CRITICALITY CONTROL OF FISSILE MATERIALS

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EXPERIMENTAL AND CALCULATED SYSTEM CRITICALITY*

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Abstract

EXPERIMENTAL AND CALCULATED SYSTEM CRITICALITY. A continuing programme directed to the neutronics of uranium-235 system criticality has been in progress for a number of years in the United States of America. The experiments were designed to provide basic criticality data describing arrays susceptible to simple description: elementary geometry and homogeneous fuel regions. Such data fall naturally into a class which lends itself directly to analyses and, at the same time, may be applied to nuclear safety evaluations. The component variables examined included mass, shape, moderation, and the ^{235}U content of the uranium, while the array variables studied were shape, moderation, degree of reflection and the number of units and their spacing. Data from four series of experiments and representative calculations utilizing two Monte Carlo codes are given.

Series I. Unreflected two- and three-component critical assemblies of coaxial U(93) metal discs with diameters between 17.78 cm and 31.10 cm were studied. The unit mass as a function of separation was determined.

Series II. Critical three-dimensional arrays of as many as 64 units of U(93.2) metal cylinders ranging from 10.5 to 26.2 kg, in five sizes, were employed to examine the effects of array moderation, reflection, shape and unit perturbation.

Series III. Five-litre components of aqueous $U(92.6)O_2(NO_3)_2$ solution, at a concentration of 415 g of uranium per litre, contained in right circular cylinders of methacrylate plastic were assembled in three-dimensional arrays. As many as 125 units were used to determine the number required for criticality as a function of their spacing and the degree of array reflection.

Series IV. Cylinders of aqueous $U(5)O_2F_2$ solution, at a concentration of 901 g of uranium per litre, 24.1 cm diam. and up to 142.0 cm high were assembled to criticality in triangular and square patterns in planar geometry.

Calculations with O5R, a General-Purpose Monte Carlo Neutron Transport Code developed at the Oak Ridge National Laboratory, yields multiplication constants in the range 0.971 to 1.028; GEM, a Monte Carlo Neutronics Code developed by the Authority Health and Safety Branch (U.K.A.E.A.), yields values in the range 1.000 to 1.032.

1. INTRODUCTION

The fountain head for specifications of the safe handling of fissile materials is the definition of their criticality. The framework for evaluating the degree of safety that has evolved over the past few years circumvents the natural starting point for safety analysis, criticality, by specifying factors and conditions to be applied to a given number of units producing a fictitious system which must be demonstrably subcritical. Such an approach can result in fertile ground for disputations between mutually interested parties, even within managements of installations.

Knowledge of systems criticality and of the magnitudes of factors affecting their criticality directly applied to problems in safety is a preferable, less controversial approach. During the past decade the United States of America

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has actively supported an experimental programme on systems criticality. The purpose of the programme has been to produce information (a) on systems that are as free from extraneous materials as possible, that are free from geometric complexities, and that may serve as a basis both for evaluating calculative techniques or models, and (b) for direct application to nuclear safety problems. Monte Carlo calculative techniques combined with the existing system-criticality data can provide a necessary basis for a more favourably oriented delineation of safety evaluations.

Data representative of the contributions of the Oak Ridge National Laboratory Critical Facility are presented in this paper together with the results from three methods of calculation. The experiments deal exclusively with uranium having two greatly differing values of ^{235}U enrichment, a wide range of uranium densities, and extensive variations in size and geometry of units used. In addition to the effect on array criticality of unit variation, other array effects examined included array shape, degree of reflection and/or moderation, and combination of arrays with different neutron energy spectra. Typical Monte Carlo and neutron current calculations of these data are compared.

2. MATERIAL

A general description of materials is presented in this section. Specific details necessary to characterize criticality are given with the experimental data for each series. The principal mode of assembly and control is stated; particular details of the apparatus are to be found in the reference cited.

