

PROCEEDINGS SERIES

CRITICALITY CONTROL
OF FISSILE MATERIALS

PROCEEDINGS OF THE SYMPOSIUM ON
CRITICALITY CONTROL OF FISSILE MATERIALS
HELD BY THE INTERNATIONAL ATOMIC ENERGY AGENCY
AT STOCKHOLM, 1 - 5 NOVEMBER 1965

CRITERIA AND EVALUATION FOR THE STORAGE OF FISSILE MATERIAL IN A LARGE AND VARIED REACTOR RESEARCH AND DEVELOPMENT PROGRAMME

D. R. SMITH
UNIVERSITY OF CALIFORNIA,
LOS ALAMOS SCIENTIFIC LABORATORY,
NEW MEX. , UNITED STATES OF AMERICA

Abstract

CRITERIA AND EVALUATION FOR THE STORAGE OF FISSILE MATERIAL IN A LARGE AND VARIED REACTOR RESEARCH AND DEVELOPMENT PROGRAMME. Precise analysis of the neutron interaction between fissile material containers is possible through the application of Monte Carlo techniques such as the GEM code of the U. K. A. E. A. or O5R developed by Oak Ridge National Laboratory. While this is appropriate for well-defined and inflexible arrays, many problems of practical materials storage do not require this rigor, nor are many materials storage configurations sufficiently well defined to permit full advantage to be derived from such treatment. An analysis which is amenable to slide rule calculation has been found sufficient for most of the problems that arise in a laboratory which has a large inventory of fissile material in the many forms required for a large, extensive, and varied reactor research and development programme (including fuels and materials development). This presentation is directed toward the nuclear safety specialist who must, with limited support facilities, derive criteria for the safe storage of fissile material without undue economic penalty.

The Los Alamos Scientific Laboratory, a facility of the United States Atomic Energy Commission, is situated in the mountainous northern portion of the state of New Mexico. This laboratory has been quite active in development work on nuclear reactors. Largely as a result of the somewhat remote nature of this facility, the laboratory has chosen to restrict its reactor research activities to those of a specialized nature appropriate to the unique capabilities of the organization. This intentional involvement with advanced reactor concepts, as manifested by the Los Alamos Molten Plutonium Reactor (LAMPRE) and the Ultra-High Temperature Reactor Experiment (UHTREX), makes it inevitable that operations will be somewhat flexible and changeable.

Another major Laboratory effect is directed toward the development of a functional propulsion reactor for space missions. The nine reactor cores which the laboratory has operated for this purpose have consisted of graphite loaded with highly enriched uranium. A single core may contain more than 200 kg of uranium. When one recognizes the need for storing several such cores; and in addition spare elements, fuel scrap, and feed material, it is perhaps not surprising that the total Laboratory inventory is approximately ten thousand kilograms of highly enriched uranium and plutonium.

The bulk of this material is to be found in half a dozen storage vaults. Thus when we consider the problems of safe storage of fissile material, we are involved with large quantities of material in flexible and diversified storage.

It is considered significant that we have not found it necessary to utilize sophisticated reactor physics analyses to provide confidence in the reasonableness of desired storage configurations. Questions of nuclear safety are not commonly so marginal as to require such refinement. We find ourselves utilizing a "density analogue" approach as the first step in considering any storage proposal.

The data which are available for interacting arrays demonstrate that the mass in a bare critical array is essentially a power function of the material density in the array and that the value of the exponent is dependent on the size of the units comprising the array. The array is then analogous to a low-density homogeneous system where, instead of a density exponent of 2, one may conservatively use $2(1-f)$, where f is the "fraction critical" for an individual unit. This "fraction critical" is defined as the ratio of the effective mass of the unit to the critical mass of a unit of similar shape and composition. The effective mass is the actual mass increased as is appropriate for any reflector associated with the unit. While approaches of this kind have been used for many years, the practice in the past has been to consider a smaller value of the density exponent for reflected arrays.

