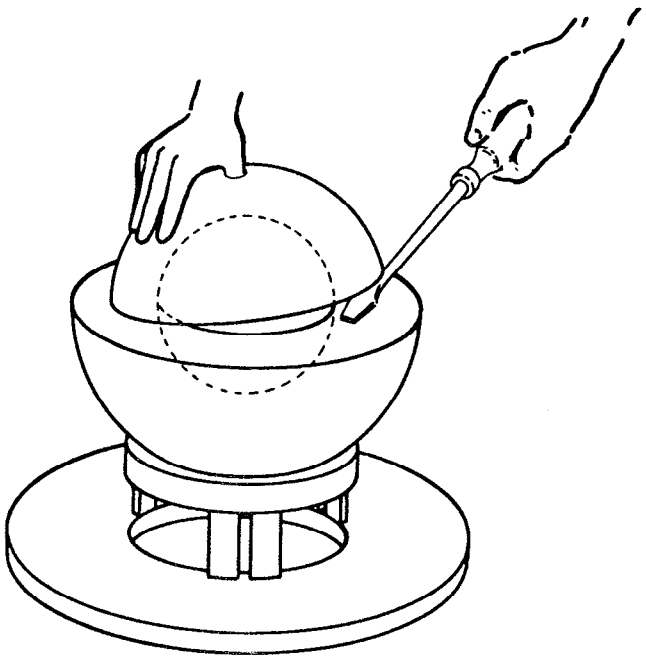


Fig. 3-2. Configuration for the critical-experiment accident at LASL involving a plutonium sphere reflected with hemispherical beryllium shells.

Y-12 Plant

The first of the plant excursions occurred on June 16, 1958, in the Y-12 plant at Oak Ridge, Tennessee. The facility has responsibility for fabricating components from highly enriched uranium.

The accident occurred in an area of the plant where highly enriched uranium was being recovered from scrap. The recovery solution was kept in storage vessels that were designed to be of favorable geometry.

During a material inventory, a bank of storage vessels was emptied, disassembled, and cleaned. They were to be leak-tested with water following reassembly and the water was to be drained into a 208-ℓ (55-gal) drum, as shown in Fig. 3-3. However, before the leak testing, uranium solution accumulated in the manifold under the tanks through

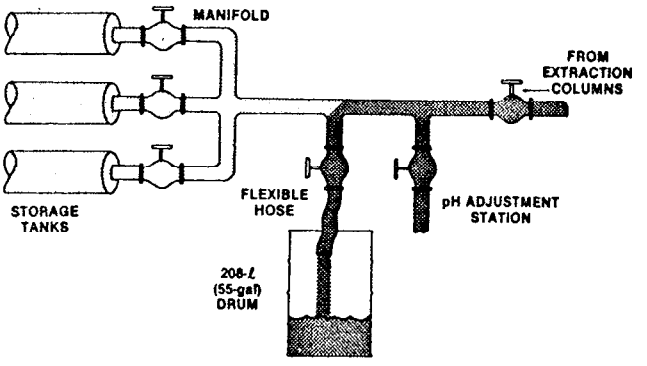


Fig. 3-3. Geometrically favorable storage pipes draining into a large drum—the cause of the Y-12 criticality accident.

a leaking valve that was intended to isolate the tanks from the upstream process operations. The solution was subcritical in the tanks and manifold, but not so when it drained into the unfavorable geometry of the drum and was followed by fresh leak-test water.

Initial criticality occurred with ~2.1 kg of ^{235}U in 56 ℓ of solution. A succession of pulses produced a total of 1.3×10^{18} fissions over a period of ~2.8 min before the continuing flow of leak-test water diluted the solution to a subcritical level. There was no evidence that the multiple excursions splashed solution from the open container.

Eight individuals received significant radiation exposures, which, however, were limited by prompt evacuation. It was reported that the employee, seeing the liquid change color from gray to yellow, recognized a potential problem and began moving away. (Specific radiation doses for this and other accidents are identified and correlated with excursion energy and distance in the last part of this section.)

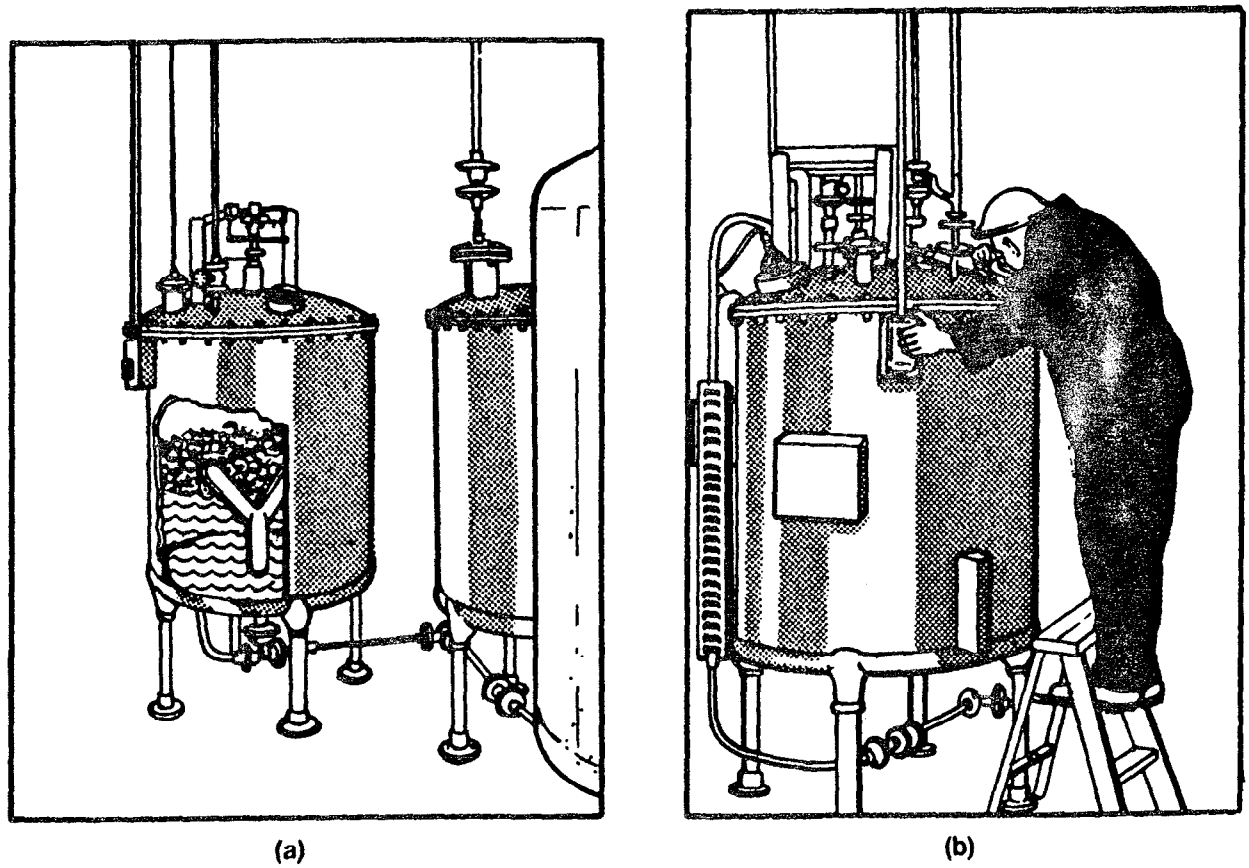
As a result of the Y-12 experience, several corrective measures were adopted by the facility (as well as by the more general nuclear fuel processing community). Transfer lines that could contain fissile solution were disconnected rather than merely being valved-off. Only geometrically favorable containers were permitted in the facility, thereby treating all solutions and other material forms as if they contained highly enriched uranium. Administrative responses included requirements for written operating procedures, a comprehensive accident analysis for and criticality safety reviews of plant operations, and designation of emergency response teams.

Los Alamos Scientific Laboratory

The second excursion occurred at LASL in New Mexico on December 30, 1958. The facility in question was charged with recovering plutonium from various laboratory operations.

The accident occurred in an area of the plant where residual plutonium (typically at ~0.1 g/ℓ) and americium were recovered from dilute raffinate, i.e., from process solutions that could be recycled to the operation after the recovery was complete. Because of the low concentration and a plutonium inventory of only ~0.1 kg, solvent extraction between aqueous and organic phases was conducted in large closed tanks. During a material inventory, it was intended that each tank be emptied and cleaned individually. Instead, residues and acid solutions from four vessels were transferred via interconnecting lines to a single 850-ℓ, 96.5-cm-diam tank as shown in Fig. 3-4a. A critical excursion occurred when the stirrer in the tank was started.

Just before the excursion, the tank contained plutonium solids that had presumably accumulated in the four



(a) (b)

Fig. 3-4. The plutonium-processing tank (a) cutaway and (b) with the operator in position at the time of the LASL criticality accident.[11]

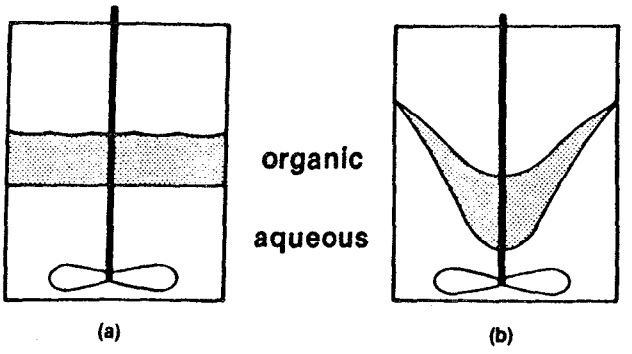


Fig. 3-5. The plutonium-containing organic layer involved in the LASL process criticality accident: (a) initially and (b) as thickened by the onset of stirring.[7]

vessels during 7.5 yr of operation. It also had organic floating on top of the acidic-aqueous solution content of the tank. The great affinity of the organic solution for plutonium (as is, of course, the basis for solvent-extraction with the two immiscible liquids) resulted in extraction of the plutonium to the organic layer shown in Fig. 3-5a. The layer is thought to have been roughly 20 cm thick and to have contained ~3.3 kg of plutonium in 160 ℓ of solution. Although subcritical in its initial disk geometry, the onset of stirring briefly thickened the layer to a supercritical configuration as shown in Fig. 3-5b. Continued stirring mixed the organic and aqueous phases to reduce the plutonium concentration and produce a return to subcriticality.

The excursion of 1.5×10^{17} fissions produced a flash that was seen from an adjoining room and activated a radiation alarm 53 m away. The operator, who was looking into the tank through a sight glass as shown in Fig. 3-4b, received a lethal exposure. Two others who sought to aid him received significant doses. There was no contamination or damage to equipment even though the tank support was displaced ~10 mm by the shock.

The entire plutonium recovery plant had been scheduled to operate for only six more months before it was to be rebuilt. Thus, after the excursion, it was retired immediately. The new facility was constructed with geometrically favorable equipment and other corrective measures were adopted. Written procedures and nuclear safety training were improved. Unnecessary solution transfer lines were blocked. Auxiliary vessels of large volume were "poisoned" with borosilicate-glass Raschig rings. Portable survey instruments were employed to detect the buildup of plutonium in various portions of the process. Radiation alarms were installed to warn of possible criticality and signal evacuation.

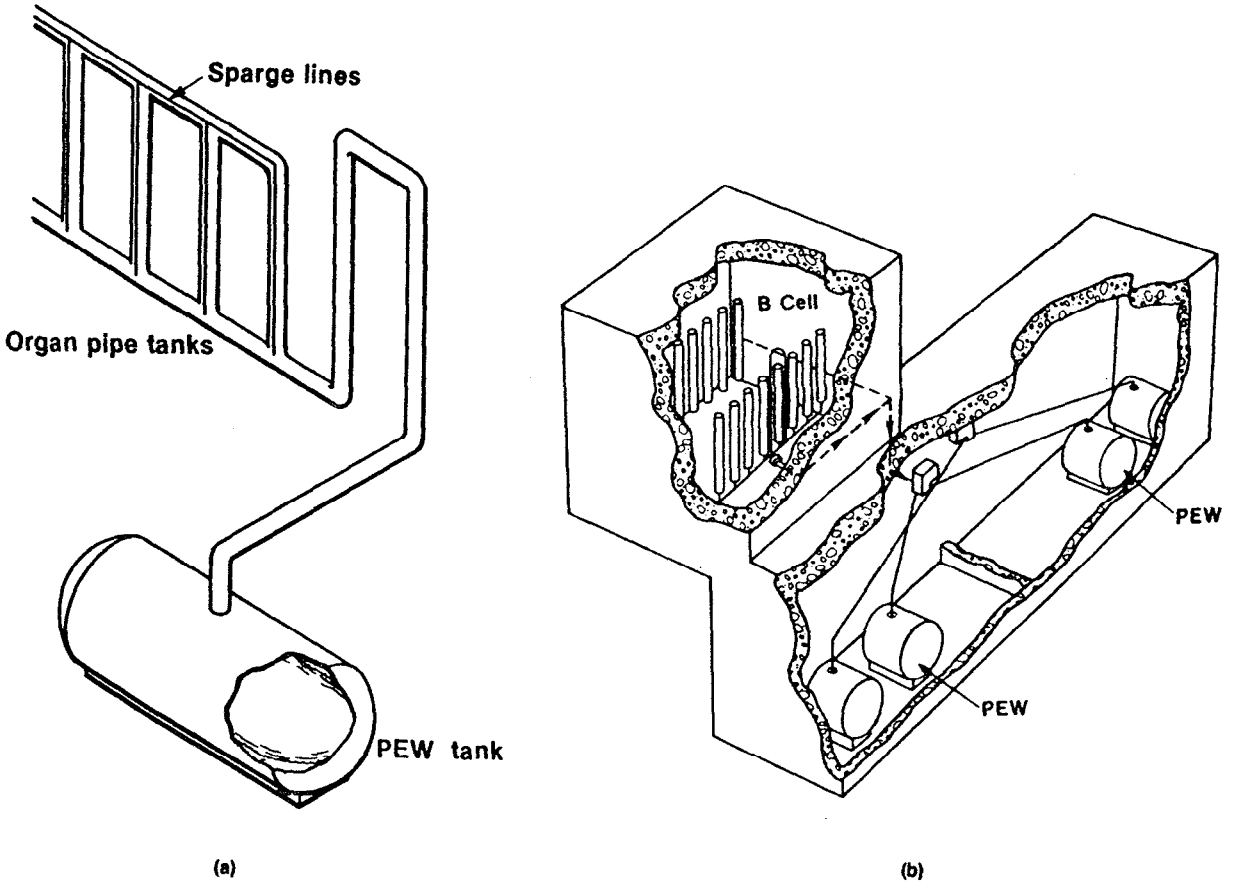


Fig. 3-6. Storage cylinder or "organ pipes" and the PEW tank involved in the first criticality accident at ICPP: (a) basic features and (b) layout cutaway.[7]

Idaho Chemical Processing Plant—First Excursion

The first of three excursions at the Idaho Chemical Processing Plant (ICPP) at the National Reactor Testing Station (now the Idaho National Engineering Laboratory) near Idaho Falls occurred on October 16, 1959. The facility was designed to reprocess ^{235}U bearing irradiated reactor fuel.

The excursion was a result of inadvertently siphoning highly enriched uranium solution from a bank of geometrically favorable storage cylinders (nicknamed "organ pipes") to a process equipment waste (PEW) tank as shown in Fig. 3-6. On the day of the accident, some of the storage cylinders were nearly filled and thus required sampling before their contents could be removed. Since the pump normally used for recirculation mixing was inoperable, the air sparge line in each cylinder was used instead. Excessively vigorous sparging, however, caused solution to flow over the siphon break directly to the PEW tank (Fig. 3-6).

Over a period of ~15 min, ~200 ℓ of solution containing 34 kg of ^{235}U were transferred to the 19 000-ℓ (500-gal) PEW tank and mixed with the 600 ℓ of water that was already there. It is postulated that the ensuing excursion caused solution to splash up the sides of the tank and into lines connecting to other tanks. This apparently broke the siphon and stopped further drainage. However, as the solution flowed from the lines back to the tank, additional fission pulses are thought to have occurred and to have been followed by more or less stable boiling for ~20 min. After nearly 400 ℓ of water boiled off to another tank, the system became subcritical.

A large total yield of 4×10^{19} fissions resulted from the combination of the large solution volume and the long duration of the excursion. The initial pulse itself was thought not to have been especially intense, consisting of $\sim 10^{17}$ fissions.

Since the facility had heavy shielding for handling irradiated fuel, personnel were well protected from direct radiation exposure. Fission products vented into a working area along the evacuation route did result in some beta exposure to two workers. There was no equipment damage.

The incident disclosed the need for improved evacuation procedures and demonstrated the value of radiation alarms in areas that might be affected by an excursion occurring elsewhere. Equipment and procedures were modified to establish several lines of defense against inadvertent transfer of fissile materials.

Idaho Chemical Processing Plant—Second Excursion

The second excursion at the ICPP occurred on January 25, 1961. The location was an evaporator designed with a geometrically favorable 12.7-cm-diam working section. However, it also had a 61-cm-diam vapor-disengagement cylinder above the solution level as shown in Fig. 3-7.

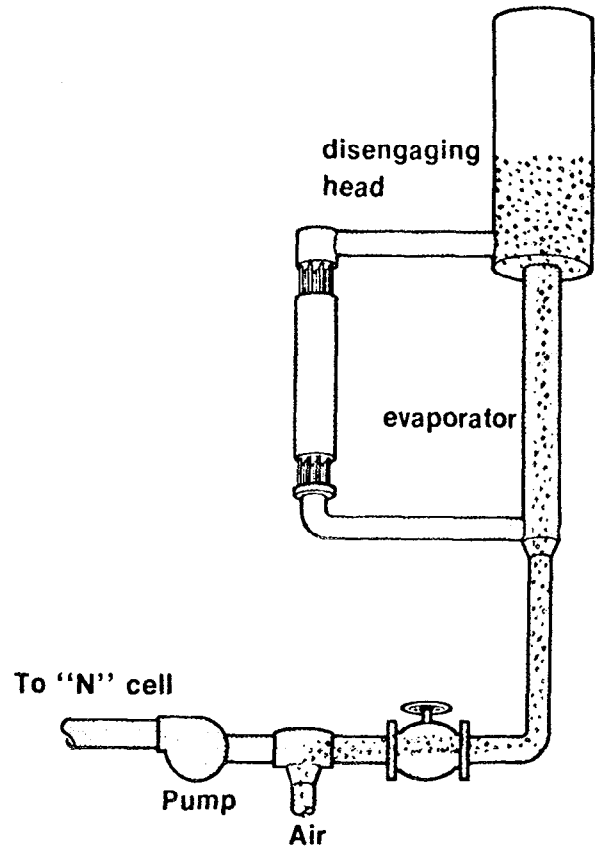


Fig. 3-7. Evaporator involved in the second criticality accident at ICPP.[7]

After several unsuccessful attempts to pump evaporator product solution to a storage bank, pressure was applied through the line at the bottom of the apparatus (Fig. 3-7) in an attempt to clear the postulated blockage. It is thought that an air bubble forced solution from the evaporator into the larger diameter head section. About 40 ℓ of solution containing 8 kg of ^{235}U produced the excursion. The reaction occurred very quickly as the air bubble passed through the cylinder and allowed the solution to drain back into the lower region and a subcritical configuration.

The incident produced 6×10^{17} fissions but resulted in no substantial radiation doses to personnel because of the shielding present for the operations with irradiated fuel. Fission product release was also prevented by systems installed after the first excursion at this facility.

The overall effect of the excursion was sufficiently minor that recovery operations began within an hour. Afterward, steps were taken to prevent the inadvertent introduction of air into solution lines. Soluble poison was also added to process solutions before they entered the evaporator. The upper cylinder was replaced with one poisoned by a group of stainless steel plates containing 1% boron. (The cylinder was later replaced with a thin slab geometry.)

Recuplex Plant

The fifth plant criticality (within a span of slightly <4 yr since the Y-12 incident) occurred at the Recuplex Plant on the Hanford Reservation, Washington, on April 7, 1962. The facility was used to recover plutonium from various processes conducted on the reservation.

The excursion took place in a solvent-extraction area that was enclosed in a room-sized glove box. A general cleanup operation was in progress for a variety of reasons including to help restore visibility in the 7-yr-old facility. Wash solutions were collected by suction and deposited in a 69-ℓ, 45.7-cm-diam vessel (Fig. 3-8), which was normally used to store a dilute solvent-extraction side stream prior to secondary recovery (in a process similar to that involved in the Los Alamos accident).

Apparently, concentrated plutonium solution overflowed from a geometrically favorable tank somewhere in the room while the cleanup operations were still in progress. It is thought to have collected in the sump from where it then was sucked into the vessel through a temporary line used for the cleanup operations. When the vessel accumulated between 1.4 and 1.5 kg of plutonium in a volume of 46 ℓ, the excursion began. An initial pulse was followed for ~20 min by smaller pulses.

Most descriptions state that general boiling occurred for nearly 37 h before enough water was distilled off to bring the solution to subcriticality.[2] Another attributes the final shutdown to a chemical reaction with the plutonium settling out.[12]

The total yield was 8.2×10^{17} fissions of which ~20% occurred in the first half hour. Since the initial pulse triggered a radiation alarm, prompt evacuation followed. Only three nearby operators received significant radiation exposures.

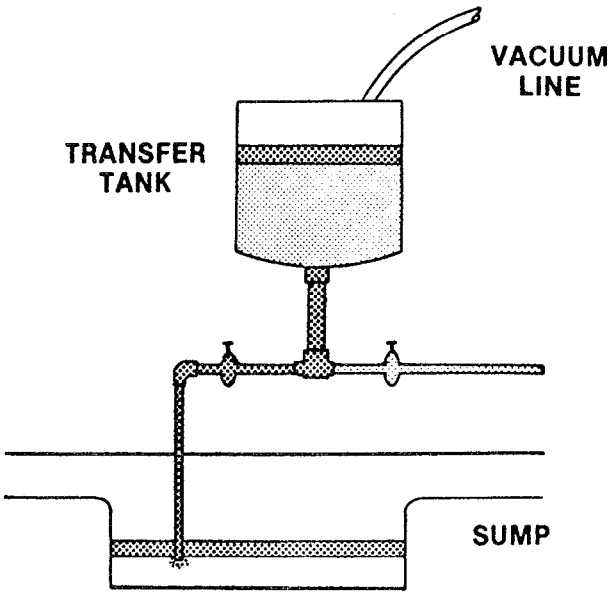


Fig. 3-8. Transfer tank and sump involved in the criticality accident at the Hanford Recuplex Plant.

Operations were never resumed in the original facility. A new plant, which had been authorized before the accident, included design features aimed at preventing similar excursions. Vessels of nonfavorable geometry were usually poisoned with neutron absorbers. The new plant was also more readily adaptable to multiple uses and easier to keep clean. Realistic, up-to-date written procedures were recognized as being especially important to the flexibility needed in the salvage operations conducted by the plant.

Wood River Junction Plant

The only criticality accident in a commercial nuclear facility occurred in the Wood River Junction Plant in Rhode Island on July 24, 1964. The facility was designed to recover unirradiated enriched uranium from solid scrap and solutions generated by fuel fabrication activities.

The facility experienced some difficulties in startup operations, which resulted in the presence of large volumes of trichloroethane (TCE) with low uranium concentrations. Small amounts of uranium were recovered by mixing the solutions with sodium carbonate in 11-ℓ, 12.7-cm-diam bottles and agitating the contents by manual shaking. Since the procedure was quite tedious, an alternative method was improvised whereby the TCE was treated in a 45.7-cm-diam tank intended only for sodium carbonate solution preparation. This procedure was instituted with the knowledge of two of the three shift supervisors but unknown to the plant superintendent and the remaining supervisor. Although not approved, the method would have been safe for the low uranium concentrations initially envisioned.

The cleanout of plugged equipment elsewhere in the plant produced high-concentration uranium solutions, which were stored in 11-ℓ bottles identical to those used for contaminated TCE. Since these latter bottles were unmarked, it is presumed that one was mistakenly emptied into the sodium carbonate solution being stirred in the make-up tank as shown in Fig. 3-9a. The critical excursion that resulted knocked the operator to the floor, splashed ~20 ℓ of the solution out of the tank (some as far as the ceiling ~4 m above), and triggered a radiation alarm.

The final content of the tank (~2 kg of uranium in 41 to 42 ℓ of solution) appears to have been subcritical with the vortex produced by the automatic stirrer. However, when the stirrer was turned off ~2 h later, a second, much less energetic excursion is thought to have occurred as the solution settled to the configuration shown by Fig. 3-9b. The radiation alarm, still sounding after the first excursion, was not able to respond to this second event.

The first excursion appears to have consisted of a single pulse of 10^{17} fissions. One operator received a lethal dose from it. The smaller second excursion, bringing the combined yield to $\sim 1.3 \times 10^{17}$ fissions, caused significant radiation doses for two other workers.

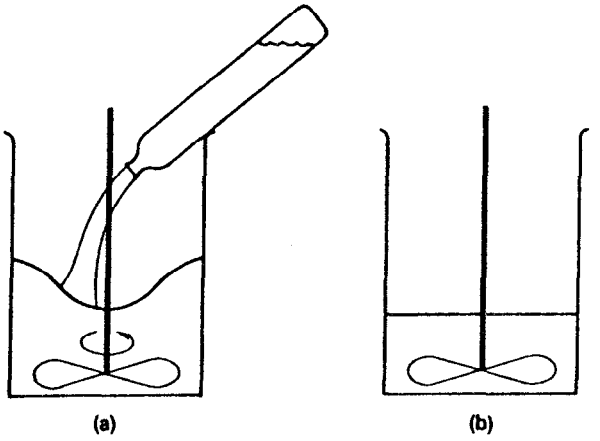


Fig. 3-9. Solution bottle and mixing tank involved in the criticality accident at the Wood River Junction Plant: (a) during solution addition and (b) after stirrer was turned off.[7]

The entire process at Wood River Junction was analyzed extensively before operations were resumed. Emergency procedures, criticality limits and controls, uranium accountability and material balance practices, health physics procedures and controls, and operator training were all reviewed thoroughly and modified. Geometrically favorable equipment for recovering uranium from TCE was put into operation (as had been planned even before the accident).

Windscale Works

The only reported [13,14] criticality outside of the United States occurred at the Windscale Works of the United Kingdom Atomic Energy Authority on August 24, 1970. The facility was charged with processing several types of spent reactor fuel.

The excursion took place at the head end of a solvent-extraction process employed to recover plutonium (in the north cell shown in cutaway in Fig. 3-10a). Under normal conditions, an aqueous solution at 6 g/ℓ of plutonium from a dissolver and a "conditioner" (used for feed adjustment) was raised by vacuum into a transfer tank. Solution then flowed from the transfer tank (Fig. 3-10b) by gravity through a U-shaped trap into a feeder tank that in turn supplied metered solution to geometrically favorable pulsed extraction columns.

When 40 ℓ of organic solvent from an unknown source entered the transfer tank, its lower density caused it to float in a layer on top of the aqueous solution (Fig. 3-11). The depth of the trap allowed the organic material time to float to the surface even when there was some mixing with the incoming aqueous solution. With continuing flow of the aqueous solution, the organic extracted plutonium until the concentration reached 55 g/ℓ of plutonium. It appears that an aqueous-organic emulsion band between the two phases led to an excursion during the brief period after the

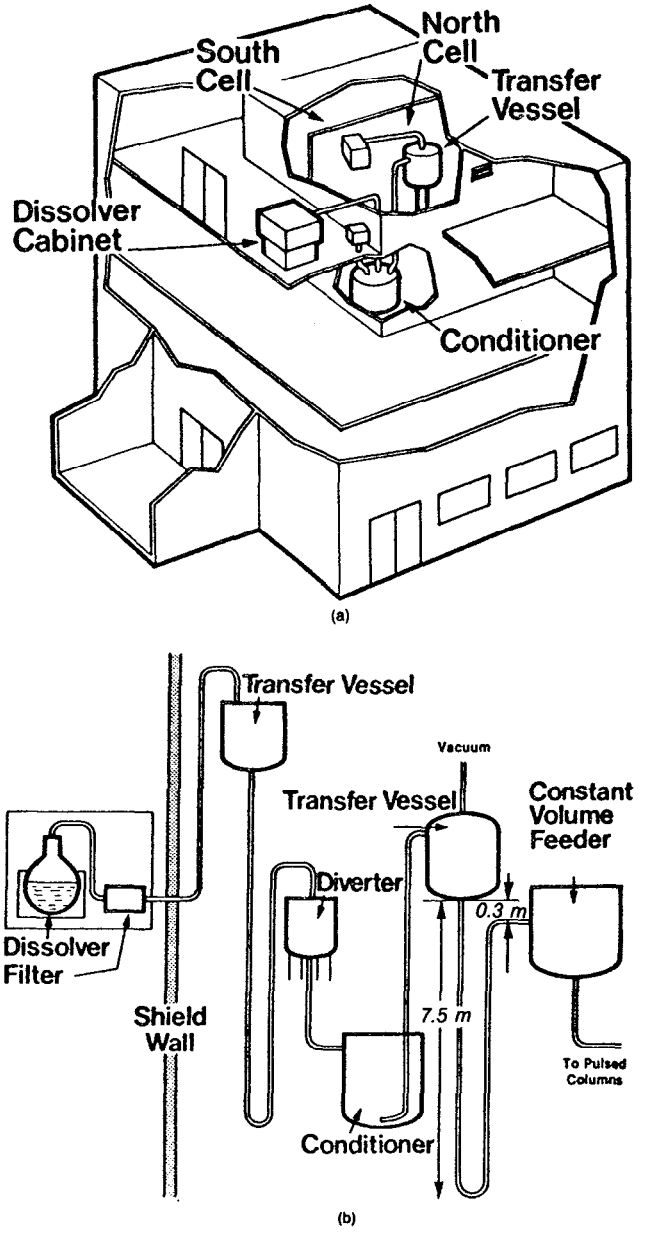


Fig. 3-10. Relative locations of key components involved in the Windscale criticality accident shown in (a) cutaway and (b) schematic representation.[14]

flow stopped and before the emulsion constituents separated as shown in Fig. 3-11c. This sequence of events was later reconstructed and demonstrated by means of an inactive transparent replica of the transfer system.

The excursion produced only on the order of 10^{15} fissions because the excess multiplication of the emulsion band was low and its time of existence short. Although similar in principle to the earlier Los Alamos accident, radiation exposure from this excursion was negligible as a result of the protection afforded by shielding.