2.1. Fissile

The physical forms of the fissile materials used were either metal or aqueous salt solutions. The low-^{235}U enrichment material was a fluoride solution used only at a single concentration near that which would produce the minimum critical volume measured in a single vessel. The material at a higher ^{235}U content was utilized both as nitrate solution of various uranium concentrations and as a metal. There was a negligible amount of excess fluoride ion in the fluoride solutions; the total nitrate in the nitrate solution corresponded to an $N/^{235}U$ ratio of 2.006. No other impurities were present in significant quantities in the fissile materials. The isotopic content of the uranium for the various physical forms is given in Table I.

2.2. Hydrogenous materials

Hydrogenous materials were used as reflectors and moderators in the arrays. Paraffin, in various thicknesses from 1.3 to 15.2 cm, and polyethylene of 15.2 cm thickness, were used as reflectors. Plexiglas, a methacrylate plastic, was used as a moderator in the arrays. The physical properties of these materials are listed in Table II.

TABLE I

WEIGHT PERCENT OF URANIUM ISOTOPES PRESENT IN THE FISSILE MATERIALS

Uranium Isotope	$U(5)O_2F_2$	$U(92.6)O_2(NO_3)_2$	Metal
234	0.03	1.0	1.0
235	4.97	92.6	93.2
236	0.05	0.5	0.2
238	94.95	5.9	5.6

TABLE II

PHYSICAL CHARACTERISTICS OF REFLECTOR AND MODERATOR MATERIALS

Material	Chemical Form	Density
Paraffin	$C_{25}H_{52}$	0.93[a]
Polyethylene	CH_2	0.916
Plexiglas	$C_5H_8O_2$	1.18

a. An exception to the paraffin density given is 0.88 g/cm³ for the 1.3-cm-thick reflected experiments.

2.3. Iron

A number of experiments with metal units were performed with the units in iron containers. The containers consisted of suitable lengths of 14.130 cm o.d. pipe with a 0.655 cm wall thickness (5 in. schedule 40 pipe), having 0.635 cm thick end plates. The density was 7.85 g/cm^3.

2.4. System assembly

The method of assembly for solution units was to space the proposed system of units by hand, taking (depending upon the total number of units in the array) three, four or five centrally located empty containers which could be filled remotely. Reactivity control and criticality were achieved by varying the common solution height in these central units. The spacing was adjusted to attain criticality when the control containers were indistinguishable from the hand-placed units. A system of 125 units is shown in Fig. 1; visible at the top of the array are five vent lines from the control units in the central plane of the array.