This resulted in an over conservative evaluation for large arrays while losing applicability to some arrays of only a few units. With the availability of the Oak Ridge data on bare and reflected critical arrays, it was observed that, for the larger systems, the density exponent of the reflected array approached that of the bare array so that, in the limiting case, there is a constant mass ratio between the two systems. This is demonstrated in Figs. 1 and 2. One-dimensional transport calculations, as shown in Fig. 3, have demonstrated the same effect in homogeneous low-density cores reflected by water. As the core density becomes small, the core density exponent approaches two, and the ratio of bare to reflected critical mass becomes constant. The value of this ratio is found to be in reasonable agreement with the limiting values inferred from the experimental data for arrays of the same material. We then have a means for making a conservative approximation of the critical size for an array of fissile units of interest.

One first evaluates the "fraction critical" of an individual unit by determining the ratio of the effective mass of the unit to the mass of a critical unit of the same shape and composition. As an example, the fraction critical for a bare sphere of fissile material is the ratio of the mass of the sphere to the bare spherical critical mass. For a cylindrical unit, one uses the ratio of the unit mass to the mass of a critical cylinder of the same material and the same height-to-diameter ratio. If the individual unit is enclosed in a container, one first increases the unit mass as indicated by the degree of reflection associated with the container. This consideration is more frequently significant in evaluations for transportation than for storage.

Having obtained the fraction critical and the associated density exponent $s = 2(1-f)$, we find the mass in the conservative array to be $m = m_0 (\rho_0 / \rho_e)^s$ where m_0 is the bare spherical critical mass at density ρ_0 , and ρ_e is the effective mass of a unit divided by the unit cell volume. We find it convenient to use an array capacity C which is the number of "spherical crits" in our conservative bare array where $C = (m/m_0) = (\rho_0 / \rho_e)^s$. The array capacity at constant ρ_e may then be reduced by a factor which takes into