Before the plant was returned to service, neutron

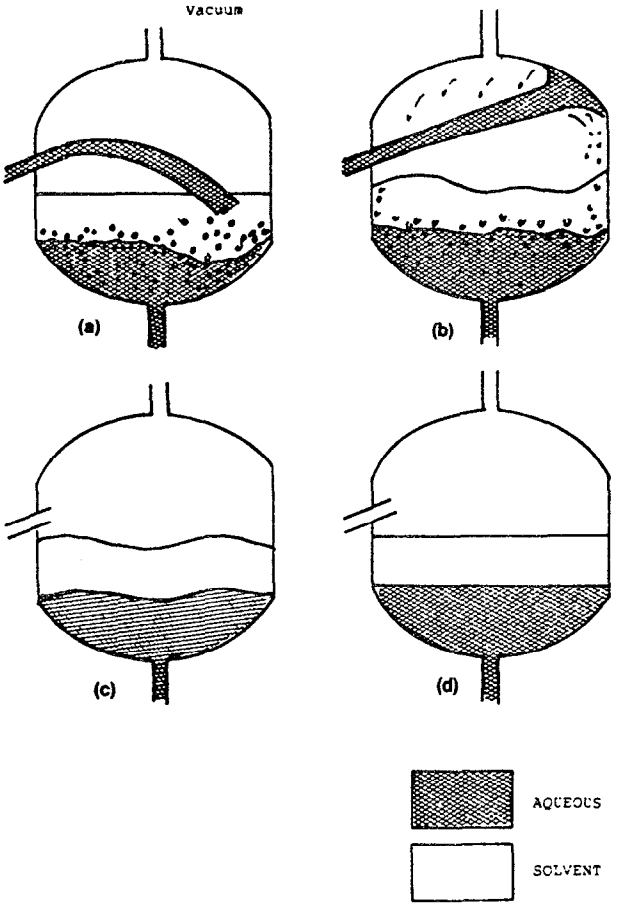


Fig. 3-11. Plutonium-containing organic layer and emulsion layer in the Windscale transfer tank during (a) filling, (b) inrush, (c) separation, and (d) draining. Criticality is believed to have occurred in a configuration similar to that in (c).[14]

monitors for detecting plutonium buildup were installed on all vessels of nonfavorable geometry. The drain traps were also modified to permit positive drainage and to facilitate washout procedures.

Idaho Chemical Processing Plant—Third Excursion

The most recent criticality accident,[7,15] the third at the ICPP, took place on October 17, 1978. It occurred in the first solvent-extraction cycle (shown in Fig. 3-12) where the uranium content of spent fuel is normally extracted, scrubbed, stripped, and washed in separate process columns in order to separate it from the fission products. In the first step, a nitric-acid aqueous solution of the spent fuel ⑤ is contacted with a tributylphosphate (TBP) and kerosene organic solvent ⑥ in such a manner that the organic extracts the uranium and leaves the fission products in the aqueous. The resulting organic stream then enters the scrub column ⑧ where additional fission products and some entrained nitric acid are removed to an aqueous phase ⑨ that is "salted" with aluminum nitrate. Because the scrubbing process removes some uranium, its aqueous phase is recycled ④ to the extraction column. The uranium-rich organic from the scrub column is fed to the next column ⑩ where the uranium is stripped to a weak, non-salted nitric acid stream ⑪. This new, uranium-rich aqueous stream ⑯ is washed of its remaining organic solvent in the wash column.

The specific location of the excursion was the H-100 scrub column shown as part of the cycle in Fig. 3-12 and in more detail in Fig. 3-13. A leaking valve in the water line to the aluminum nitrate makeup tank caused the solution to be diluted from 0.7 to 0.08 *M*. This dilution went

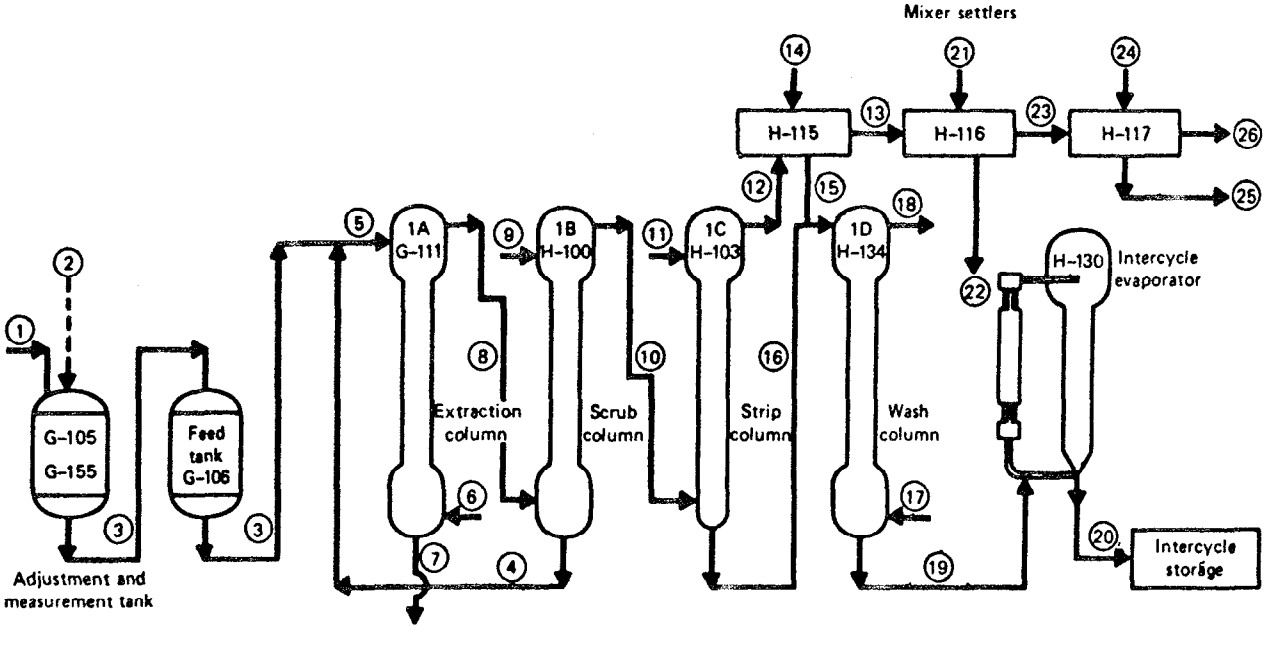


Fig. 3-12. First solvent-extraction cycle for ICPP.[15]

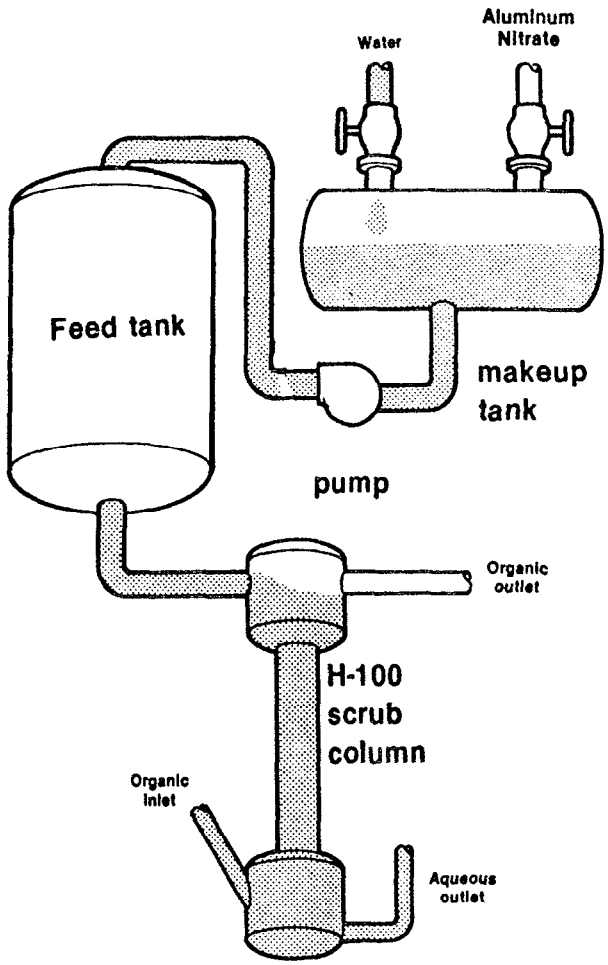


Fig. 3-13. Scrub column H-100 involved in the third criticality accident at ICPP.[7]

unnoticed because a low-density-solution alarm had become inoperable and the latest version of the operating procedure, which called for periodic sampling, was not being used.

The very low aluminum nitrate concentration caused the resulting aqueous solution in the scrub tank to act as a stripping agent rather than for its intended purpose. Thus, as organic solvent moved through the scrub column, much of its uranium content was left behind in the aluminum-nitrate-poor solution. Approximately a month's buildup brought the uranium concentration from its usual 0.3 g/ℓ in the aqueous and 1.8 g/ℓ in the organic to ~22 g/ℓ in the aqueous in the lower head region of the column. This concentration resulted in a configuration that was apparently slightly delayed supercritical over an extended time. The inherent negative reactivity feedback effect of increasing temperature must have been counterbalanced by enhanced uranium extraction for the supercritical condition to have been maintained. Eventual operator action in response to the resulting pressure buildup resulted in a radiation spike, which may have signaled a fission pulse. Ultimately, the excursion probably terminated due to the effect of the operator action, the temperature feedback effects, or both.

The accident produced ~3×10^{18} fissions without any release of solution or damage to equipment. The inherent shielding prevented any substantial radiation exposure to personnel.

Following a formal investigation[15] and before restarting the affected part of the plant, a plant protective system was installed to provide automatic response to specified off-normal process parameters (in much the same manner as power and research reactors are typically operated). Operator training and certification were also greatly enhanced. Plant safety limits were all reevaluated,[16] revised, and incorporated into a technical specification format.

Summary of Consequences

The general features of the eight plant criticality accidents are summarized in Table 3-I. The radiological consequences of the incidents in unshielded facilities may be noted to have been limited by evacuation of personnel alerted by alarms. Especially for the prolonged excursions, evacuation may be credited with saving lives. Radiation-initiated alarms, then, should be installed where there is significant potential for an accidental excursion. Criticality alarms are discussed further in Appendix E.

The two fatalities were suffered by persons within 1 m of an excursion. Significant exposures were received by others at distances extending to 15 m (50 ft). Figure 3-14 generalizes this observation. Personnel doses normalized to a "reference excursion" of 10^{17} fissions and "crudely adjusted to exposure times of 15 s" correlate roughly to source distances[2] as shown in the figure. The center band corresponds to LD 50/30, the range of doses that would be expected to be lethal to 50% of a general population within 30 days of exposure.

Based on Fig. 3-14, it may be concluded that within ~3 m of an excursion, lethal exposures may be expected; at 20 m, the exposure is ~25 rad, a level at which effects are generally not medically detectable. These distances are quite comparable to those considered dangerous for moderate chemical explosions.

General Observations

A large number of changes, recommendations, and observations have been engendered by the accidents, their investigations, and related calculations and experiments. A few of the significant observations, primarily from Refs. 2 and 5, are summarized below.

The concentration of accidents, five in the 1958–1962 period, is partially attributed to increased production of highly enriched uranium and plutonium without growth in the facilities. Plants originally designed for moderate capacity and with minimal criticality safety guidance were

TABLE 3-I
Criticality Accidents in Processing Plants*

Date	Plant	Total Fissions	Fissions	Doses (Rads)	Cause
6/16/58	Y-12	1.3×10^{18}	7×10^{16}	365, 339, 327, 270, 236, 69, 69, and 23	^{235}U solution washed into drum
12/30/58	LASL	1.5×10^{17}	1.5×10^{17}	4400 (fatal), 135, and 35	Plutonium concentrated in solvent layer
10/16/59	ICPP	4×10^{19}	10^{17}	50 and 32 (primarily beta)	^{235}U solution siphoned into tank
1/25/61	ICPP	6×10^{17}	6×10^{17}	None	^{235}U solution forced into cylinder by air
4/7/62	Hanford Recuplex	8.2×10^{17}	10^{16}	87, 33, and 16	Plutonium solution in sump sucked into tank
7/24/64	Wood River Junction	1.3×10^{17}	10^{17}	10 000 (fatal) Two 60 to 100	^{235}U solution poured into tank
8/24/70	Windscale	10^{15}	10^{15}	Negligible	Plutonium concentrated in trapped solution
10/17/78	ICPP	3×10^{18}	Unknown	None	^{235}U buildup due to diluted scrub solution

*From Ref. 6.

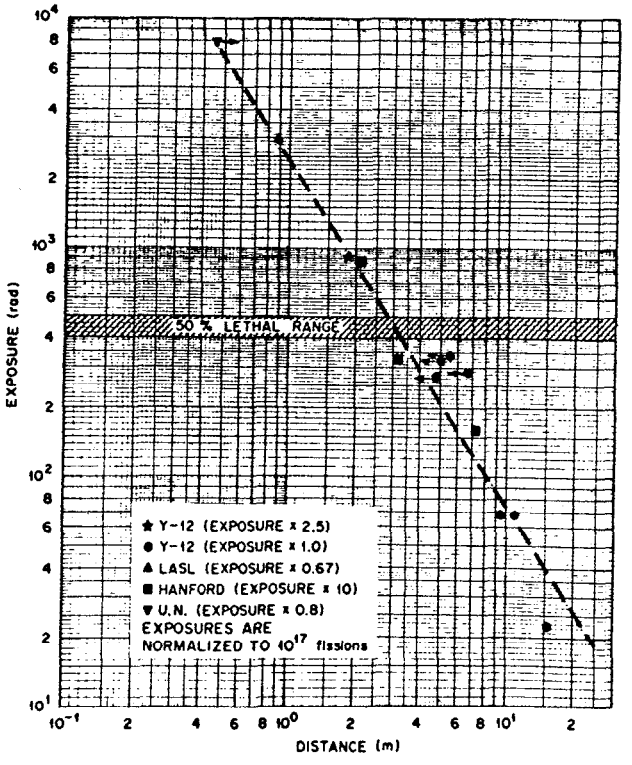


Fig. 3-14. Approximate correlation of exposure with distance from a solution excursion of 10^{17} fissions. Arrows appear where it is believed that available estimates are displaced from the probable values.[2]

used for increased throughput and a wider variety of operations. Thus, the accident potential increased even during the long accident-free period before 1957. There was little incentive, for example, to increase criticality safety and modernize the Los Alamos or Hanford plutonium facilities until after the accidents occurred there. Under the influence of the cluster of accidents, more precise guiding data were collected and techniques for criticality control were refined. Criticality safety became a respected field at this time. Improvement of the accident record was a natural consequence of the transition.

That the excursions have occurred in solutions of highly enriched uranium and plutonium is not unexpected. Small critical masses coupled with the mobility and ease of solution exchange (which, of course, make solutions desirable for chemical processing in the first place) invite criticality in unexpected locations. Solids, by contrast, have larger critical masses and their movement is more apparent and more easily controlled. Criticality control for solids may be enhanced by use of sealed, oversized storage cans present in limited numbers in a given vault. For example, 125 units of 10 kg each of enriched uranium in 20.3-cm-diam × 24.1-cm-deep containers would remain subcritical if tumbled together on a concrete floor (as might occur with an earthquake). Based on energetics, it is more important that criticality control be effective for certain solids than for solutions. Solutions, however, have been noted to present significant and often subtle problems.

None of the processing plant accidents involved the slightly enriched uranium used in light water reactor (LWR) fuel. Even at the maximum enrichment of 4 wt% ^{235}U, both moderation by water and very large solution volumes are required for criticality. For example, the minimum critical volume for an aqueous solution of 4 wt% ^{235}U in the form of uranyl nitrate solution is 100 ℓ at a concentration of 1000 g/ℓ of uranium. This critical volume increases rapidly for decreasing concentration. At concentrations below 400 g/ℓ of uranium (a rough upper limit on

usual working solutions), the critical volume is so large that criticality is essentially unattainable.

Slightly enriched uranium in powder or solid oxide form can be critical under certain moderation conditions. Finished fuel assemblies, for example, would be critical by design if spaced and moderated by water in the manner intended for the reactor core or under certain conditions of wider spacing and low-density moderation (e.g., such as could occur with fire fighting as discussed in Chapter 9). Again, however, the solid forms are readily observed and potential problems are anticipated more easily than for solutions.

The solution accidents that have occurred to date have produced minimal damage to equipment and no radiation exposure to the general public. Sufficient energy to cause widespread dispersion of fission-produced contamination would require unusual circumstances. Properties of excursions with solutions are well illustrated by a series of experiments in a program known as "Consequences Radiologiques d'un Accident de Criticité" (CRAC) conducted at the Valduc Laboratory of the French Commissariat á l'Energie Atomique.[3,17] These experiments, considered further in Chapter 5 along with some more recent experiments, have also provided a basis for calculations on other systems.[18]

Certain types of accidents with solid fissile material (especially ^{235}U metal) are more likely to be violent.[5] It is fortunate that conditions leading to such criticality excursions are generally foreseen readily. Control of conditions, such as large metal pieces falling together, is usually straightforward and can be emphasized in plant design and operations.

Criticality Accident Risk

To quantify the risk from accidental criticality excursions, it is useful to consider accident experience at the facilities operated by the U.S. Atomic Energy Commission (AEC) and its successors, the Energy Research and Development Administration (ERDA) and the Department of Energy (DOE). The simplest comparison is the number of fatalities that have occurred in the various accident categories shown in Table 3-II. Plant criticality produced a single death (recalling that the other such death was in a commercial facility) while two others were associated with the critical-assembly exposures described earlier. The SL-1 reactor explosion caused three fatalities. Each of these ranks with gunshot, poison, and drowning. Risks are seen to be much higher for the other more common industrial hazards such as motor vehicles and aircraft, electric shock, falls and falling objects, burns, and explosions.

Although the favorable record speaks well for the effectiveness of criticality controls, continued attention is required (especially because the public perception of nuclear accident risk has been noted to be somewhat distorted). Maintaining the record as the worldwide demand for fissile material increases will require improved control techniques, especially for process designs and operations. A further discussion of risk assessment is contained in Chapter 10.

TABLE 3-II
Fatalities in Contractor-Operated AEC-ERDA-DOE Plants and Laboratories 1943 through 1979*

Accident Category	Fatalities
Motor vehicle, aircraft	71
Electric shock	23
Falls, falling objects	23
Burns	17
Chemical explosion	12
Asphyxiation, suffocation	12
Drowning	4
Poison	3
Reactor explosion	3
Critical assembly exposure	2
Plant criticality exposure	1
Gunshot	1

*From Ref. 10.

TABLE 3-III
Frequent Elements and Factors in Criticality Accidents in U.S. Processing Plants*

Causal Element or Contributing Factor	Number of Criticality Accidents Involved
Critical configuration of liquids	7
Bulk transfer to unsafe vessel	6
Unintended transfer	3
Ignorance of concentration in intended transfer	3
Valve problems	5
Motive force due to high-pressure air	2
Poor operational communication	≥2[a]
Lack of current knowledge of system configuration	≥2
Development of dangerous routine practices	≥2
Errors of commission by operators	≥2
Errors by supervisors and managers	≥2
Existence of "abnormal" situations	≥2

*From Ref. 10.
[a]≥ means a contributing causal factor in at least two or more accidents.

Exercises

3-1. The information in Table 3-III compiled by Lloyd[10] relates U.S. process criticality accidents and their general causes. Using only the accident descriptions contained in

this chapter, determine independently the number of accidents that fall into each cause/contribution category. Compare these results with those shown by Table 3-III.

3-2. Use the same classification scheme as in exercise 3-1 for the two LASL critical-experiment accidents and the Windscale process accident.

3-3. Considering the order in which the process criticality accidents occurred, list the principle "new" (i.e., those not explicitly associated with any previous accidents) lessons learned from each. Did any accident(s) result in no "new" lessons from an industry standpoint?

3-4. Explain how a vortex in a process solution played opposite roles in accidents at LASL and Wood River Junction.

3-5. Identify the process criticality accidents where

a. criticality alarms saved lives
b. shielding prevented significant radiation exposures to personnel.

3-6. Describe the correlation between distance and radiation exposure from criticality accidents. Explain the hazard comparison between criticality and chemical explosion.

3-7. Calculate the fraction of deaths in AEC/ERDA/DOE facilities attributed to criticality accidents.

3-8. Identify the differences between the fuel material that has been involved in the process criticality accidents and that found in the LWR fuel cycle. Explain the significance of these differences.

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Standards 4

Every person and organization uses formal and informal standards against which performance and achievement can be measured or otherwise assessed. Standards developed and adopted by an entire industry are often particularly effective in promoting sound, yet flexible, practices. The following discussions of nuclear and criticality safety Standards are based primarily on Refs. 1 through 11.

According to McLendon,[2] criticality safety may be thought of as a road that crosses the hazards of criticality on a bridge that, as depicted in Fig. 4-1, rests on support pillars of "technical specifications of flawless quality" and "good reliable practices." The strengths of these supports provide protection from the hazards of accidental criticality. It should be apparent that a failure in either component weakens the structure so that perfection in the other is wasted effort. Thus, it is necessary for each pillar:

1. to incorporate strength and quality into its basic design characteristics and nature
2. to be built with assurance to the same specifications
3. to assure the ability to withstand the trials and tribulations of use and abuse without significant failure.

Although the solution sounds simple, there is a problem. One person's ideas of what it takes to accomplish this may be different from those of others. Even so, the problem is not insurmountable if the interested parties work together and set down the best of their collective knowledge, ex-

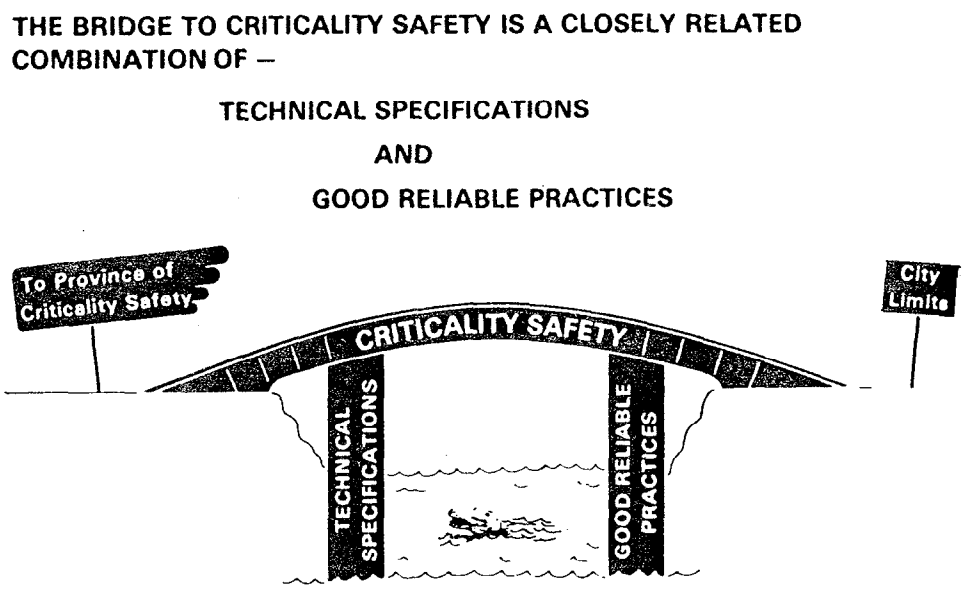

Fig. 4-1. The bridge to criticality safety.[2]

perience, and successful practices. If they can agree on, and use, a set of proven, voluntary, consensus standards, the likelihood of achieving the goal of criticality safety can be greatly enhanced.

General Features

Standards when spelled with a capital "S" imply voluntary industry Standards "promulgated through the American National Standards Institute (ANSI) consensus procedure whereby all interested parties have an opportunity to comment on and contribute to the content in a manner avoiding any conflict of interest."[3] The purpose of a Standard is to set forth acceptable practices, procedures, dimensions, material properties, specifications, or other considerations that have been agreed on by representatives of a broad segment of the subject activity. As voluntary Standards, they are by definition not legally binding (unless otherwise incorporated in local or national laws or regulations).

The management organization of Nuclear Standards has accepted definitions propounded by an international standards organization as[3]:

> *Standardization:* The process of formulating and applying rules for an orderly approach to a specific activity for the benefit and with the cooperation of all concerned and in particular for the promotion of optimum overall economy taking due account of functional conditions and safety requirements. It is based on the consolidated results of science, technique, and experience. It determines not only the basis for the present but also for future development and it should keep pace with progress.
>
> *Standard:* The result of a particular standardization effort approved by a recognized authority.

According to Landis,[4] a past president of ANSI, "Standards are one of the main foundations of modern civilization in that they condense in documentary form the vast technological experience that man has accumulated. This experience has been obtained primarily from commercial operations, not experimentation in laboratories. Thus, standards guide much of what we do each day and determine the nature and quality of much of the materials, equipment and facilities we use." He further notes that properly developed standards produce great economic benefits by:

1. establishing, and gradually raising, acceptable levels of quality, performance, reliability, and safety
2. increasing productivity and reducing overall costs
3. simplifying and routinizing many commercial and technical operations so that maximum effort can be devoted to improvement and innovation
4. minimizing environmental effects
5. expediting licensing
6. providing a rational basis for contracts
7. reducing misunderstandings between suppliers and users
8. expanding both international and intranational trade
9. fostering new applications for existing technology

and that "Standards are not used, except in very unusual circumstances, to introduce new ideas, methods, materials, equipment or facilities. This would be a clear case of 'putting the cart before the horse.' Since the quality of modern life, not to mention modern life itself, depends in large measure on the validity of the standards we adopt, we must be extremely careful to base them on germane experience, rational interpretation and logical generalization. This is especially true since many standards developed by the voluntary consensus standards system are incorporated into codes used by local, state and/or federal government agencies to regulate various business activities."

Historically, efforts such as the standardization of railroad track width greatly enhanced national and international commerce. The development of the boiler codes increased safety, as noted previously in Chapter 2. The use of "ASA numbers" to quantify the light-capturing characteristics (or "speed") of photographic film is the well-known result of another standardization effort. The electrical codes for residential wiring are given legal stature when adopted by municipalities.

Inherent features in the commercial marketplace have brought about such standardizations as the sizes and revolution speeds of stereophonic "records." These same forces are probably working at this time to select a standard from among the several formats for the newer "videotape" and "videodisk" recording materials and equipment, as well as from "floppy disks" used to store microcomputer data.

History and Organization

The American Engineering Standards Committee was founded in 1918 by the five leading U.S. engineering societies to coordinate development of material Standards. Having in the interim been joined by three federal agencies, the Committee was reorganized ten years later and renamed the American Standards Association (ASA). The ASA ultimately grew into a national federation of more than 100 technical societies and trade associations and more than 2200 corporate members. Although it did develop over 2300 standards, the ASA was less productive than might have been expected perhaps because of cumbersome procedures and ill-defined and overlapping responsibilities. Lack of industry and government support, particularly financial, also cut the efficiency and effec-

tiveness with which ASA could handle responsibilities and assignments.

Based in part on the recommendations of a 1963 study by the U.S. Department of Commerce, the ASA was replaced in the same year by a new organization known as the United States of America Standards Institute (USASI). The output of the institute was intended to serve the entire economy by establishing a consensus of applicability on proposed Standards among producers, users, regulators, and others concerned with a product. Concern (probably by the Federal Trade Commission) that the USASI designation could indicate governmental sanction was responsible for a 1969 name change to the American National Standards Institute (ANSI). (If the term American Standard had not previously been copyrighted by a plumbing manufacturer, the seemingly redundant word National would probably not be part of the name.) Currently ANSI is the organization within the United States responsible for promulgation of voluntary Standards and for providing the nation's focus on international standardization projects.

It is important to recognize that ANSI does not write and formulate any Standards. Instead, ANSI has an administrative and judicial function. Standards developing organizations (SDOs), including technical societies, have the duty and responsibility for the actual preparation of Standards. ANSI is charged with the identification of a single consistent set of voluntary Standards. ANSI approval of these Standards is intended to verify that the principles of openness and due process have been followed in the approval procedure and that a consensus of those directly and materially affected by the Standards has been achieved.

ANSI is administered by a Board of Directors to which 16 Standards Boards and several staff groups report. Each Standards Board is concerned with Standards in a particular discipline, such as electrical and electronics, safety, mechanical, photography, and, as is of interest here, nuclear. The membership of each board is comprised of technical, engineering, and professional societies; governmental agencies; labor organizations; and other interested parties.

One especially important ANSI group is the Board of Standards Review (BSR). After the technical content of a Standard is approved by the cognizant consensus committee, the BSR must determine whether there is a general consensus in favor of it and that it meets the many procedural requirements established to ensure fairness and balanced input by all affected parties. The approval process includes a period for public review and comment. Objections must be considered and resolved before final approval as an American National Standard is granted.

This consensus approach is especially important to the nuclear industry as a viable way to attempt to satisfy the multitudinous federal regulations.[4] For it to be successful, it is essential that feedback from the use of Standards be meaningful and continuous. The needed communications are best maintained by having the users serve on the Standards writing committees and by encouraging them to submit inquiries.

Nuclear Standards

The Nuclear Standards Board (NSB) has 25 members, including the National Bureau of Standards, the U.S. Nuclear Regulatory Commission (NRC), and the American Nuclear Society (ANS). There are also some individual members. The routine affairs of the NSB are administered by an Executive Committee elected by and from the NSB. The other constituent units of NSB are its Operations Committee, Planning Committee, and Nuclear Technical Advisory Group. The NSB currently meets biannually.

The function of the NSB is totally administrative. It manages the Nuclear Standards activities by recognizing needs for Standards, assigning priorities, and seeing, generally, to their preparation. It has no judicial powers or responsibilities. The latter reside with the BSR.

Membership on the NSB consists of interests directly and materially affected by Nuclear Standards activities. The interests include SDOs, Standards users, individual members, and other interest categories as may be appropriate.

Each organization that assumes responsibility for development of Standards within the cognizance of the NSB is concerned with a particular element of the nuclear industry. Consensus committees are established to develop evidence of consensus for approval of the Standards within each of their scopes. The technical societies or other SDOs that sponsor the consensus committees are designated as the secretariat for them. The ANS, the Institute of Nuclear Materials Management (INMM), and the Health Physics Society (HPS), among others, serve as secretariats. For example, ANS serves as secretariat for four consensus committees: N16 (nuclear criticality safety), N17 (research reactors, reactor physics, shielding, etc.), N48 (radioactive waste management), and the Nuclear Power Plant Standards Committee; HPS for N13 (dosimetry and radiation protection); and INMM for N15 (accountability of nuclear materials). The consensus committee's function is primarily judicial. It votes on proposed Standards and recommends to the BSR approval of new American National Standards. In short, a consensus committee receives a draft of a proposed Standard, reviews it for technical content, requests revisions as necessary, approves, and recommends. A Standard that has been issued also requires maintenance at least every five years and is reviewed for reaffirmation, revision, or withdrawal.

Any knowledgeable individual or organization who foresees a need for a Standard can make a proposal described by a title and a general scope. After review by the NSB to assure that it is not a duplication, a draft Standard is prepared by experts on the subject who are organized

into a subcommittee or into a small working group. A working group is generally a segment of the Standards effort of a professional, technical, or engineering society. It should be noted that the persons comprising such a group serve as individuals because of their particular expertise, and not as representatives of their employers. The latter is basic to the consensus principle and the need for Standards to be, if anything, impartial within the industry.

When a working group has completed preparation of a proposed Standard, the document is sent to the cognizant consensus committee for consideration. Each member organization of the consensus committee establishes its own procedures for approval. Thus, while some representatives are empowered to vote directly on behalf of their organizations, others must take the proposal back to their standards subcommittee and then vote according to the consensus of that body.