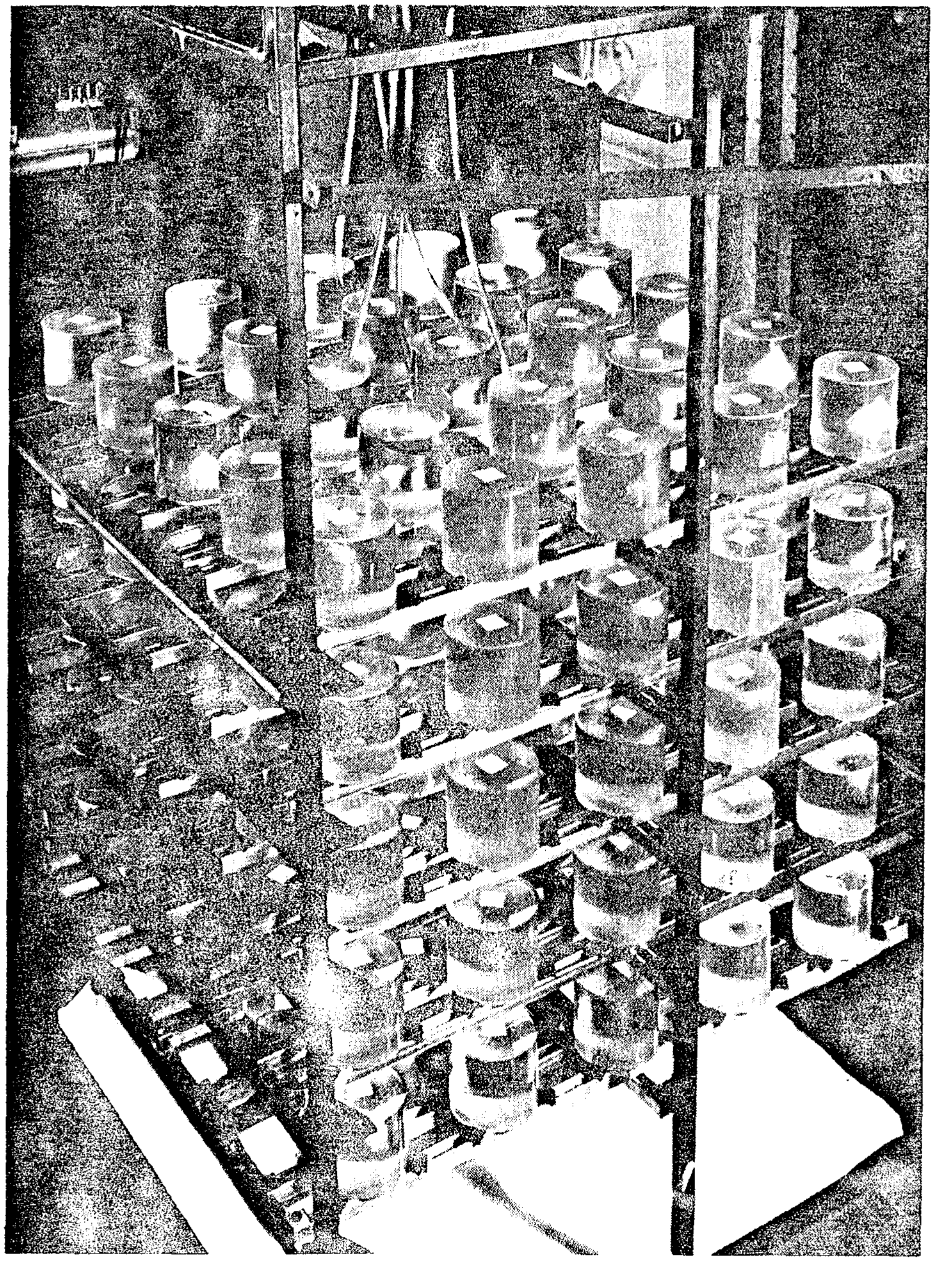

FIG. 1. A view of a critical system of 125 F^1-units of Series III experiments. Each unit contains 1.92 kg of ^{235}U as uranyl nitrate solution.

The two component metal assemblies were conducted on the Criticality Testing Unit [1], a double platform device in which the lower platform is movable vertically by an air-actuated hydraulic system bringing the parts of an assembly together. Reactivity control was obtained by varying the distance separating the units.

The multiple component metal systems were conducted on the Split Table Apparatus [1], two tables in the same horizontal plane, one of which can be moved by an air-hydraulic cylinder regulated by an electric motor drive providing variable closure speed; the other table is fixed. A suitable portion of an array was assembled on each of the tables and reactivity control was effected by varying the table separation.

3. SERIES I: U(93.2) METAL DISCS[1,2]

The neutron interaction was studied between two and three component systems [2] utilizing uranium metal cylinders of varying thicknesses to determine the critical spacing of identical pieces. The units were U(93.2) metal with a density of 18.7 g/cm^3. The unit surface separation between the large, parallel, flat surfaces of the cylinders as a function of their geometry and thickness is shown in Fig. 2. The insert on the figure gives the data for the critical height of the individual cylinders of various diameters used in the experiments [3] and provides asymptotes for the curves shown. The asymptotic behaviour is typical of unreflected and unmoderated arrays limited to one or two dimensions.