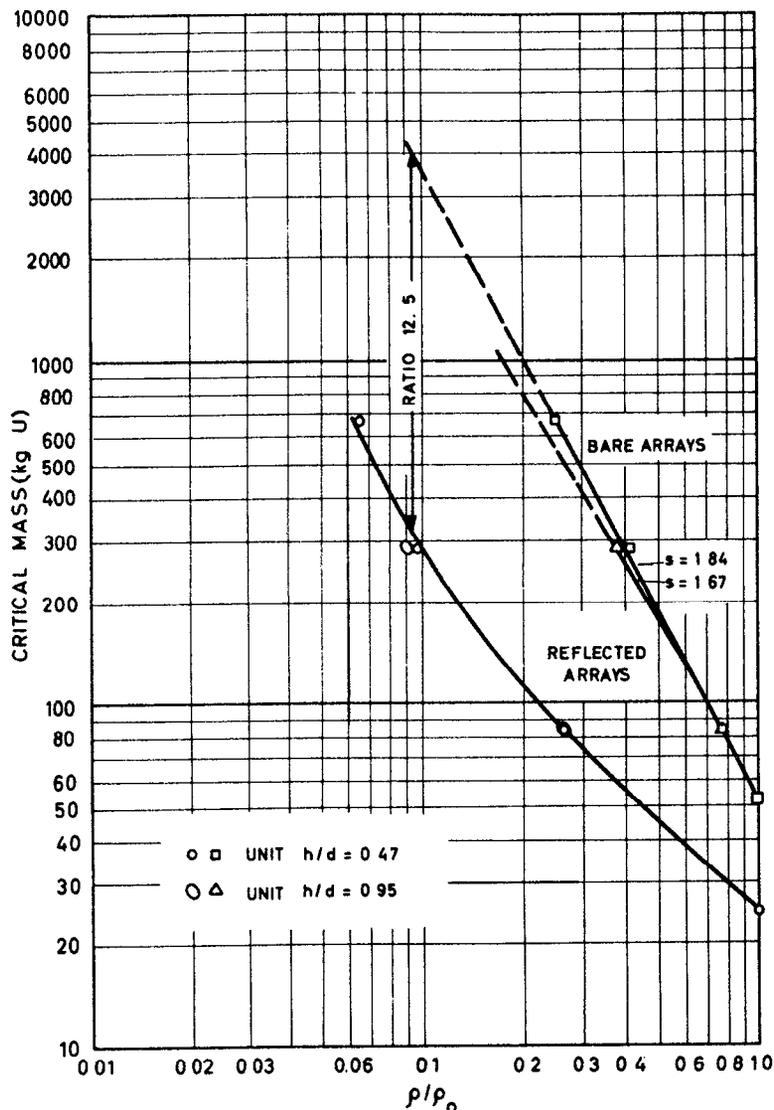


FIG. 1. Critical masses, air-spaced arrays of 10.5 kg cylinders of U(93) metal. There are the same number of units along each of the three principal axes, surface-to-surface spacing is uniform (ρ/ρ_0 is the fraction of the array volume that is occupied by material).

account the effect of array reflection. Factors appropriate for thick water reflection of low density arrays are given in Table I. These values represent the ratio (r) of bare to reflected array capacity at constant density. The ratio is evaluated at a low density.

The application of the density analogue technique to available array data has indicated that the density exponent which we derive is less than the exponent which is measured, so that we calculate arrays smaller than those which are critical. This conservatism is indicated in Fig. 4.

Figure 5 illustrates the applicability of the density analogue to arrays of long cylinders filled with solution. This is perhaps an extreme test, and we find that the results are acceptably conservative.

Though we recognize that this procedure is only an approximation, and in some cases a very conservative one, useful and illustrating implications may be drawn if we treat the technique as being more precise.

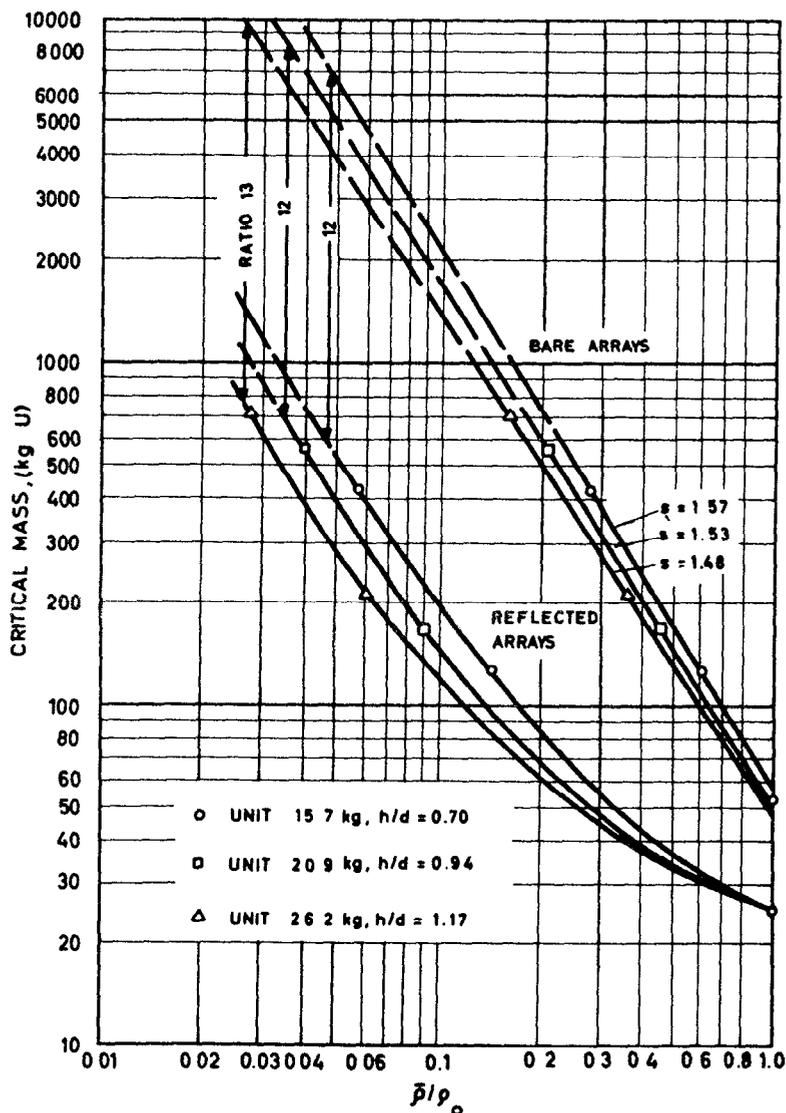


FIG. 2. Critical masses, air-spaced arrays of 15.7 kg, 20.9 kg, or 26.2 kg cylinders of U(93) metal. There are the same number of units along each of the three principal axes, surface-to-surface spacing is uniform.

