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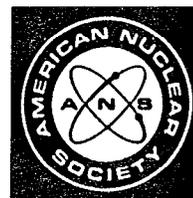
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## Surface Density and Density Analog Models for Criticality in Arrays of Fissile Materials

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*A surface density model based on experimental and calculated criticality data is developed for finite water-reflected arrays and results in semiempirical analytic expressions describing criticality. The relations provide information on the reactivity associated with such perturbations to arrays as changes in unit shapes, cell volumes, array shapes, and array reflectors. Equivalence between different fissile materials in a critical array is defined. The surface density and density analog models are shown to be in correspondence when applied to the same data. The density analog model is expressible as  $f(N) = g(m)\rho^{-2}$ . The functions  $f(N)$  and  $g(m)$  are explicitly given, and the constant exponent has general applicability.*

### INTRODUCTION

The body of information on the criticality of individually subcritical components of fissile materials arranged in reflected critical arrays has grown sufficiently in the past 10 yr to warrant examination of density techniques and of an understanding of concepts employed in nuclear criticality safety. The available experimental data<sup>1-3</sup> are limited to small numbers of units in necessarily high fissile material density systems relative to practical situations encountered in operations. These data have proven to be calculable by Monte Carlo codes.<sup>4</sup> As a verified method of calculation, it is available to extend the data to

regions of interest and to investigate factors affecting the criticality of arrays. The criticality of single units is also calculable by the Monte Carlo code. A verified  $S_n$  transport<sup>5</sup> code, however, is often employed, and satisfactory comparison of the results from the two codes has been observed.

While detailed examination of individual cases is possible by computation, there remains the desire to express criticality information both systematically and comprehensively by means of models. A model should embody the following criteria:

1. expressible in terms of measurable quantities, e.g., mass and dimension, yet be consistent with what is known of neutron behavior
2. contain as few auxiliary rules or supplemental restrictions as possible consistent with its intended area of applicability
3. exhibit relatively good agreement with data and verifiable calculation and be consistent with limiting values of parameters

<sup>1</sup>H. C. PAXTON, J. T. THOMAS, A. D. CALLIHAN, and E. B. JOHNSON, "Critical Dimensions of Systems Containing U-235, Pu-239, and U-233," TID-7028, U.S. Atomic Energy Commission (1964)

<sup>2</sup>H. F. FINN, N. L. PRUVOST, O. C. KOLAR, and G. A. PIERCE, "Summary of Experimentally Determined Plutonium Array Critical Configurations," UCRL-51041, Lawrence Livermore Laboratory (1971).

<sup>3</sup>J. T. THOMAS, *Nucl. Sci. Eng.*, **52**, 350 (1973).

<sup>4</sup>L. M. PETRIE and N. F. CROSS, "KENO IV—An Improved Monte Carlo Criticality Program," ORNL-4938, Oak Ridge National Laboratory (1975)

<sup>5</sup>W. W. ENGLE, Jr., "A User's Manual for ANISN," K-1693, Oak Ridge Gaseous Diffusion Plant (1967).

4. display the ability to relate different systems and perturbations to a system.

Development of a model begins with simple, describable systems of maximum symmetry and progresses to and through added complexities. The simplest three-dimensional array geometry is the cubic array composed of cubic cells. The unit, considered centered in a cell, is fissile material with a spherical shape. These regular arrays are ideal data to investigate the density analog and surface density models and will make evident, through more complex developments, the complementary information derivable from both methods when applied to the same data.

In storage or handling operations, consideration is usually given to possible neutron reflectors. Concern in the following, therefore, is with the characterization of criticality for closely reflected arrays, since this will produce more generally applicable results for practical nuclear criticality safety. The reflector materials completely surround the arrays, being located at the boundaries of the peripheral cells.

Let us review the common expression of surface density. The projected surface density,  $\sigma$ , is the mass,  $m$ , of a unit multiplied by the number,  $n$ , of units in a column divided by an area defined by the dimension,  $d$ , of a cube:

$$\sigma = \frac{nm}{d^2} \quad (1)$$

For noncubic arrays, the value of  $\sigma$  depends on the plane of projection and so is not unique. In this case, we consider only the minimum value of  $\sigma$  corresponding to the smallest column. Multiple values of  $\sigma$  are also associated with arrays comprised of noncubic cells. To alleviate this difficulty and at the same time to establish a unique relation between the average volume density of fissile material and the surface density, we assign the cell dimension of a cube equal in volume to the noncubic cell to determine the dimension used in Eq. (1).

These constraints are not serious. They support our requirement for consistency and for the removal of ambiguity. Having established a common beginning, let us examine the density analog models.

#### DENSITY ANALOG MODELS

The density analog representation of array criticality depicts the number of units, or total mass, in a critical reflected array as a function of the average fissile material density in the array, given by the relations

$$N = g(R, f)\rho^{-s} \quad (2)$$

$$\rho = \frac{m}{d^3}, \quad \text{and} \quad s = 2(1 - f) \quad (3)$$

The number of units,  $N$ , varies inversely with the average density,  $\rho$ , to a power,  $s$ , multiplied by a coefficient,  $g(R, f)$ , expressed as a function of the reflector effect,  $R$ , and a quantity,  $f$ , related to the reactivity of the unit. In the model,  $f$  is the ratio of the mass of the unit to the mass of a critical single unit of the same shape and material. The density exponent,  $s$ , is approximated by a relation suggested by Paxton.<sup>6</sup> The expression is applicable to unreflected arrays where the coefficient 2 is the limiting value of  $s$  as  $f$  approaches zero, corresponding to the behavior of the critical mass of an unreflected unit as its density varies. The influence of a reflector on a bare array is approximated by a reflection factor,  $R$ , reported by Smith.<sup>7</sup> Applicable values of  $R$  were determined from  $S_n$  calculations of reflector effects on single homogeneous low-density units.

Comparison of the model with some experimental data<sup>3</sup> for uranium metal cylinders having a <sup>235</sup>U enrichment<sup>8</sup> of 93 wt% is made in Fig. 1. The cylinders are of different height-to-diameter ratios, spaced in paraffin-reflected arrays at equal surface separation and equal numbers of units along the three edges of arrays. Typical of such representation is (a) the larger total mass required for criticality as the unit mass diminishes, (b) the mild effect of unit shape represented by the upper two curves for 10.5-kg units, and (c) the nonlinearity in the high-density region. The two dashed lines are conservative envelopes of these data. The line with an exponent of 1.2 is the Smith expression,<sup>7</sup> while that with an exponent of 1.8 is the Paxton later extension<sup>9</sup> guided by Monte Carlo calculations of arrays at lower densities.

Proper choice of coefficients of Eq. (2) permits reasonable estimates of criticality for a specific unit over a limited density range. Outside the defined parameter range, the model will provide conservative estimates, i.e., describe systems known to be subcritical. Because of the linear

<sup>6</sup>H. C. PAXTON, "Correlations of Experimental and Theoretical Critical Data," *Proc. Symp. Criticality Control in Chemical and Metallurgical Plant*, Karlsruhe, pp. 173-205, Organization for Economic Cooperation and Development, European Nuclear Energy Agency (1961).

<sup>7</sup>D. R. SMITH, "Criteria and Evaluation for the Storage of Fissile Material in a Large and Varied Programme," *Proc. Symp. Criticality Control of Fissile Materials*, Stockholm, 1965, p. 667, International Atomic Energy Agency, Vienna (1966).

<sup>8</sup>The composition of <sup>235</sup>U-enriched uranium is denoted, for example, by U(93).

<sup>9</sup>H. C. PAXTON, "Density-Analog Techniques," in *Proc. Livermore Array Symposium*, CONF-680909, p. 6, U.S. Atomic Energy Commission (1968).