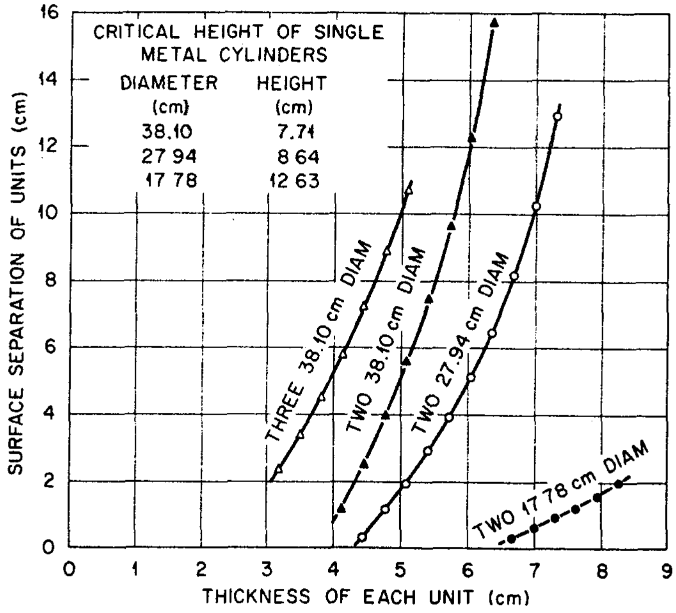

FIG. 2. Critical unreflected and unmoderated linear arrays of uranium metal.

4. SERIES II: U(93.2) METAL CYLINDERS

For convenience in reference and description, average dimensions and masses of the units utilized in the arrays constructed in these experiments have been collected in Table III. The experimental results obtained from assembling the units into arrays are grouped according to the effects investigated. The largest group comprises regular three dimensional reflected and unreflected arrays. Other groupings are partially reflected, cubic and rectangular parallelepiped lattice cells, unit shape, array shape, moderation and mixed arrays.

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[1] This series of experiments was conducted by Mihalczo [3].

[2] U(93.2) designates uranium containing 93.2 wt.% ^{235}U.

TABLE III

DIMENSIONS OF AVERAGE U(93.2) METAL UNITS CONSTITUTING ARRAYS

Uranium density = 18.76 g/cm^3

Unit Designation	Mass (kg of U)	Diameter, d (cm)	Height, h (cm)	h/d
A^1	10.480	11.506	5.382	0.47
A^2	10.484	11.509	5.382	0.47
A^3	10.507	9.116	8.641	0.95
A^4	10.489	9.116	8.641	0.95
A^5	10.458	11.494	5.382	0.47
A^6	10.434	11.481	5.382	0.47
A^7	10.384	11.454	5.382	0.47
B^1	15.692	11.494	8.077	0.70
B^2	15.683	11.490	8.077	0.70
B^3	15.696	a		
B^4	15.807	9.116	12.962	1.42
C^1	20.805	11.464	10.765	0.94
C^2	20.960	11.506	10.765	0.94
C^3	20.877	11.484	10.765	0.94
C^4	20.896	11.488	10.765	0.94
C^5	20.892	b		
C^6	21.008	9.116	17.282	1.90
D^1	26.218	11.509	13.459	1.17
D^2	26.113	11.486	13.459	1.17
E^1	5.225	11.494	2.691	0.23
E^2	5.254	9.116	4.320	0.47
E^3	5.245	9.116	4.320	0.47

a. This unit consisted of one E^3 mounted coaxially with and between two E^1's.

b. This unit consisted of one A^7 mounted coaxially with and between two E^2's.

4.1. Regular three-dimensional arrays

Arrays having an equal number of units along the three directions of the array are referred to as regular arrays. Data obtained from regular arrays of 8, 27 and 64 units, both unreflected and reflected (using various thicknesses of paraffin) are given in Table IV. Each entry represents a critical array with the two notable exceptions. The array description, column 1, utilizes the letter and superscript of Table III to identify the average unit in the array, the subscript is the total number of units in the array, and the numbers in parentheses are the number of units along each of the three directions. The uniform paraffin reflector thickness surrounding the array is given in column 2. The surface separation of units, equal in three directions, and the average uranium density appear in columns 3 and 4, respectively. Column 5 gives an indication of the array shape expressed as the ratio of the array height to the square root of its base area.