It is interesting to consider the significance of the various parameters which influence the quantity of material which can be stored in a particular vault.

Consider an array of volume V in a vault characterized by a reflection ratio r containing units of effective mass m_e . The material of the units has a bare spherical critical mass m_0 and a bare spherical critical volume V_0 .

Find N , the number of units in the critical vault, assuming

$$m = \frac{m_0}{r} \left(\frac{\rho_0}{\rho_e} \right)^s \quad \text{where } \rho_e = \frac{N m_e}{V}$$

By definition

$$N = m/m_e = \frac{1}{r} \frac{m_0}{m_e} \left(\frac{m_0}{V_0} \frac{V}{N m_e} \right)^s$$

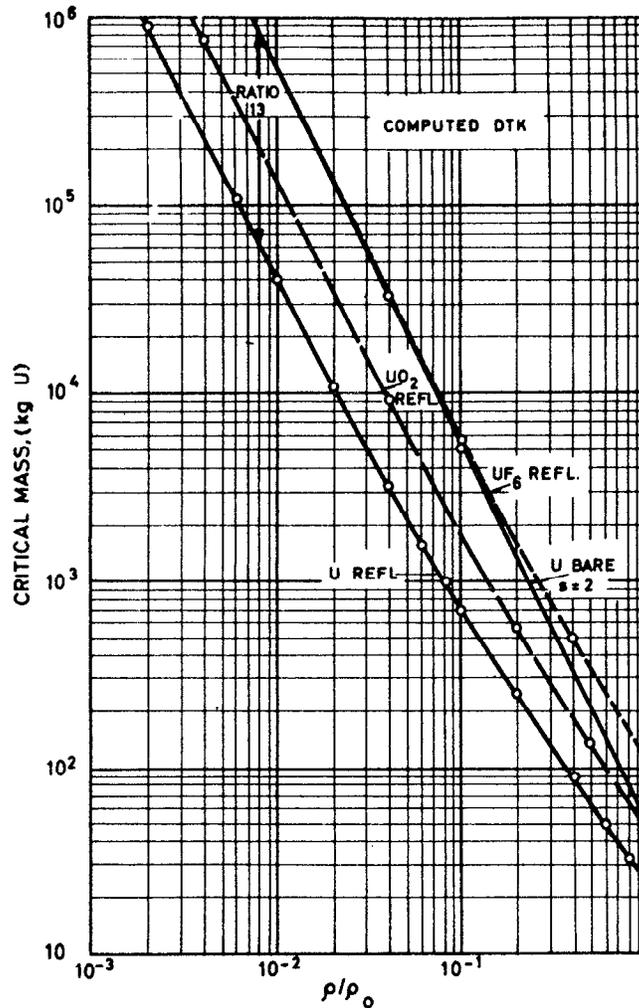


FIG. 3. Computed critical masses of spheres with core densities reduced by the fraction ρ/ρ_0 ; water-reflected U(93) metal, U(93)O₂, or U(93)F₆, and unreflected U(93) metal.

so

$$N^{s+1} = \frac{1}{r} \left(\frac{m_0}{m_e} \right)^{s+1} \left(\frac{V}{V_0} \right)^s$$

and

$$N = \frac{1}{r^{1/(s+1)}} \frac{m_0}{m_e} \left(\frac{V}{V_0} \right)^{s/(s+1)}$$