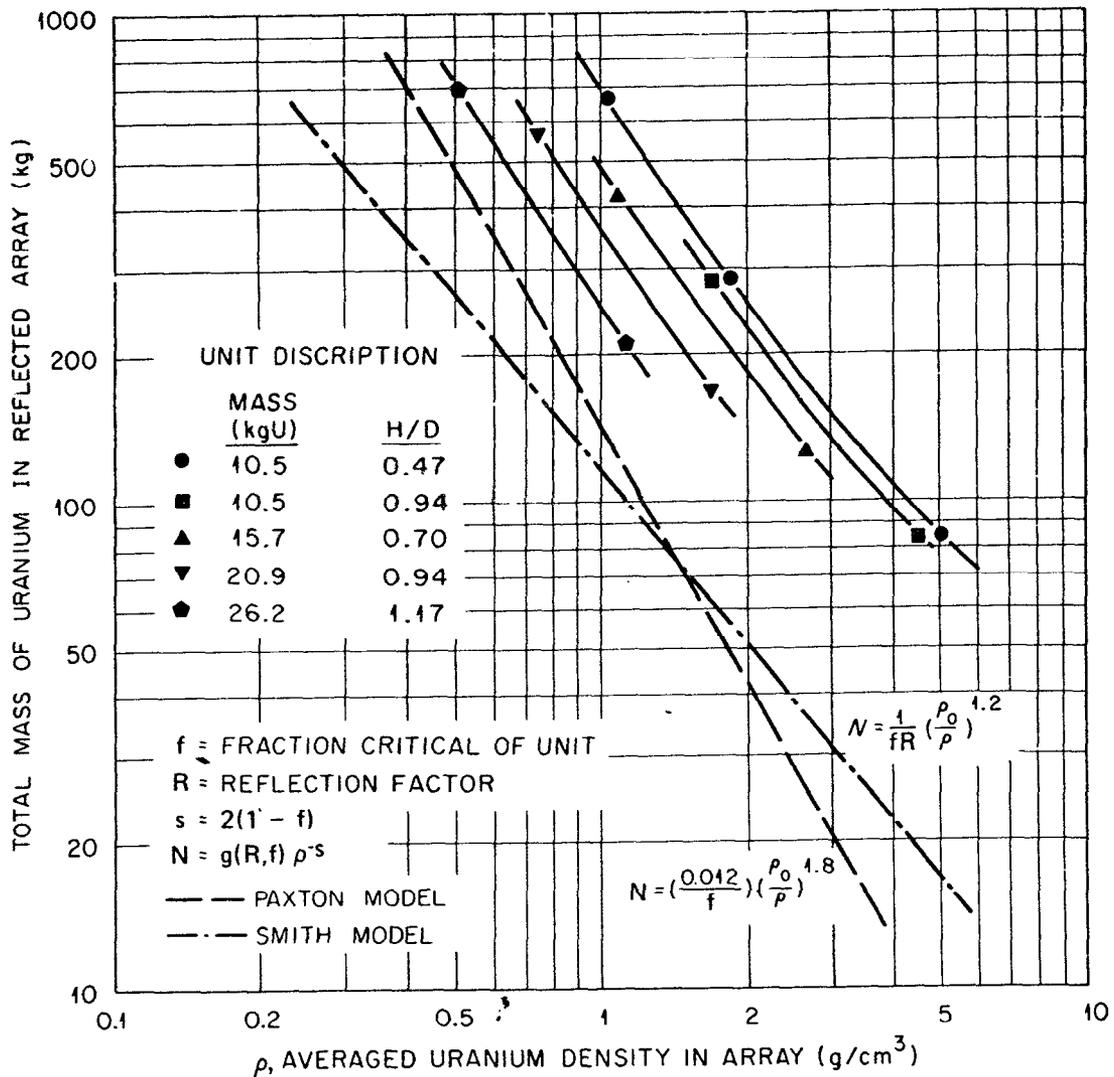


Fig. 1 Density analog models and measured reflected critical arrays of  $^{235}\text{U}$ -enriched uranium metal cylinders.

approximation in a log-log plot, the model cannot respond properly to the limiting values of the parameters, in particular, the maximum average metal density and the very low densities. A particular question is what critical mass is represented by each experimental curve as the average density achieves the fissile material density, which may involve integral and fractional parts of units? The array reflector is always located at the boundaries of the peripheral cells; thus, when fissile metal density and average density are equal, the reflector and metal are in contact. If we suppose this view to be correct, one would expect the critical mass at full density to be strongly dependent on the unit shape simply from our experiences with reflected single-unit criticality. Furthermore, is it correct to impute to single-unit criticality the neutronics of array

criticality? The points, however, are academic, since the information does not enhance the utility of the model, which clearly approximates existing data over a limited density range.

If we depart from cubic array geometry, we abandon the guidance afforded by Eq. (3) for the slope, for example, representations with a slope  $>2$  are possible. Reliable estimates of criticality of arrays of different shape, even maintaining constant the mass of the units, are difficult in the density analog model.

#### SURFACE DENSITY MODELS

The importance of criticality information on infinite-slab thicknesses of fissile materials for various reflector conditions was recognized early in nuclear criticality safety practices. If the

physical height of fissile materials being handled or stored in planar arrangements does not exceed the slab thickness required for criticality of the material, the result is subcritical. Such a dimension limitation, although occasionally useful, is not often practical. Experiments with finite arrays of cylindrical units of aqueous fissile materials were correlated by Paxton<sup>6</sup> in 1961 by smearing the fissile material in an array over the base area of the array and representing that "height" as a function of the ratio of the "effective fissile material height" to the "diameter of the array." The representation dramatically demonstrated that criticality was possible with effective surface densities, Eq. (1), less than those of uniform critical slabs, and indicated that the reactivity of an individual unit when located in an array was a significant parameter.

Stevenson and Odegaard<sup>10</sup> used Monte Carlo calculations to examine several parameters influencing the neutron multiplication factor of infinite, as well as some large, water-reflected planar arrays of uranium as aqueous solutions, as oxides and as metal. Parameters considered were uranium enrichment and concentration, reflector location, and the reactivity of units. Variation in the reactivity of the unit was effected by changes in the height-to-diameter ratio of cylindrical units expressed as the fraction critical,  $f$ .

Thirty-three calculated critical arrays had neutron multiplication factors ranging from 0.978 to 1.033, with an average of 1.005. The surface density of some of the computed arrays, normalized to the surface density of calculated reflected infinite slabs of the same materials, is presented in Fig. 2 as a function of the fraction critical,  $f$ , of a unit in the array. Also shown on the figure are calculated data for water-reflected infinite planar arrays of cubes of metal with a water reflector in contact with the units. The normalization to single-unit criticality is intended to define a region within which criticality is not possible.

The location of a curve in Fig. 2 is sensitive to the accuracy with which the critical dimensions of the normalizing unit is known. The choice of the parameter for the abscissa can lead to the inconsistent treatment of data, as illustrated by the array data of metal cubes. The parameter  $f$  in

the present model requires arrays with a receding reflector as  $f$  increases to achieve a value of  $f = 1$ . This behavior is not possible whenever reflector or other materials are continuously associated with the units in the arrays. Also excluded is information on conditions affecting criticality, such as interstitial moderation from sprinklers or moderation present inherently in some insulating materials of packages. If, in application, the surface density and unit size are to be limited so as to define subcritical planar arrays based on a demonstrated knowledge of two critical dimensions of single units for the fissile materials, then the area of applicability of the model is limited to air-spaced units in the arrays. This would provide a valid point of departure for the exploration of other factors of interest affecting planar array criticality.

The density analog and surface density models described above serve a purpose in nuclear criticality safety in that they can be made to be conservative in their application to plant problems, i.e., they define systems for which  $k_{eff}$  is known to be less than unity. Their reliability, of course, is established by comparison with calculated and experimental criticality data.

In the work reported here, there has resulted, from several thousand calculated reflected arrays with different forms of fissile materials, a correlation that implicitly contains the above two models. The analytic representation of the calculated data is presented in the following section.

#### ANALYTIC REPRESENTATION OF ARRAY CRITICALITY

The calculated critical array data<sup>11</sup> considered in this work embraced a wide variety of fissile materials: metals and oxides of <sup>233</sup>U, <sup>235</sup>U in uranium enriched from 30 to 100 wt%, and <sup>239</sup>Pu containing from 0 to 20 wt% <sup>240</sup>Pu. Dry and damp oxides having H/U or H/Pu atomic ratios ranging from 0 to 20, with the ratio expressed in terms of total uranium or total plutonium, were also examined. Critical dimensions of arrays of spherical units arranged in reflected cubic arrays were calculated.