According to Callihan,[5] the BSR evaluates the development of consensus on the proposed Standard, the organizations and individuals involved in the consensus committee, and the basis for any negative votes by members of the latter. If it is then judged that all with an interest in the Standard have been heard and that no proprietary considerations have been violated, the document is approved as an American National Standard.

A description of a typical Standards organization is illustrated by the activities within the field of nuclear criticality safety. The N16 consensus committee is devoted to this subject. ANS is its secretariat while its membership includes the Atomic Industrial Forum, the NRC, the DOE, four technical societies, and two individuals. Although N16 itself meets infrequently, it has been very productive because it processes the output of the active ANS-8 Standards subcommittee, which is described later in this chapter.

Most of the Nuclear Standards currently on the books are intended to standardize[4]:

1. definitions, terminology, symbols, and abbreviations
2. design, materials, parts, equipment, systems, and processes
3. performance characteristics of equipment, systems, processes, and facilities
4. procedures for determining performance characteristics, including reliability
5. methods for testing, analysis, rating, and application
6. safety and health requirements
7. procedures for operating, maintaining, and decommissioning equipment, systems, processes, and facilities
8. methods for selecting, training, and evaluating operators of equipment, systems, processes, and facilities.

All segments of the nuclear industry have recognized the value of Standards. If each activity had to start from basic principles and devise suitable testing and inspection programs for each of the thousands of materials, parts, pieces of equipment, systems, and processes utilized in their facilities, it would be prohibitively difficult, time-consuming, and costly. Without proven practices in the form of Standards, each company or organization involved would need to have extensive experience and a great store of knowledge in the behavior of materials, in the design and quality control of purchased components, and in many other technical specialties in order to make sure that the materials and components produced, purchased, assembled, or operated were adequate for their intended uses. A broad recognition of the imperative need has been the cornerstone of the national Nuclear Standards program, which includes participation by some 4000 individuals "volunteered" by their respective organizations (including government agencies).

Over 3500 standards are now available for use in the design, manufacture, construction, testing, operation, maintenance, and decommissioning of nuclear facilities. These are the fruits of a vast and prolonged labor by more than 50 organizations coordinated by ANSI. The 1980 annual cost of the program was ~45 million dollars, with the industry shouldering almost 80% of it.

International Standards

According to Smith,[6] "international standards provide many virtues, services, and advantages." The development of such standards may avoid, or at least reduce, duplicative effort on the part of many national standards bodies. In particular, for safety standards, which are frequently developed from experience, the broader base existing internationally may be expected to lead to a better product. Consensus standards may also facilitate cooperative international trade and other ventures while enhancing trust and understanding among nations.

Development of international standards is carried out through one of two organizations—the International Electrotechnical Commission (IEC) and the International Organization for Standards (ISO). The IEC is responsible for worldwide standardization in the electrical and electronics fields. The Commission operates through some 70 technical committees, and more than 100 subcommittees. About 100 000 electrical and electronics experts participate annually in its standardization programs.

Smith notes that "the IEC produces consensus recommendations that are intended to provide the basis for national standards and are frequently adopted verbatim for this purpose." Membership in the Commission is held by 43 national committees that represent 80% of the world's population. ANSI serves as the U.S. national committee.

The IEC is financed by dues from the member committees and by the sale of publications.

The scope of IEC Technical Committee 45 is nuclear instrumentation. Subcommittees 45A and 45B deal with instrumentation for reactors and radiation protection, respectively. A working group sponsored by the latter is responsible for developing "Recommendations for Warning Equipment for Criticality Accidents."

ISO has a broader scope[11] than the IEC, essentially being concerned with "everything else." The ISO is comprised of some 160 technical committees, 600 subcommittees, and 1200 working groups. Membership in ISO now consists of 88 countries, each represented by a member body that is "most representative of standardization in its country." ANSI serves as the U.S. member body.

ISO Technical Committee 85, which concerns itself with nuclear energy, has four subcommittees: SC 1—Terminology, Definitions, Units, and Symbols; SC 2—Radiation Protection; SC 3—Power Reactor Technology; and SC 5—Nuclear Fuel Cycle. The last of these, in turn, has seven working groups, one of which (WG 6) has for several years been developing a draft standard for "Criticality Accident Alarm Systems." This effort reached the status of a draft proposal—DP7753—which is being distributed for ballot and, if the result is successful, will become a Draft International Standard for a period of time before being redesignated as an International Standard.

Subcommittee 5 has also acted to initiate a writing group that would address "Standardization of Calculations, Procedures, and Practices Related to Criticality Safety." If this proposal is endorsed by the participating member countries in SC 5, it is expected that the U.S. ANSI will serve as convenor for this new working group.

Close cooperation exists between ISO and IEC, even to the point of sharing a common address in Geneva. Both also have close ties with those agencies of the United Nations that demonstrate significant interest in international standardization. Strong efforts are made to avoid duplication of efforts and to prevent conflicts or inconsistencies between the products of the two organizations. As an example, the chairman of the ISO/TC 85/SC 5 working group developing the standard for "Criticality Accident Alarm Systems" was designated to provide liaison to the IEC working group developing the standard "Recommendations for Warning Equipment for Criticality Accidents." This was very effective in enhancing the consistency of these two complementary efforts.

The role of international standards is gaining increased recognition. Such standards frequently precede national developments in many countries.

Nuclear Criticality Safety Standards

McLendon[2] notes that many years ago the early travelers of the criticality safety road of Fig. 4-1, led by pioneers Callihan at Oak Ridge and Paxton at Los Alamos, undertook the task of providing standard blueprints and materials for a strong bridge to avoid the pitfalls of the criticality abyss. It is clear that the guidance was well founded, although a few travelers have fallen off the bridge. However, after 40 years it can still be said that the accidents that did happen were not the result of failures in the technical specifications, but were the results of human error, which merely proves once more that "Physics is still physics and people are still fallible."

As time progressed and experience was gained, the first steps toward standard practices were taken by initiating the series of Industrial Criticality Safety Meetings described previously in Chapter 2. They began in 1955 and continued until the formation of the ANS Nuclear Criticality Safety Technical Group in 1967. This group began to address standards in a formal way under Callihan's leadership in the late 1950s. Retrospectively, the original version of the Nuclear Safety Guide, LA-2063 (1956), which was promoted by this group, might be considered the "grandaddy" standard in the field of criticality safety. An unclassified version of the guide was issued one year later as TID-7016. Subsequent revisions were issued in 1961 and most recently in 1978 (Ref. 12).

Under the standards program umbrella of ANS and the American Standards Association (now ANSI), ANS Subcommittee 8 and ASA N.6 (now N16) undertook the preparation of standards for "nuclear criticality safety in operations with fissionable materials outside reactors" (see Fig. 4-2). The latter term may be recognized not only as the title of ANS-8 and N16, but also as the title of one of

Fig. 4-2. ANSI/ANS Standards umbrella.[2]

the earliest American National Standards in the nuclear field. This standard, ASA N6.1-1964, sponsored by the ANS and approved by the ASA, was published by ASME. Its revision, ANSI N16.1-1969, was the first nuclear ANSI Standard published by ANS. (Other codes of good practice were published earlier by ANS including the first, which was the original version of ANS Std-1, "Safety Guide for the Performance of Critical Experiments,"[13] with principal author Callihan.)

Those early years of standards writing and establishment of consensus were satisfying, but they were also somewhat painful. The development and final acceptance of N6.1 and its first revision N16.1, for example, took place over a period of more than ten years. As a matter of fact, the draft for N6.1 became the basis for the international standard, ISO Recommendation R-1709, "Principles of Criticality Safety in Handling and Processing Fissile Materials," in 1963. Although it looked for a while like the international version would be published before the U.S. product came to fruition, it turned out that bureaucracy was not confined to one country with the result that the ISO Recommendation was issued seven years later in 1970.

From this beginning with ANSI N16.1, identified successively as ANSI/ANS-8.1/N16.1 and now as ANSI/ANS-8.1 using the ANS priority numbering system,[14] a reasonably well coordinated program for criticality safety Standards has been developed. That program was intended to look like the well-constructed wheel shown in Fig. 4-3. The hub of the wheel is ANS-8.1, which is generic in nature in that it identifies general administrative and technical criticality safety practices and addresses single-parameter limits for fissile nuclides. It is the natural center from which the supporting standards radiate to form an integrated end product. (Since the time this wheel was published, several of the Standards have been reviewed and reissued; the ANS-8.11 "validation" Standard was incorporated into ANS-8.1.) Although the program contains a good cross section of types of Standards, no pretense is made that all important areas have been addressed. The needs for additional Standards to enhance and strengthen the program at large have been identified in a number of areas, as considered later in this chapter.

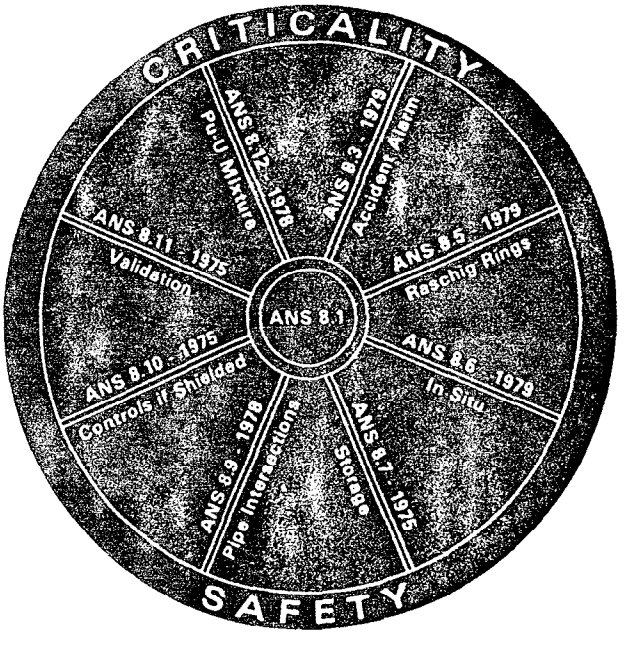

Fig. 4-3. The criticality safety Standards plan.[2]

The General Criticality Safety Standard

The "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," ANSI/ANS-8.1-1983, presents generalized basic criteria and specifies numerical limits for certain simple single fissile units.[14-16] As noted previously, the original version of the Standard was adopted in 1964. An expanded version was approved as N16.1-1969, revised and renamed ANS-8.1 in 1975 with minor changes, and revised and expanded again to its present form. Thus, the basic Standard now has nearly two decades of practical use and experience (in addition to the two decades accumulated before the original version was adopted).

Basic Philosophy

A very important element in this and all other Standards is the differentiation between requirements and recommendations. In this regard, three key words are defined as[14]:

> The word "shall" is used to denote a requirement, the word "should" to denote a recommendation, and the word "may" to denote permission, neither a requirement nor a recommendation. In order to conform with this Standard, all operations shall be performed in accordance with its requirements but not necessarily with its recommendations.

The basic features of these definitions are quite uniform among ANSI Standards, despite some differences of phraseology. In any event, the user must establish a special sensitivity to their restrictive nature.

The key to the Standard's philosophy is contained in its introduction, which states:

> Operations with fissionable materials introduce risks of a criticality accident resulting in the release of radiation that may be lethal to nearby personnel. However, experience has shown that extensive operations can be performed safely and economically when proper precautions are exercised. The few criticality accidents that have occurred show frequency and severity rates far below those typical of nonnuclear accidents. This favorable record can be maintained only by continued adherence to good operating practices, such as are embodied in this standard; however the standard, by itself, cannot establish safe processes in an absolute sense.

Good safety practices must recognize economic considerations, but the protection of operating personnel and the public must be the dominant consideration.

Administrative Practices

The ANS-8.1 Standard encompasses many of the safety principles described in Chapter 2. Substantial flexibility is maintained, for example, where the following administrative practices are required, but their methods of implementation are not prescribed:

1. safety responsibility and criteria established by management
2. process analysis that includes the effects of credible abnormal conditions
3. written procedure for pertinent operations
4. controlled movement of fissionable materials including labeling and posting of limits
5. prompt reporting, investigation, and correction of process deviations that affect safety
6. frequent independent operational reviews
7. emergency procedures and training.

Similarly, the ANS-8.1 Standard requires (but does not specify values for) margins of safety to allow for process uncertainties and accidental conditions.

The administrative practices required by the ANS-8.1 Standard are consistent with the lessons learned from the four most serious of the plant accidents reviewed in the previous chapter. In each case, recognized administrative controls broke down and procedures were improvised without due allowance for the unique features of nuclear systems.

The four accidents all occurred in the course of unusual cleanup operations. Under such extraordinary conditions, effective and practical procedures were difficult to maintain. Improvisation, unfortunately, provided less cumbersome means for adopting procedures to new conditions. The Standard recognizes that a high degree of managerial wisdom is required to establish procedures that maintain effective criticality control while accommodating process difficulties and maintenance.

Double-Contingency Principle

In recognition that improbable operational abnormalities cannot be ignored, the ANS-8.1 Standard delineates the double-contingency principle as a generally accepted guide to the proper degree of protection. The principle calls for controls that assure no single mishap—regardless of its probability of occurrence—can lead to criticality. Stated another way, it requires that two unlikely, independent, and concurrent changes in process conditions occur before criticality is possible.

An important example of application of the double-contingency principle may be found in fabrication facilities for light water reactor (LWR) fuel. Since the slightly enriched uranium can be critical only if appropriately moderated or reflected, full water flooding is the "first contingency" for nearly all analyses. Then, even if flooded, an additional independent change, e.g., major geometrical rearrangement, would be required for criticality under the double-contingency principle.

The principle is rather subjective and does little more than establish a point of view about criticality control. Experience and common sense usually provide the only bases for classifying a mishap as "likely" or "unlikely," or for ruling it out as an impractical concept. An interesting discussion of such contingencies and related risks is contained in Ref. 17.

In shielded facilities, the principle may be relaxed to allow what is effectively single-contingency control. Alternatively, the shielding itself may be considered as one unbreachable contingency.

Geometry Control

The ANS-8.1 Standard recommends that, where practicable, criticality control be based on limited geometrical size rather than on administrative procedures. Even when geometrically favorable equipment is used, however, care is required to assure maintenance of dimensions and nuclear properties. Administrative controls may still be necessary for certain loading and/or unloading operations.

While geometrically favorable equipment is a comforting means of criticality control, sizing equipment for all conceivable compositions and forms of fissile material is often economically intolerable. Large volumes of solution, for example, are much more readily handled by positive control of concentration or fissile mass. However, it may be recalled that several accidents considered in the previous chapter were based on failure of concentration controls. On the other hand, the Wood River Junction accident occurred partially as a result of an attempt to impose an especially inconvenient form of geometry control (by processing large solution volumes by hand agitation in small containers).

Control by Neutron Absorbers

Due in part to the lessons learned from criticality accidents, there is a trend toward poisoning large vessels for which geometry control is impractical. The ANS-8.1 Standard permits reliance on neutron-absorbing materials in process equipment provided their effectiveness is confirmed by available data. Commonly used poisons include cadmium, boron, and gadolinium. Although not stated directly in the Standard, fixed poisons are less favored than geometry, but generally preferred to administrative procedures, for criticality control.

A simple and often effective means of preventing criticality in a large vessel is to pack it with borosilicate-

glass Raschig rings—small, hollow cylinders typically ~3 cm high with a 3-cm outside diameter and a 0.5-cm wall thickness. Such rings of ordinary glass are sometimes used in nonnuclear chemical processing as a simple means of increasing vessel surface area to enhance reaction rates. More detailed guidance for permissible usage, degree of protection, and appropriate surveillance is provided by Standard ANS-8.5 "American National Standard Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Materials."[18,19] The surveillance function sets forth the need for administrative controls to ensure that the absorber is present and properly located, e.g., that the rings have not had their boron content leached out or otherwise dissolved.

The straightforward possibility of loss or redistribution of soluble poisons limits their reliability as criticality controls. Since the single act of adding the wrong reagent could precipitate soluble poison and allow criticality, the double-contingency principle would not be satisfied. Thus, they are generally used only as a secondary control, e.g., in an auxiliary vessel where solution would be expected to appear only after an unlikely ("first-contingency") mishap.

In shielded operations, soluble poisons are sometimes employed as primary controls since accidental excursions are not expected to be destructive, e.g., when pressure buildup can be kept small by allowing extra volume or venting to an empty vessel. Spent fuel processing operations may make especially good use of such a relaxation of the double-contingency principle.

Subcritical Limits

Standard ANS-8.1-1983 defines[14] a subcritical limit as:

> The limiting value assigned to a controlled parameter that results in a subcritical system under specified conditions. The subcritical limit allows for uncertainties in the calculations and experimental data used in its derivation but not for contingencies; e.g., double batching or failure of analytical techniques to yield accurate values.

Its predecessor[20] contained the slightly different definition:

> A limit to a specified variable for the optimum values of all unspecified variables that keeps the system subcritical by a margin of reactivity sufficient to compensate for inexactness in experimental data and calculations, but that contains no allowance for operating contingencies (e.g., double batching) or for inaccuracies in the values of process variables (e.g., mass or concentration).

Subcritical limits may be specified in terms of mass, dimensions, concentration, or a combination of these and other parameters (e.g., isotopic, physical, or chemical composition). Composite limits, of course, are valid only if the conditions on all controlled parameters are maintained. This topic is the subject of Chapter 7. Some of the single-parameter limits for fissile nuclides and multiparameter limits for slightly enriched uranium contained in the ANS-8.1 Standard are listed or displayed there.

Subcritical limits incorporate uncertainties in experimental and/or calculational results in a manner designed to assure that the system will not be critical if the specified conditions are met. This is distinctly different from the common "best-estimate" critical parameters reported for many experiments and calculations. Thus, if the latter data are to be employed, the user must take the uncertainties into account, e.g., as described by Clark.[21]

The subcritical limit, as such, is not adjusted to include any margin for misapplication. Thus, the user must account for the possibility of "double batching" (i.e., as the term implies, using twice as much material as intended) and for anticipated uncertainties in measuring controlled parameters.

The Standard requires that subcritical limits be established on bases derived from experiments wherever applicable data are available. In the absence of such data, it is permissible to derive the limits from validated calculations, as described more fully in Chapter 6. Allowances for uncertainties in the data and the calculations must be applied as appropriate in either case.

Safety Margins

The Standard requires that subcritical limits be modified by "adequate" safety margins to establish working limits for plant applications. Selection of these unspecified margins calls for judgment based on experience. An appendix to the Standard, which is not officially part of it, contains a listing of the types of contingencies that need to be considered; these are summarized in Chapter 7.

Safety limits are commonly established to coincide with a fraction of a critical mass or a specified subcritical k value and then converted to masses, volumes, or dimensions. Safety factors are applied to the subcritical limits (which already include allowance for data uncertainties) appropriate to configurations that account for "contingency" conditions. For example in LWR fuel fabrication where full water flooding is credible, safety margins must produce working limits based on dimensions or k values appropriate to this contingency. Contingency-based limits are seldom approached in actual operations with fissionable materials outside of nuclear reactors.

Additional Guidance

More detailed guidance than that in the general criticality safety Standard is available in specialized Standards, guides, and manuals. In all cases, the desired goal is to provide substantive recommendations without introducing arbitrary constraints, inconsistencies, or inaccuracies.

Specialized Standards

ANS has issued the nuclear criticality safety Standards listed in Table 4-I (some of which are shown by the "wheel" in Fig. 4-3). Since the current priority numbering system dates back only to about 1980, original designations are also shown in the table to facilitate comparison to references that pre-date this change. (It may be noted that certain of these retain both numbers. This may or may not change as mandatory five-year reviews are completed.)

ANS-8.1 is described above (with the portion on computer code validation, formerly contained in ANS-8.11/N16.9, deferred to Chapter 6). Seven of the others in Table 4-I are considered briefly below. The remaining three are discussed elsewhere—ANS-8.3 (criticality alarms) in App. E, ANS-8.5 (Raschig rings) in this chapter, and ANS-8.19 (administration) in Chapter 10.

TABLE 4-I
Nuclear Criticality Safety Standards Developed by ANS

Current Designation	Original Designation	Title
ANS-8.1-1983	N16.1	Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors
	N16.9	Validation of Calculational Methods for Nuclear Criticality Safety
ANS-8.3-1979	N16.2	Criticality Accident Alarm Systems
ANS-8.5-1979	N16.4	Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material
ANS-8.6-1983	N16.3	Safety in Conducting Subcritical Neutron-Multiplication Measurements *In Situ*
ANS-8.7-1982	N16.5	Guide for Nuclear Criticality Safety in the Storage of Fissile Materials
ANS-8.9-1978		Nuclear Criticality Safety Guide for Pipe Intersections Containing Aqueous Solutions of Enriched Uranyl Nitrate
ANS-8.10-1983	N16.8	Criteria for Nuclear Criticality Safety Controls in Operations with Shielding and Confinement
ANS-8.12-1978		Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors
ANS-8.15-1981		Nuclear Criticality Control of Special Actinide Elements
ANS-8.17-1984		Criticality Safety Criteria for the Handling, Storage, and Transportation of LWR Fuel Outside Reactors
ANS-8.19-1984		Administrative Practices for Nuclear Criticality Safety

The ANS-8.1 Standard requires that before a new operation is started, or an existing one changed, it be determined that the entire process will be subcritical under both normal and credible abnormal conditions. In some cases, it may be desirable to employ *in situ* neutron-multiplication measurements to confirm that the proposed configuration is subcritical. For example, a series of measurements made as the quantity of fissile material is increased can be extrapolated to establish the reference critical quantity. Standard ANS-8.6, "Safety in Conducting Subcritical Neutron-Multiplication Measurements *In Situ*," enumerates[22,23] equipment criteria and other good practices to protect against criticality in the course of such measurements. *In situ* measurements are especially useful to confirm the safety of expensive operations with equipment and surroundings that are not amenable to mock-up in critical facilities, to correlation with known critical systems, or to reliable modeling in computer calculations.

Another product of the Standards working groups is the "American National Standard Guide for Nuclear Criticality Safety in Storage of Fissile Materials," ANS-8.7 (Ref. 24). The document contains subcritical limits for many water-reflected cubic arrays of spherical fissile units. In addition to considering a number of physical and chemical forms for the units, it also addresses limiting cases for the effects of moderation between units, displacement of units, distortion of units and cells, substitution of concrete reflector for water, and interaction of arrays through concrete. The tabulations of subcritical limits in this guide do not include arrays of solution cylinders or units of low-enriched uranium.

Standard ANS-8.9, "Nuclear Criticality Safety Guide for Pipe Intersections Containing Aqueous Solutions of Enriched Uranyl Nitrate," provides[25,26] general guidance specifically for a single, unlimited length, large-diameter pipe (column) intersected by numerous lesser diameter pipes (arms). Such geometries are characteristic, for example, of manifolds for transfer and storage. Restrictions are provided in terms of arm length-to-column diameter, total intersection areas of the arms, reflection and moderation conditions, and analysis requirements. As the title implies, of course, ANS-8.9 is applicable only to homogeneous aqueous solutions of enriched (not exceeding 93.5 wt%) uranyl nitrate. Expansion of the Standard to solutions of uranium and plutonium, ^{233}U, and slightly enriched uranium will be possible only with appropriate experimental and calculational support.

Standard ANS-8.10, "Criteria for Nuclear Criticality Safety Controls in Operations with Shielding and Confinement," defines[27–29] the conditions under which the double-contingency control requirements of ANS-8.1 can be relaxed for shielded facilities. The criteria for transfer to single-contingency control are based primarily on limiting the radiation doses and the radioactive releases from postulated criticality accidents. If the criteria are met, any

of the following are acceptable as primary criticality controls: geometry, spacing, isotopic composition, concentration, and soluble poison content. More restrictive controls are required, however, where improper performance alone might lead to criticality, i.e., so that "no single administrative error on the part of any one individual leads to criticality."

The ANS-8.1 Standard provides single-parameter limits for operations with ^{235}U, ^{233}U, and ^{239}Pu, but does not address their combinations. Standard ANS-8.12, "Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors," serves[30] to expand the data base by specifically addressing plutonium-uranium fuel mixtures that contain no more than 30 wt% plutonium combined with uranium containing no more than 0.71 wt% ^{235}U (i.e., less than or equal to enrichment of natural uranium). Mixed-oxide fuels of plutonium and depleted uranium for LWRs and liquid-metal fast breeder reactors (LMFBRs) meet these restrictions. The ANS-8.12 guidance is limited, however, to defined homogeneous mixtures and explicitly excludes heterogeneous arrangements such as fuel rod lattices and other lumped-fuel geometries.

Although current criticality safety concerns are focused primarily on ^{235}U, ^{233}U, and ^{239}Pu, other fissile transuranic nuclides are also of interest.[31,32] Some of the unique concerns of such nuclides are addressed[33,34] by Standard ANS-8.15, "Nuclear Criticality Control of Special Actinide Elements." Although subcritical mass limits have been developed for 14 new nuclides (including ^{251}Cf whose minimum critical mass is thought to be <5 g), major applications for the Standard are expected to be in facilities that handle 6 important ones—^{237}Np, ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, and ^{244}Cm. The stated limit values are estimates, however, since critical masses have not been measured. Future extension of the Standard to address isotopic mixtures of plutonium in a more formal manner may be of particular benefit to fuel recycle operations in both LWR and LMFBR systems.

TABLE 4-II
Proposed Nuclear Criticality Safety Standards

Designation	Title
In Preparation	
ANS-8.14	Use of Soluble Neutron Absorbers for Criticality Control
ANS-8.18	CPCV as a Neutron Absorber in Solutions of Fissile Material
Awaiting Experimental Data	
ANS-8.12.1	Nuclear Criticality Control and Safety of Heterogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors
ANS-8.16	Maximum Subcritical Limits for Slightly Enriched Uranium Compounds Processed in the LWR Fuel Cycle

Standard ANS-8.17, "Criticality Safety Criteria for the Handling, Storage, and Transportation of LWR Fuel Outside Reactors," provides[35,36] guidance for ex-reactor handling, storage, and transportation of LWR fuel elements and components such as individual fuel rods. Both fresh and irradiated fuel assemblies are considered. Principal applications are expected to include fuel fabrication plants, dry storage areas, on-site and away-from-reactor spent fuel pools, head end operations in reprocessing plants, and shipping casks. The Standard specifically addresses guidance and criteria for basic criticality control, use of and control over neutron absorbers, need for and use of criticality alarms, and allowance for verifiable presence of burnable poisons and fuel burnup.

Standards Needs

Needs for additional Standards have been identified in a variety of areas.[1,2] Those that have received some attention are identified in Table 4-II. The top entries in the table are under consideration by working groups. Efforts on the last two standards await availability of experimental data.

Further extension of existing Standards could also benefit from additional experimental data. The many areas related to administrative practices are also perceived as worthy of substantial standardization efforts.

Working Groups

Two proposed standards in preparation are listed in Table 4-II. They are described briefly below.

The initial development of proposed Standard ANS-8.14, "Use of Soluble Neutron Absorbers for Criticality Control," is emphasizing homogeneous mixtures of soluble boron and ^{235}U solutions.[37,38] Subcritical limits as a function of fissile concentration are presented, but detailed specification and control requirements are left to the user to provide site-specific flexibility. The Standard affirms that soluble neutron absorbers can be used readily as both primary and secondary criticality controls. Future plans include extension to other soluble poisons, such as cadmium and gadolinium, and to other fissile materials, namely, ^{233}U and plutonium.

The recognition that Raschig rings are chemically incompatible with some process solutions employed in fuel plants has led to use of chlorinated polyvinyl chloride (CPCV) in some operations. A natural follow-up has been the work on the proposed Standard ANS-8.18, "CPCV as a Neutron Absorber in Solutions of Fissile Material."[39] The initial version of the Standard is restricted to low-enriched uranium solutions and to applications as a secondary control (i.e., another parameter such as concentration must serve as the primary control required by the double-contingency principle stated in the ANS-8.1 Standard).

Experimental Needs

The two proposed Standards listed in the last section of Table 4-II specifically await further experimental data.[2] In addition, lack of data has limited the scope of a number of the Standards that have already been issued or drafted.

Among these, the "Raschig ring" Standard ANS-8.5 is being reissued to include low-enrichment uranium as though the ^{238}U were not present.[18,19] It could be improved in the direction of reduced conservatism if there were experimental data to account explicitly for the effect of ^{238}U. There is still no experimental basis for correcting other deficiencies, namely, inapplicability to plutonium-uranium solution except by ignoring the ^{238}U, to ^{233}U solution, and to absorbers such as polyvinyl chloride and boron steel that would be suitable for solutions that attack glass.

The "storage guide" Standard ANS-8.7 is being expanded to include enriched-uranium solutions as well as solids.[24] Applications to storage of plutonium-uranium solutions, ^{233}U solutions, or undermoderated hydrogenous mixtures might be derived from calculations verified for individual units, but would be strengthened if based on array experiments. Furthermore, there is insufficient experimental information for generalizing moderation effects, such as those of an activated fire sprinkler, on array criticality.

The "pipe intersections" Standard ANS-8.9 originally included ^{235}U, ^{239}Pu, and ^{233}U solutions and was revised to include low-enrichment uranium in solution.[25,26] An experimental basis for extension to plutonium-uranium and ^{233}U-Th solutions would be desirable.

The ANS-8.12 Standard for homogeneous plutonium-uranium fuel mixtures[30] may be excessively conservative because of considerable reliance on computed data. Improved experimental guidance should be useful for PuO_2-UO_2, and would be essential for a companion standard on ^{233}U-Th.

The draft Standard ANS-8.14 for soluble neutron absorbers[37,38] has been prepared for boron in enriched uranium solutions, and is being extended to gadolinium in plutonium solutions. More experiments are required for application to other absorbers and to solutions of plutonium-uranium and ^{233}U-Th.

Finally, the ANS-8.15 Standard for special actinide elements[33,34] applies to all transuranic elements except ^{239}Pu. Because of scanty integral experimental guidance and reliance on measured cross sections, its critical-mass estimates are extremely conservative. These estimates should be subject to improvement as significant quantities of the elements become available and experiments are performed.

Administrative Needs

According to McLendon,[2] the greatest remaining area of need may be to address the "how to" standards in greater detail. Although many of the Standards identified in Tables 4-I and 4-II have some administrative aspects, only ANS-8.19 (a draft of which is contained in Chapter 10) has administrative practice as a primary focus. Concerns not yet addressed include specification of:

1. acceptance criteria for safety analyses
2. margins of safety required for a proposed condition or action with fissile nuclides
3. verification and confirmation of decisions
4. decision responsibilities by managers, staff members, committees, and others
5. authority for final approval
6. reconciliation of differences in judgmental conclusions on the adequacy of the controls specified
7. systems for tracing experience, detecting infractions of procedures or limits, and implementation of follow-up actions
8. proper levels of auditable documentation.

A more detailed formulation of administrative practices into codes of good practice would likely be of great service to the nuclear community. It seems clear that government regulatory bodies also have such a need. In satisfying the latter, a consensus of good practice developed and tested by the nuclear community would appear to be far preferable to dependency on persons less involved and less experienced. It is obvious, however, that the needs of all should be recognized and interrelated to the maximum extent possible. Regulatory interfaces are described in Chapter 9.