From this it can be seen that even for metal units which are "half critical", an extreme case, the effect of wall reflection equivalent to thick water (4 to 6 in. of concrete) reduces the unreflected vault capacity by less than a factor of five. In the extreme case of plutonium metal, $r = 20$ from the table above, and $s = 1$, so $r^{1/(s+1)} = 4.5$. Calculations indicate that large thicknesses of concrete (~ 12 in.) may have a reflection ratio twice that for thick water, and so reduce vault capacity by about a factor of six below the unreflected value, for very large storage units. For storage units of more practical

TABLE I

FACTORS FOR THICK WATER REFLECTION
OF LOW DENSITY ARRAYS

Fuel ^a	Experiment	Transport calculation
Plutonium metal (α phase)		20
U(93) metal	12.5	13
U(93)O ₂		8
U(93)-water H/X = 60	~5.4	5.4
U(93)F ₆		6
U(93)-water H/X = 400		2.75
U(93)-C ₈₀		2.4
U(4.9)O ₂ F ₂ solution, H/X = 500		2.0

^a U(93) indicates 93 wt. % enrichment of ²³⁵U.

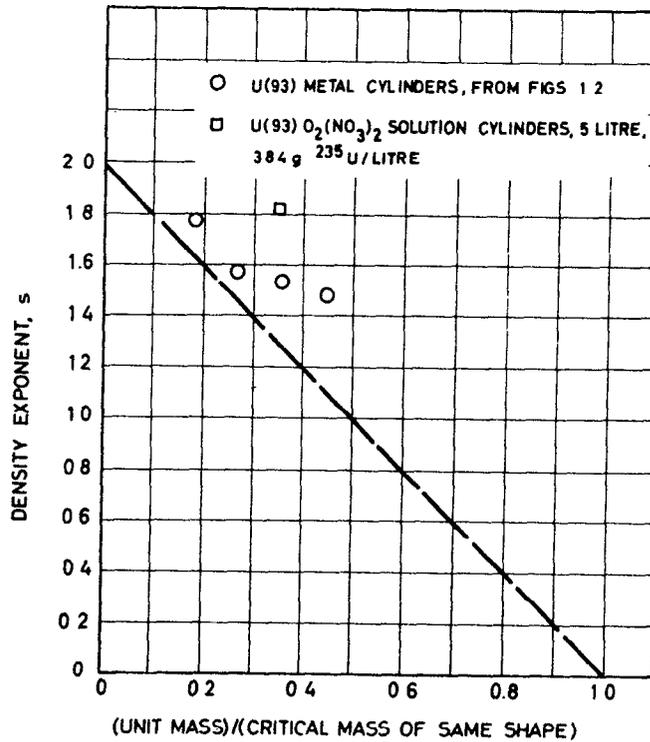


FIG. 4. Density exponents of bare air-spaced equilateral arrays versus "fraction critical" of fissile units.

size, a factor of two to four seems typical for wall effects on the vault capacity.

Two laboratory storage facilities are chosen as illustrative of the application of the density analogue approach.