The actual surface density,  $\sigma$ , and a limiting surface density,  $\sigma(m)$ , of critical cubic arrays having units of mass  $m$  are related by the following semiempirical equation:

$$\sigma = \frac{nm}{(2a_n)^2} = \sigma(m)/(1 - c/\sqrt{N})^2 \quad , \quad \text{for } N \geq 64$$

or

$$\sigma(m) = \frac{nm}{(2a_n)^2} (1 - c/\sqrt{N})^2 \quad , \quad (4)$$

<sup>10</sup>R. L. STEVENSON and R. H. ODEGAARDEN, *Trans Am Nucl. Soc.*, **12**, 890 (1969).

<sup>11</sup>J. T. THOMAS, "Uranium Metal Criticality, Monte Carlo Calculations, and Nuclear Criticality Safety," Y-CDC-7, Oak Ridge Y-12 Plant (1970); see also, J. T. THOMAS, "The Criticality of Cubic Arrays of Fissile Materials," Y-CDC-10, Oak Ridge Y-12 Plant (1971); see also, J. T. THOMAS, "Generic Array Criticality," in *Nuclear Criticality Safety*, TID-26286, p. 66, U.S. Atomic Energy Commission (1974).

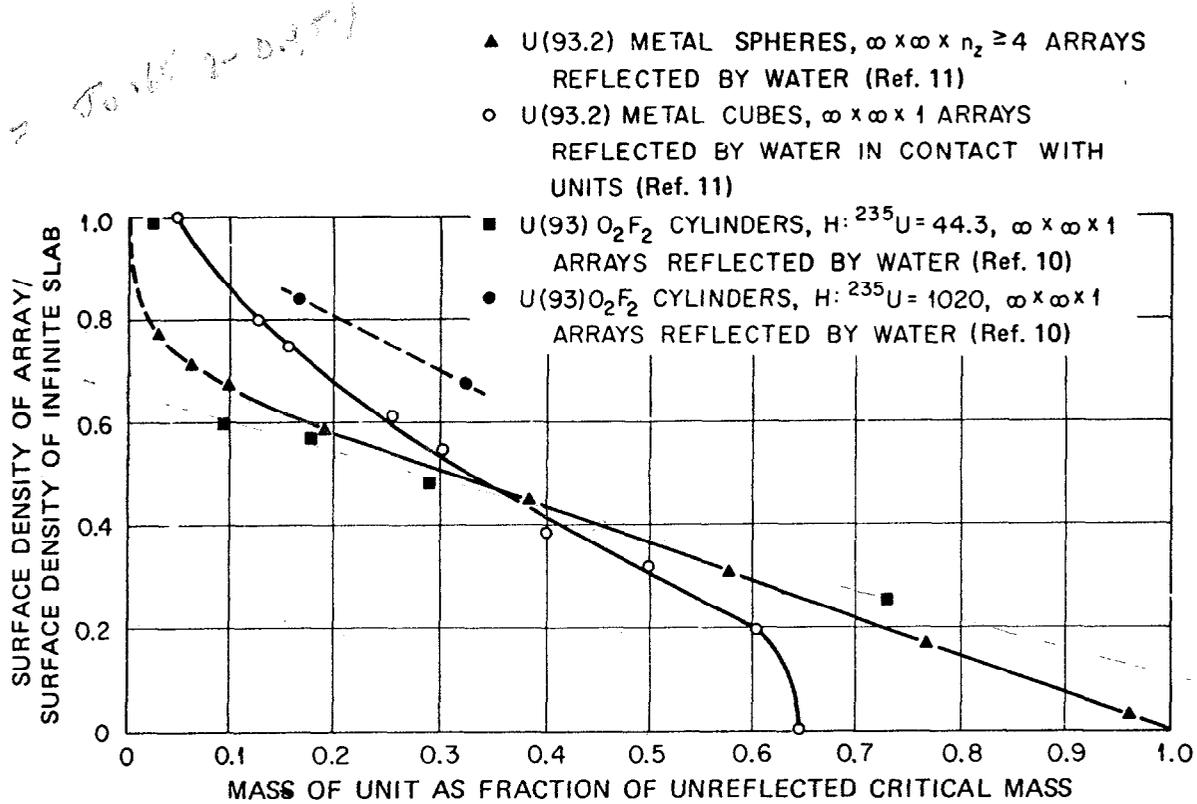


Fig. 2. Surface density representation of reflected planar arrays of U(93.2) metal and solution. The abscissa is the mass of a unit expressed as the fraction of the unreflected critical mass for the shape. With the exception of the metal cube data, the reflector is located at the peripheral cell boundaries of the arrays.

where  $a_n$  represents the half-cell dimension,  $n$  is the number of units along each edge of the array  $n^3 = N$ ,  $m$  is the mass of unit expressed as total uranium or plutonium, and  $c$  is a constant, independent of the array size, equal to  $0.55 \pm 0.18$  as determined by a least-squares fit to families of calculated arrays. The value of  $\sigma(m)$  is valid for all  $n \geq 4$  for a given mass,  $m$ , and in the limit of arbitrarily large  $n$  agrees with the surface density parameter given by Eq. (1).

The dependence of  $\sigma(m)$  on  $m$  is adequately expressed by the empirical linear relation

$$\sigma(m) = c_2(m_0 - m) \quad (5)$$

where  $m_0$  is the critical mass of an unreflected sphere of the fissile material and  $c_2$  is a constant characteristic of the material in water-reflected arrays. This linearity is illustrated in Fig. 3, where results typical of the calculations performed are represented in the format of Eq. (4) for four materials. We note this linearity breaks down for units of small mass as shown by the sharp rise in the data for <sup>235</sup>U. This behavior was observed for cubic arrays of all materials examined. As the mass of units in an array becomes small, we may assume they approach atomic di-

mension in the limit defining a cube of homogeneous low-density U(93.2) metal surrounded by water, which is represented by the point plotted at zero mass. There is no change in the calculated  $k_{eff}$  of this system, since the size of the cube is increased provided that value of surface density is retained.

Rewriting Eq. (4),

$$\frac{\sigma(m)}{m} = \frac{n}{(2a_n)^2} (1 - c/\sqrt{N})^2 \quad (6)$$

shows  $\sigma(m)/m$  to be constant for all critical cubic arrays of 64 or more units of mass  $m$  and effects separation of material and geometry characteristics of arrays. A straight line through the origin, therefore, intersects the lines representing different fissile materials at points defining equivalent spheres, as illustrated by the line in Fig. 3. These spheres of different materials may be substituted in an array, and criticality is maintained without change in array size. Substituting the expression for  $\sigma(m)$ , Eq. (5), yields an explicit relation for equivalent masses in the same critical array, namely,

$$m' = m_0' \left[ 1 + \frac{c_2}{c_2'} \left( \frac{m_0}{m} - 1 \right) \right]^{-1} \quad (7)$$

LIMITING SURFACE DENSITY,  $\sigma(m)$  (g/cm<sup>2</sup>)

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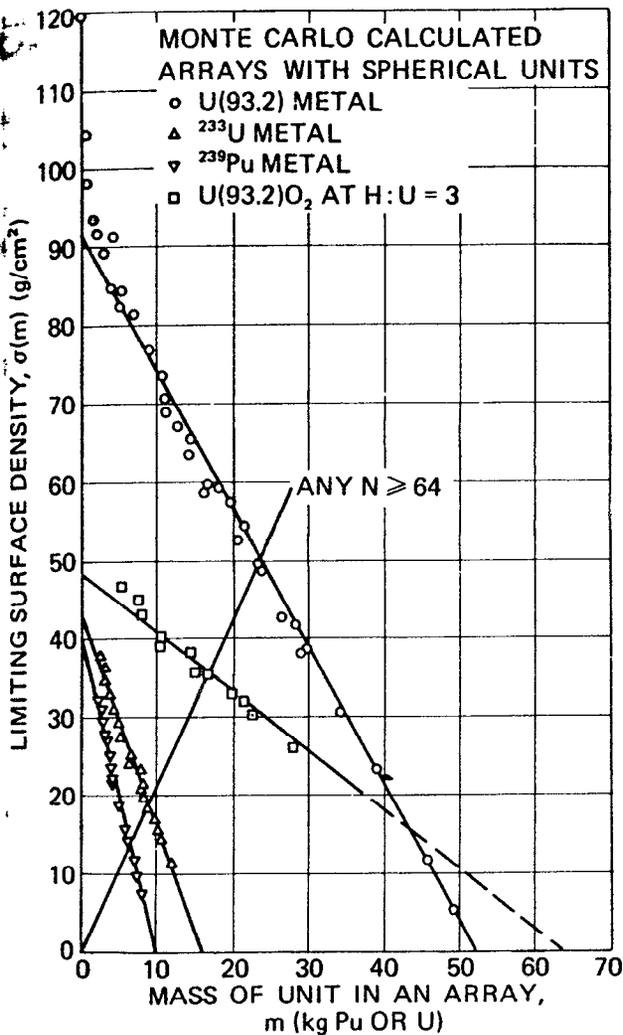