To simplify reference to the critical arrays of Table IV and to reduce their recurring descriptions the following notation will be used:

$$x^{i}_{n}[t;\ \delta;\ \rho;\ r]$$

where

x^i = average unit in array described in Table III;

n = total number of units in the array;

t = paraffin reflector thickness (cm);

δ = surface separation of units (cm);

ρ = average uranium density in array ($g(U)/cm^3$);

r = ratio of array height to the square root of its base area.

Comparison of arrays with equal numbers of units and the same reflector conditions reveals the expected inverse relation between the average uranium density and the unit shape, the array shape and the mass of the unit.

4.2. Partial reflection

The effect of a 15.2 cm thick reflector on three sides of an array, 'corner reflection', was investigated in two assemblies of units having average masses of 20.9 kg and a height-to-diameter ratio of 0.94. The results are given in Table V. The average densities of these two arrays may be compared to that of the critical array $C^2_8[2.5;\ 5.710;\ 4.292;\ 0.96]$ from Table IV and the interpolated[3] array, C^3_{27} [2.5; 11.53; 1.87; 0.96], which allows of the conclusion that the thick reflector on three sides of the array was slightly less effective than one 2.5 cm thick completely surrounding the arrays.

4.3. Comparison of array patterns

Eight and twenty-seven unit arrays were employed to explore the effect of changing the lattice cells in regular arrays from rectangular parallelepipeds to cubes. Four arrays were constructed, each with the units located at the corners of a cube. These arrays are described in Table VI where, for

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[3] This critical array is an interpolation between critical arrays of twenty seven C^3 units with 1.3 and 3.8 cm thick paraffin reflectors.

TABLE IV

CRITICAL CONDITIONS FOR REGULAR THREE DIMENSIONAL ARRAYS WITH VARIOUS PARAFFIN REFLECTORS

Array Description[a]	Paraffin Reflector Thickness (cm)	Surface Separation[b] of Units (cm)	Average Uranium Density in Array (g/cm^3)	Ratio of Array Height to $\sqrt{\text{Base Area}}$
A_8^1 (2x2x2)	0	0[c]	14.709	0.47
	1.3	0.229	13.563	0.48
	3.8	1.981	7.825	0.55
	7.6	3.416	5.350	0.59
	15.2	3.696	4.995	0.60
A_{27}^2 (3x3x3)	0	2.007	7.767	0.55
	1.3	2.992	5.954	0.58
	3.8	5.872	3.085	0.65
	7.6	8.258	1.967	0.69
	15.2	8.689	1.826	0.70
A_8^3 (2x2x2)	0	0[d]	14.632	0.95
	1.3	0.602	12.037	0.95
	3.8	2.362	7.248	0.96
	7.6	3.970	4.865	0.96
	15.2	4.308	4.503	0.97
A_{27}^4 (3x3x3)	0	2.436	7.096	0.96
	1.3	3.426	5.526	0.96
	3.8	6.579	2.798	0.97
	7.6	9.017	1.807	0.97
	15.2	9.434	1.686	0.97
A_{64}^6 (4x4x4)	0	3.952	4.693	0.61
	15.2	12.360	1.035	0.74
B_8^1 (2x2x2)	0	0.902	11.374	0.73
	1.3	1.905	8.756	0.75
	3.8	4.961	4.445	0.79
	7.6	7.391	2.845	0.82
	15.2	7.823	2.645	0.82
B_{27}^2 (3x3x3)	0	4.204	5.185	0.78
	1.3	5.677	3.869	0.80
	3.8	10.190	1.827	0.84
	7.6	13.693	1.137	0.86
	15.2	14.194	1.067	0.87
C_8^1 (2x2x2)	0	2.217	8.562	0.95
C_8^2 (2x2x2)	0	2.248	8.514	0.95
	1.3	3.678	6.295	0.95
	2.5	5.710	4.292	0.96
	3.8	8.207	2.843	0.96
	7.6	11.509	1.777	0.97
	15.2	11.986	1.669	0.97

TABLE IV (cont.)