One of the major shortcomings in the nuclear community may be that too little information has been published on administrative-control methodology and experience. As a result of proprietary or other concerns, many organizations appear to be reluctant to share experience that is needed to define the "voluntary common practice" that could be the basis of a consensus Standard.

Recognizing the need for administrative criticality safety Standards has led to the following recommendations[2]:

1. preparation of additional administrative Standards representing the best of practices and recognizing both user and regulator needs
2. extensive use of the Standards to build and extend a common experience base
3. publication and other widely based sharing of information on experience
4. efforts made to improve industrial and public confidence through utilization of Standards and promotion of successful practices.

Accomplishment of such objectives would likely be quite difficult. However, the potential value of the product is apparent. The nuclear community now has over four decades of experience, a substantial base upon which to build administrative Standards.

Guides and Manuals

The Nuclear Safety Guide/TID-7016, revision 2, is intended to provide guidance for the practice of nuclear criticality safety.[12,40] It contains a substantial amount of background information as well as providing subcritical limits for a very wide range of material compositions. Limits and hand calculation methods are addressed for arrays in storage and/or transportation. The final chapter considers nuclear criticality safety applications to processing plants. As expressed by the preface to its original version, the TID-7016 report is not extensive enough to be a handbook; it is also not specific enough to be a manual for a particular installation. Instead its authors hope that "the Guide will continue to serve immediate needs and will encourage continuing and more comprehensive efforts toward organizing nuclear criticality safety information."

Manuals for nuclear criticality safety combine appropriate data with numerical guidance for operations in specific plants. Such manuals, for example, have been prepared for Oak Ridge's Gaseous Diffusion Plant[41] and Y-12 Plant,[42] the Savannah River Laboratory,[43] and Hanford's fuel reprocessing[44] and plutonium processing[45] operations. Other compilations from around the world include the United Kingdom Atomic Energy Authority's "Manual of Experimental Criticality Data,"[46] the "Guide de Criticité" from the French Commissariat á l'Energie Atomique,[47] the German "Handbuch zur Kriticalitat,"[48] the Soviet Union's "Critical Parameters of Fissionable Material Systems and Nuclear Safety,"[49] and Japan's "Criticality Safety of Nuclear Fuel."[50]

Guides and manuals are of most help for nuclear criticality safety problems that are straightforward or have been solved for standardized operations. They are much less useful in the design of new operations where economic considerations dictate an optimization approach. Guidance based too heavily on past experience can create stereotyped criticality control. By evaluating each major problem on its own merits, more innovative practical solutions may be found.

Exercises

4-1. Explain the intent of generalized standards and define ANSI Standard.

4-2. Describe the role of each of the two pillars in the criticality safety "bridge" of Fig. 4-1.

4-3. Explain the role in Standards development of each of the following: ANSI, NSB, consensus committee, ANS-8, working group, BSR, IEC, and ISO.

4-4. Considering Standard ANSI/ANS-8.1/N16.1,

a. compare its statement on risk to that from Paxton in Chapter 2
b. define the specialized meanings for the words "shall," "should," and "may"
c. compare the safety principles to those of Reider and Paxton stated in Chapter 2
d. explain the double-contingency principle
e. establish the hierarchy of preference for administrative practices, neutron poisons, and geometry in criticality control
f. explain the role of safety margins
g. define subcritical limit
h. describe the new relationship with Standard ANSI/ANS-8.11/N16.9.

4-5. Summarize the focus of each of the specialized Standards shown as spokes of the wheel in Fig. 4-3. By comparing these to the list in Table 4-I, identify and summarize the Standards issued since the wheel was drawn.

4-6. Identify the topic areas that are the subjects of criticality safety Standards working group activity.

4-7. Summarize the needs for Standards related to administrative practices for criticality safety.

4-8. Identify the U.S. facilities and other countries that have developed manuals for criticality safety.

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Experiments 5

The data generated by experiments are important bases for nuclear criticality safety. When the experimental arrangements provide a close match to process conditions, results may be applied directly. The more general case, however, usually requires the data to be used in conjunction with calculational procedures. It is important that the calculations be validated and their biases determined by comparison with experimental results.

Additional tools for criticality evaluation are semiempirical approximations or hand calculation methods. When they contain factors of conservatism based on generic studies, the methods may often be applied directly. Other situations may call for verification by a combination of experimental data and calculations.

Ideally, calculational methods would be used only to interpolate between experimental points. Currently, however, many regions of practical interest are not bracketed experimentally. If new experiments are not feasible within the constraints of time and economics, extrapolation is necessary, used with increased safety margins to allow for the additional uncertainty.

This chapter describes experimental methods used for nuclear criticality safety. The next chapter is devoted to the computer methods. The subcritical limits and operating limits, which are usually developed from a combination of experimental and computer-generated data, are the subject of Chapter 7. Validated hand calculation procedures are described in Chapter 8.

Methods

Gathering experimental data is generally time-consuming and expensive. Thus, it is important to select those experiments that have the most general applicability. Benchmark experiments with well-defined compositions, precise dimensions, and regular geometries are especially important to the extent that they may be accurately modeled and, thus, used to validate computer codes. The care with which such experiments are designed and conducted must be complemented by accurate and complete reporting of results for their usefulness to be maximized.

Valuable data have been generated by both critical and subcritical measurements related to typical process conditions. Some consideration has also been given to simulation of accident conditions. Dissemination of the results has been facilitated by issuance of reports, compilations, and bibliographies.

Critical Facilities

Critical measurements have been performed at one time or another at most of the U.S. government's nuclear research laboratories. Other governments and numerous private research organizations have also found it necessary to conduct experimental critical-mass studies. Today, experimental studies in support of nuclear criticality safety are performed at three U.S. government laboratories and at facilities in France and the United Kingdom (plus, presumably, a facility in the Soviet Union for which no specific details are available). A few other programs tied principally to reactor design are also in progress, e.g., Argonne National Laboratory's Zero-Power Reactor and Zero-Power Plutonium Reactor used in support of breeder reactor design. An overview of the facilities and programs related specifically to criticality safety is provided in a variety of references[1–12] and summarized below.

Many critical experiments of general interest have been conducted by facilities at the Oak Ridge National Laboratory (ORNL) in Tennessee and the Battelle Pacific Northwest Laboratory (PNL) on the Hanford Reservation near Richland, Washington. Unfortunately, the very productive ORNL facility was closed in 1975.

The ORNL Critical Measurements Laboratory performed a multitude of experiments with ^{235}U and ^{233}U. Some efforts addressed quick determination of whether a certain piece of apparatus or material or configuration was critical under the conditions of intended use. Many other experiments addressed basic criticality concerns both of obvious and immediate application and of more fundamental interest. Array studies, including those for 93 wt% enriched ^{235}U metal with various reflection and interspersed materials, were very important to the initial development of the ANS-8.7 Standard for storage of fissionable materials described in Chapters 4 and 7. A variety of experiments with 5 wt% ^{235}U fluoride solutions and metal rods were also conducted.

PNL's Plutonium Critical Mass Laboratory, designed especially to handle that element, has conducted several thousand experiments with plutonium systems of almost every description (e.g., geometry, moderation, isotopic composition, density, nitrate concentration, and presence of neutron poisons). More recently, attention has been focused on plutonium-uranium mixtures and on low-enriched uranium lattices in support of liquid-metal fast breeder reactor (LMFBR) and light water reactor (LWR) fuel cycle operations, respectively.

Critical-measurements facilities at the Los Alamos National Laboratory (LANL) in New Mexico and the Rocky Flats Plant in Colorado have focused most of their attention on specialized problems associated with highly enriched uranium and plutonium used for components of nuclear explosives. However, much information of general interest has come from these operations both in the form of data and of general methods for nuclear criticality safety. A third facility, located at what is now the Lawrence Livermore National Laboratory, conducted a number of plutonium-metal array and other experiments during a relatively brief period of operation.

The Los Alamos Critical Assemblies Laboratory[11] is the oldest of the operating facilities and is, perhaps, the only one to handle routinely all of the three major fissile species in a multitude of configurations involving all physical forms (solid, liquid, and gaseous). It is probably best known for the general-purpose critical assembly machines—with names like Godiva, Jezebel, Flattop, Comet, Honeycomb, and Mars—that have provided many useful benchmark experiments and continue to be used. A recent addition to the group is called "Sheba," a 5 wt% ^{235}U solution system designed to benchmark the response of criticality alarm detectors (as considered further in Appendix E).

The Rocky Flats Critical Mass Facility, like its Battelle counterpart, was designed to handle plutonium. Experiments with this element have been conducted for metals and oxides with various reflectors and in arrays. A number of other measurements with high-enriched uranium metal and solution have emphasized interaction among units; one experimental series, for example, examined the interaction of uranium solution in vertical cylinders and horizontal slabs. More recently, measurements with low-enriched uranium oxides have also been performed.

The remaining U.S. critical facility was operated by the Babcock and Wilcox (B&W) Company in Lynchburg, Virginia. After a period of dormancy, it conducted important experiments with slightly enriched uranium lattices. Steps for its closure were initiated during the early 1980s.

There have been two critical-measurements programs in Europe (or, perhaps, three if the Soviet Union were included). The United Kingdom completed experiments related to plutonium-uranium mixtures before entering a period of dormancy in the late 1960s. Programs were reactivated, however, around 1980 with a series of *in situ* measurements on low-enrichment, low-moderation uranium at the British Nuclear Fuels plc (BNFL) facility in Springfields.[13] Recommissioning of the DIMPLE and other critical facilities at Winfrith was the next step in the process.

Starting in 1960, the French conducted their ALECTO experiments with nitrate solutions of ^{239}Pu, ^{235}U, and ^{233}U in Saclay. Three years later, the Critical Mass Laboratory, or "Station de Criticité," at Valduc began operation. Experimental programs have considered plutonium and high-enriched uranium solutions including interaction effects, isolating effects, and neutron poisoning. Recent studies have focused on problems in support of LMFBR and LWR fuel processing. As noted in Chapter 3 and described later in this one, the French study "Consequences Radiologiques d'un Accident de Criticité" (CRAC) and subsequent programs have contributed greatly to the general understanding of solution criticality excursions.

In line with lessons learned from the fatalities that accompanied early critical-measurements accidents, all experiments are performed remotely (at least in their concluding stages). Most of the facilities rely on heavy shielding during experiments to protect personnel in adjacent control rooms and offices. The LANL operations conduct experiments in one of three structures called "kivas," named after buildings used historically by native Pueblo Indians for ceremonial purposes. The kivas are located away from a central complex of control rooms so that distance rather than shielding protects personnel from potential radiation exposure.

Critical Experiments

One important class of experiments includes those in which critical configurations are established directly. Some experiments determine critical dimensions for individual units. Others are concerned with measuring the critical spacing for a given number of units in an array or the critical number for a given spacing. Determination of the

effects of various reflector, poison, and moderator materials is also important.

In the earlier experiments at ORNL, criticality was attained whenever possible. Most of the current experiments approach the critical condition as closely as practicable, but may or may not actually reach or exceed criticality. For critical experiments, it is desirable to avoid supercritical configurations that result in excessive neutron levels, fission product production, and heat generation. These factors may complicate the experimental results and also cause material handling problems.

Since precise criticality is such a delicately balanced condition, frequently it must be achieved with the aid of one or more external reactivity-control devices, which may also serve safety functions by having the capability to shut down quickly the neutron chain reaction. Ingenuity is required so that such systems have minimal effects on the final critical configuration. Although certain empirical corrections are readily applied to experiments to produce valid estimates of critical conditions, they are often not so easily accommodated in the corresponding benchmark computer calculation models. Overall, precision in constructing geometric configurations, establishing material compositions, and reporting both are vital to the effective use of these experiments.

The standard procedure for critical experiments is to measure inverse multiplication (1/M). This method (also commonly used in university reactor laboratory courses,[14–16] for example) relies on the fact that a given neutron source generates a fission-neutron population whose size increases with the multiplication factor k of the material in which the source is placed. At precise criticality each source neutron may start a sustained chain reaction producing an average of one neutron per fission. Continued presence of the source, then, would in theory cause the neutron population to grow without bound. The reciprocal or inverse of this neutron number, thus, approaches zero as criticality is approached. Using this fact, the inverse multiplication method relies on plotting the reciprocal neutron counting rate (or actually its ratio to a reference initial count rate) against fuel loading. Extrapolation to zero predicts critical loading. This same extrapolation procedure may be used in the course of the experiment to estimate the "safe" material additions to be made without causing criticality and, as appropriate, to establish when hands-on operations should be ceased in favor of remote ones. A number of representative examples of critical measurements based on this method are provided in the following paragraphs.[4,5]

A system used at ORNL for surveying the critical heights of ^{235}U and ^{233}U solution cylinders is illustrated in Fig. 5-1. The solution and neutron source in the test vessel are used for inverse-multiplication measurements. Solution from geometrically favorable storage containers is pumped into the vessel in a controlled manner. The

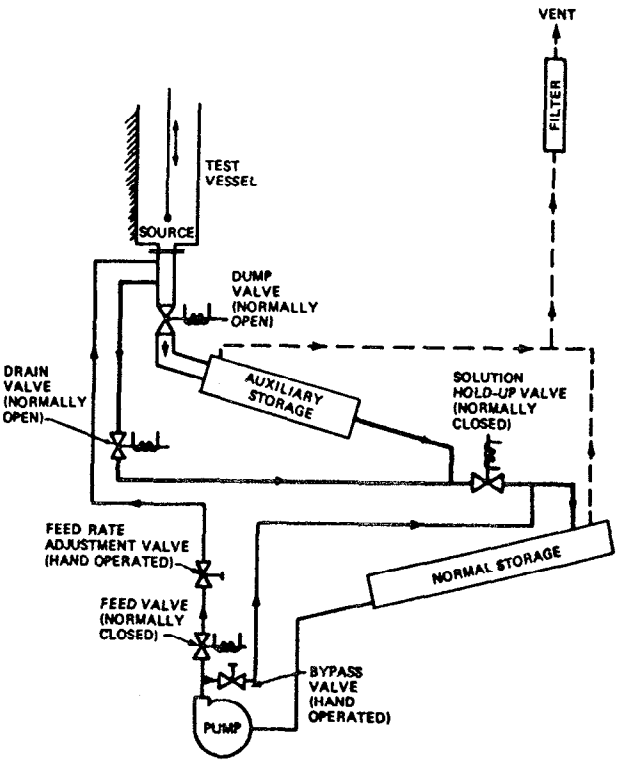

Fig. 5-1. Experimental arrangement for the survey of critical heights for ^{235}U and ^{233}U solutions performed at the ORNL Critical Measurements Laboratory.[4]

dump valve provides for removal of solution. Since the valves for adding solution are "normally closed" (i.e., must be held open by an electric current to their solenoid coil, and thus "fail" to a closed position) while those for draining are "normally open," the design is "fail-safe" to interruption of electrical current caused either by a power failure or by a designated signal associated with violating one or more predetermined parameter limits. The latter, of course, correspond closely to the reactor "scram" or "trip" function.

Experimental configurations for water-reflected plutonium-uranium nitrate solution measurements are shown in Fig. 5-2. The experiments, conducted at PNL, employed cylinders and spheres of various dimensions. By varying solution concentration, criticality was approached as closely as possible for the particular reflected geometry. The presence of the containers, piping, and control rods emphasizes the requirements of real experiments in contrast to the idealized situation often assumed in simple calculational models.

Similar experiments in slab geometries have been conducted using the assembly shown in Fig. 5-3. The adjustable bellows allows measurements for a range of slab thicknesses. The "egg-crate" design was established to provide strength for maintaining flat surfaces while minimizing the amount of material available to act as an unwanted neutron reflector.

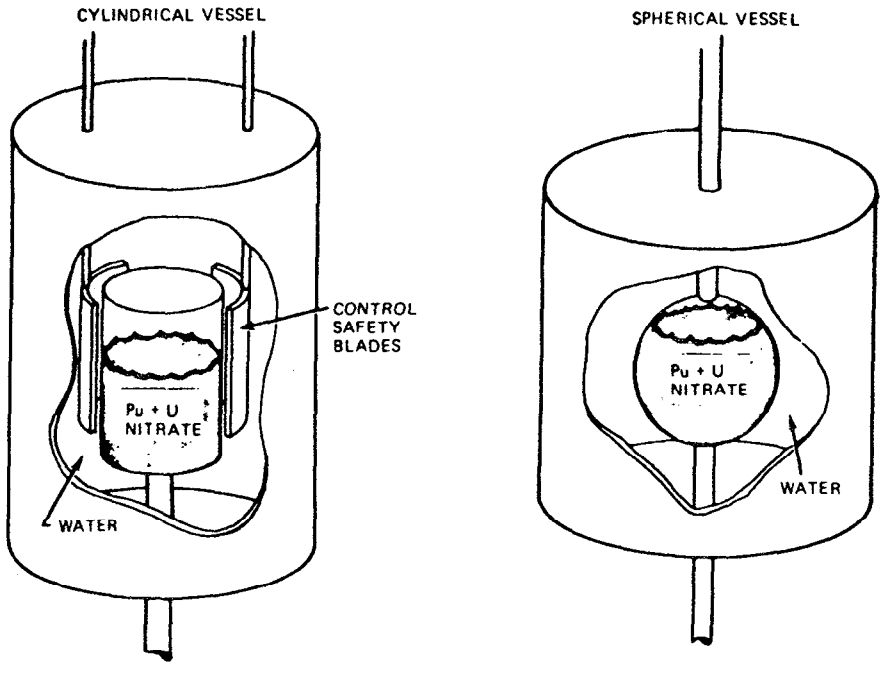

Fig. 5-2. Experimental configurations for reflected cylinder and sphere measurements performed at the PNL Plutonium Critical Mass Laboratory.[4]

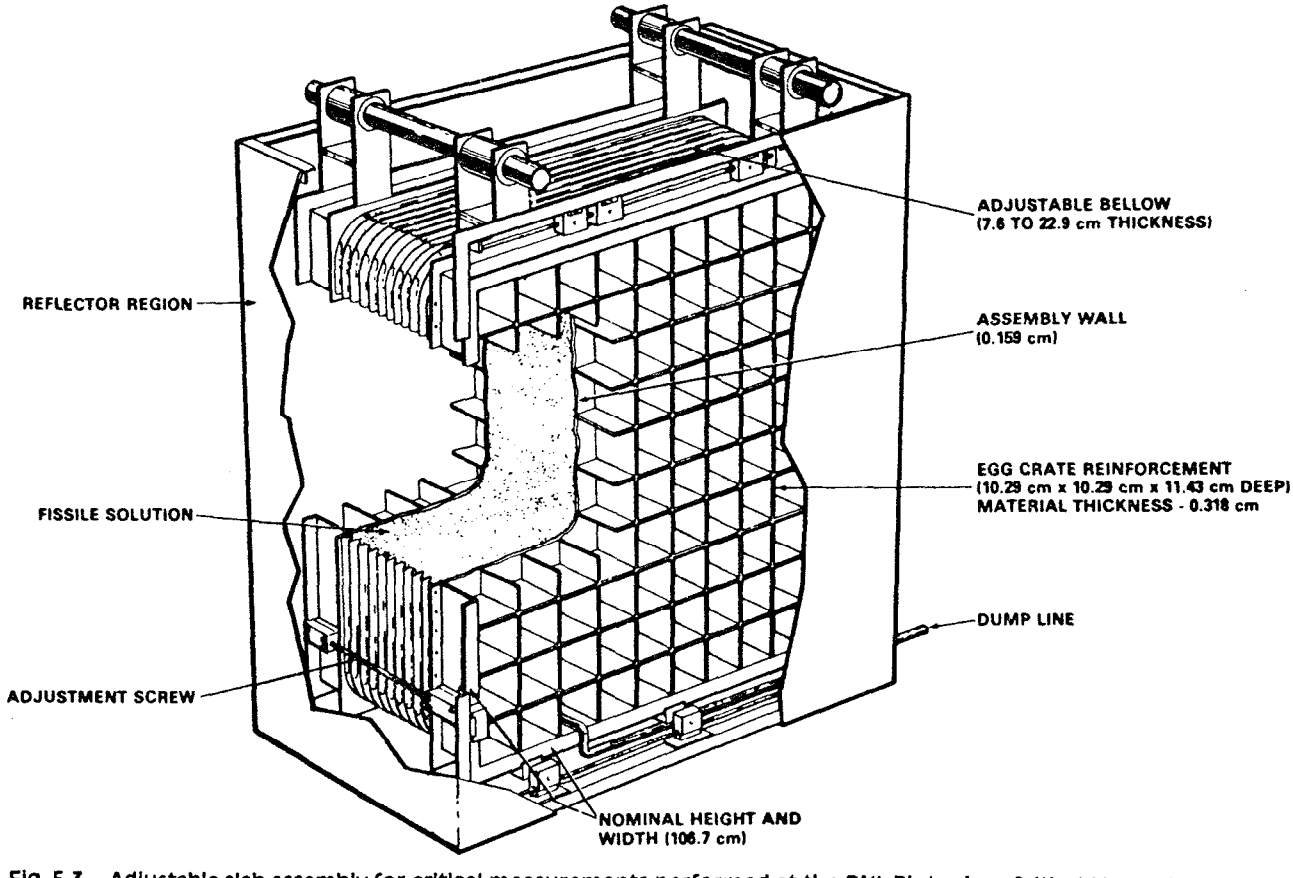

Fig. 5-3. Adjustable slab assembly for critical measurements performed at the PNL Plutonium Critical Mass Laboratory.[17]

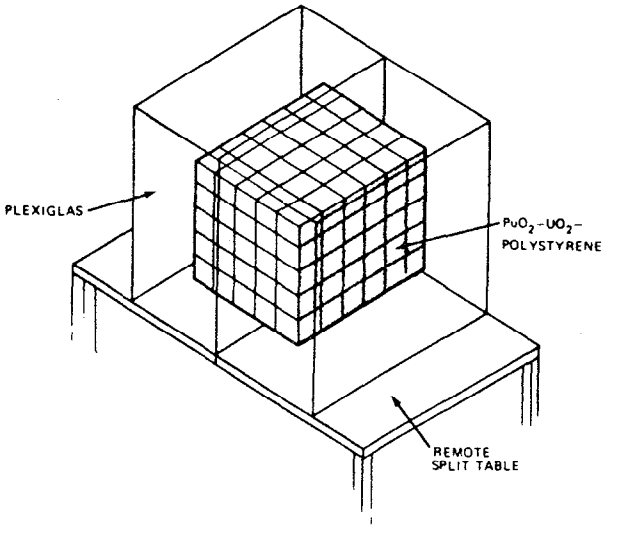

Fig. 5-4. Split table (closed position) as used for mixed-oxide experiments performed at the PNL Plutonium Critical Mass Laboratory.[4]

The split table is a useful device for performing certain critical measurements with solids. As implied by the name, it is based on splitting an array of the fissionable material into two parts. The material may be placed manually on tables that are initially separated and in a configuration that is substantially subcritical. The two halves are later moved together remotely to approach criticality. A vertical lift is similar to the split table except that, as its name implies, the final configuration is achieved by moving the separated portion of fuel upward.

A series of experiments with mixed oxides performed at PNL used a split table as shown in Fig. 5-4. Solid, cube-shaped compacts of PuO_2, UO_2, and polystyrene were used to represent mixed plutonium-uranium oxide in water solution. The surrounding Plexiglas approximated a close-fitting water reflector. The arrangement was designed for ease of handling and to eliminate the relatively massive containers that would be required if actual liquid solutions were used. The absence of containers made the experiments relatively "cleaner" benchmarks than they would have been otherwise. It should be noted, however, that the aluminum and/or plastic coatings on each compact (used primarily to contain the plutonium) do tend to complicate modeling for purposes of calculation.

Other experiments have been used to correlate numbers and dimensions for critical arrays of units. A critical configuration consisting of a double-tier array of 13-ℓ polyethylene bottles of highly enriched uranyl nitrate solution is shown by Fig. 5-5. (Actually, five of the bottles were used like control rods and could be moved into final position after the departure of the person shown in the foreground.) Although the individual cylinders are equivalent to favorable geometry bottles for the uranium solution, the array itself is critical because of strong interaction among units and, therefore, not an overall "favorable" geometry. This, of course, reemphasizes the possibilities for misunderstanding in use of the term "safe geometry" for these or similar individual containers (see also Chapter 2).

Subcritical Experiments

The inverse-multiplication technique can provide useful information without the attainment of (or close approach to) criticality. In the ORNL single-cylinder surveys of Fig. 5-1, for example, very valuable information was obtained by noting when a cylinder diameter was small enough to be subcritical for all heights with a given solution concentration. Such results were an important basis for defining favorable geometry configurations.

In some other cases, criticality may be avoided and the experiment simplified when count data can be extrapolated with sufficient accuracy for defining "safe" equipment. The subcritical experiments may also eliminate the need for specially designed and expensive critical experiments when equipment can be tested directly in its intended location. If nothing else, such testing removes any lingering doubt about subcriticality. Safe conditions for this type of measurement are outlined by the Standard for performing *in situ* measurements[18] introduced in the last chapter.

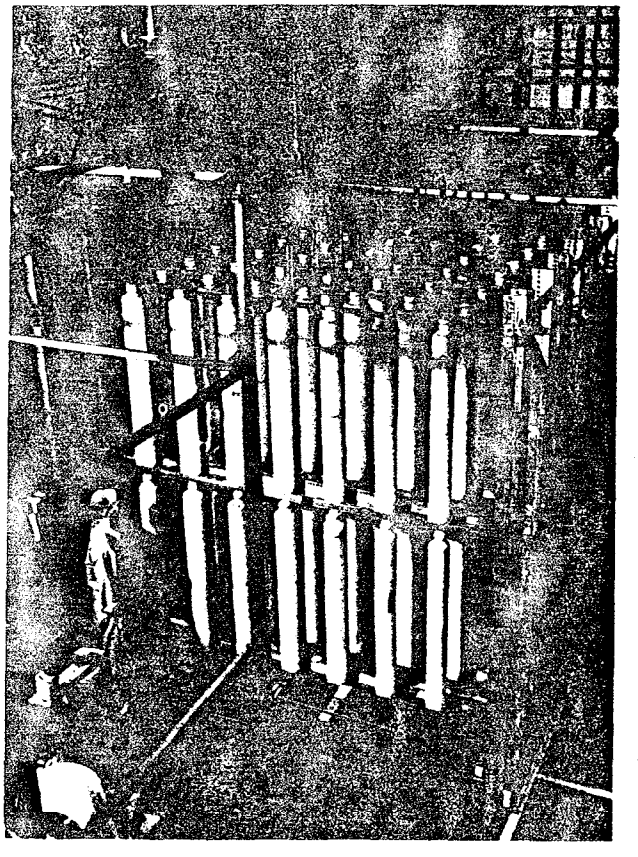

Fig. 5-5. Double-tier array of 13-ℓ polyethylene cylinders containing highly enriched uranyl nitrate solution in a critical experiment conducted at the ORNL Critical Measurements Laboratory.[4]

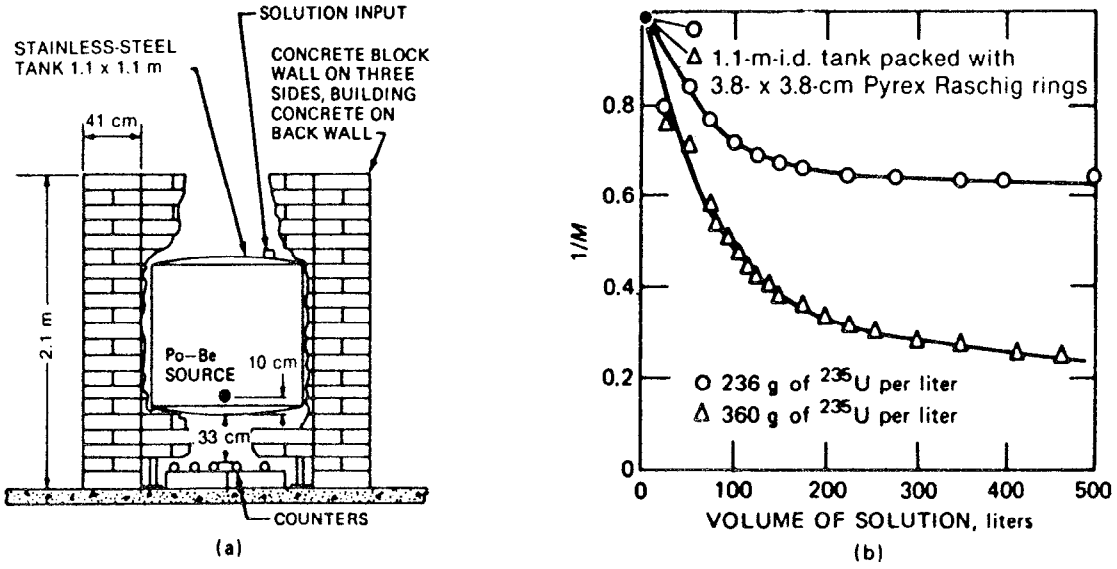

Fig. 5-6. Pyrex-Raschig-ring-filled vessel at Rocky Flats: (a) experimental arrangement and (b) inverse multiplication versus uranyl nitrate solution volume for two concentrations.[4]

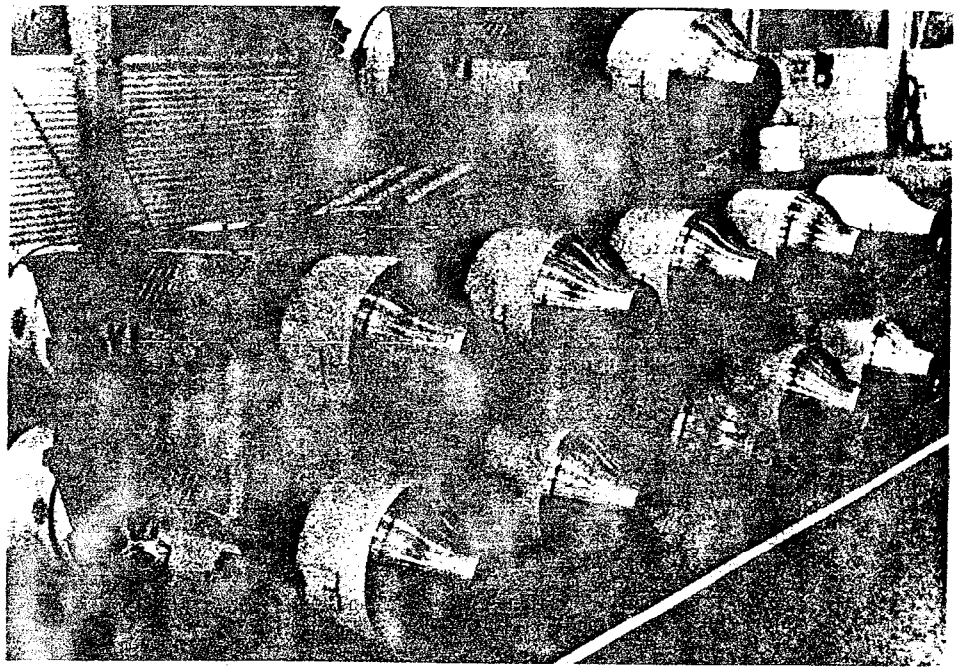

Fig. 5-7. Experimental arrangement used by LANL to simulate shipboard storage of weapons containers.[19]

The arrangement for a subcritical measurement performed at Rocky Flats is shown in Fig. 5-6. The large tank packed with Pyrex Raschig rings was being considered for storage of 93 wt% enriched ^{235}U solution. The inverse multiplication was calculated from the neutron counting rate as a function of the amount of solution added to the tank. Figure 5-6b shows that for both 236 and 360 g/ℓ of ^{235}U solution, the vessel was substantially subcritical even when filled.