Fig. 3. Surface density representation of critical water-reflected cubic arrays of fissile materials as spheres centered in cubic cells.

where primes distinguish different fissile materials. This relation demonstrates that equivalent masses for array criticality of different fissile materials do not, in general, have the same reactivity nor are they the same fraction of a critical mass.

A line in the  $m, \sigma(m)$  plane defining criticality need not be linear, but Eq. (5) may be applied to a limited mass range of interest, i.e., criticality can be linearly approximated. Just as there are different  $c_2$  values for different fissile materials, there are different values for the same fissile material for specific consistent conditions. For example, concrete-reflected arrays of U(93.2) metal units would have a value of  $c_2$  distinct from that for water-reflected arrays. Comparisons of two conditions, or perturbations to an array, become susceptible to analytic expression by

establishing the values of  $c_2$  characterizing the conditions. This in itself is useful, but it becomes significant when associated with a change in array reactivity. A general result derived by Avery<sup>12</sup> for coupled reactors provides this association. For an array of identical units, the concept can be expressed by the following: For an array to experience a change of  $(\Delta k/k)_{\text{eff}}$ , it is sufficient that the reactivity of each unit in the array be changed by the same  $(\Delta k/k)_{\text{eff}}$ .

The reactivity corresponding to  $\Delta m/m$  for simple geometries is not difficult to determine and, in many instances, can be adequately estimated for nuclear criticality safety purposes. The relationship will depend on the energy spectrum of neutrons producing fission. An approximate linear relation exists between  $k_{\text{eff}}$  and a dimension of a unit when a majority of fissions is produced by neutrons of greater than thermal energy. In the case of fissile metal spheres, the  $k_{\text{eff}}$  of a unit is approximated by the ratio of radii  $r/r_0$ , where  $r_0$  is the unreflected critical radius and  $r$  the radius of the unit. The resultant  $k_{\text{eff}}$  of an array perturbed from criticality by a change in unit mass from  $m$  to  $m'$  is

$$k_{\text{eff}} \cong \left(\frac{m'}{m}\right)^{1/3} \quad (8)$$

These concepts and relationships are illustrated in the following sections.

#### ARRAY PERTURBATIONS

##### Reflector

The influence of changing from water-reflected arrays to arrays reflected by different thicknesses of concrete is indicated in Fig. 4, where the units of all arrays are U(93.2) metal spheres. The evaluated slope,  $c_2$ , for each thickness of reflector may be used in Eqs. (7) and (8) to define the change in  $k_{\text{eff}}$  associated with the reflector change. Note that the magnitude of  $(\Delta k/k)_{\text{eff}}$  is not constant but is dependent on the mass of the units in an array. For example, the reactivity increase may range from 0 to 18% upon substitution of a 40.6-cm-thick concrete reflector for a water reflector.

##### Unit Shape

The influence of unit shape on array criticality is exemplified by the results depicted in Fig. 5. These calculated data represent reflected cubic arrays with cylindrical units of U(93.2) metal

<sup>12</sup>R AVERY, "Theory of Coupled Reactors," *Proc 2nd UN Int Conf Peaceful Uses At Energy*, Geneva, 12, 182, United Nations, New York (1958)

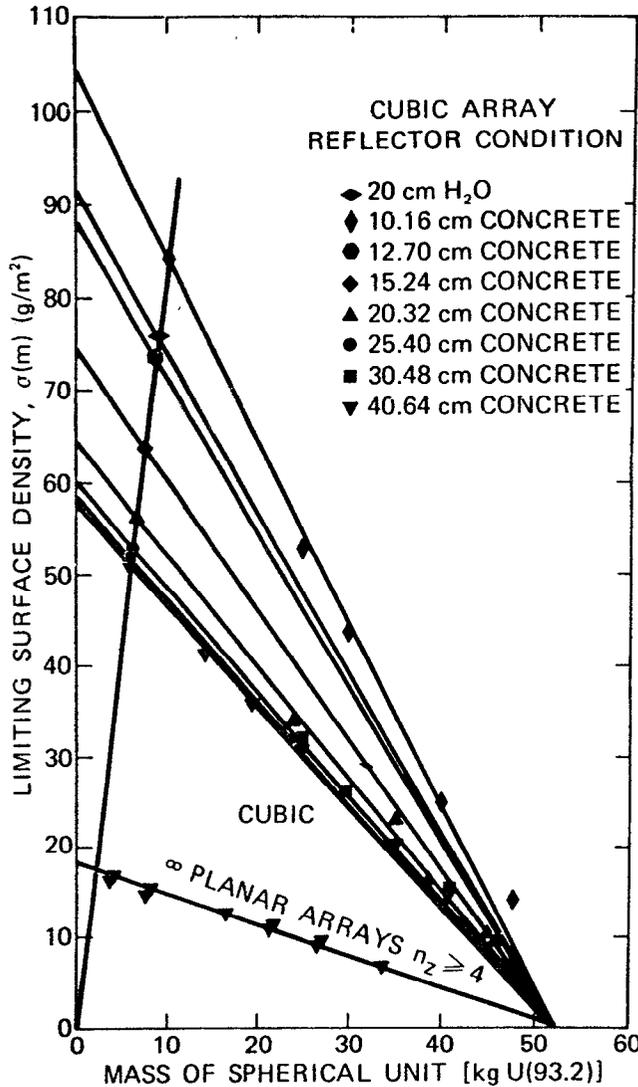


Fig. 4. Surface density representation of critical cubic arrays reflected by various thicknesses of concrete and infinite planar arrays with  $n \geq 4$  reflected by 40.64-cm-thick concrete

having three different shapes described by their height-to-diameter ratios,  $h/d$ . Along each line, the shape of units is maintained (constant  $h/d$ ) as mass increases to, at the intercept with the abscissa, the unreflected critical mass of a single unit of that shape. There is a range of height-to-diameter ratios in which the linearity characteristic of spheres is observed, as shown here by the  $h/d$  values of 1 and 0.3. That range is indicated to be from 0.3 to 3. Outside this range, two or more linear regions may be necessary to define criticality, each range of mass being characterized by a slope  $c_2$ . The data for an  $h/d$  ratio of 0.2 are examples of this behavior.

An illustration of the application of Eq. (8) is

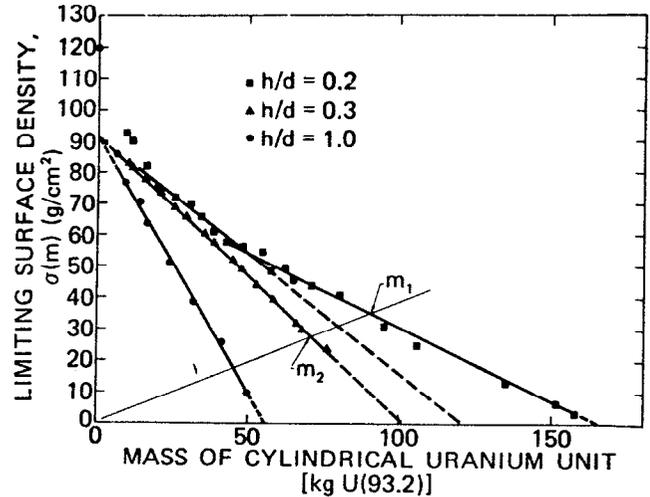


Fig. 5. Surface density representation of critical water-reflected cubic arrays of U(93.2) metal cylinders of various height-to-diameter ratios.

as follows. The masses  $m_1$  and  $m_2$ , indicated in Fig. 5, are equivalent masses for criticality in any selected array provided  $n \geq 4$ , as described in Eq. (6) and represented in Fig. 5 by the line through the origin. If the mass  $m_1$  in a critical array were reduced to  $m_2$ , maintaining  $h/d = 0.2$ , then the resulting array would be subcritical with a  $k_{eff}$ , from Eq. (8), of approximately  $(m_2/m_1)^{1/3}$ . If, on the other hand, the mass,  $m_1$ , were maintained but the shape of each unit changed to  $h/d = 0.3$ , then the resultant array would be supercritical and the  $k_{eff}$  would be estimated by  $(m_1/m_2)^{1/3}$ .