C_{27}^{3} (3x3x3)	0	6.363	3.827	0.96
	1.3	8.574	2.683	0.96
	3.8	14.764	1.187	0.97
	7.6	18.720	0.776	0.98
	15.2	19.147	0.744	0.98
D_{8}^{1} (2x2x2)	0	3.543	6.806	1.18
	1.3	5.423	4.843	1.12
	3.8	11.532	1.976	1.09
	7.6	15.697	1.215	1.07
	15.2	16.378	1.130	1.07
D_{27}^{2} (3x3x3)	0	8.494	2.980	1.10
	1.3	11.323	2.025	1.09
	3.8	19.606	0.817	1.06
	7.6	24.498	0.531	1.05
	15.2	24.991	0.510	1.05

a. The letter and the superscript identify the average unit in the array described in Table III; the subscript is the number of units in the array; the numbers in parentheses are the horizontal and vertical dimensions, respectively, of the array expressed in number of units.

b. Errors on all surface separations are ± 0.013 cm for unreflected arrays and ± 0.026 cm for reflected arrays.

c. Array was subcritical with an apparent neutron source multiplication of ~ 3.

d. Array was subcritical with an apparent neutron source multiplication of ~ 10.

TABLE V

CRITICAL CONDITIONS FOR ARRAYS PARTIALLY ENCLOSED IN A REFLECTOR

Array Description[a]	Paraffin Reflector	Surface Separation of Units (cm)	Average Uranium Density in Array (g/cm^3)	Ratio of Array Height to $\sqrt{\text{Base Area}}$
C_{8}^{2} (2x2x2)	b	5.398 ± 0.013	4.538	0.96
C_{27}^{3} (3x3x3)	c	10.541 ± 0.013	2.028	0.97

a. The letter and the superscript identify the average unit in the array described in Table III; the subscript is the number of units in the array.

b. The dimensions of the base reflector were 76.2 x 76.2 x 15.2 cm and of the two sides were 76.2 x 45.7 x 15.2 cm.

c. The dimensions of the base reflector were 106.7 x 106.7 x 15.2 cm and of the two sides were 106.7 x 76.2 x 15.2 cm.

TABLE VI

COMPARISON OF URANIUM DENSITIES OF UNREFLECTED CUBIC AND RECTANGULAR PARALLELEPIPED ARRAYS

Array[a] Description	Centre Spacing[b] (cm)	Surface Spacing[b] (cm)		Average Uranium Density in Array (g/cm^3)	Ratio of Array Height to $\sqrt{\text{Base Area}}$	Ratio of Unit Height to Diameter
		Horizontal	Vertical			
A^2_{27} (3x3x3)	11.509	0	6.127	6.877[c]	1.00	0.47
A^2_{27}	-	2.007	2.007	7.767	0.55	0.47
B^1_8 (2x2x2)	11.494	0	3.417	10.334[d]	1.00	0.70
B^1_8	-	0.902	0.902	11.374	0.73	0.70
C^2_8 (2x2x2)	13.503	1.997	2.738	8.513	1.00	0.94
C^2_8	-	2.248	2.248	8.514	0.95	0.94
C^3_{27} (3x3x3)	17.602	6.118	6.837	3.828	1.00	0.94
C^3_{27}	-	6.363	6.363	3.827	0.96	0.94
D^1_8 (2x2x2)	15.778	4.269	2.319	6.675	1.00	1.17
D^1_8	-	3.543	3.543	6.806	1.13	1.17

a. The letter and the superscript identify the average unit in the array described in Table III; the subscript is the number of units in the array.

b. The error on all spacing values is ± 0.013 cm.

c. Array subcritical, maximum apparent source neutron multiplication ~ 70.

d. Array subcritical, maximum apparent source neutron multiplication ~ 81.

comparison, are also the dimensions of arrays of the same units located at the corners of rectangular parallelepipeds.

The arrays of A^2 and B^1 units in cubic pattern could not be made critical. As expected, arrays of C^2 and of C^3 units were critical at substantially the same density in both patterns since the units were of approximately equal height and diameter. The uranium density in the array of D^1 units at equal centre spacing, however, was less than that in the array at equal surface spacing.