An example of a simulated-environment subcritical experiment conducted[5,19] by LANL is shown in Fig. 5-7. Nuclear weapons were to be stored or shipped in proximity to each other. Experiments with simulated ship transport arrays demonstrated that regular 12-unit arrays, two layers deep, were comfortably subcritical even if an additional unit was added, steel plate was nearby, or slight geometric changes were made. Figure 5-8 shows the inverse-multiplication curve generated as the units were stacked sequentially. The figure also includes an arbitrarily selected "safe storage" limit comparison line as $1/M_x = 0.5$. Multiplication values are also shown for conditions that, for example, could be achieved when a unit is moved over

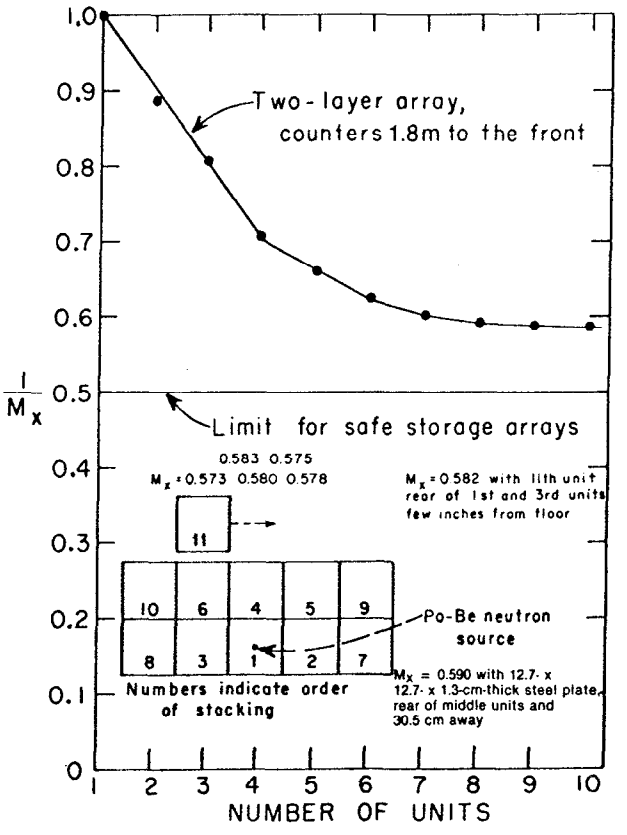

Fig. 5-8. Inverse-multiplication data for LANL experiment simulating shipboard storage of weapons containers. The lower portion shows a loading sequence for regular two-layer arrays plus "11th unit" position and associated inverse multiplication.[19]

the top of the storage array to its final resting place.

The use of approach-to-critical *in situ* measurements has tended to decline over the years due in part to safety concerns. However, other types of *in situ* measurements based on parameters that correlate to reactivity have been receiving additional attention. Prompt-neutron decay constants from pulsed neutron experiments are reactivity indices somewhat like neutron multiplication. Neutron flux distributions from exponential experiments give material buckling values from which critical dimensions for simple geometries may be deduced.[20,21] Active measurements with a ^{252}Cf neutron source[22] and passive methods[23–25] have also been used or proposed for determining multiplication.

Accident Simulations

The French CRAC program[26,27] consisted of experiments in which aqueous solutions of 93 wt% enriched uranyl nitrate were injected into a large-diameter pipe to heights in excess of that required for criticality. By using small (300-mm) and large (800-mm) pipe diameters and a wide range of concentrations, supercritical configurations were obtained with shapes from flat disks to long cylinders. Other experimental parameters included solution addition rates (from roughly 2 to 30 ℓ/min), subcritical or low-power critical initial condition, and presence or absence of an external neutron source.

In typical CRAC experiments, a supercritical solution volume produced an initial power pulse (e.g., like that of Fig. 2-4) consistent with the negative reactivity feedback effects of thermal expansion and gas formation. Additional peaks of generally decreasing size usually followed until some combination of heating, material ejection, and gas formation balanced the system's excess reactivity to produce a more or less stable power level. An example of such behavior is shown in Fig. 5-9. As noted in Chapter 3, these experiments have provided data useful for assessing the risks of potential criticality accidents.

Another French experimental program[28] is being carried out with a small solution reactor known as "SILENE." Employing high-enriched uranium nitrate solution, the pulse reactor has been used to study the phenomenology of boiling-type experiments (e.g., similar to the effects in the CRAC experiment shown in Fig. 5-9), fission gas release rates during excursions, and the pressure wave associated with the radiolytic gas formation of the first pulse.

The Sheba assembly at LANL may also be applied to simulation of criticality accidents in low-enriched uranium systems. It is considered further in Appendix E.

Recent Directions

During the mid to late 1960s, the impressive number and variety of completed experiments coupled with great advances in the computational tools led some to an impression that additional critical measurements were not needed.[7] This, coupled to the high relative expense of experiments, was partially responsible for the closing of the Oak Ridge and other facilities and has continued to threaten the vitality and productivity of the laboratories still in operation. Funding uncertainties along with pressures for return of all unused fissionable materials threaten to limit flexibility dramatically.

Some current needs for experimental data were identified in the previous chapter in the context of extending or adding to Standards. Other needs are based on economic impacts of excessive conservatism.[7] Present research efforts in the United States are directed to projects funded by the U.S. Department of Energy (DOE) and U.S. Nuclear Regulatory Commission (NRC). These and three international programs are described briefly below.

The PNL Plutonium Critical Mass Laboratory is expected to focus much of its attention on support for the Consolidated Fuel Reprocessing Facility program. A broadly based effort is envisioned to obtain data on almost all forms in which plutonium-uranium mixtures may exist in reprocessing plants. Additionally, BNFL has funded a series of experiments related to use of soluble poisons in a reprocessing plant dissolver. Other research relating to the stor-

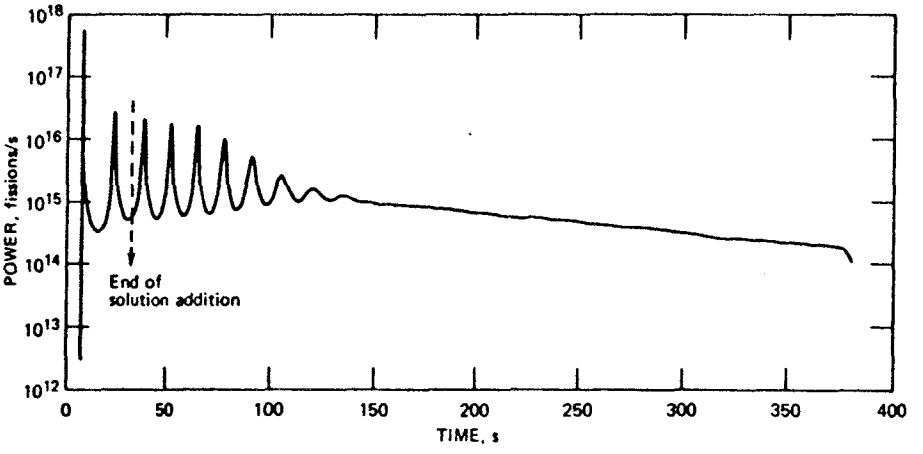

Fig. 5-9. Power trace for the French CRAC 13 experiment showing initial and successive pulses followed by eventual quiescence. Solution was added through the completion of the second pulse.[26]

age and handling of spent LWR fuel may be funded by the NRC.

The Los Alamos Critical Assemblies Laboratory is scheduled to conduct a few damp mixed-oxide experiments for the Consolidated Fuel Reprocessing Facility program referred to above. It is also constructing a device known as "SKUA" of 93 wt% ^{235}U, internally moderated by $ZrH_{1.6}$, and reflected by copper. It is intended to operate in a burst mode that can vaporize a ^{235}U foil to simulate fuel pin behavior under intense heat. Extension of the Sheba experiments (mentioned earlier and considered further in Appendix E) is also anticipated. Plans for use of the facility in training, safety, and design evaluation have also been reported.[29]

The Rocky Flats Critical Mass Facility continues to conduct experiments focused on resolving production concerns. One such program was aimed at finding substitute methods for storing concentrated fissile solutions. The high cost of purchasing and maintaining Raschig rings (in addition to other concerns) has prompted experimental attention to annular tanks and "poisoned tube" vessels. The former consist generally of several annular regions with interspersed neutron-absorbing walls. The latter rely on neutron poison rods clad in metal and distributed through the vessel in much the same geometry that characterizes a heat exchanger.

A smaller effort at Rocky Flats that was funded by the NRC addressed low-enriched uranium oxides in low-moderation systems with various reflectors and interspersed materials. The experiments were intended to support criticality safety of large volumes of material through moderation control.

The Valduc facility in France has been conducting experiments with low-enriched uranium dioxide rods in uranium nitrate solution to study effects expected in nitric acid dissolution. The MARACAS program[30] has been initiated to study criticality of 5 wt% enriched uranium oxide at low hydrogen-to-uranium ratios. A separate part of the same study simulated accidental moderation during fresh fuel storage. The facility is also continuing the SILENE experiments described in the previous section.

The United Kingdom is recommissioning its DIMPLE facility at Winfrith.[13] Its initial program is to address critical measurements for water-moderated arrays of low-enriched uranium fuel pins clad in stainless steel.

The Japan Atomic Energy Research Institute has announced its intention to construct a critical facility[31] by 1987. Their ambitious plans call for three experimental cells capable of handling ^{235}U, ^{233}U, and plutonium in almost any form. Also to be included is the ability to simulate accident and other transient conditions. A small-capacity fuel processing plant is planned on the same site for source material preparation.

Criticality Data

Data for criticality safety guidance are distributed through a number of publications. An early data summary, "Critical Dimensions of Systems Containing ^{235}U, ^{239}Pu, and ^{233}U" is a convenient but somewhat out-of-date source[32] that is, however, scheduled to be updated by 1987. The more recently published "Nuclear Safety Guide/TID-7016/ Revision 2" contains or references a substantial amount of general information.[33]

Paxton's "Experimental Criticality Specifications: An Annotated Bibliography" and its update[34] list most of the important documents through 1979 that contain criticality data. It is well enough annotated that a user should have little problem determining applicability of a specific entry. Battelle's "Handbook of Critical Experiment Benchmarks" is another potentially useful reference.[35]

The Lawrence Livermore National Laboratory (LLNL) has developed an extensive annotated bibliography of experimental data.[36] This computer-generated document has the advantage of including lookup-table and subject index volumes. It has also served as the starting point for a criticality safety data base[37,38] tied to DOE's Technical

Information System for rapid computer-based access around the United States and in other parts of the world. A directory of criticality safety personnel[39] has been issued and plans for additional capabilities reported.[40]

Results of new critical measurements are often the subject of papers whose summaries appear in the *Transactions* of the American Nuclear Society. In many cases, such publication precedes eventual dissemination in technical report documents or journals. The *Transactions* has also typically provided periodic updates on current and proposed experimental programs (e.g., as in Refs. 2, 3, 6, 9, and 10). These and other summaries through 1982 have been compiled and published in conjunction with the LLNL data base described above.[41]

Exercises

5-1. Identify the locations and principal capabilities of the critical-measurements facilities in the world. List factors that tend to lead to the closing of such facilities.

5-2. Describe the method used in the ORNL solution cylinder experiments including solution entry and discharge provisions and shutdown mechanisms.

5-3. Describe the use of split tables for critical measurements.

5-4. Distinguish between roles of critical and subcritical measurements in nuclear criticality safety.

5-5. Describe the focus of the CRAC, SILENE, and Sheba accident simulation experiments.

5-6. Identify the principal compilations of experimental critical data. Describe the recent computer-based compilation.

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6 Computer Methods

Whenever experimental data are not available to establish critical conditions for a given system, it may be necessary to employ a calculational method. A computational procedure that qualifies as a substitute for experiments must apply to a wide variety of compositions and must reproduce the effects of all neutronic processes that occur in real systems. The methods capable of such versatility are conceptually straightforward but are so complex numerically that a large-capacity, high-speed computer is required. (Although the non-computer "hand calculation" methods described in Chapter 8 may appear at first to be exceptions, they too must be verified by comparison with experimental data and computer calculations.)

The two general approaches to criticality calculations are diffusion theory and transport theory. The former is used extensively in reactor situations, but has some limitations for criticality safety purposes (as previously noted in Chapter 2 and an example of which is provided in Appendix G). Transport theory is usually applied through discrete ordinates or Monte Carlo formulations. (Not infrequently, however, the terms "transport" and "discrete ordinates" are used interchangeably with the other formulation designated as simply "Monte Carlo.") Although both methods are also used for gamma-ray and other transport calculations, only neutronics applications are described here. Each of the two methods has a different role for applied calculations and must be verified against benchmark or other experiments.

The following discussions assume that the reader is familiar with basic reactor theory including reaction rates, fluxes, cross sections, and fundamental concepts of neutron transport. A review[1] or more detailed description[2–7] may be warranted for the reader needing additional background. Other general discussions of nuclear criticality safety calculations, including those by O'Dell[8] and from an American Nuclear Society (ANS) "tutorial" session,[9] also provide valuable insight. Fortunately for criticality safety specialists, it is important that they understand the basic theory rather than the intricacies of its computer implementation. They do need to be able to run and verify computer codes developed by experts.

Transport Theory

A complete description of each neutron in a system requires knowledge of the:

1. position in space r, requiring up to three variables for a full description, e.g., cartesian coordinates x, y, z or spherical coordinates r, θ, and ϕ
2. direction of motion $\mathbf{\Omega}$, requiring two variables
3. kinetic energy E, or equivalently its speed v or lethargy u
4. time t when the neutron is at position r moving in direction $\mathbf{\Omega}$ with energy E.

The choice of using direction $\mathbf{\Omega}$ and energy E rather than vector velocity $\mathbf{v}$ is somewhat arbitrary but has advantages in computational strategies. A full description of neutrons within a given system is obtained (at least in principle) by solution of the Boltzmann transport equation (usually referred to simply as the "transport equation") for each of the seven variables.

For neutron flux $\Phi(r,E,\mathbf{\Omega},t)$, one form of the Boltzmann transport equation may be written as

$$\frac{1}{v(E)}\frac{d\Phi(r,E,\mathbf{\Omega},t)}{dt} = -\mathbf{\Omega}\cdot\nabla\Phi(r,E,\mathbf{\Omega},t) - \Sigma_t(r,E,\mathbf{\Omega})\Phi(r,E,\mathbf{\Omega},t) + S(r,E,\mathbf{\Omega},t) + \int_{E'}dE'\int_{\Omega'}d\Omega'\left[\Sigma_s(r;\,\mathbf{\Omega}'\rightarrow\mathbf{\Omega};\,E'\rightarrow E)\times\Phi(r,E',\mathbf{\Omega}',t)\right], \tag{6.1}$$

representing term-by-term that the net rate of neutron accumulation is equal to the algebraic sum of the (negative)

leakage and interaction rates and of the (positive) source and in-scattering rates. The interaction term contains the total macroscopic cross section Σ_t in recognition that any interaction will result in a change in the neutron parameters (between the extremes of complete disappearance by absorption to change in energy and direction as a result of scattering). The in-scatter term has a double integral over the differential scattering cross section $\Sigma_s(r; E'\rightarrow E; \Omega'\rightarrow\Omega)$ to account for scattering of neutrons at a range of initial energies E' and directions Ω' to produce those at reference energy E and direction Ω.

Although it is not shown explicitly by the form of the Boltzmann equation in Eq. (6.1), the portion of the source term S that represents the fission source S_f is the double integral

$$S_f(r,E,\Omega,t) \tag{6.2}$$

$$=\chi(E)\int_{E'} dE' \int_{\Omega'} d\Omega'\ \nu\Sigma_f(r,E',\Omega')\ \Phi(r,E',\Omega',t)$$

for fission neutron spectrum $\chi(E)$, average number of neutrons per fission $\nu(E')$, and fission cross section $\Sigma_f(r,E,\Omega,t)$. This equation accounts for the fact that neutrons of arbitrary energy E' and direction Ω' may cause the fission event that ultimately produces fission neutrons of energy E and direction Ω. In well-moderated systems, for example, thermal neutrons *cause* most of the fissions, but fast neutrons are always the *result*.

In principle, the transport equation could be solved by suitable numerical procedures after insertion of appropriate cross sections and a geometric description of the system. Due to the extreme complexity of the cross sections and of the geometries of most real systems, however, such an approach is not practical. Even with the very large, high-speed computers available today, simplifications and approximations must be employed to make the problem tractable.

Removal of the time dependence from Eq. (6.1) results in a formulation suitable for static calculations. This is usually very well suited to criticality safety calculations since only accident conditions produce significant time-varying neutron populations.

The next step in simplification is based on grouping the neutrons by energy and defining the group flux ϕ_g by

$$\phi_g = \int_{\Delta E_g} dE\ \Phi(E) \quad , \tag{6.3}$$

and the group cross section Σ_{rg} for reaction r by

$$\Sigma_{rg} = \frac{\int_{\Delta E_g} dE\ \Sigma_r(E)\Phi(E)}{\int_{\Delta E_g} dE\ \Phi(E)} \quad , \tag{6.4}$$

where ΔE_g is the energy range for group g (noting that the energy dependence of the neutron flux and cross section has been indicated even though position and other dependencies may also be present). It should be noted that the definitions in Eqs. (6.3) and (6.4) assure that the reaction rates remain unchanged. Another very important feature to recognize is that the group cross sections are dependent on the neutron flux of the system in which they are to be employed. Thus, for example, a set based on the flux in a fast system should not be assumed to be valid for a thermal system, unless demonstrated to be so by careful validation procedures. Employing a larger number of energy groups usually minimizes the actual energy spectrum dependence of each group cross section. For example in the limit where each data point from the underlying cross-section library represents a group, the neutron spectrum dependence disappears completely (or at least within the accuracy of the data themselves).

Discrete Ordinates

The multigroup energy method described above is well known in both diffusion theory and transport theory methods. The former, introduced in Chapter 2 and noted to include no explicit directional representation, is useful in reactor design but not for the more nonuniform geometries of concern to criticality safety. The discrete ordinates method, however, carries the multigroup principle a step further by also dividing the neutron directional dependence into groups. The most commonly used discrete ordinates method is called S_n, from the n'th order expansion used to describe the direction of neutrons emitted from the scattering source. The interested reader will find further details on this method in textbooks on the subject[2–7] and in a report by Lathrop[10] that addresses criticality applications.

Computational time and, therefore, cost tend to scale directly with the numbers of energy groups, angular directions, and spatial mesh points. Thus for purposes of economy, it has been common to use one-dimensional codes with cross sections of 16 energy groups and a low-order angular approximation. Such codes as ANISN (Ref. 11) and DTF-IV (Ref. 12) are well suited to problems that are simple enough to be represented in a one-dimensional geometry, i.e., spheres, infinitely long cylinders, infinite slabs, and certain infinite arrays of these shapes.

Two-dimensional discrete ordinate codes, such as DOT (Ref.13) and TWOTRAN (Ref. 14) may be quite expensive to run. The regular rectangular or cylindrical mesh arrangements also limit the ability to represent the irregular shapes and patterns often encountered in process facilities. Three-dimensional discrete ordinates codes have not been developed primarily since the problems of economy and accurate representation may be even more acute than for the two-dimensional case.

Despite the limitations noted above, the discrete ordinates methods are useful for certain criticality calcula-

tions. They are often a simple and quick means of bounding a problem by looking at extreme situations. Infinite arrays of long cylinders or fuel pins can be calculated quite well. In a water-moderated system, for example, the difference between its k_∞ (calculated by the model) and actual k_{eff} becomes increasingly smaller as dimensions increase above ~2 m. Correction terms based on buckling calculations can reduce discrepancies if used properly. In any case, if inferred material limits are not overly restrictive, more detailed calculations may not be required. Discrete ordinates methods can also serve in many situations as important cross-checks of other computer calculations.

A possible misuse of the discrete ordinates method that is worthy of note relates to mesh spacing that is too large. Although increased size (and, thus, a smaller number) saves computer running time, calculational accuracy for k_{eff} or critical dimensions may decrease significantly. A good "rule of thumb" for mesh size is that it be less than one-half of the neutron mean free path in the material of interest.

Monte Carlo

The Monte Carlo method considers neutrons as individual particles that interact with nuclei on a random basis obeying certain fundamental laws of probability. Although very different in form, the procedure can be shown to be equivalent to using a random-sampling technique to evaluate the double integral in the Boltzmann equation (e.g., as demonstrated by Schaeffer[15] and Carter and Cashwell,[16] both of which provide good general references on the method).

The Monte Carlo method generally receives much less emphasis than transport theory (if, indeed, any at all) in most nuclear engineering curricula. As a result of this and the importance of the method to criticality calculations, it is described in more detail here (following the development in Ref. 1) and in Appendix A.

In the context of nuclear criticality safety, the Monte Carlo procedure is applied by developing a model that is capable of "tracking" individual (fictitious) neutrons through a material medium containing fissile and other species. A complete description of neutron travel paths accounts for: types of and distances between interactions; energies and directions of travel; production from fission or other reactions; and loss by capture or leakage. It is well known that a precise outcome for any such "event" cannot be identified beforehand. Instead, each may be described by a probability distribution that quite accurately predicts the composite behavior of a very large number of events. These "laws" are usually described or otherwise characterized by such familiar parameters as reaction cross sections, mean free paths, scattering distributions, and energy spectra. Since the outcome of an event is random (within the bounds of the governing laws), the Monte Carlo method makes event selection equivalent to choosing a random number. The overall results (usually in terms of reaction rates) of following a number of neutrons through their "paths of events" in a particular medium provide a reasonable approximation to the behavior of the actual system represented by the model.

One fundamental requirement for developing a Monte Carlo procedure is to relate physical events to random numbers in an unambiguous manner that is firmly tied to the governing laws of probability. This may be accomplished by computing a probability density function $p(x)$ for each possible interaction parameter x defined by

$$p(x)\,dx = \text{probability that the interaction parameter will lie between } x \text{ and } x + dx \quad , \tag{6.5}$$

and normalized such that

$$\int_{-\infty}^{\infty} dx\, p(x) = 1 \quad . \tag{6.6}$$

In practice, the function may be continuous as defined above, or it may be discrete when only a finite number of outcomes is possible. The fission-neutron energy spectrum $\chi(E)$, for example, is a continuous probability density function. An example of a discrete function may be constructed by noting that Σ_a/Σ_t and Σ_s/Σ_t are the probabilities for neutron absorption and scattering, respectively, in a material with total macroscopic cross section Σ_t.

The cumulative probability distribution function $P(x)$ is defined by

$$P(x) = \int_{-\infty}^{\infty} dx'\, p(x') \quad . \tag{6.7}$$

It is single-valued, ranges from zero to unity, and preserves probability. Thus, it allows a number selected randomly on the 0–1 interval to be assigned unambiguously to event x. Most importantly, the probability of occurrence of any given event is identical to the probability that the corresponding random number will be selected.

The basic components of a Monte Carlo code are related to random numbers, geometry, tracking, and scoring. Random numbers are selected on the 0–1 interval. In practice, a mathematical algorithm that generates "pseudorandom" numbers is often preferred for simplicity and for the ability to repeat calculations in an exact manner.

The geometry identifies the location of each material type in the system. Macroscopic cross sections describe the nuclear characteristics of each region. The boundaries that separate regions often serve as convenient neutron "scoring" locations. For example, neutrons leaking from one material to another or from the system as a whole may be counted, or "scored," as they cross boundaries.

A useful system in many codes, sometimes called "nesting," allows a material and shape to be set at particular coordinate locations such that other additions of material will be considered to exist only in nonintersecting areas; e.g., in the KENO code,[17] if a large cylinder is

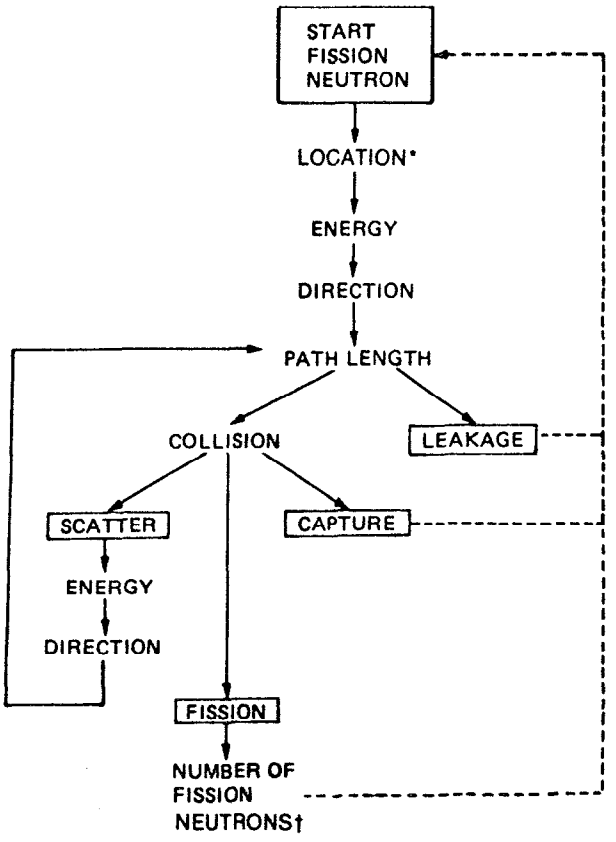

Fig. 6-1. Flow diagram for an analog Monte Carlo method used to calculate the effective multiplication factor k_{eff} (Ref. 1).
*Locations for generation N based on fission points from generation $N - 1$.
†Record neutron number and fission location for generation $N + 1$ starting locations.

placed over a smaller cylinder, the new composition is considered to be only in the annular region bounded by the outer surfaces of each figure. Options that allow automatic repetition of geometries to make up arrays are also common.

A simplified flow diagram for a multiplication-factor calculation is shown in Fig. 6-1. The procedure, a form of unbiased or "analog" Monte Carlo, follows "generations" of neutrons and compares the number started to the number produced to compute k_{eff}. In an arbitrary generation, the locations for starting individual neutron histories are selected from those of the previous generation, the first generation having started neutrons from an arbitrary distribution. The energy and direction are selected randomly from appropriate cumulative distribution functions. Neutron path lengths between collisions depend, of course, on the total macroscopic cross section $\Sigma_t(E)$. The geometry determines whether a neutron leaks or experiences a collision at the end of its path length. Collision types are selected randomly in accordance with the appropriate reaction cross sections. Scattering events change the energy and direction of the neutron before it continues through the system. Leakage, capture, and fission terminate the history and signal the start of the next fission neutron. For fission reactions, the number of neutrons is randomly selected with the resulting number and the location of the event stored for use in starting neutrons of the next generation. Since it is typical to start a fixed number of neutrons in each generation N, the number of fission points in generation $N - 1$ may be adjusted upward or downward by random duplication or elimination, respectively.

As more specific examples, the expressions that follow lead to valid selection of the physical parameter correlated to random number n. Isotopic emission of a fission neutron in spherical-coordinate directions θ and ϕ (two random numbers n and n' are required) may be selected from

$$\cos\theta = 2n - 1 \qquad (6.8)$$

and

$$\phi = 2\pi n' \quad . \qquad (6.9)$$

Path length x in a homogeneous region with total cross section Σ_t may be selected from

$$n = 1 - \exp(-\Sigma_t x) \quad . \qquad (6.10)$$

For interaction type, the interval 0–1 may be arbitrarily divided into parts associated with each fractional constituent of the total cross section (e.g., the ratios Σ_c/Σ_t, Σ_f/Σ_t, and Σ_s/Σ_t sum to unity), so that the subinterval into which n falls specifies the interaction type.

By considering "neutron weight" rather than whole particles, the biased Monte Carlo methods enhance computational efficiency. The situation sketched in Fig. 6-1, for example, can be modified by considering each collision to be a partial absorption and a partial scatter in proportion to the respective cross sections. Likewise, fission can produce a neutron weight equivalent to the average number of neutrons per fission rather than an integral number of neutrons. Application of region-wise biasing is considered further in Appendix A. Calculations other than those for k_{eff} can be biased by other procedures to enhance efficiency.

The Monte Carlo method is particularly useful in performing criticality calculations since it can handle many combinations of regular and irregular shapes. Although substantially more costly than one-dimensional discrete ordinates calculations, three-dimensional Monte Carlo is comparable in cost with the two-dimensional versions. Little can be saved by reducing the dimension of the Monte Carlo calculations since the more time-consuming operations tend to be relatively geometry independent. A good overview of Monte Carlo applications to nuclear criticality safety is provided in a report by Whitesides.[18]

The most widely used Monte Carlo codes for criticality safety are KENO (Refs. 17, 19, and 20) and MONK (Refs. 21 through 24) from the United States and United Kingdom, respectively. Each has been validated exten-

sively against benchmark experiments such as those described in Chapter 5. More specific information on the two codes is provided in Appendix A.

Cross Sections

The most readily available cross sections for general purpose calculations have been the 6- and 16-energy-group Hansen-Roach sets.[25] Since they have been in use for a considerable time, their ranges of validity and biases have been established for many systems. Calculations with the set tend to agree with experimental critical masses to better than 10% for spheres of uranium at enrichments >5 wt% ^{235}U in combination with many other materials.[26] Similar agreement has been noted for hydrogen-moderated plutonium or ^{233}U with or without water reflection. The Hansen-Roach set has nonconservative biases, i.e., it overcalculates critical masses or undercalculates k, for hydrogen-moderated uranium with enrichments below ~5 wt%. Discrepancies of ~15 and 30% in critical mass or k have been noted for 2 wt% and 1.4 wt% enriched uranium, respectively.

Cross-section sets with relatively few groups, like those of Hansen-Roach, are especially favored for discrete ordinates calculations since the running time and number of energy groups are roughly proportional. The Monte Carlo method, however, does not share this limitation since it employs the cross sections to compute interaction rates, and does not require evaluation of the complex double integrals in Eqs. (6.1) and (6.2). Thus, a number of larger sets have been and are being developed, primarily for Monte Carlo calculations.

Of current special interest is work being done on a criticality safety reference library (CSRL). A recent version,[27] CSRL-V consists of a 227-group library developed from ENDF/B-V data processed through the AMPX system.[28] The Evaluated Nuclear Data File (ENDF) is one of the world's most extensive compilations of nuclear cross-section data encompassing over 80 nuclides, all important interaction types, and essentially continuous energy dependence.[29] That this computer-based file is subject to periodic updating is especially valuable. The CSRL-V set is intended as a data base for a subsequent generation of problem-dependent fine- and broad-group cross sections that can be used in both discrete ordinates and Monte Carlo codes. A key feature of this library is that its energy structure has as subsets those of the earlier 16-group Hansen-Roach, 27-group SCALE (which itself uses the first 15 Hansen-Roach groups plus 12 new thermal groups),[30–32] and 218-group CSRL cross-section sets.[33] The library is also designated to be used with codes such as KENO-IV (Ref.17), ANISN (Ref. 11), DOT (Ref. 13), and XSDRNPM [a cross-section processing code with one-dimensional S_n calculation capability derived from XSDRN (Ref. 34) and included in the AMPX (Ref. 28) package].