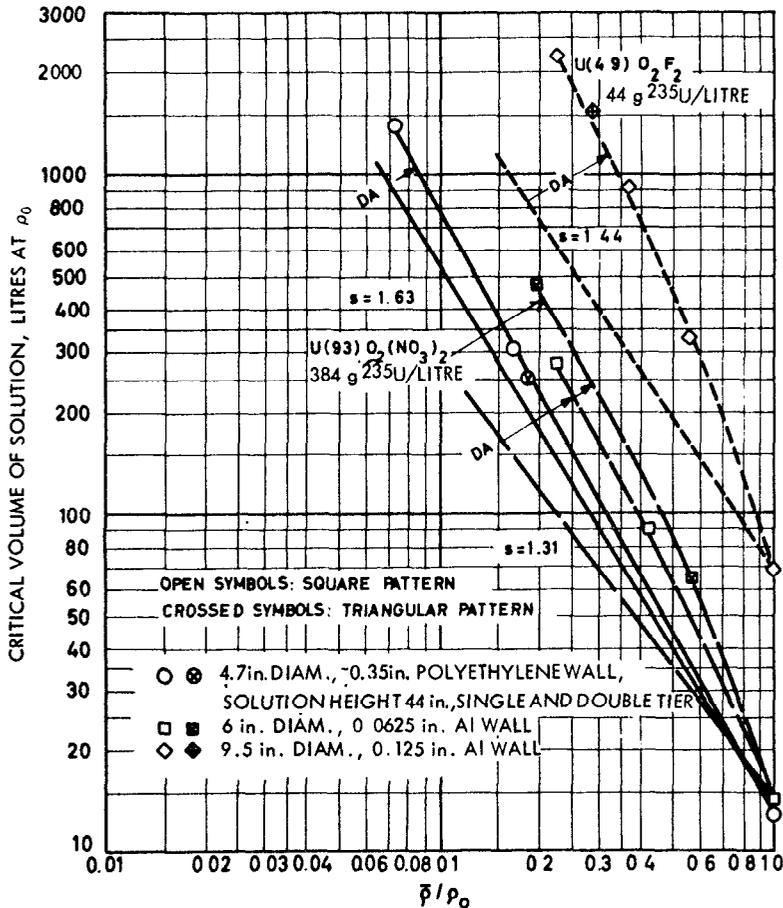


FIG. 5. Relative critical masses, unreflected air-spaced arrays of long ²³⁵U-solution cylinders. Solution heights are adjusted so that each array is equilateral. Density-analogue reconstructions are included.

First is a large general storage building which is used chiefly for graphite loaded with highly enriched uranium. The storage volume is approximately forty feet by one hundred feet and six feet high. This is 6.8×10^5 litres. In LAMS-2955, Stratton has tabulated critical dimensions of uranium (93.5)-graphite-water system. The UHTREX fuel elements are loaded to a C/²³⁵U ratio of about 300, so the values for C/²³⁵U = 316 will be considered. Here the bare critical volume is 714 litres and the corresponding critical mass is 89.3 kg of enriched uranium. The capacity of a bare cubic array of 6.8×10^5 litres is then

$$C_{\text{bare}} = \left(\frac{6.8 \times 10^5}{714} \right)^{s/(s+1)}$$

The fraction critical for a storage unit is very small (< 0.01) so that a density exponent of 1.9 is conservative. Then

$$C_{\text{bare}} = 952^{0.655} = 89.5 \text{ spherical crits or } 7992 \text{ kg, and in the reflected array,}$$

$$C_{\text{ref}} = C_{\text{bare}} / r^{1/(s+1)} = 7992 / 2.4^{1/2.9} = 5920 \text{ kg.}$$

The maximum quantity which has been stored in this area is approximately 1500 kg.

One may estimate that the array shape will increase the capacity by a factor of about three, so the margin of safety in our storage practice probably exceeds a factor of ten. This margin is perhaps slightly generous, but we try to maintain something like an indicated factor of five for normal storage.

A second example is the storage of similar material in a higher density configuration at a critical mass laboratory. Here access is restricted to those who are engaged in critical assembly measurements.

A storage array five feet by eight feet by fifteen feet is used. The capacity of a 600 cubic feet bare cubic array would be

$$C = \left(\frac{1.7 \times 10^4}{714} \right)^{0.655} = 23.8^{0.655} = 8 \text{ spherical crits}$$

$$= 714 \text{ kg bare or } 590 \text{ kg reflected.}$$

Storage criteria limit the amount of material in this array to about $\frac{1}{3}$ of this value. Some additional benefit is obtained from the shape of the array, but this remains the least conservative storage with which I am familiar.

No claim can be made that this array would be safe if flooded. Criticality would be expected at an H/²³⁵U ratio of about 50, or 20 g of water per litre. However, to achieve this would require the retention of about 100 gallons or 370 litres of water in the array. The probability of flooding this laboratory is small, but not zero. As the location is in an open canyon at an elevation of about 7000 ft, some catastrophic occurrence would be required to cause flooding, and it follows that the liberation of a few kilowatt-hours of energy in a criticality accident would be of minor concern.