The results suggest that, given a number of units, if the maximum achievable density with equal centre spacing is less than the critical density at equal surface spacing, the array at equal centre spacing cannot be made critical.

4.4. Unit shape

A brief study was made to determine the effect of changing the shape of the units on the critical density in eight-unit arrays. For one experiment, B^3 units were constructed from three 5.2 kg cylinders arranged coaxially, with one of smaller diameter (9.1 cm) between two larger one (11.5 cm). The critical density in both reflected and unreflected arrays of these irregularly shaped units was greater than that in arrays of regular B-units. In an unreflected array of C^5 units, formed with a piece 11.5 cm in diameter, between two smaller pieces 9.1 cm in diameter, the critical density was also larger than in the unreflected array of C^2 units. The densities of arrays of C^2 and C^5 units with a paraffin reflector 15.2 cm thick were approximately equal.

In another experiment the units were the C^6 units of Table III, having a mass of 21 kg and a height-to-diameter ratio of 1.90. The resulting critical densities of both the reflected and unreflected arrays were greater than those of arrays of cylinders with h/d ratio more nearly equal to unity. The data are compared in Table VII. Figure 3 shows the average uranium density as a function of the surface-to-volume ratio of the units. In each case the change in geometry of the unit resulted in an increase of the surface area

TABLE VII

COMPARISON OF CRITICAL DENSITIES FOR VARIOUS UNIT SHAPES

Array Description[a]	Paraffin Reflector Thickness (cm)	Surface Separation of Units[b] (cm)	Average Uranium Density in Array (g/cm^3)	Ratio of Unit Height to Diameter	Ratio of Array Height to $\sqrt{\text{Base Area}}$
B^1_8	0	0.902	11.374	0.70	0.72
	15.2	7.823	2.645	0.70	0.82
B^3_8	0	0.229	11.497		0.85
	15.2	6.904	2.792		0.90
C^2_8	0	2.248	8.514	0.94	0.95
	15.2	11.986	1.669	0.94	0.97
C^5_8	0	1.013	8.941		1.21
	15.2	10.945	1.668		1.12
C^6_8	0	1.466	10.002	1.90	1.77
	15.2	10.328	2.013	1.90	1.42

a. The letter and superscript identify the average unit in the array described in Table III; the subscript is the number of units in the array. The irregular shapes of the B^3 and C^5 units are also described in Table III.

b. The error in the separation of units in the unreflected arrays is ± 0.013 cm; in the reflected arrays it is ± 0.026 cm.

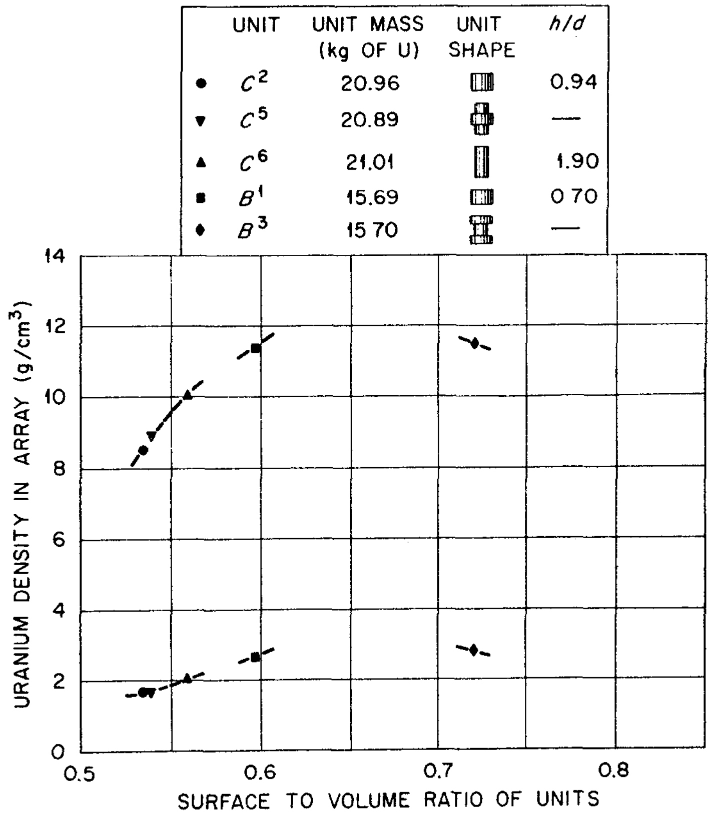

FIG. 3. Effect of the shape of units on the critical density in reflected and unreflected eight-unit arrays.