As may be suspected, the intercepts with the abscissa trace the familiar curve of cylindrical critical mass as the shape changes, approaching the minimum and receding toward larger values as the  $h/d$  ratio increases through unity. These arrays may be related to the arrays of spheres or other shapes for which the  $c_2$  characteristic values are known.

Array Shape

Application of these concepts to cubic arrays provides the criticality safety specialist with a powerful tool for the evaluation of many situations involving neutron interacting systems. The number of units in a cubic array may be conservatively applied to similar units in similar cells arranged in noncubic arrays, with the degree of conservatism dependent on complex factors. Insight into the influence of array shape on array criticality may be had by examining the perturbation of cubic arrays and interpreting the effect in terms of the characteristic slope,  $c_2$ . It is known that the rearrangement of cubic cells of a critical

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cubic array into an array having an unequal number of units along the three edges of an array, i.e., into a cuboidal shape, will result in subcriticality because the neutron leakage increases. Criticality of the resultant subcritical array may be restored in several ways, for example, by increasing the unit mass, by reducing the cell volume, or by increasing the number of units.

We begin the development of an analytic expression by assigning a unique numerical value corresponding to possible array shapes with cubic cells. The surface-to-volume ratio,  $s/v$ , of a noncubic arrangement of cubic cells is

$$(s/v)_A = \frac{1}{a_n} \left( \frac{1}{n_x} + \frac{1}{n_y} + \frac{1}{n_z} \right),$$

and for a cubic array is

$$(s/v)_c = \frac{3}{na_n}.$$

We eliminate the dependence on cell dimension by normalizing the  $s/v$  ratio for noncubic to that for cubic arrays of the same  $N$ , and define a shape factor for the noncubic array as

$$R = \frac{(s/v)_A}{(s/v)_c} = \frac{N^{1/3}}{3} \left( \frac{1}{n_x} + \frac{1}{n_y} + \frac{1}{n_z} \right). \quad (9)$$

Note that  $N^{1/3}$  need not be an integer, i.e., Eq. (4) is a continuous function of  $N$ .

To maintain criticality of an array, originally cubic and comprised of units of mass  $m$ , as cells are rearranged to form a cuboidal array of shape factor  $R$ , the mass of each unit must be increased to  $m'$ . The mass,  $m'$ , necessary to maintain criticality for all possible rearrangements of the cells in a reflected array were calculated<sup>11</sup> beginning with the  $a_0 = 16.454$ -cm array of 10.4-kg U(93.2) metal spheres in a cubic array. The resultant values of  $m'$  satisfy the relation

$$m' = 10.4R^{0.672}, \quad (10)$$

where the exponent was determined by a least-squares fit to the data and has a standard deviation of  $\pm 0.013$ . The maximum value of  $R$  for the  $a_0$  array is 5.34, corresponding to a linear-reflected array of 512 units. These data for various array shapes may be related to cubic array data and, thus, define the characteristic slope,  $c_2$ , as a continuous function of  $R$ .

A function is determined by assuming that reflected cuboidal arrays, having a shape factor  $R$ , satisfy Eqs. (4) and (5). The  $n$  appearing in Eq. (4) is the number of units in the  $z$  direction designated as  $n_z = \min(n_x, n_y, n_z)$ . The  $c_2$  of Eq. (5) is interpreted as the value characteristic of arrays with shape factor  $R$ . The mass,  $m'$ , necessary for criticality will satisfy

$$\frac{1}{c_2'} \frac{\sigma(m')}{m'} = \frac{m_0}{m'} - 1 = \frac{n_z}{c_2'(2a_n)^2} (1 - c/\sqrt{N})^2. \quad (11)$$

The line through the origin having a slope  $\sigma(m)/m$  for the cubic array of 10.4-kg U(93.2) units will be related to the line through the origin having slope  $\sigma(m')/m'$  for the cuboidal array by the simple ratio of the number of units in the  $z$  direction for the two arrays, i.e.,

$$\frac{\sigma(m')}{m'} = c_2' \left( \frac{m_0}{m'} - 1 \right) = \frac{n_z}{n} c_2 \left( \frac{m_0}{m} - 1 \right) = \frac{n_z}{n} \frac{\sigma(m)}{m}, \quad (12)$$

since the cell volume is maintained. Substituting the expression for  $m'$  from Eq. (10) and solving for  $c_2'$ , we obtain

$$c_2' = \frac{n_z}{n} \frac{4c_2}{5R^{-0.672} - 1}. \quad (13)$$

Inserting this expression with  $c_2 = 1.762 \times 10^{-3} \text{ cm}^{-2}$  for U(93.2) metal spheres into Eq. (11) and dropping the prime from  $m$  yields the semi-empirical equation

$$\frac{m_0}{m} - 1 = \left[ \frac{5.962(\sqrt{N} - c)}{na_n} \right]^2 (5R^{-0.672} - 1), \quad (14)$$

applicable to reflected U(93.2) metal arrays of any shape  $R$  provided  $R \leq 5.34$ . Arrays with shape factors  $> 5.34$  are correctly evaluated by setting  $R = 5.34$ .

Equation (14) is examined in Fig. 6, where the limiting surface density,  $\sigma(m)$ , of Eq. (4), is shown as a function of the mass of the units in an array. The upper line with the greatest negative slope represents water-reflected cubic arrays of metal spheres. Note that each of the three points represents three different cubic arrays having  $N = 1728, 512,$  and  $216$  units of equal mass but different spacing. The three curves emanating from this cubic array line in a downward direction, curving to the right, represent rearrangements of those units into noncubic arrays. That is, units are removed from the  $z$  direction of the array, added to the  $x$  and  $y$  directions, and the resulting array of shape  $R$  is closely reflected by water. Changes in the shape of nine arrays, therefore, are presented. Arrays with the same shape factor,  $R$ , lie on a line having a slope,  $c_2'$ , satisfying Eq. (13).

The changes in the number of units in any dimension of cubic array by a factor of 2 has little effect on the  $k_{eff}$  of the array. This is illustrated in Fig. 6 by the small increase in the mass of the units required to maintain criticality when the array of 10-kg U(93.2) units is changed from a  $12 \times 12 \times 12$  to an  $18 \times 16 \times 6$  arrangement. The