Validation

Like experimental results, computed critical conditions must be evaluated for reliability before being accepted.[26,35–39] Indices of accuracy, such as probable error or standard deviation, are not as directly available from calculations as they are from experiments. Although there is some exploration in this direction, the present lack of indices leaves comparison with experimental data as the only means of judging the reliability of a computational scheme.

Before proceeding, it is important to differentiate between the precision of a calculation and its accuracy. The precision is based on the effectiveness of the code in using the input data to obtain approximate solutions to the Boltzmann equation. In discrete ordinates codes, the iterative-solution process is capable of high precision provided that convergence criteria and mesh spacing are properly specified and employed.

The Monte Carlo method is statistical in nature such that results from the analog method are characterized by a standard deviation proportional to the square root of the number of events, e.g., similar to the typical $n \pm \sqrt{n}$ reported for a radiation counting experiment. For biased Monte Carlo, the same principle applies even though treating "weight" is less direct than counting particles. Biasing is ultimately intended to strike a favorable balance between precision and computation costs. In any case, the precision of Monte Carlo calculations depends on the number of histories considered in the frustrating "squared" manner where a factor of 2 in precision costs a factor of 4 in computer time. The statistical nature of the Monte Carlo approach indicates that results with large deviations cannot be discounted even though not highly probable.

The overall accuracy of most calculations is controlled by the accuracy of the cross-section set (including the theory and compromises employed to make it usable) and by the degree of complexity of the geometry. A significant concern with complex geometries is correct modeling of resonance capture [related to the factor p in Eq. (2.1)]. The process may be assisted by use of such codes as "Nordheim's integral treatment and working library" (NITAWL) in the AMPX system.[28] On the whole, comparison with experiments provides one of the best measures of overall calculational accuracy.

The ANS-8.1 Standard,[39] described earlier in Chapter 4, includes provision for use of calculations validated against experimental measurements for establishing subcritical limits. Previously, this subject was addressed by a separate standard, "American National Standard Validation of Calculational Methods for Nuclear Criticality Safety."[40]

The ANS-8.1 Standard defines a bias as "a measure of the systematic disagreement between the results calculated by the method and experimental data" and an uncertainty in the bias as "a measure of both the precision

of the calculations and the accuracy of the experimental data." It then requires that biases be established by correlating results of criticality experiments with the results of the calculation method of interest applied to the same systems. This serves to normalize a method over its areas of applicability (i.e., ranges of material compositions and geometric arrangements under consideration). Use of the method may be extended beyond the range of experimental conditions by employing the trends in the bias.

The Standard requires that the uncertainty in the bias contain allowances for uncertainty in the conditions of the experiment, for lack of precision in the calculation method, and for extension to conditions outside of those covered by the experiments. When adequate allowance is made for the uncertainty in the bias, conditions predicted to be subcritical would be expected to be subcritical.

For the usual situation where calculations are performed with a computer program, the ANS-8.1 Standard requires checks to confirm that mathematical operations are performed as intended. This includes reconfirmation following each change in the computer program.

One reason that the KENO code is widely used for criticality safety studies is that it has been extensively validated against experimental data.[41–43] The critical system pictured in Fig. 6-2 (and sketched by computer in Appendix A) represents an example included for testing purposes in the KENO-IV manual.[17] In this case, the calculated values of 0.987 ±0.016 for 1700 neutron histories and 0.989 ±0.011 for 4000 happen to agree particularly well with the $k_{eff} = 1.0$ critical condition.

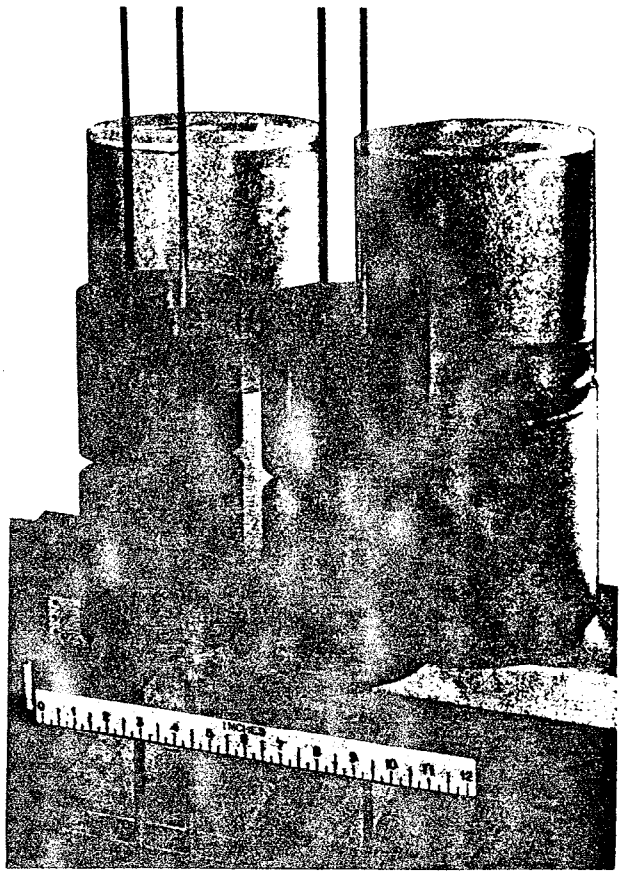

Fig. 6-2. Critical eight-unit array experiment compared to KENO-IV calculations. The four units in the foreground are uranium metal, the four in the background are uranyl nitrate in Plexiglas cylinders.[17]

Quality Assurance

The validation standard does not address the always-present concern over errors generated by improper use of the code. The "one knowledgeable in the field" assumed to be using the code would be expected to uncover such errors as a matter of course. Common sources of input errors include improper cross sections, material designations, and dimensions. Machine errors are generally identified by self-monitoring features of the computers.

The persons who perform computer calculations are not necessarily the criticality safety specialists or plant management personnel who wish to employ their results. It is then especially important that the suppliers be convinced of the validity of their own calculations and be able to reassure the users that the results represent the situation initially requested. The users, on the other hand, should understand enough of the basic features of the calculational procedure to make independent assessments. In any case, current practice suggests that at least one person in addition to the supplier should check the calculation for consistency. This operation, referred to as quality assurance or simply QA, may be performed by one within the same organization, by a member of a different group, or by an outside consultant, as appropriate. A totally independent verification might employ different models, codes, cross sections, and computers used by persons who do not know of the original results. Such extreme measures are not usually justified except for especially difficult cases or where safety margins and conservatisms are small. A more general approach is described below.[36–38,44]

Fundamental steps in a QA program for computer calculations may include:

1. verification of accurate specification of the physical system to be modeled
2. verification of appropriate assumptions included in the computer model (e.g., presence of moderator and reflector materials under nominal and, if appropriate, postulated accident conditions; other conditions and contingencies such as those identified in Chapter 7)
3. code input checks (usually made from the computer listing of geometry, material, cross-section, and option data)
4. code convergence verification
5. comparison of results to benchmark criticality data from other sources and to parametric cal-

culations run by the same calculational procedure
6. examination of calculated reaction rates and neutron spectra for general consistency
7. check of code options to assure compatibility with those used to validate the method against experimental data.

By using the computer output rather than the actual input data from cards or a remote terminal, the person(s) tasked with quality assurance can avoid some of the problems the originator may have experienced. A good QA procedure is to work backwards by determining the features of "what was actually calculated" and then comparing this to "what was desired."

Specific "pitfalls" and QA practices for computer work are described for several nuclear facilities in Ref. 44. Additional "food for thought" to the interested reader is provided by Whitesides' "A Difficulty in Computing k-effective of the World,"[45,46] and Clark's "Snake Bites from Code Misuse and Overuse."[47]

A final comment on the subject is "a horrible example" retold by Paxton, the former head of the Los Alamos critical facilities. It emphasizes that even very knowledgeable persons can be "fooled" by computer codes, especially when time pressures are great. (As noted in Chapter 2, such willingness to admit mistakes is important to overall safety of the facility and the industry as a whole.) According to Paxton[48]:

> Although it should be apparent that even validated computational methods cannot be substituted blindly for experiments, it took a shocking error to impress me with this fact. Let me share with you an experience that may help some of you to avoid the sort of mistake that I made. This example arose in response to an army inquiry, passed on by the U.S. Atomic Energy Commission (AEC), about increasing the efficiency of routine air transport of a certain weapon component. This component of enriched uranium, cadmium-plated, was packaged in the container shown in [Fig. 6-3] consisting of a 7.4-in.-diam. steel cylinder within a 16-in.^{2} tubular framework. During transport these packages were secured so as to provide extra spacing, and the number per aircraft was limited to 12. The army asked for an increase of this limit to 16.
>
> Originally, in the days before arrays could be computed in detail, the following extreme accident conditions were assumed. The tubular framework and the cadmium were lost, and components within cylindrical containers were packed together in a closely reflected array. A density-analog scheme established the acceptable number (12 seemed more than adequate at that time). We believed that uncertainty in the criticality estimate was more than offset by the farfetched accident mode.
>
> Once established, however, an accident model is

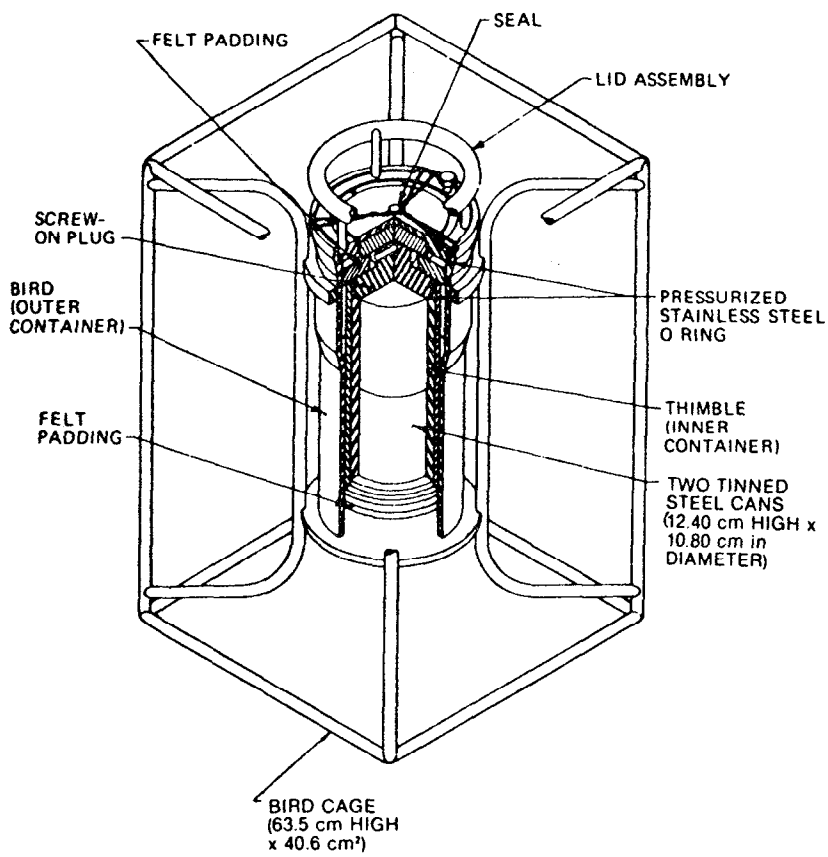

Fig. 6-3. "Birdcage" used to maintain safe spacing of fissionable material during storage and transport.[48]

unlikely to be made less conservative. For reconsideration, the same array was assumed except that it was flooded with aircraft fuel. By this time we thought we were able to promise realistic Monte Carlo results, and colleagues at Lawrence Radiation Laboratory (LRL) and the Y-12 Plant offered to check our calculation.

To satisfy an impatient General, however, we submitted the k_{eff} values shown in the top line of [Table 6-I] without waiting for confirmation and recommended the increased limit. Then LRL and Y-12 showed that we were wrong; we tracked down a code difficulty and came into line. But, as indicated by the rest of the table, the accident model was too extreme for even one component; so the AEC and the army threw up their hands and left things unchanged.

It was especially embarrassing to admit that we had used a subroutine that always gave cosine(scattering angle) = 1. The KENO code for an IBM machine was imported from Oak Ridge, and the seemingly innocuous subroutine did not come through on a CDC machine at Los Alamos. It is clear that validation at Oak Ridge did not help us—that we should not have relied on the code before proper confirmation under our condition. [At Los Alamos the code did reproduce criticality of a bare U(94) sphere.] We do not now consider it a luxury to run a problem of a new class on two different codes, such as KENO and the Monte Carlo ANDY, developed at Los Alamos.

TABLE 6-I
Values of k_{eff} for Flooded Systems* (Precision about ±0.02)

	Single Unit Water Inside	Array 3 × 3 × 3 Dry Inside	Array 3 × 3 × 3 Water Inside
Los Alamos (initial)	0.89	0.92	1.00
LRL (MORSE)[a]	0.96[b]	0.99	0.97
Y-12 (KENO)	0.95	1.00	0.99
Los Alamos (KENO)	0.97	0.98	1.04
Los Alamos (TWOTRAN)[c]	0.97		

*From Ref. 48.
[a]Similar values from the 05R code.
[b]For two flooded units with bases in contact, k_{eff} = 1.00.
[c]A two-dimensional transport code.

Exercises

6-1. Describe the roles of the diffusion theory, transport, and Monte Carlo methods in criticality safety.

6-2. Explain why larger cross-section sets such as those in the SCALE system generally may be expected to result in greater accuracy than smaller ones such as the Hansen-Roach sets.

6-3. Distinguish between precision and accuracy in computer calculations.

6-4. Describe the necessity for and the general process of computer code validation.

6-5. Explain the basic steps in QA of computer calculation output.

6-6. Consider a bare sphere of uranium with number (nuclide) densities 4.480×10^{-2} or 4.480(−2) at/b·cm for ^{235}U, 2.658(−3) for ^{238}U, 4.827(−4) for ^{234}U, and 9.57(−5) for ^{236}U. Determine:

a. the composition by atom percentage (at.%)
b. the composition by weight percentage (wt%).

See Appendix A.

6-7. Consider a computer model for a light water reactor (LWR) fuel pin with three compositions (in at/b·cm):

Composition 1	^{235}U	6.3552(−4)
	^{238}U	2.1782(−2)
	oxygen	4.4834(−2)
Composition 3	hydrogen	6.6888(−2)
	oxygen	3.3444(−2)
Composition 2	zirconium	4.2510(−2)

Convert each composition to a weight percentage basis and describe the physical nature of each. Describe and sketch the geometry for the following sequential overlays (with dimensions in centimeters and referenced to the center point as origin of the axis system):

Composition 1	cylinder	radius 5.08(−1)
		height ±1.8288(+2)
Composition 2	cylinder	radius 5.69(−1)
		height ±1.83(+2)
Composition 3	cuboid	sides ±7.67(−1)
		height ±1.85(+2)

6-8. Consider a computer model for an LWR fuel pin with the following compositions:

Fuel region	uranium dioxide (UO_2) pellet
	3.0 wt% ^{235}U
	97.0 wt% ^{238}U
	10.06 g U/cm
Cladding region	Zircaloy (6.5 g/cm^3)
Moderator	water

Calculate the number densities for each constituent in the three separate compositions.

6-9. The KENO model for the "double 2 × 2 array" in Fig. 6-2 (and Fig. A-2 in Appendix A) is developed with the following:

Composition 1	^{238}U	3.2275(−3)
	^{235}U	4.4802(−2)

Composition 2	hydrogen	5.8100(−2)
	nitrogen	1.9530(−3)
	oxygen	3.6927(−2)
	^{235}U	9.8471(−4)
	^{238}U	7.7697(−5)
Composition 3	carbon	3.5552(−2)
	hydrogen	5.6884(−2)
	oxygen	1.4221(−2) .

Convert each composition to a weight percentage basis and describe its nature and general location.

6-10. A computer model of a two-region subcritical assembly has the following compositions:

^{235}U	8.193(−4)	9.879(−5)
^{238}U	1.608(−2)	1.362(−2)
Oxygen	3.380(−2)	2.744(−2)
Aluminum		6.941(−3)
Iron	8.958(−3)	9.975(−3)
Nickel	1.339(−3)	1.687(−3)
Chromium	2.474(−3)	2.698(−3)
Manganese	1.301(−4)	1.502(−4)
Molybdenum		2.150(−4) .

Calculate weight percentages of each constituent and of the uranium dioxide, aluminum, and stainless steel mixtures.

6-11. Research reactor fuel in a shipping cask has a composition:

^{235}U	9.62(−4)
^{238}U	7.15(−5)
Aluminum	5.86(−2)
Iron	1.43(−4)
Silicon	2.84(−4) .

Calculate weight percentages of each constituent.

6-12. The following composition of liquid-metal fast breeder reactor fuel pellets represents a volume fraction of 0.328 of a smeared combination of fuel, cladding, and coolant:

^{239}Pu	2.205(−3)
^{240}Pu	2.970(−4)
^{241}Pu	4.306(−5)
^{242}Pu	5.050(−6)
^{238}U	4.750(−3)
^{235}U	1.068(−5)
Oxygen	1.462(−2) .

Calculate the number densities for the fuel pellets. Determine the weight percentages of each composition.

6-13. Consider the following composition:

^{235}U	7.481(−3)
^{238}U	5.164(−4)
Hydrogen	3.943(−2)
Oxygen	3.448(−2) .

Calculate the weight percentages of each constituent separately and of the uranium dioxide and water mixtures.

6-14. Resonance absorption characteristics incorporated into cross-section libraries are indexed in terms of a parameter SIGP (as described in Appendix A), which may be calculated from

$$\text{SIGP}_i = \frac{\Sigma_s^{\text{mix}}}{n_i} = \frac{\sum_{j=1}^{j=M} \sigma_{sp}^{j} n_j}{n_i}$$

for mixture macroscopic scattering cross section Σ_s^{mix}, number density n_i, and microscopic scattering (potential) cross section σ_{sp}^{i} for constituent i, and summation over all M constituents. Using the following σ_{sp} values:

Plutonium	12 b	Nitrogen	10.0 b
Uranium	12 b	Aluminum	1.4 b
Hydrogen	12 b	Silicon	2.2 b
Oxygen	3.7 b	Iron	11.4 b ,
Carbon	4.6 b		

calculate SIGP for each fissile constituent in all fuel compositions of exercises 6-6, 6-7, 6-9, 6-11, 6-12, and 6-13.

6-15. Consider a random number generator that has produced values 0.619, 0.832, 0.657, 0.069, 0.433, 0.352, 0.521, and 0.330. For each of the numbers, determine the parameter it would select from the equations in this chapter and plot the distribution of results on a separate linear scale for:

a. polar angle
b. azimuthal angle
c. path length for thermal neutrons in ^{235}U ($\Sigma_t = 1.47\ \text{cm}^{-1}$)
d. interaction type and nuclide in the uranium dioxide composition of exercise 6-7
e. interaction type and nuclide in the mixed-oxide composition of exercise 6-12.

NOTE: For parts d and e use the data in Table 6-II and select both "events" using only one random number.

TABLE 6-II
Cross Sections for Selected Materials

Nuclides	σ_c	σ_f	σ_s
Oxygen	—		3.8
^{235}U	95	584	8.9
^{238}U	2.7	—	8.9
^{239}Pu	270	742	7.7
^{240}Pu	290	0.05	7.7
^{241}Pu	360	1010	7.7
^{242}Pu	19	—	7.7

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7 Subcritical Limits

A combination of the experimental and calculational methods considered in the previous chapters may be used to determine subcritical limits from which facility operating parameters can be generated. Limits for individual units may be established in terms of a single parameter or a combination of parameters. Other factors such as the presence of poisons or reflectors also influence limits.

Subcritical limits have been developed for certain arrays of units in storage or transportation. Neutron interaction algorithms are available for performing hand calculations for various array applications, for example, as described in the next chapter.

General Limits

The definition of "subcritical limit" from Standard ANSI/ANS-8.1-1983 is[1]:

> The limiting value assigned to a controlled parameter that results in a subcritical system under specified conditions. The subcritical limit allows for uncertainties in the calculations and experimental data used in its derivation but not for contingencies; e.g., double batching or failure of analytical techniques to yield accurate values.

The previous definition[2] was:

> A limit to a specified variable for the optimum values of all unspecified variables that keeps the system subcritical by a margin of reactivity sufficient to compensate for inexactness in experimental data and calculations, but that contains no allowance for operating contingencies (e.g., double batching) or for inaccuracies in the values of process variables (e.g., mass or concentration).

The first portion of an alternative definition[3] is:

> The limiting value assigned to a controlled parameter that results in a system known to be subcritical provided the limiting value of no other controlled parameter of the system is violated. . . .

In all three cases, emphasis is placed on one or more parameters subject to control—recognizing that all others must be assumed to take on their least favorable (i.e., most reactive) values in order for subcriticality to be maintained under all credible process conditions. It is important to recognize the difference between a subcritical limit and a best-estimate minimum critical mass. While the latter would most usually be reported as a value plus-or-minus a standard deviation, the subcritical limit could be expected to be less than the smallest critical mass estimate due to conservative application of all experimental and calculational uncertainties.

The subcritical limits must be recognized as the absolute maximum values that would ever be applied to actual process operations since they contain no allowances for any operational contingencies. For actual plant applications, the appropriate requirement stated by Standard ANS-8.1 is that:

> Process specifications shall incorporate margins to protect against uncertainties in process variables and against a limit being accidentally exceeded.

Process specifications, or "plant operating limits," are thus derived from subcritical limits by applying allowances for credible contingencies, i.e., to be in line with the "double-contingency" philosophy of criticality control.

Single-Parameter Limits

Single-parameter limits are applied to processes in which only one parameter, such as fissile mass, is controlled to prevent criticality. Thus, the definition provided above is modified to[3]:

> single-parameter limit (single-parameter subcritical limit): the limiting value assigned to a controlled parameter that results in a system known to be subcritical provided the conditions under which it applies are maintained

where allowance is still made for uncertainties in calculations and experimental data but not for operating contingencies. Specific contingencies are identified later in this chapter.

The process for establishing subcritical limits from experimental and calculational results has been described by Clark.[4] The key role of such limits in the guidance provided by Standards is reflected in studies conducted by Clark for ^{235}U (Ref. 5), ^{233}U (Ref. 6), and plutonium[7] in support of the most recent revision[1,8] of Standard ANS-8.1.

The following discussion of specific subcritical limits is based primarily on the guidance of Standard ANS-8.1 (Ref. 1) and of TID-7016, revision 2 (Ref. 3). Examples have been selected from these documents to highlight important features rather than to try to make this chapter a definitive guide by itself. For actual applications, the reader is best advised to refer to actual Standards, reports, or other documents to assure proper identification and consideration of all conditions and restrictions that apply to specific data.

Aqueous Solutions

Subcritical limits for uniform aqueous solutions of the three principle fissile species are provided in Table 7-I. The values assume reflection by an unlimited thickness of water but, of course, include no allowances for contingencies. All of these values represent higher values of k than are generally specified for other subcritical limits because of the especially detailed studies on which each is based. The limits expressed in linear dimensions apply to the diameter of a uniform cylinder of unlimited length and to the thickness of a uniform slab of unlimited area.

Of the sphere, infinite cylinder, and infinite slab dimensions included in Table 7-I, the latter two are of the most direct use. However, as considered in the next chapter, all three are especially convenient reference shapes that can be readily related to many "real" geometries. "Areal" density is defined as the product of the thickness of a uniform slab and the density of the contained fissile material. Equivalently, it is the fissile mass per unit area of the slab.

The mass, concentration, and areal density limits for plutonium in Table 7-I apply to the sum of ^{239}Pu and ^{241}Pu when the ^{240}Pu content exceeds that of ^{241}Pu (a typical situation in commercial nuclear fuel cycles). The limit on atomic ratio is equivalent to the limit on solution concentration, but the limit may also be applied to nonaqueous solutions regardless of the chemical form of the fissile nuclide.

The limits in Table 7-I are appropriate for many reflector conditions that are common to process facilities. Examples of such reflectors are the metal/water combination of a cooling jacket and a steel wall of moderate thickness. Water flooding may be a reasonable design contingency, but the assumption in the limits also allows for the unknown neutron reflecting properties of nearby concrete walls, floors, neighboring water lines and process vessels, and transient personnel. The limits are inappropriate for close-fitting reflectors of thick beryllium, BeO, D_2O, concrete, lead, or graphite. Composite reflectors (e.g., thin hydrogenous material surrounded by thick steel) may be more effective than water and, thus, also require explicit evaluation.

The limits in Table 7-I are also applicable to homogeneous mixtures and uniform slurries provided certain minor restrictions are met.[1] For nonuniform slurries, the limits must be reduced to assure subcriticality.

TABLE 7-I
Single-Parameter Limits for Uniform Nitrate Aqueous Solutions* of Fissile Nuclides

Parameter	Subcritical Limit ^{235}U (Ref. 5)	^{233}U (Ref. 6)	^{239}Pu (Ref. 7)
Mass of fissile nuclide (kg)	0.78	0.55	0.48
Solution cylinder diameter (cm)	14.4	11.7	15.4
Solution slab thickness (cm)	4.9	3.1	5.5
Solution volume, (ℓ)	6.2	3.6	7.3
Concentration of fissile nuclide (g/ℓ)	11.6	10.8	7.3
Areal density of fissile nuclide (g/cm^2)	0.40	0.35	0.25
Atomic ratio of hydrogen to fissile nuclide (lower limit)	2250	2390	3630

*$UO_2(NO_3)_2$ or $Pu(NO_3)_4$.

Metal Units

Single-parameter subcritical limits for water-reflected metal units are provided in Table 7-II. As for the solution values in Table 7-I, these represent relatively higher values of k than may be common elsewhere.

All of the mass limits in Table 7-II apply to a unit

TABLE 7-II
Single-Parameter Limits for Metal Units

Parameter	Subcritical Limit ^{235}U (Ref. 5)	^{233}U (Ref. 6)	^{239}Pu (Ref. 7)
Mass of fissile nuclide (kg)	20.1	6.0	5.0
Cylinder diameter (cm)	7.3	4.5	4.4
Slab thickness (cm)	1.3	0.38	0.65
Uranium enrichment (wt% ^{235}U)	5.0	—	—
Maximum density for which mass and dimension limits are valid (g/cm^3)	18.81	18.65	19.82

without reentrant space that can be occupied by water or other moderator. They may also be extended to a group of small pieces having the same total mass provided the moderator is excluded from between the pieces.

The ^{233}U and ^{235}U limits in Table 7-II may be applied to mixtures of either isotope with ^{234}U, ^{236}U, and ^{238}U provided that the ^{234}U is considered to be ^{233}U or ^{235}U, respectively, in computing mass. For typical plutonium, i.e., where the ^{240}Pu content exceeds the ^{241}Pu content, the total plutonium mass should satisfy the listed limit.

Multiple-Parameter Limits

Compliance with the single-parameter limits contained in Tables 7-I and 7-II would assure subcriticality for any applicable operation. The limits for metal units of tens of kilograms or less and those for solutions of <1 kg are quite small compared to the fissile material inventories necessary for efficient and economic operation of fuel cycle facilities. They seem especially tiny when contrasted to the ~100 tonnes of uranium dioxide that must be fabricated into fuel assemblies for an initial core loading of a typical light water reactor. The multiple-parameter limits described below provide substantial relief, but at the expense of requiring additional administrative controls to verify that the necessary conditions are met.

Concentration-Dependent Limits

Control of solution concentration is one of the most useful methods for relaxing single-parameter limits. As examples, Figs. 7-1 and 7-2 show spherical mass and infinite slab thickness subcritical limits, respectively, as a function of uranium concentration for aqueous solutions, metals, and homogeneous metal/water mixtures of ^{235}U. It may be noted that compositions are expressed in terms of the hydrogen-to-uranium ratio (H:U), in contrast to the volume or weight fractions more typically employed in reactor applications. The source of these curves, Ref. 3, also provides subcritical limits based on mass, volume, cylinder diameter, and slab thickness for each of the ^{235}U, ^{233}U, and ^{239}Pu nuclides.

The minima of the curves in Figs. 7-1 and 7-2 may be noted to be lower than the corresponding single-parameter limits in Tables 7-I and 7-II, respectively. This occurs because the single-parameter limits were established to an accuracy not readily achieved over the entire concentration/density ranges.

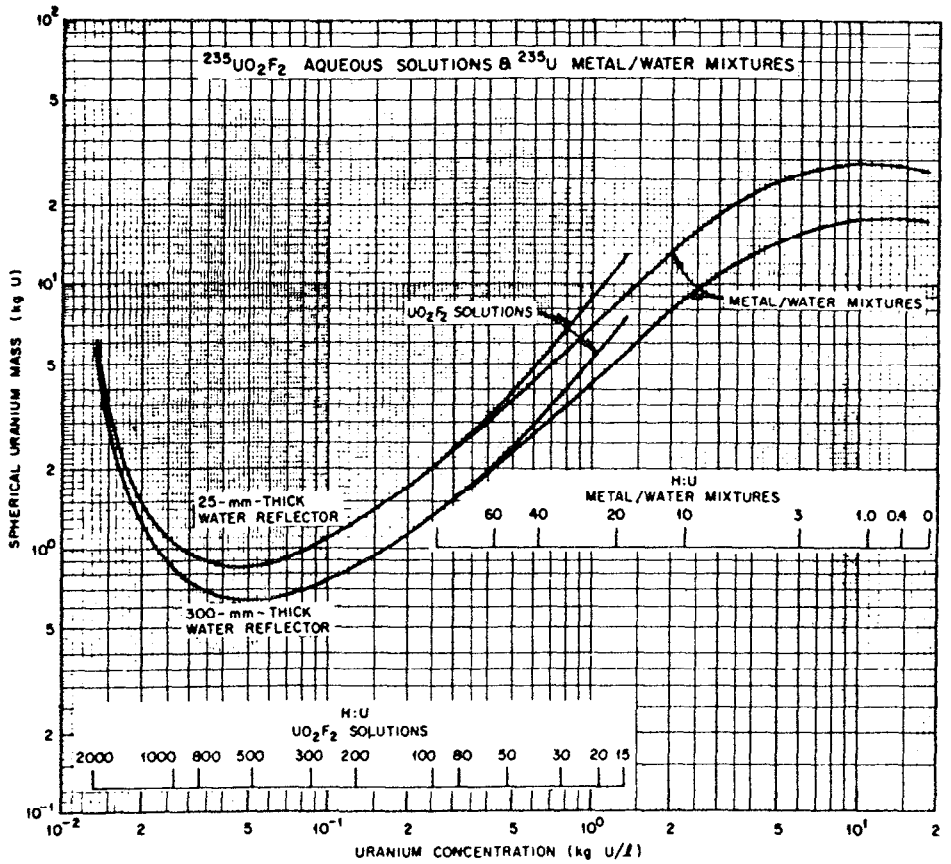

Fig. 7-1. Subcritical mass limits for individual spheres of homogeneous water-moderated and water-reflected ^{235}U (Ref. 3).