Where convenient, we plan storage arrangements such that water flooding could be tolerated. This is thought of as a handy way to build in a desirable margin of safety. Of course, this does not work for storage of units which are at or near optimum moderation, nor does it seem fruitful to add cadmium, say, to make safe an array against flooding when there is no significant chance that flooding will occur. The use of boron as a fixed poison can provide k_{∞} less than unity for any degree of moderation, and for some activities we consider this of sufficient value to justify the expense. In particular, such a criterion is used for the storage of fuel elements which have been subjected to power runs in the experimental propulsion reactors. This material is stored outdoors in large quantities with modest environmental protection so that the provision of safety under all conditions of moderation is justified.

A problem which has been the subject of much discussion involves the degree of isolation provided by concrete walls between storage facilities. Data in TID-7028 (p.109) are relevant and indicate that while 8 in. of concrete is not as effective an "isolator" as is 8 in. of water, it does rather well and for many purposes provides effective isolation. Measurements at Los Alamos with 21 in. diameter plates of U(93) metal have also indicated that 8 in. of concrete greatly reduces the interaction between two stacks of the plates. The interaction through 8 in. of concrete was about $\frac{1}{4}$ of that through 8 in. of air.

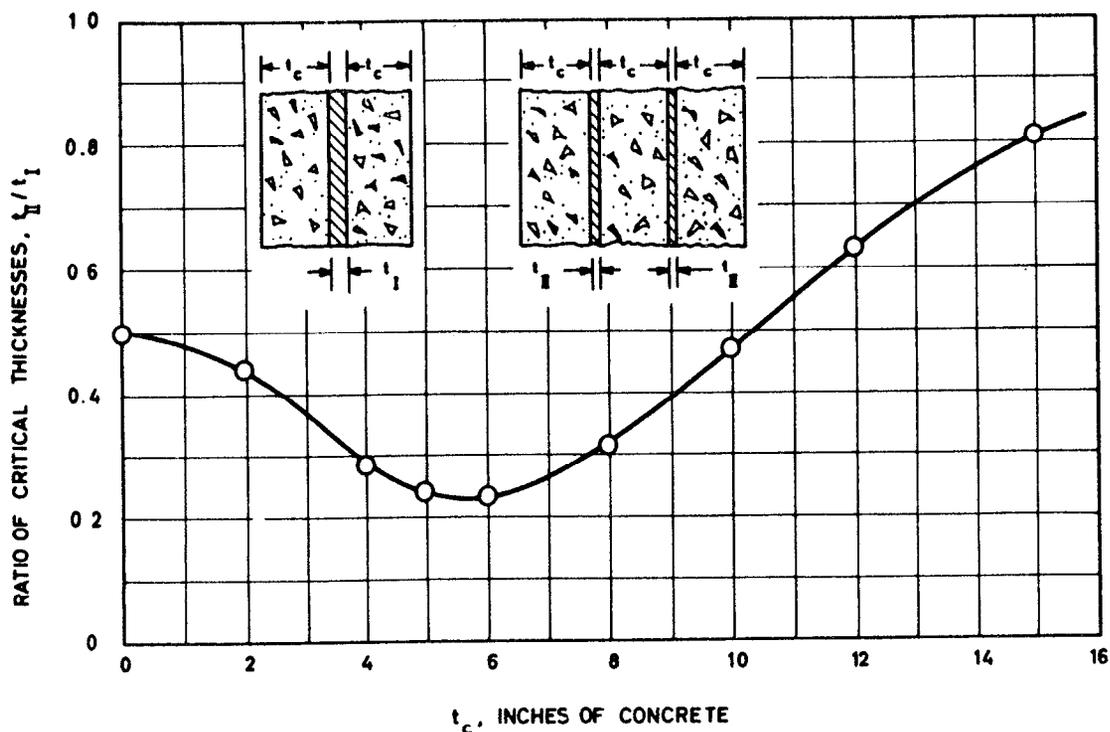


FIG. 6. Concrete-reflecting critical infinite slabs U(93.5) metal, DTK calculations