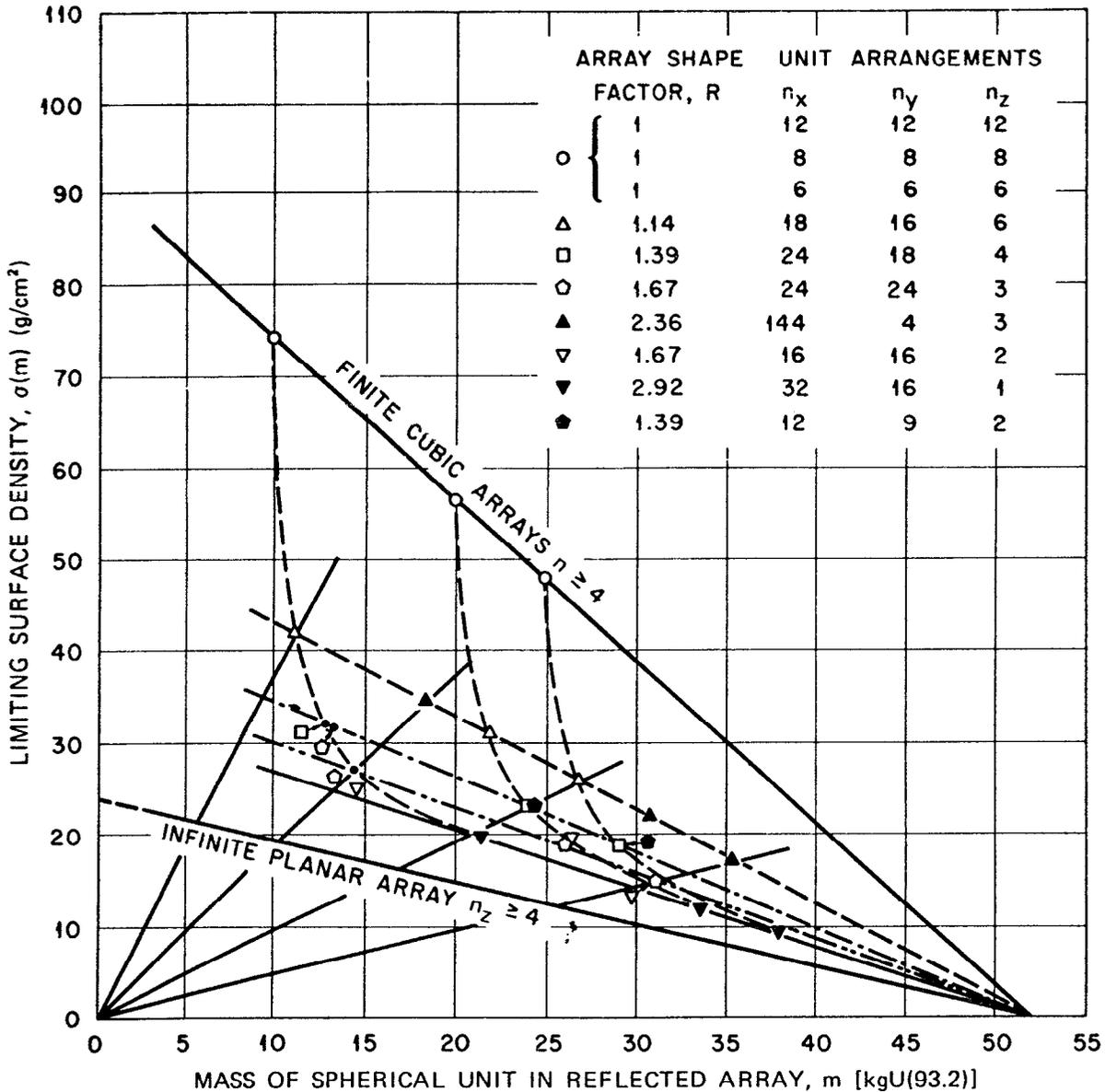


Fig. 6. Surface density representation of critical, noncubic water-reflected arrays of U(93.2) metal spheres centered in cubic cells.

rearrangement of arrays having units of large mass obviously will result in only a small change in the array  $k_{eff}$ .

Criticality in reflected cuboidal arrays of fissile materials other than U(93.2) metal spheres is properly specified by application of the equivalent mass relation, Eq. (7), to the spherical mass of U(93.2) satisfying Eq. (14) for the cuboidal array of shape factor  $R$ . This procedure avoids the necessity of developing for each fissile material a relation characterizing array shape effects on criticality. The applicability of Eq. (7) to other fissile materials was confirmed by Monte Carlo calculations, and the comparison is given in

Table I. The 512-unit cubic array of 10.4-kg U(93.2) units ( $a_s = 16.454$  cm) was subjected to extreme shape changes. The radius of U(93.2) spheres necessary for criticality in each case was specified by Eq. (14). This radius, in turn, was converted to a radius of a plutonium metal sphere by Eq. (7). The average of the ratios of radii predicted by Eq. (7) to those calculated by Monte Carlo for the arrays is  $0.999 \pm 0.003$ .

*Infinite Planar Arrays*

Let us now return to the characterization of criticality of reflected infinite planar arrays. The

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TABLE I

Comparison of Monte-Carlo-Calculated Critical Water-Reflected Arrays of Various Shapes with Estimates from Application of Eqs. (7), (9), and (14) to Plutonium Metal Spheres

Unit Arrangement			U(93.2) Radius, by Eq. (14) (cm)	Equivalent <sup>239</sup> Pu Radius, by Eq. (7) (cm)	KENO Calculation <sup>a</sup>	
n <sub>x</sub>	n <sub>y</sub>	n <sub>z</sub>			Converged Radius (cm)	k <sub>eff</sub>
16	16	2	5.635	3.81	3.821	1.000
32	4	4	5.542	3.76	3.757	0.995
32	8	2	5.740	3.86	3.877	1.004
64	8	1	6.488	4.19	4.182	0.998
128	2	2	6.403	4.16	4.180	0.995
128	4	1	6.729	4.29	4.294	0.997
256	2	1	7.062	4.42	4.405	0.999

<sup>a</sup>Iteration of radius to give k<sub>eff</sub> of unity within a standard deviation of ±0.005.

relation applicable to infinite planar arrays is derived as follows. When dealing with units of equal mass in different reflected arrays, we noted in Eq. (6) that the cell dimensions and numbers of units relate in the manner given by the equation

$$\left(\frac{a_n}{a'_n}\right)^2 = \left(\frac{n}{n'}\right)_z \left(\frac{1 - c/\sqrt{N'}}{1 - c/\sqrt{N}}\right)^2 \quad (15)$$

If (n/n')<sub>z</sub> is maintained and N is allowed to increase indefinitely (i.e., n<sub>x</sub>, n<sub>y</sub> → ∞), then the cell dimensions vary directly as the square root of the ratio of units in the z direction,

$$\frac{a_n}{a'_n} = \left[\frac{n}{n'}\right]_z^{1/2} \quad (16)$$

Consequently, as with reflected infinite slabs of uniform fissile materials, the surface density does not change with separation for a given value of sphere mass.

This behavior is confirmed in Table II. The radius of the unit in each of six sets of arrays of different size was determined to test Eq. (16). In each set, the critical condition n<sub>z</sub> = 4 was used in Eq. (16) to estimate the necessary increase in spacing for n<sub>z</sub> = 6, 8, and 10. The predicted cell dimension of infinite critical arrays was an input parameter in Monte Carlo calculations to determine the critical radii of spheres to within one standard deviation. The variation of radii within each set is negligible, differing by <1% or about the statistical precision expected.

The data of Table II appear as the lower envelope in Fig. 6. When normalized to the value of the surface density for a reflected infinite uniform slab of U(93.2) metal (32 gU/cm<sup>2</sup>), the data appear as shown in Fig. 2.

Similar results for concrete-reflected infinite planar arrays also sustain Eq. (16), and these data

TABLE II

Monte-Carlo-Calculated Critical Water-Reflected Infinite Planar Arrays of U(93.2) Metal Spheres Centered in Cubic Cells

n <sub>x</sub> = n <sub>y</sub> = ∞ n <sub>z</sub>	Predicted Half-Dimension of Cell, a <sub>nb</sub> by Eq. (16) (cm)	KENO Calculation <sup>a</sup>	
		Converged Sphere Radius, r (cm)	k <sub>eff</sub>
4	7.897	2.718	1.001
6	9.672	2.715	0.996
8	11.169	2.710	0.999
10	12.487	2.700	0.998
4	11.764	3.452	0.996
6	14.408	3.451	1.002
8	16.637	3.448	1.002
10	18.601	3.452	1.002
4	15.364	4.058	1.001
6	18.817	4.045	1.005
8	21.728	4.024	1.002
10	24.293	4.027	1.002
4	19.029	4.592	1.004
6	23.305	4.566	1.001
8	26.911	4.542	1.001
10	30.087	4.591	1.005
4	23.082	5.057	0.996
6	28.269	5.057	1.001
8	32.643	5.057	0.998
10	36.496	5.057	0.997
4	27.873	5.566	0.996
6	34.138	5.554	1.003
8	39.419	5.550	1.003
10	44.072	5.600	1.004

<sup>a</sup>Iteration of radius to give k<sub>eff</sub> equal to unity within a standard deviation of ±0.005.