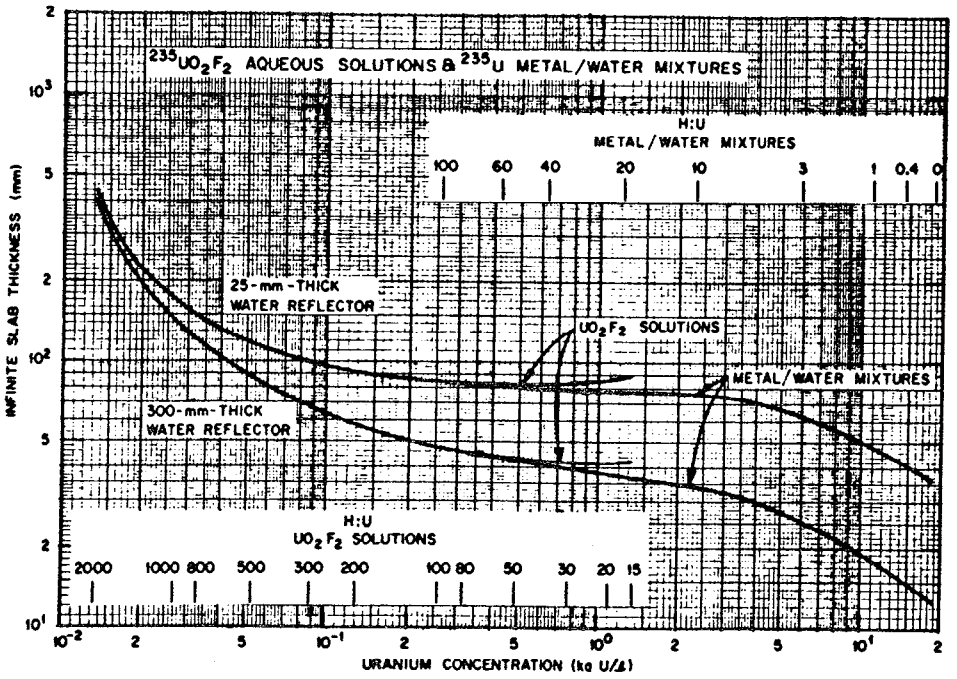

Fig. 7-2. Subcritical thickness limits for individual slabs of homogeneous water-moderated and water-reflected ^{235}U (Ref. 3).

Limits for systems with water reflectors of both 25 and 300 mm are shown in Figs. 7-1 and 7-2. The latter represents an effectively infinite thickness. When reflector conditions can be rigidly controlled, the limits for 25-mm-thick water generally provide a sufficient margin of subcriticality to compensate for water jackets on piping and for concrete 300 mm or more distant. Otherwise, the limits for 300-mm-thick water are appropriate when reflection control cannot be assured.

Materials such as beryllium, D_2O, uranium, tungsten, and concrete (especially important due to its abundance) can be more effective thick close-fitting reflectors than water. Thus, if such materials are present, the limits in Figs. 7-1 and 7-2 may not be adequately conservative. Further detail on reflection and other specific limitations on the use of data from these, and other members of the family of curves, is provided in their source document.[3]

Slightly Enriched Uranium

Light water reactor (LWR) fuel cycle facilities contain uranium of <5 wt% ^{235}U. Application of the limits in Table 7-I and Figs. 7-1 and 7-2 leads to subcriticality, but in an especially overconservative manner. More realistic limits for uranium of low enrichment can be used to advantage when strict administrative controls establish the enrichment and maintain material identification. (Both economic and safeguards considerations do make receipt of overenriched uranium highly unlikely.) Since criticality is not possible in unmoderated uranium with <5 wt% ^{235}U, moderator exclusion provides positive subcriticality of LWR fuel material.

The critical mass for moderated uranium of up to ~6 wt% ^{235}U may be significantly lower for heterogeneous arrangements than for homogeneous solutions. (This principle, of course, is well known and employed to advantage in design of fuel pin lattices for light water and other thermal reactors.) Thus, for any given enrichment in this range, subcritical limits are greater for an aqueous solution than for heterogeneous systems. The latter are generally defined as any mixtures with uniformly distributed particles larger than ~0.1 mm.

It may be possible to make natural uranium (0.711 wt% ^{235}U) metal rods critical in water if they are of the appropriate diameter and spacing (and if all constituents are of extremely high purity). On the other hand, criticality for a homogeneous mixture requires ~1 wt% ^{235}U. The subcritical limits shown in Table 7-III are appropriate to ho-

TABLE 7-III
^{235}U Enrichment Limits for Uranium Mixed Homogeneously with Water*

Material	Subcritical Limit (wt% ^{235}U)
Uranium metal	0.93
UO_2, U_3O_8, or UO_3	0.96
$UO_2(NO_3)_2$	1.96

*From Ref. 5.

mogeneous aqueous mixtures or solutions regardless of the values of other controlled parameters.

Subcritical limits on masses and dimensions of U(<5)—read as "uranium enriched to less than or equal to 5 wt% in ^{235}U"—metal and oxide rods of any diameter or latticing in water surrounded by a thick water reflector have been calculated.[9] Reactivity of a heterogeneous array depends on the surface-to-volume ratio of uranium pieces and their spacing. Therefore, limits derived for rods with diameters and lattice spacings that maximize reactivity can also be applied to other sizes, shapes, or distributions. Experiments indicate that a random arrangement is less reactive than a uniform array at optimum spacing.

Subcritical limits for uranium metal and uranium dioxide in homogeneous and heterogeneous mixtures have been calculated. As one example, Fig. 7-3 shows such limits for spherical mass as a function of uranium enrichment. (Similar curves based on volume, cylinder, and slab parameters are also available in Ref. 3.) These limits are applicable regardless of the size or shape of the metal or oxide pieces. They also apply if the environment of an aggregation of pieces does not return neutrons to the system more effectively than a closely fitting water reflector.

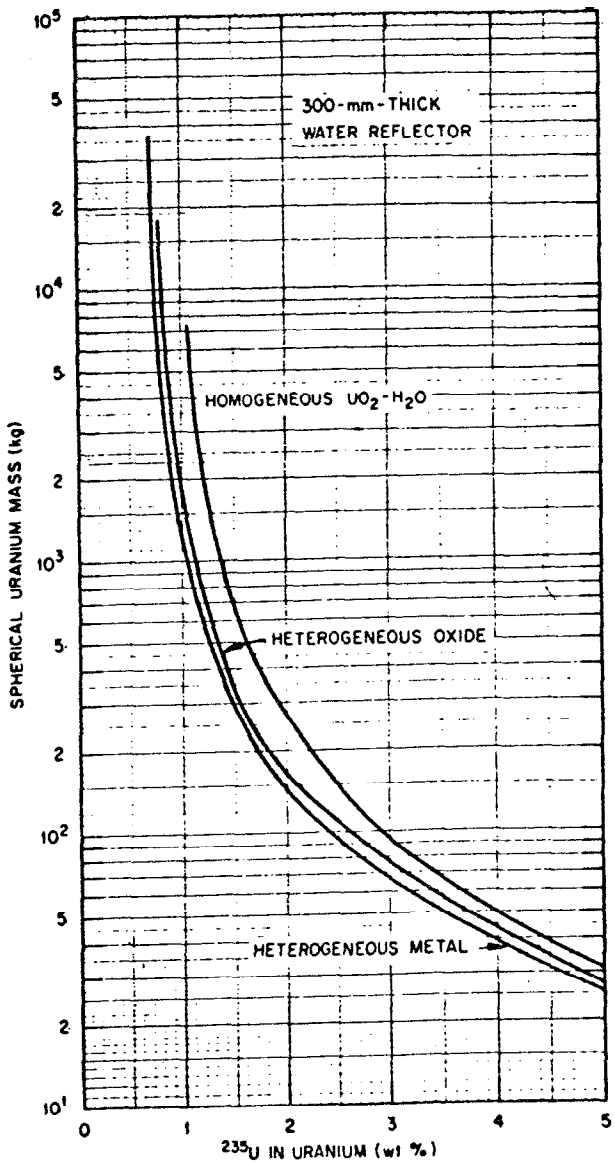

Fig. 7-3. Subcritical mass limits for individual spheres of water-moderated and water-reflected uranium enriched to 5 wt% or less in ^{235}U (Ref. 3).

Other Considerations

The subcritical limits introduced up to this point have been described in some detail to emphasize the relative complexity involved in their development and use. Other subcritical limits provided in Ref. 3 include those for:

1. mixtures of uranium and plutonium
2. mixtures of ^{233}U, carbon, and water with ^{232}Th
3. mixtures of ^{235}U, water, and graphite
4. special geometries—annular cylinders and pipe intersections.

As one example, Fig. 7-4 shows a complex set of parametric curves for ^{233}U mass as a function of both hydrogen and carbon content.[3,10]

Operating Limits

The differences between the subcritical limits provided in the first part of this chapter and limits for actual plant operations are twofold. Consideration of contingencies requires incorporation of safety factors and, therefore,

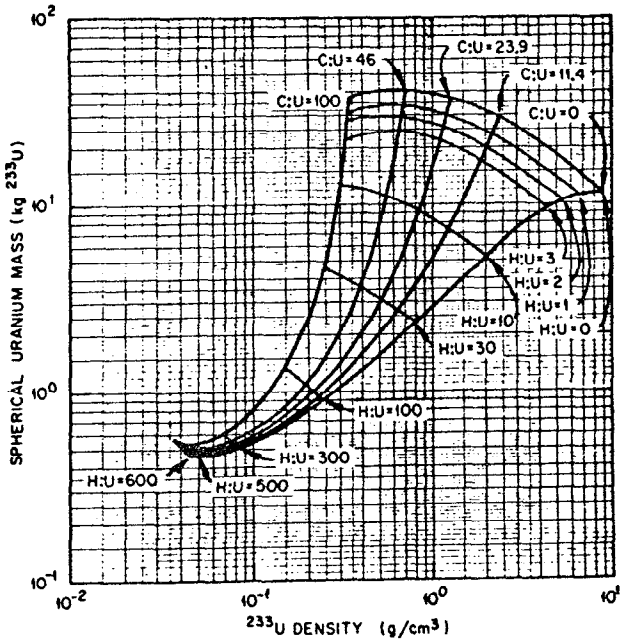

Fig. 7-4. Subcritical mass limits for water-reflected individual spheres of a homogeneous $^{233}UO_2$-carbon mixture containing various amounts of water.[3]

results in lower limits. On the other hand, features of actual systems may allow limits to be increased. Both possibilities must be resolved by careful study that includes evaluation of normal and credible abnormal conditions.

Fissile Units

An appendix to Standard ANS-8.1 (which although not formally a part of it, is present for information purposes) provides a list of examples of process condition changes that should be considered in establishing operating limits. Those applicable to single units[1,3] are:

1. a change in intended shape or dimensions, resulting from bulging, corrosion, or bursting of a container or failure to meet specifications in fabrication
2. an increase in the mass of fissionable material in a location as a result of operational error, improper labeling, equipment failure, or failure of analytical techniques.
3. a change in the moderator-to-fissionable material ratio resulting from:
    a. inaccuracies in instruments or chemical analyses
    b. evaporating or displacing moderator
    c. precipitating fissionable material from solutions
    d. diluting concentrated solutions with additional moderator
4. a change in the fraction of the neutron population lost by absorption resulting from:
    a. loss of solid absorber by corrosion or by leaching
    b. loss of moderator
    c. redistribution of absorber and fissionable material by precipitation of one but not the other from a solution
    d. redistribution of solid neutron absorber within a matrix of moderator or solution by clumping
    e. failure to add the intended amount of neutron absorber to a solution or failure to add it with the intended distribution
    f. failure of analytical techniques to yield correct amounts or concentrations
5. a change in the amount of neutron reflection resulting from:
    a. an increase in reflector thickness by addition of material (e.g., water or personnel)
    b. a change in reflector composition such as loss of absorber (e.g., by corrosion of an outer casing of absorber)
6. an increase in the density of fissionable material.

Analysis of these and other concerns, applying to both the individual units considered previously in this chapter and to the arrays discussed in the next part, tends to result in the use of safety factors or margins and produce lower limits. As stated by Paxton[11]:

> The requirement that adequate but unspecified safety margins be applied to subcritical limits is anything but straightforward. Again, judgment based on experience is called for. Some organizations have selected certain absolute safety limits, the most common for individual units being 75% or 80% of the appropriate critical mass, corresponding to dimensional limits or k values of 0.90 or 0.95. It is understood that these extreme limits apply after allowances for uncertainties of data, of analysis, and of conditions that may be encountered. Very seldom are such limits actually approached in operations outside of reactors. For example, in plants with an absolute limit of 75% of the critical mass, 50% of a critical mass will be encountered only rarely.

Extended subcritical limits described and quantified in Ref. 3 include:

1. densities reduced below the theoretical values on which the limits are usually based (applied to unmoderated fissile compounds only when accidental compaction and moderation are excluded)
2. dilution of fissile metals by nonmoderating, nonfissionable elements ($11 < Z < 83$)
3. less than full enrichment in ^{235}U of uranium metal
4. dilution of ^{239}Pu with nonfissile ^{240}Pu
5. addition of solid neutron absorbers in the form of borosilicate-glass Raschig rings
6. addition of soluble neutron absorbers (gadolinium for plutonium solutions, boron for uranium solutions)
7. allowances for special shapes that enhance neutron leakage, e.g., elongated or squat cylinders
8. close-proximity concrete reflection.

All but the last of these may provide bases for increasing the tightly defined limits described earlier in this chapter. One example of this process is illustrated by Fig. 7-5, which shows factors for increasing subcritical mass limits as a function of the volume fraction of fissile metals when dilution is by nonmoderating elements $11 < Z < 83$.

Concrete is significant in criticality safety due to its relative abundance in facilities and because its effectiveness as a neutron reflector may exceed that of water. Thus its presence, notably in a close-fitting configuration, may require reduction of associated limits. One recommendation[3] is that for closely fitting concrete (of >230 kg/m^2 areal density), full-water-reflector limits be multiplied by the following factors:

1. 0.90 for mass and volume
2. 0.80 for the diameter of infinite cylinders
3. $0.44\ \rho^{-0.155}$ for thickness of infinite slabs with fissile material density ρ in grams per cubic centimeter.

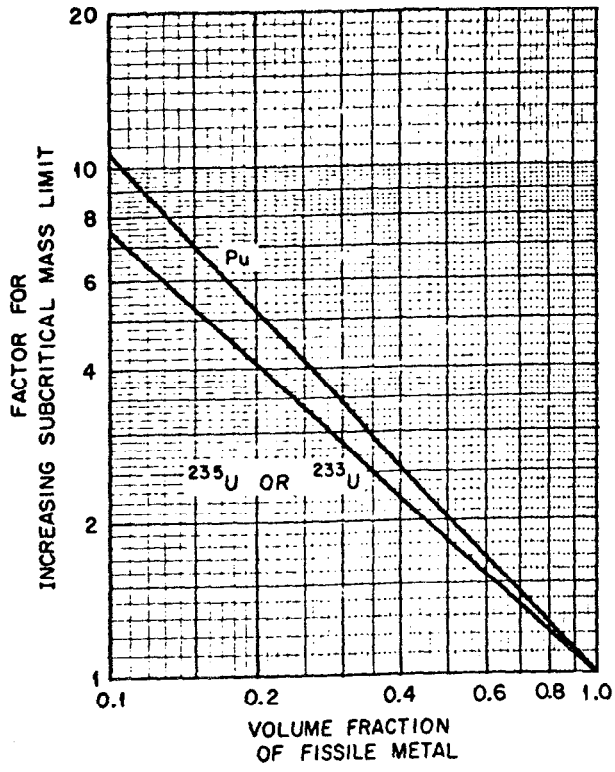

Fig. 7-5. Factors by which subcritical mass limits for fissile metals may be increased as a result of dilution by non-moderating elements $11 < Z < 83$ (Ref. 3).

Arrays

Single fissile units are appropriately considered for individual process steps, but it is extremely rare when multiple units are not stored at some point, especially just before and after particular operations. Transit between operations or facilities is also typically performed with more than a single fissile unit. Thus, subcritical limits for arrays are essential for virtually all actual operations.

Establishing the "best" nuclear criticality safety practice for storage of fissile material is highly dependent on the purpose of the storage area. Service areas for temporary storage of materials in process, areas for transient materials in transport, and long-term storage areas each generate different requirements. The spacing of such materials must be determined by accounting for the number of units, their mass and other properties, the necessary accessibility, and the desired margin of subcriticality.

The Standard "Guide for Nuclear Criticality Safety in the Storage of Fissile Materials," ANS-8.7 (Ref. 12), provides mass limits for spherical units of fissile materials assembled in cubic arrays reflected by thick water. The Standard is applicable to:

1. plutonium, ^{233}U, and uranium containing > 30 wt% ^{235}U
2. metals and wet and dry oxides
3. oxides with water content between 1.4 and 40 wt%.

Since a sphere has the minimum surface-to-volume ratio and, thus, the lowest leakage for a given mass and density, the limits are conservative for nonspherical shapes of the same composition. The low-neutron-leakage cubic array is also conservative compared to other arrays of the same number, type, and center-to-center spacing of units.

The simple prescriptions available in the Standard and elsewhere, however, cannot readily account for the introduction of arbitrary amounts of hydrogenous materials into the space between units—although the Standard does allow for the incidental moderation that would result in enclosing the units in plastic bags of no more than 10 g of polyethylene per kilogram of fissile material. If interspersed moderation is credible, e.g., that from fire protection systems, the effect must be evaluated by a validated computational technique. It is found that the reactivity effect is a complex function of fissile form, mass and spacing of units, and the amount and distribution of moderator.

Each unit in an array must be subcritical if immersed in water as a contingency against flooding. Of course, the same must be true of a double-batched unit if it is possible for this to occur. While administrative controls can reduce the likelihood of double batching, use of limited-capacity containers and innovative storage cell designs may essentially eliminate the concern, as noted by examples provided in Chapter 10.

As described above for individual units, normal and credible abnormal storage conditions should be considered when they may affect array subcriticality. Changes in operating conditions for arrays that bear examination include[1,3]:

1. a change in the moderator-to-fissionable material ratio resulting from:
   a. flooding, spraying, or otherwise supplying groups of units with water, oil, snow (i.e., low-density water), cardboard, wood, or other moderating materials
   b. introducing air bubbles between rows of fuel assemblies in a storage basin
2. a change in the interaction between units and reflectors resulting from:
   a. the introduction of additional units or reflectors (e.g., personnel)
   b. improper placement of units
   c. loss of moderator and neutron absorber between units
   d. collapse of a framework used to space units
   e. an increase in the density of fissile material during storage

TABLE 7-IV
Factors and Examples Thereof Considered in Assessing Plant Criticality*

	Fissile Material				Geometry	Moderator		Reflector		Neutron Interaction		Neutron Absorbers	General Emergencies Affecting Criticality
	Type	Chemical Form	Physical Form	Quantity		Type	Amount	Type	Size and Configuration	Spacing and Configuration	Shielding		
	A	B	C	D	E	F	G	H	I	J	K	L	M
Basic parameters affecting criticality	Fissile isotope Enrichment Density (plutonium)	Metals Alloys Chemical compounds Mixtures	Massive solid Heterogeneous solid Particulate Suspensions Slurries Solutions	Mass Volume Concentration/ density Quantity in: process storage movement	Spherical Cylindrical Slab Undefined	Hydrogenous Graphite Beryllium D_2O	Moderator-to-fissionable volume ratio Moderator-to-fissile atomic ratio	Hydrogenous Graphite Beryllium D_2O Metals Concrete Human bodies	Thickness Proximity	Separation between fissile materials Lattice pitch Basic geometry of assembly: slab-like cubic random	Type Thickness Location Neutron absorbers: cadmium sheet	Type Distribution: heterogeneous homogeneous Quantity Location	
Change of Basic Parameters During Normal Processing	Change of fissile isotope or enrichment: enrichment blending mixing plutonium and uranium	Alteration of chemical form: oxidation reduction precipitation dissolution	Alteration of physical form: machining crushing grinding casting sintering centrifuging	Change of quantity: batch size quantity in storage accountable losses	Change of geometry: transfer of liquor between vessels of different shape or size drainage ventilation	Change of moderator: leaching of graphite crucibles	Change of moderation: dissolution evaporation precipitation drying deliquescence	Change of reflector: casting into graphite mold precipitate reflected by fissile liquor	Change of reflector thickness or amount: movement of reflector during process flooding steam jacket with cooling water	Change of fissile or reflector configuration during process: approach of other fissile material during transportation fissile material in adjacent processes	Change of shielding configuration during process: removal of fissile material from transport containers	Change of absorber/fissile ratio: precipitation of absorber or fissile material change of absorber location	
Control of basic parameters during normal processing	Isotopic and/or chemical analysis Enrichment or material identification: color coding	Analysis Instrumentation: pH conductivity Visual observation	Size measurement: particle grading Instrumentation: concentration viscosity density Visual observation	Check and process weighing Analysis Accountancy Visual observation	Administrative control Instrumentation: flowmeters level indicators contents gauges	Administrative control of moderator materials	Analysis pH control Concentration control Control of quantity	Administrative control of reflector materials	Control of quantity Control of proximity to fissile materials: physical barriers	Administrative control of movement: tally or baton systems "captive key" physical barriers special porterage	Administrative control of fissile material movement in relation to shielding	Chemical analysis Instrumentation: pH control concentration control	
Contigencies (equipment malfunction, maloperation, emergencies)	Wrong selection Incorrect identification Incorrect analysis	Accidental precipitation or dissolution: accidental neutralization of acid solution Chemical changes: hydration crystallization	Change of particle size Change of concentration: deposition on vessel walls sludging	Accidental double-batching Accountancy errors Unaccountable losses	Accidental alteration of geometry: erroneous transfer syphoning fissile material entering drains vessel rupture spillages redispersion of settled sludges	Flooding: fire fighting	Accidental precipitation, dissolution or evaporation Flooding	Flooding Fissile material entering reflector: leakage into water jacket	Flooding Accidental approach of reflector materials	Accidental approach of other fissile materials Change of configuration: collapse of shelving	Accidental removal of shielding: fire or other damage corrosion of absorber materials	Accidental removal of absorber: precipitation or dissolution of absorber precipitation or dissolution of fissile material	Loss of services: electricity water compressed air Plant or site evacuation
Safety measures designed to prevent or indicate contingency	Geometric constraint: containers of special size or shape flange pegs	Interlocks Instrumentation: pH or concentration alarms	Instrumentation: pH or concentration alarms α and γ probes neutron detectors	Interlocks Surveys: γ visual	Interlocks Pipe blanks and removable sections Anti-syphons Sump probes Radiation detectors and alarms	Bund-walls Alarms	Instrumentation: pH and concentration alarms Mass control of moderator materials	Instrumentation: radiation alarms on coolant Metallurgical examination of vessels	Physical barriers Mass control of reflector materials	Physical barriers Interlocks "Captive key" Special transfer equipment Movement control organization	Fire-proof shielding Protective coating to absorber	Instrumentation: pH alarms concentration alarms "Immutable" absorbers: Raschig rings stainless steel vessels	Service failure alarms Stand-by supplies Evacuation drill
	Non-flooding locations, adequate drainage, regular patrols during silent hours, prohibition of fire-fighting with water												

*From Ref. 15.

f. the substitution of units containing more fissile material than permitted in operations as a result of operational error or improper labeling.

An increasingly useful extension of the information in the ANS-8.7 Standard is provided by the criticality indicator (CI) system.[3,13] In this method, each unit that meets specified composition requirements is assigned a CI based on its content and cell or container volume. The sum of all CIs for units in a given storage area must not exceed 100 if subcriticality is to be assured. This method can be particularly useful for mixed arrays since, once assigned, criticality indicators are not dependent on the type of fissile material.

The transportation index (TI) plays essentially the same role for arrays of fissile units in transit.[3,14] However, transport regulations (considered further in Chapter 9) distinguish between "undamaged" and "damaged" packages. The condition of the undamaged package is established by tests that simulate the effects of dropping during handling, of rain, and of anticipated seasonal temperature extremes. The damaged package is defined by a sequence of severe tests for impact, fire, and flooding. A single package must remain subcritical when immersed in water, thus in-leakage of water is assumed unless it can be specifically demonstrated before use that in-leakage cannot occur. As would be expected, the TI value assigned to a package must meet the criteria for both undamaged and damaged packages.

Summary

Practical operating limits for nuclear criticality safety must be set at levels that provide an adequate margin of subcriticality for credible contingencies. A variety of applicable factors, and examples thereof, to be considered in establishing such limits have been delineated by Chalmers[15] and are shown in Table 7-IV. The table may provide useful guidance for formulating realistic analytical models of operations (e.g., as described in the previous chapter). It may be equally valuable in setting actual limits based on the results from experiments or calculations.

Exercises

7-1. Define "subcritical limit" and distinguish between single-parameter and multiple-parameter limits.

7-2. Describe each of the four main categories of "process condition changes that should be considered in establishing operating limits" for single units as stated in the appendix to Standard ANSI/ANS-8.1.

7-3. Explain the effect of each of the "extended subcritical limits" described in Ref. 3.

7-4. Describe the changes in operating conditions that should be analyzed in establishing operating limits for storage arrays.

7-5. Differentiate between the criticality indicator (CI) and transportation index (TI) systems for establishing array subcriticality.

7-6. Review Table 7-IV. Identify the five basic levels of controls and the 13 categories and characteristics that make up the matrix.

7-7. Using data from Fig. 7-1, determine the volume fractions equivalent to atomic ratios H:U of 1000, 100, 10, 1, and 0.1.

7-8. Explain the significance of the "atomic ratio" limitation in Table 7-I. Convert this atomic ratio to a volume ratio for each fissile nuclide.

7-9. Explain the intended roles of the 25- and 300-mm water reflection in Figs. 7-1 and 7-2. Identify the concern associated with close-fitting concrete reflection of individual units and arrays.

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8 Hand Calculation Methods

A variety of hand calculation methods have been developed to simplify the process of establishing limiting conditions for individual fissile units and arrays thereof. Some have been developed specifically for nuclear criticality safety applications, while others such as the six-factor formula of Eq. (2.3) and migration-area formulation of Eq. (2.9) have been extended on a limited basis from reactor application (as described previously in Chapter 2). The methods may be used directly for design purposes or merely to provide a starting estimate for more detailed calculations. No matter what their use, validity must be established by extensive comparison to experiments and computer calculations.

Among the numerous hand calculation methods, this chapter considers only four—buckling conversion and recent formulations of surface density, density analog, and solid angle. Examples of applications of surface density and solid angle are provided in Appendixes B and C, respectively. A fifth method, known as "limiting surface density" or "NB_N^2" is addressed in Appendix D. Additional details and alternative approaches have been provided at a symposium on the subject[1] and by Clark,[2] Paxton,[3] Schuske,[4] Thomas,[5-8] and Evans.[9]

Buckling/Shape Conversion

Diffusion theory treats leakage as being proportional to the geometric buckling B_g^2 of the system of interest. Various geometries with the same buckling are assumed to have the same leakage, and vice versa. The method of buckling conversion,[10,11] or shape conversion, equates buckling values to establish equivalent dimensions for various geometries. Expressions for bucklings of spheres, cylinders, and cuboids (i.e., rectangular parallelepipeds) are provided in Table 2-I. Those for other geometries (e.g., hemisphere, annulus, and elliptical cylinder) are also available.[12]

Since diffusion theory calculations overpredict critical dimensions, an extrapolation distance δ [the extrapolation length d of Eq. (2.4) or its sum with the reflector savings δ_r of Eq. (2.5) for bare and reflected systems, respectively] must be subtracted to obtain an estimate of actual values. The buckling contains diffusion theory-calculated dimensions requiring that the actual dimensions be adjusted upward. Table 8-I shows the buckling formulation for each regular geometry as based on actual dimensions and extrapolation distances. The cylinder height and cuboid dimensions must be extrapolated at both ends as indicated by the factor of 2 preceding the applicable δ values.

In practice, extrapolation distances are determined empirically by fitting data obtained from experiments and calculations.[10] Effective extrapolation distances applicable to uranium solutions and to uranium and plutonium metals are shown in Figs. 8-1 and 8-2, respectively. The abscissa may be noted to depend on the height h and the diameter

TABLE 8-I
Expressions for Geometric Buckling in Terms of Actual Dimensions and Extrapolation Distances

Geometry	Buckling B_g^2
Sphere of radius r	$\left(\frac{\pi}{r + \delta}\right)^2$
Cylinder of radius r and height h	$\left(\frac{2.405}{r + \delta}\right)^2 + \left(\frac{\pi}{h + 2\delta}\right)^2$
Cuboid of dimensions a, b, and c	$\left(\frac{\pi}{a + 2\delta}\right)^2 + \left(\frac{\pi}{b + 2\delta}\right)^2 + \left(\frac{\pi}{c + 2\delta}\right)^2$

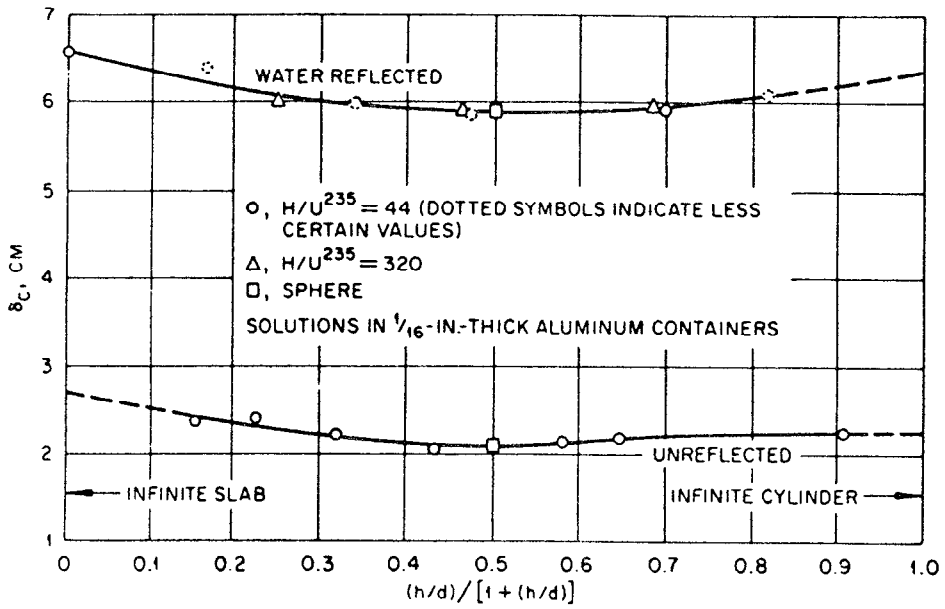


Fig. 8-1. Effective extrapolation distances for cylinders of height h and diameter d containing U(93.2)O_2F_2 solutions.[10]

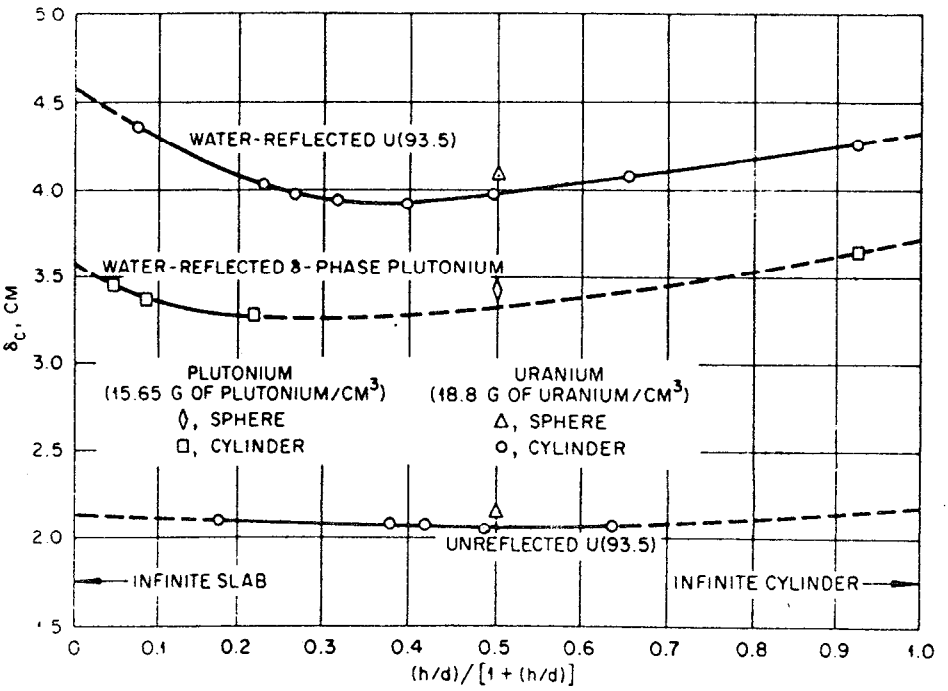


Fig. 8-2. Effective extrapolation distances for cylinders of height h and diameter d containing U(93.5) metal and δ-phase plutonium metal.[10]

d of a cylinder according to $(h/d)/[1+(h/d)]$. This particular formulation was designed to allow a finite representation of an essentially infinite range of height-to-diameter ratios. It also provides a single, averaged extrapolation distance to be applied to both the radial and axial dimensions (in contrast to the real situation where the curved and flat surfaces have slightly different values).