We have attempted to apply the Los Alamos transport code to calculations of interaction through concrete. The results of infinite slab calculation are shown in Fig. 6. The indications here are that for thicknesses of concrete of less than ten inches, the presence of concrete increases the interaction between two infinite slabs. This appears to be contradicted by the available data. Perhaps the laboratory measurements, because of the modest size of the experiments, are greatly influenced by scattering effects which remove neutrons from the experiment and are not significant for the calculated infinite slabs. The calculations do, however, indicate a need for caution in evaluating the safety of adjacent storage rooms of great size. Even here the factor of ten which we find common in establishing storage limits is sufficient to provide for interaction effects. For storage areas of modest size, as usually encountered, the assumption of isolation from 8 in. concrete walls is valid for all but very high capacity storage.

The indicated discrepancy in the comparison of calculations with the results of measurements is considered interesting, but of little practical safety significance. When material becomes sufficiently abundant to make the construction of adjacent large storage facilities desirable, it will be convenient to monitor neutron levels during initial occupancy and observe any significant interaction effects.

Though many uncertainties exist with regard to interaction effects, and the gradual reduction of these uncertainties is desirable for more economical storage and transportation of fissile material, we are able today to achieve a reasonable level of economy on the basis of simple and convenient analysis that is applicable to the great majority of problems which we encounter.

DISCUSSION

K. J. ASPINALL: With regard to Figs. 1 and 2 in your paper, which show the critical masses for bare arrays and reflected arrays, and yield a figure of 12:1 or 13:1 for the ratio between them, I should like to know the actual reflection conditions. Was it six-sided reflection by thick water? And if you had concrete reflection would you expect the ratio to be increased by a factor of 5?

D. R. SMITH: The data were obtained by Thomas of Oak Ridge, and represent reflection equivalent to thick water. I would consider an additional factor of 2 more than generous for reflection by very thick concrete.

O. SCHAFFER: As the criticality of the stored array depends mainly on the "fraction critical" in the exponent of the formulas given in the paper I would like to ask what maximum figure of this fraction, f , seems to be acceptable in the special cases mentioned, what will then be the loading procedure and what is the maximum reactivity addition applied to the system by any loading step? Obviously a value of less than 0.01 for the critical fraction f , as mentioned in the case of the Los Alamos storage units, seems to be very conservative.

D. R. SMITH: This value is appropriate for some specific units, and was not intended to represent all situations. In practice, unit sizes may vary up to 40% or 50% of the critical size. I would tend to question the need for placing a limitation on the size of units in storage, though obviously, as the size of units increases, the need for restrictions on the manner of storage increases commensurately.

B. G. OWEN: Are there any restrictions on storing adjacent to the walls?

D. R. SMITH: I am unable to recall any cases where it was necessary to impose restrictions of this sort.

A. THOMAS: In the Oak Ridge experiments reported by J. T. Thomas* the space between the outer units and the reflector was at least half the lattice spacing. Would you not lose some of your conservatism if units were stored right up against the walls?

D. R. SMITH: That is quite true, but we feel that our safety margin is large enough to cover this eventuality.

J. LECLERC: Have you studied the neutronic properties of concrete as a function of its composition? If so, did you find, for example in the experiments referred to in the last section of your paper, that the hydrogen content of the concrete exerts a great influence?

D. R. SMITH: For the experiments which were performed at Los Alamos we have only an approximate indication of the composition of the concrete, and do not know how much water was present.

The calculations were made using a sixteen-group set of cross-sections based on the most simple chemical analysis of concrete which I found.

Additional hydrogen in the composition would increase the capture cross-section, and would be expected to reduce the interaction.

E. D. CLAYTON: In regard to the subject of neutron isolation I would like to mention that a paper will be presented at the November 1965 meeting of the American Nuclear Society detailing the results of recent experiments

* These Proceedings, paper SM-70/29.

in the Hanford Plutonium Critical Mass Laboratory. In the experiments to be reported, effective isolation thicknesses were determined for several materials such as polyethylene, borated polyethylene, compressed wood, concrete, borated concrete and lead. The effect of cadmium sheet on surfaces of the polyethylene was also measured.