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TABLE III  
Unreflected Spherical Critical Masses and Characteristic Array Constants  
for Some Fissile Materials in Water-Reflected Cubic Arrays

Number	Material	Atomic Ratio, <sup>a</sup> H/U or H/Pu	Spherical Unit Unreflected Critical Mass <sup>a</sup> (kg)	Characteristic Constant for Criticality of Water-Reflected Arrays (10 <sup>-3</sup> cm <sup>2</sup> )	
				c <sub>2</sub>	±
1	Metal, U(100)	0	45.68	1.806	0.036
2	Metal, U(93.2)	0	52.10	1.762	0.017
3	Oxide, U(93.2)O <sub>2</sub>	0.4	90.24	0.854	0.007
4		3.0	63.59	0.758	0.008
5		10.0	31.43	0.778	0.007
6		20.0	17.34	0.805	0.004
7	Metal, U(80)	0	69.89	1.359	0.012
8	Oxide, U(80)O <sub>2</sub>	0.4	111.36	0.780	0.006
9		3.0	74.08	0.713	0.006
10		10.0	36.16	0.725	0.006
11		20.0	18.67	0.779	0.005
12	Metal, U(70)	0	89.16	1.192	0.018
13	Oxide, U(70)O <sub>2</sub>	0.4	133.39	0.723	0.006
14		3.0	83.44	0.686	0.006
15		10.0	36.89	0.735	0.004
16		20.0	19.30	0.793	0.004
17	Metal, U(50)	0	159.60	0.901	0.008
18	Oxide, U(50)O <sub>2</sub>	0.4	207.73	0.589	0.005
19		3.0	112.82	0.594	0.004
20		10.0	55.14	0.520	0.006
21		20.0	21.48	0.777	0.005
22	Metal, U(40)	0	228.06	0.787	0.016
23	Metal, U(30)	0	379.70	0.589	0.007
24	Oxide, U(30)O <sub>2</sub>	0.4	409.60	0.450	0.003
25		3.0	150.01	0.603	0.005
26		10.0	54.01	0.636	0.004
27		20.0	25.15	0.744	0.005
28	Metal, Pu(100)	0	9.95	4.346	0.112
29	Oxide, Pu(100)O <sub>2</sub>	0.4	26.66	1.542	0.015
30		3.0	28.65	1.113	0.010
31		10.0	20.21	0.965	0.007
32		20.0	14.05	0.885	0.008
33	Metal, Pu(94.8)	0	10.34	4.138	0.091
34	Oxide, Pu(94.8)O <sub>2</sub>	0.4	27.93	1.561	0.013
35		3.0	32.78	1.097	0.011
36		10.0	28.74	0.817	0.007
37	Metal, Pu(80)	0	11.69	4.261	0.099
38	Oxide, Pu(80)O <sub>2</sub>	0.4	32.14	1.529	0.023
39		3.0	42.43	1.022	0.013
40		10.0	47.81	0.679	0.005
41	Metal, <sup>233</sup> U	0	15.75	2.751	0.022
42	Oxide, <sup>233</sup> UO <sub>2</sub>	0.4	34.46	1.199	0.008
43		3.0	31.69	0.939	0.008
44		10.0	17.64	0.907	0.010
45		20.0	10.28	0.947	0.009
46	Metal U(93.2)-10 wt% Mo	0	73.06	1.305	0.009

<sup>a</sup>Total uranium or total plutonium.

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are shown in the lower portion of Fig. 4. Calculated critical surface densities for a concrete-reflected uniform slab of U(93.2) metal range from 12 to 20.6 gU/cm<sup>2</sup> (Refs. 13 and 14, respectively) depending on cross sections, concrete composition, and code options. Were the concrete-reflected planar array data normalized to a surface density in the range given, the line depicting criticality would appear in Fig. 2 above the water-reflected data.

Application of the equivalent mass relation, Eq. (7), to the data for infinite planar arrays of U(93.2) metal spheres will, in general, lead to conservative estimates of criticality of other fissile materials. Presented in Table III is a summary of characteristic constants for spherical units of some fissile materials. The apparent general applicability of the U(93.2) spherical data in Fig. 2 to the variety of fissile materials listed in Table III strongly supports the concept that a suitably limited area of the figure can be defined within which subcriticality will be specified.

#### COMPARISON OF DENSITY ANALOG AND SURFACE DENSITY MODELS

Having established some understanding of density representations of array criticality, let us now relate the two density models. This is accomplished by cubing the surface density equation for criticality, Eq. (4), and interpreting the quantities in the following manner. The cube of the edge dimension of a cubic cell squared,  $(2a_n)^2$ , becomes the cell volume squared and when divided into the unit mass squared gives the average fissile material density in the array,  $\rho$ , to the second power. The cubed  $n$  becomes the total number of units in the array,  $N$ , and the binomial factor is raised to the sixth power. Rearranging terms and substituting Eq. (5) for  $\sigma(m)$  gives

$$N(1 - c/\sqrt{N})^6 = \frac{[c_2(m_0 - m)]^3}{m\rho^2} \quad (17)$$

The result is an explicit form of the coefficient to be used in the density analog model and an unambiguous second power for the density exponent. Similar treatment of the relation for criticality of arrays with shape  $R$ , Eq. (14), gives

$$N(1 - c/\sqrt{N})^6 = \frac{(m_0 - m)^3}{(5R^{-0.672} - 1)(11.924)^6 m\rho^2} \quad (18)$$

An illustration of possible interpretation of Eqs. (17) and (18) is given in Fig. 7, where the lines depict reflected critical arrays of 20-kg U(93) metal spheres. Beginning with the first line on the left, it can be seen that the cubic arrays, represented as the total mass reduced by the fraction  $(1 - c/\sqrt{N})^6$ , have a density exponent of 2.0. The line immediately to the right represents the same data as actual total mass in the arrays, revealing the variable density exponent behavior typically reported. The line tangent to the cubic arrays at 160-kg U(93.2) exhibits the result of maintaining only two cells in the  $z$  direction as  $N$  increases, i.e., arrays of variable shape. The slope of this line has already achieved a value  $>2$ , and the line would, if continued, ultimately approach a limiting average density below which criticality would not be possible. The arrays represented by the upper curve, to the right, are for arrays of constant shape,  $R$ , whose dimensions are in the ratio of those of a critical reflected slab of metal,  $63.5 \times 63.5 \times 1.8$  cm. Of significance here is the critical mass predicted as the average uranium density achieves the metal density. The curve underpredicts the experimental mass for this shape by  $\sim 8\%$ , as shown by the open triangle, one of a series reported by Mihalczko and Lynn<sup>15</sup> and by Paxton.<sup>16</sup> Other data from that series are compared in Fig. 8 with the results obtained from the application of Eq. (18). The agreement of experimental and estimated masses is well within 10%. These results show that the shape of the critical assembly as the average array density approaches the metal density is that of the array and not that of the units in the array. Similarly, consideration of cubic arrays leads to the conclusion that the geometry of the limiting assembly is cubic, independent of the shape of the units in the array.

It is informative to apply Eq. (17) to the experimental arrays of Fig. 1 considering the cylindrical masses as spherical. The points plotted in Fig. 9 represent experimental data<sup>3</sup> obtained with units of different shape in noncubic cells arranged in reflected cuboidal arrays. The lines represent calculations of arrays comprised of spherical units in cubic cells and, in accord with Eq. (17), have a slope of minus 2. The ordinate is the corrected number of units [left side of Eq. (17)] in the arrays. It is observed that the experimental data do not vary as the inverse square of the average density law.

<sup>13</sup>D. R. SMITH, Los Alamos Scientific Laboratory, Personal Communication.

<sup>14</sup>S. J. ALTSCHULER and C. SCHUSKE, *Nucl. Technol.*, **13**, 131 (1972).

<sup>15</sup>J. T. MIHALCZO and J. J. LYNN, "Neutron Multiplication Experiments with Enriched Uranium Metal in Slab Geometry," ORNL CF-61-4-33, Oak Ridge National Laboratory (1961).