Extrapolation distances for the effectively one-dimensional geometries—the sphere, infinite cylinder, and infinite slab—are read from Figs. 8-1 and 8-2 above abscissa values of 0.5, 1.0, and 0.0, respectively; the latter are also appropriate for cuboid lengths. Values are provided for both bare and reflected geometries. The latter represent the sum of the extrapolation distance and the reflector savings defined by Eq. (2.5). However, since the reflector savings has been defined elsewhere in reactor

theory as the sum itself, confusion is avoided by merely referring to the curves as the extrapolation distances for bare and reflected systems, respectively.

Shape conversions using the expressions in Table 8-I are most easily accomplished going from actual geometries to those of effectively one dimension. The reverse process, on the other hand, requires the height-to-diameter ratio for a cylinder or the ratio of sides for a cuboid to be specified in advance.

As an example of the buckling conversion method, consider a reflected cylinder of uranium solution with a height of 30 cm and a diameter of 20 cm. The abscissa of Fig. 8-1 is

$$\frac{h/d}{1 + h/d} = \frac{30/20}{1 + 30/20} = 0.6 \quad ,$$

with the result that $\delta = 5.9$ cm can be read from the top (reflected) curve. For conversion to an equivalent sphere, δ (the square at an abscissa value of 0.5 on Fig. 8-1) has essentially the same value as for the cylinder. Equating the bucklings in Table 8-I for the sphere and cylinder leads to the expression:

$$\left(\frac{\pi}{r + \delta}\right)^2 = \left(\frac{2.405}{r + \delta}\right)^2 + \left(\frac{\pi}{h + 2\delta}\right)^2 \quad .$$

Since the radius is half the diameter and all dimensions are in centimeters,

$$\left(\frac{\pi}{r + 5.9}\right)^2 = \left(\frac{2.405}{10 + 5.9}\right)^2 + \left(\frac{\pi}{30 + 11.8}\right)^2$$

$$= 2.85 \times 10^{-2} \text{ cm}^{-2} \quad ,$$

and solving for the radius of the sphere,

$$r = 12.7 \text{ cm} \quad .$$

Thus for uranium solution, a 12.7-cm-radius sphere has a leakage equivalent to that of a 20-cm-diam by 30-cm-high cylinder.

Individual unit sizes are readily compared to known critical dimensions using the buckling conversion method. Finite cylinders are generally referenced to infinite cylinders. For some of the applications described below, it is also convenient to convert unit geometries to equivalent spheres. Conversion between shapes that are extremely different (e.g., spheres and infinite slabs), however, is not advisable.

Surface Density Method

If an array of well-spaced units containing fissile material were compressed to form a slab on the floor, and that slab were in turn reflected by water, it would be desirable for subcriticality to be the result. The surface density model for calculating limiting conditions of arrays derives from analyzing of such a situation.

The surface density method views an array in terms of its fissile material density as projected onto a bounding plane. Usually this is the floor, but occasionally it may be a wall. Comparison of the resulting value to the density of a water-reflected critical slab of the same composition is then the basis for establishing material limits. The choice of plane is usually determined by whichever projection satisfies the "safety criterion" established below. If just one of the planes (e.g., x-y, x-z, or y-z) does so, then safety is demonstrated.

Since multiple formulations, referred to as "surface density models," do exist,[1,4-6] it is important that features not be mixed indiscriminately. Any modification of a single procedure or combination of portions of several procedures must be subjected to reverification.

The surface density model described in Ref. 5 is a well-verified and recent formulation. It suggests an allowed surface density expressed as

$$\sigma = 0.54\, \sigma_0\, (1 - 1.37f) \quad , \qquad (8.1)$$

where σ_0 is the surface density in grams per square centimeter of a critical water-reflected infinite slab and f is the "fraction critical," i.e., the ratio of the mass of a unit in the array to the critical mass of an unreflected sphere of the same material. [Since negative surface densities are not achievable, the fraction critical must not exceed 0.73 for this formulation to be applicable. The detailed derivation of the expression shows that k_{eff} of the unit is kept to no greater than 0.9, which, according to the expression $k_{eff} = (m/m_0)^{1/3}$ (Refs. 7 and 8), corresponds to a fraction critical of 0.73.]

If each fissile unit is considered to be located at the center of a cube and the cubes are stacked to form a regular array (one column of which may be represented by Fig. 8-3), the surface density is

$$\sigma = \frac{nm}{d^2} \quad , \qquad (8.2)$$

where n is the number of units stacked perpendicularly to the bounding plane (probably the floor), m is the mass of a unit in grams, and d is the center-to-center spacing between the units (or, equivalently, the length of each side of the cubical cell). [For noncubic cells, the d^2 term in Eq. (8.2) may be replaced by the cell area projected to the plane; its square root serves as the characteristic dimension.] Combining Eqs. (8.1) and (8.2) shows a limiting center-to-center spacing in centimeters of

$$d = 1.37 \left[\frac{nm}{\sigma_0(1 - 1.37f)}\right]^{1/2} \quad . \qquad (8.3)$$

Based on the details of its derivation, this expression is "applicable to infinite planar arrays reflected by water at least 155 mm thick or its nuclear equivalent. The reflector is located no closer to the units in the array than the boundaries of the cells associated with the units." [5]

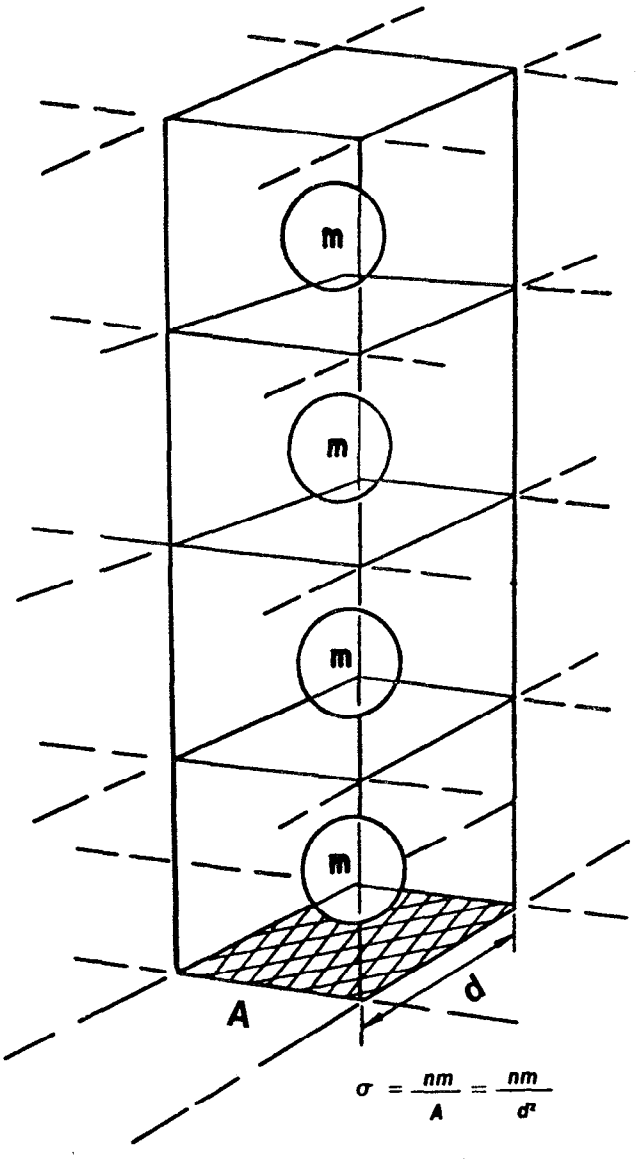


Fig. 8-3. Relationship among parameters used in surface density model.

When specific experimental data for the desired materials are available, the surface density method may be applied directly. Otherwise, subcritical limits, such as those in Figs. 7-1 and 7-2 in the last chapter, may be used for guidance as illustrated by the following example.

Consider units of fully (i.e., 100%) enriched UO_2 containing 5 kg of uranium metal mixed with water such that the hydrogen-to-uranium ratio (H:U) is 10. A conservative estimate for unreflected spherical critical mass may be established as 15 kg from the upper subcritical limit curve on Fig. 7-1. (Since the units are stated to be unreflected, use of limits for 25-mm-thick water reflection may be noted to provide some conservatism.) Thus, for this case, the fraction critical f is calculated to be

$$f = \frac{5 \text{ kg}}{15 \text{ kg}} = 0.333 \quad .$$

An estimated critical reflected slab thickness of 34 mm may be obtained from the lower curve on Fig. 7-2. This thickness may be used to calculate the critical surface density σ_0 by noting from the same figure that H:U = 10 corresponds to a uranium concentration of 2.3 kg U/ℓ:

$$\sigma_0 = (2.3 \text{ kg U}/\ell)(34 \text{ mm})$$
$$= (2.3 \text{ g U/cm}^3)(3.4 \text{ cm}) = 7.82 \text{ g U/cm}^2 \quad .$$

Then, according to Eq. (8.1), the allowed surface density is

$$\sigma = 0.54 \, (7.82 \text{ g U/cm}^2)[1 - 1.37(0.333)]$$
$$= 2.30 \text{ g U/cm}^2 \quad .$$

Applying this result to construct an arbitrary two-tier array, Eq. (8.2) recommends a spacing d of

$$d = \left[\frac{(2)(5000 \text{ g U})}{2.30 \text{ g U/cm}^2}\right]^{1/2} = 65.9 \text{ cm} = 0.659 \text{ m} \quad .$$

It may be noted that in an actual situation, calculations such as this should be supplemented by others in which variations, especially in H:U, are considered. In practice a safe separation would be based on several calculations of this type.

An important feature of the surface density method is that it is equally applicable to more irregular geometries, including those typically associated with process equipment. Whenever it can be assured that the recommended surface density will not be exceeded, units of any size or shape can be moved freely with respect to the bounding reference plane. An example of such an application is provided in Appendix B.

The surface density method compares arrays of fissile material to an infinite slab. Thus, it provides limiting conditions for an infinite array. When such an assumption does not result in overly restrictive limits and when administrative controls are readily achieved to assure that units are stacked no higher than allowed, the method may be used to very good advantage. Otherwise a different approach such as the density analog method or a detailed computer calculation may be more appropriate.

Density Analog Method

The density analog method is used to define limits that are independent of the storage arrangement. It is based primarily on observed regularities in experimental data for array criticality. Starting with the relationship between critical mass and density for bare spheres in Eq. (2.10), e.g., $m/m_0 = (\rho_0/\rho)^2$, empirical corrections were made to accommodate different shapes. The important parameters of the method are the mass, number, and spacing of units. When one of these variables is specified, an expression relating the remaining two can be used to calculate limits.

Since proposed by Paxton,[3] several formulations of density analog methods have been developed. Special care is required to assure that portions of different procedures are not mixed indiscriminately. The formulation described below is that reported in Ref. 5.

Subcritical limits for storage arrays of any shape may be defined by the following density analog relation:

$$N = \left[\frac{2.1\,\sigma_0}{m}(1 - 1.37f)\right]^3 V^2 \quad , \qquad (8.4)$$

where the critical surface density σ_0, the mass m, and the fraction critical f are as defined previously; V is the volume of the cell occupied by a unit in the array; and N is the total number of units. Reflection by at least 200-mm-thick water is an additional assumption inherent in Eq. (8.4). Procedures for extending applicability to concrete-reflected arrays have also been developed.[5]

For the special case of a cubic cell ($V = d^3$), a cubic array ($N = n^3$ for n units on a side) with unit mass m will have a minimum center-to-center spacing of

$$d = \left[\frac{nm}{2.1\,\sigma_0\,(1 - 1.37f)}\right]^{1/2} \quad . \qquad (8.5)$$

Expressions for number n and mass m may also be derived by straightforward algebraic manipulation. It should be remembered, however, that for the latter the fraction critical is also mass dependent.

As an example of this method, consider the same 5-kg unit described previously. A 125-unit ($n = 5$) array, has a recommended center-to-center spacing from Eq. (8.5) of

$$d = \left\{\frac{(5)(5000\text{ g})}{(2.1)(7.82\text{ g U/cm}^2)[1 - 1.37(0.333)]}\right\}^{1/2}$$

$$= 52.9\text{ cm} = 0.529\text{ m} \quad .$$

This spacing is substantially lower than the 0.659-m value calculated for similar units using the surface density method and does not carry the restriction that the units be stacked only two high. In fact, since the cubic array is the most reactive configuration, any other arrangement will be more subcritical. This particular analog density calculation, however, is valid for only 125 units while the surface density approach allows a two-high array of essentially unlimited extent.

It is also informative to contrast the two models as applied to a very large number of the same units considered in the previous examples. If 20 000 units were to be arranged arbitrarily, the density analog method recommends a relatively large spacing of 1.23 m for a cubic array of ~27 units on a side. The surface density method, of course, will still allow the smaller 0.66-m spacing in a roughly 141 × 142 or any other array as long as the two-tier limit is imposed by administrative control.

Solid Angle Method

The solid angle method views a reference fissile unit as having its multiplication enhanced by its neighbors in proportion to the total solid angle they subtend. For example, the central cylinder in Fig. 8-4 would have a specific solid angle subtended by the outlying units; a greater separation distance among the units would cause a decrease in this solid angle, and vice versa. The method as originally developed [13] was intended to be a quick, empirical means of evaluating interaction between small numbers of moderated fissile units.

A relationship between the effective multiplication factor k_{eff} and the maximum allowable solid angle is the basic feature of the procedure. Equally important, however, are the restrictions attached to its validity. As was noted to be the case for the two density models considered in the previous sections, several different formulations have been referred to as solid angle methods.[6,13,14] Portions of different models should never be mixed unless thoroughly verified by comparison with experiments and computer calculations. The formulation from Ref. 5 is described below.

The solid angle method as applied to an array of identical units requires knowledge of the unit's k_{eff} and the largest total solid angle subtended to any of the units by all other units in the array. When applied to an array of nonidentical units, the unit with the most limiting combination of k_{eff} and solid angle becomes the reference. Examples of both situations are provided in Appendix C.

A given array is judged to be subcritical if the actual calculated solid angle is less than or equal to the allowed solid angle given by

$$\Omega_{allowed} = 9 - 10\,k_{eff} \quad , \qquad (8.6)$$

where $\Omega_{allowed}$ is the solid angle in steradians subtended at the center of any unit by the remainder of the units in the array. The k_{eff} of a single unreflected unit may be

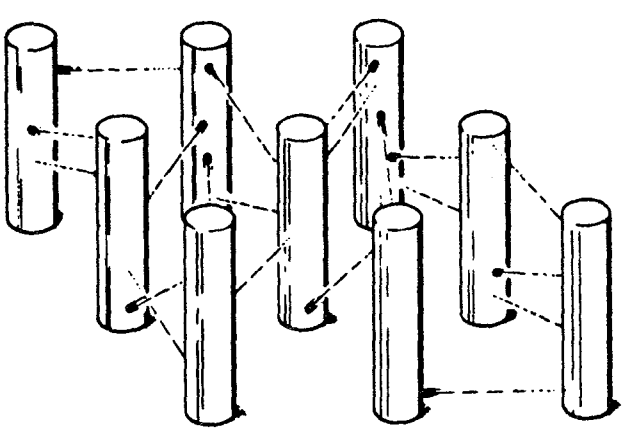


Fig. 8-4. Neutron interaction among fuel masses—basic principle of the solid angle hand calculation method.

estimated from Eq. (2.9) and from compiled data (e.g., in Ref. 12) or by using a validated calculational technique. The following conditions must be satisfied in order to apply the solid angle method [5]:

1. the k_{eff} of any unit shall not exceed 0.80
2. each unit shall be subcritical when completely reflected by water
3. the minimum surface-to-surface separation between units shall be 0.3 m
4. the allowed solid angle shall never exceed 6 sr.

The second restriction prevents criticality from the flooding of a single unit. Although it is a general requirement of the overall method, the minimum separation distance also assures that water flooding will isolate the units neutronically.

Implicit restrictions[5] on the formulation in Eq. (8.6) are (a) application of the method to other than solutions should be approached with caution, since some large arrays of metal units may not have an adequate margin of subcriticality even though the other solid angle criteria are satisfied; and (b) the method should not be applied if reflection is more effective than a thick water reflector located at the array boundary, where this boundary is assumed to be no closer to peripheral units than one-half the surface-to-surface spacing between units.

The actual solid angle, Ω, is calculated between the center of the unit chosen for reference purposes and the surfaces of other units. According to the definition,

$$\Omega = \frac{dA}{r^2} \quad , \tag{8.7}$$

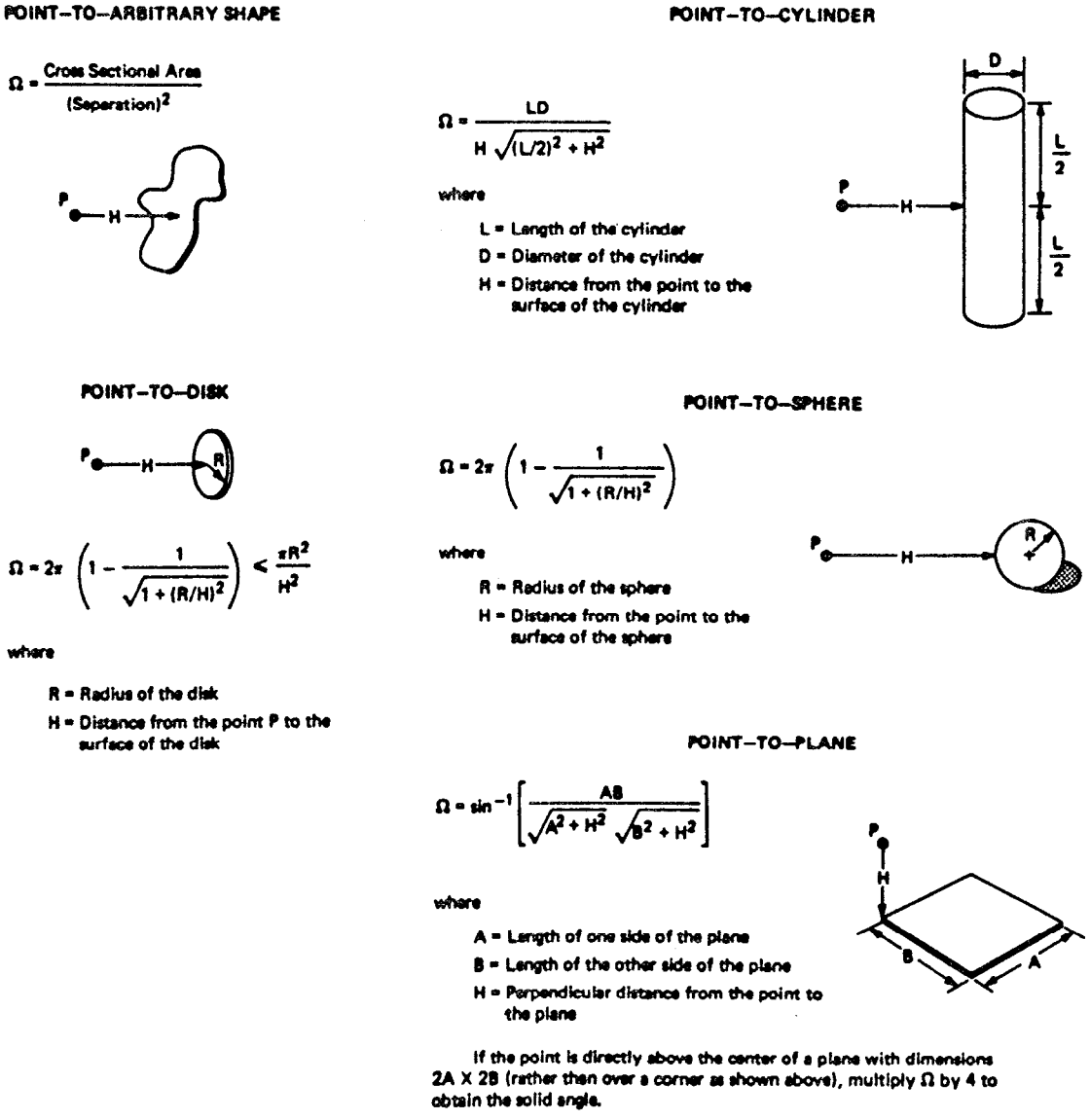


Fig. 8-5. Solid angle approximate formulas.[5]

where dA is the incremental surface area and r is the distance from the reference point to dA. Exact calculations require integration over the total area as seen directly from the reference point (e.g., "back surfaces" are excluded). Such integration is often quite complicated even for simple figures because of the manner in which the radius r changes as a function of position.

Approximate solid angles for simple geometries may be calculated from the formulas in Fig. 8-5. The approximations are based on projecting each figure's cross-sectional area onto a plane touching its front surface. Thus, for example, the expressions in Fig. 8-4 represent a cylinder and a sphere, respectively, by a rectangle and a disk. These and the other prescriptions are intended to give values greater than or equal to the actual solid angles.

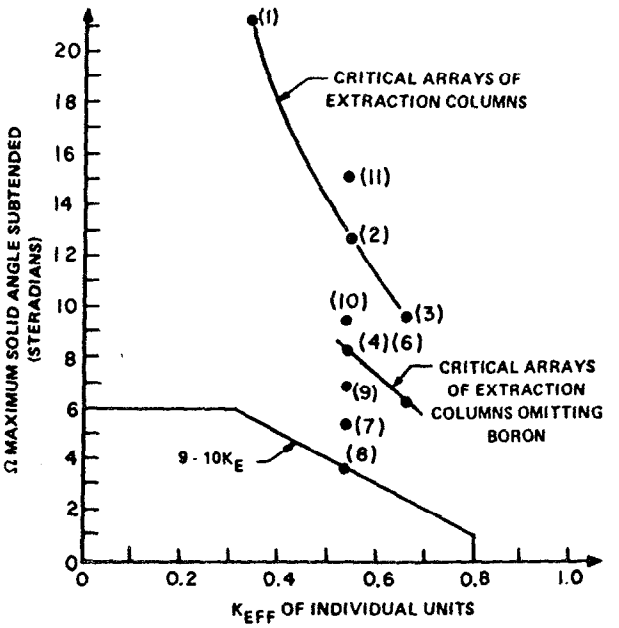
For regularly spaced arrays with identical units, solid angle calculations are based on the center-most unit. Individual solid angles need only be computed for symmetric locations with the result multiplied by the number of units located equivalently. As formulated here, the solid angle method requires consideration of all units in the array with no provision for eliminating units "shadowed" (i.e., obscured from view) by others.

Since the H values are measured center-to-surface (rather than center-to-center) and thus are not simple multiples of each other, calculations are somewhat tedious even for symmetric arrays. Computer programs, from those designed for pocket calculators to the more sophisticated SNAKE system,[15] are quite useful for these evaluations. Final results are generally specified in terms of center-to-center spacing for the units in the array. Appendix C provides two examples of use of the solid angle method in plant applications for a symmetric storage array and for a complex arrangement of equipment.

Modifications of the solid angle method as presented above may provide useful simplifications. For example, it is possible to consider only the first two "rings" of units or to exclude from consideration those that are shadowed. Such changes, however, do require thorough verification for all process conditions of interest by comparison to experiments or computer calculations.

Results of a verification study for a particular application of the solid angle method [16] are shown in Fig. 8-6. It may be observed that substantial conservatism exists between the experimental data and the solid angle algorithm [i.e., Eq. (8.6) plus its restrictions on maximum k_{eff} and total solid angle] for most of the situations that were considered.

Working Group ANS-8.13.1 attempted to write a consensus Standard on the solid angle technique.[17] Although ultimately unsuccessful, the effort did focus on key elements to be considered by anyone seeking to validate specific applications of the method. Such a Standard was intended to state the requirements for establishing, the criteria for applying, and the methods to demonstrate the



(7) CRITICAL ARRAYS, POISONED SOLUTION REFLECTOR
(8) K_{EFF} = .777, MAXIMUM ALLOWED BY SOLID ANGLE
(9) CRITICAL ARRAY, INTERSPERSED MODERATION
(10) CRITICAL ARRAY, CYLINDERS AND BOTTOM SLAB
(11) CRITICAL ARRAY MIXTURES OF CYLINDERS AND SLABS

Fig. 8-6. Results of a comparison between the solid angle technique and KENO-IV calculations.[16]

nuclear criticality safety of an array of individually subcritical units of fissile material. Although they sought initially to provide a generally applicable solid angle method, it was determined that such an approach would probably prove to be overly restrictive for most specific applications. The next thrust was to consider permitting each user to develop and implement a method tailored to specific needs. With this in mind, the working group attempted to develop guidance on allowances for such conditions as unit shape, array shape, vessel walls, interspersed moderation, reflector material, and reflector location. "Mixed arrays" (i.e., those with units of different fissionable material, shape, size, orientation, etc.) were also considered.

Exercises

8-1. State the basic principles upon which each of the following hand calculation methods is based:

a. surface density
b. density analog
c. solid angle.

8-2. Explain the need for validation and the principle limitations on use of the hand calculation methods.

8-3. Use the buckling conversion method and data from Figs. 8-1 and 8-2 to transform critical spheres in Tables 7-I and 7-II to equivalent critical infinite cylinders and slabs. Compare these results to appropriate entries in Tables 7-I and 7-II as a rough assessment of the accuracy of the buckling conversion method.

8-4. Damaged fuel from the Three Mile Island Unit 2 (TMI-2) reactor may have accumulated in ex-core locations in the form of a "mush" mixture of fuel pellet pieces and water (as considered further in Appendix G). For scoping purposes, it has been determined that a fully water-reflected infinite cylinder would have a 33-cm critical diameter for 2.96 wt% enriched pellets at their optimum (i.e., maximum reactivity) volume fraction in mixture with poison-free water. Estimate the equivalent critical:

a. sphere diameter
b. sphere volume
c. slab thickness.

From Fig. 7-3 estimate the critical mass for a sphere of material of the referenced composition. Use this result to estimate the "mush" density. Assume an extrapolation distance of 7.5 cm where appropriate.

8-5. Considering the dimensions used to model the TMI-2 submerged demineralizer system sand filters (Appendix G), estimate the equivalent diameter for an infinite cylinder. Comparing this to the limit in exercise 8-4, discuss the need for borosilicate glass as a criticality safety measure.

8-6. Criticality calculations for TMI-2 used the reflected extrapolation distance for water when configurations were known to be surrounded by water, and that of 20 cm when surrounded by reflectors of unknown composition. Estimate the decrease in the critical diameter of the infinite cylinder (exercise 8-4) when the extrapolation distance increases from ~7.5 cm for water to the value noted above. Also estimate the associated fractional decrease in spherical critical volume.

8-7. Repeat the sample surface density and density analog calculations in this chapter for units of:

a. 5 kg U metal with H:U = 60
b. 5 kg U metal with H:U = 1.0
c. 10 kg U metal with H:U = 10
d. 2.5 kg U metal with H:U = 1.0
e. 1 kg U in UO_2F_2 with H:U = 20.

8-8. Consider the reference 5-kg uranium units used in the examples in this chapter and:

a. determine the number of units and configuration for which the surface density (two-tier) and analog density models allow the same center-to-center spacing
b. repeat (a) for a four-tier surface density array
c. determine and plot surface density spacing for arrays from one to ten tiers.

8-9. Determine the maximum unit mass and recommended spacing for surface density (two-tier) calculations of:

a. uranium metal/water mixtures at H:U = 0, 1.0, 10, and 60
b. UO_2F_2 solutions at H:U = 20, 100, 200, and 1000.

8-10. Repeat the array surface density calculation in Appendix B for the composition employed in the array solid angle calculation in Appendix C. Compare the resulting allowed spacings of the two methods.

8-11. Repeat the array solid angle calculation in Appendix C for the composition employed in the array surface density calculation in Appendix B. Compare the resulting allowed spacings of the two methods.

8-12. Consider the reflected critical masses for metal spheres listed in Table 7-II. Use the surface density model described in this chapter to:

a. determine the allowed spacing for 2-kg ^{235}U metal units in a two-tier array
b. repeat (a) for 2-kg plutonium and ^{233}U metal units, respectively
c. determine the masses of plutonium and ^{233}U metal units, respectively, that could be accommodated by the spacing calculated for (a).

8-13. Repeat exercise 8-12 for 0.2-kg solution spheres and data from Table 7-I.

8-14. Repeat exercise 8-12 using the density analog model for a 1000-unit array.

8-15. Repeat exercise 8-14 for 0.2-kg solution spheres and data from Table 7-I.

8-16. Consider the unit geometry and composition for the array calculation in Appendix C and determine:

a. the cylinder size for an allowed spacing of 42 cm
b. the allowed center-to-center spacing for a 10% reduction in cylinder diameter.

8-17. Consider the geometry and unit composition for the array calculation example in Appendix C. Determine the allowed center-to-center spacing for the reference cylinders with:

a. one corner unit removed
b. extra units added to form a regular 5 × 5 array.

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