<sup>16</sup>H. C. PAXTON, "Los Alamos Critical-Mass Data," LA-3067, rev., p. 39, Los Alamos Scientific Laboratory (1975).

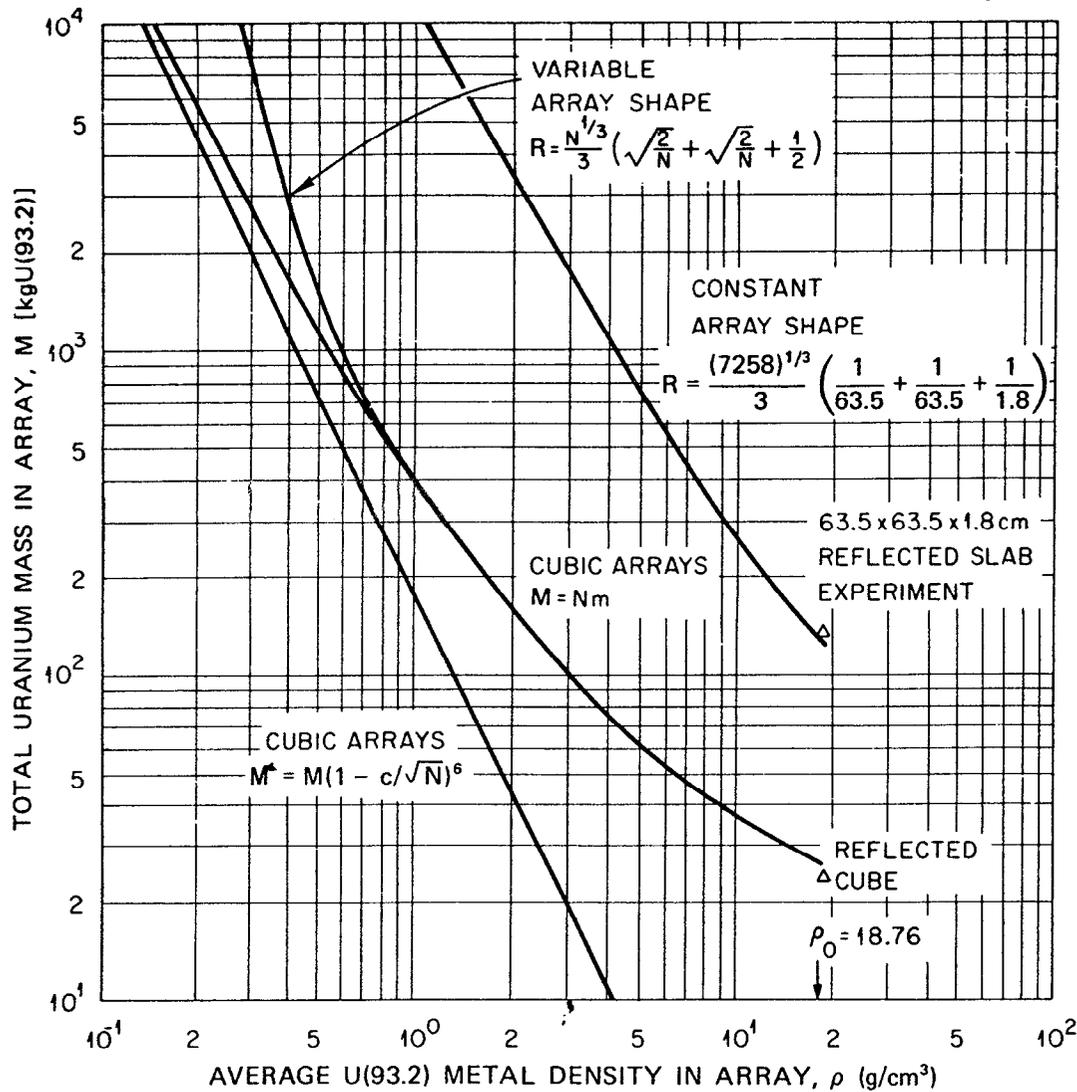


Fig. 7. Density analog representation of reflected array criticality for U(93.2) metal units as spheres.

The observed difference can be understood if we consider the results of Monte Carlo calculations of reflected arrays by the matrix method, which yield the effective unit self-multiplication within arrays. As computed by the code, the self-multiplication of a unit is the ratio of fission neutrons produced in a unit to fission neutrons born in the unit independent of their history and excludes fission neutrons from other units in the array. Calculations of 8- to 1000-unit arrays of 26-kg U(93.2) metal cylinders show that, for the 8-, 27-, and 64-unit arrays, the unit self-multiplication is, respectively, 0.74, 0.67, and 0.66, changing little from 0.66 as  $N$  increases further. The relative constancy of the effective unit self-multiplication for arrays with 64 or more units is

general. It can be concluded from these observations that there is valid reason not to expect experiments in the high-density range to follow an inverse square density relation, since the self-multiplication of otherwise identical units in various critical arrays is not constant but depends on the characteristics of the array.

CONCLUSIONS

The Paxton and the Smith density analog models are suitable for their intended purpose of specifying criteria that easily separate problems requiring no further evaluation from those needing further detailed investigation.

URANIUM CRITICAL MASS [kg U(93.2)]

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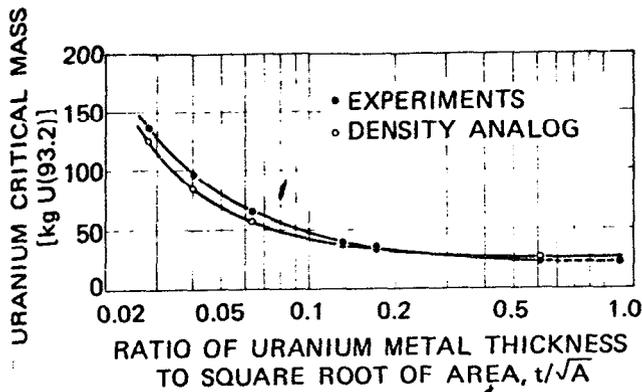


Fig. 8. Critical mass of U(93.2) metal as a function of cuboidal geometry for Plexiglas (methyl methacrylate)-reflected slabs.

The surface density representation of reflected infinite planar arrays is in need of additional definitive interpretation to clearly establish a universal safe region expressed as a fraction of the surface density and as a fraction of the unreflected critical mass for the unit shape.

The limiting surface density model and its translation to the density analog model, together present a comprehensive representation of reflected, critical arrays of air-spaced units. Having been developed from the same bases as the American National Standards Institute standard, "Guide to Nuclear Criticality Safety in the Storage of Fissile Materials," N16.5(1975), the concepts are applicable to arrays described therein.

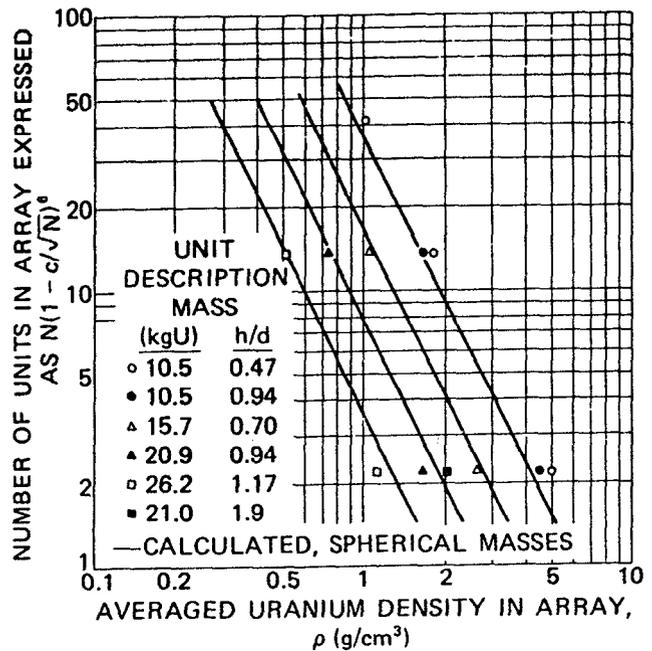


Fig. 9. Comparison of experimental data for metal cylinders with calculated metal spheres in reflected arrays.

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