and a decrease in the value of k_{eff}. If the effect of array shape is neglected, these results indicate that reducing the k_{eff} of a unit in an array will require an increase in the array density to maintain criticality.

4.5. Array shape

The effect of changing the geometry of an array is similar to that observed for changing individual critical assemblies, i. e. a change in surface-to-volume ratio is accompanied by a change in mass to maintain criticality. The shape of a critical system of units may be altered in two ways, both requiring a compensating change in the array density. The array shape, r, may be changed either by assembling different numbers of units along its three directions or by altering the h/d ratio of the units within an array.

Examples of varying the array shape by altering the height-to-diameter ratio of the units are found in Table IV where any of the A^1 or A^2 arrays may be compared to the corresponding A^3 or A^4 arrays. In the comparable arrays there has been a substantial change in the values of h/d and of r with only a slight change in array density and an insignificant change in total mass present (less than 17 g per unit). The C^4 units of Table III were used to explore the effect on the critical uranium density for values of r differing significantly from unity. The critical conditions for the various arrangements are given in Table VIII with, for comparison, the critical densities interpolated from data of Table IV, in arrays having the same total number of units, if it would have been possible to arrange them with equal numbers

TABLE VIII

EFFECT OF ARRAY GEOMETRY ON CRITICAL URANIUM DENSITIES IN UNREFLECTED ARRAYS OF U(93.2) METAL UNITS

Array Description[a]	Surface Separation of Units[b] (cm)	Ratio of Array Height to $\sqrt{\text{Base Area}}$	Average Uranium Density in Arrays of Identical Units (g/cm^3)	
			Unequal Number of Units in Three Dimensions	Equal Number of Units in Three Dimensions[c]
C_8^4 (2x4x1)	1.062[d]	0.35	12.232	8.514
C_9^4 (3x3x1)	0.658	0.31	12.400	7.83
C_{18}^4 (3x3x2)	4.641	0.64	5.212	4.97
C_{16}^4 (2x2x4)	3.907	1.91	6.008	5.38
C_{16}^4 (2x4x2)	3.891	0.67	6.027	5.38
C_{16}^4 (4x4x1)	1.516	0.24	10.059	5.38
A_{16}^1 (2x2x4)	1.349	1.05	9.442	10.50
A_{45}^5 (3x3x5)	3.442	0.99	5.313	5.70

a. The letter and superscript identify the average unit in the array described in Table III; the subscript is the number of units in the array.

b. The error on the separation values is ± 0.013 cm.

c. Interpolated values from Table IV.

d. This array consisted of two clusters of four units each with lateral surfaces in contact. This dimension is the horizontal separation between the two clusters.

in each dimension. Arrays of A^1 and A^5 units, constructed to produce r values near unity, are also presented in Table VIII.

The results indicate that the array reactivity is more sensitive to changes in array shape than to changes in the shape of the units themselves.

4.6. Array moderation

The effect of hydrogenous material, placed between adjacent units, on the critical dimensions of arrays was examined in assemblies of units having an average mass of 20.9 kg. Boxes of several sizes and wall thicknesses, fabricated from Plexiglas and described in Table IX, were mounted on the rods supporting the uranium units. In each instance the unit was centred in its container. The data for the critical configurations appear in Table X.

An investigation was made of the effect on reactivity of the thickness of hydrogenous material separating adjacent units. A system of eight C^2 units, each in a P^3 box, assembled at an average density of 1.189 g/cm^3

TABLE IX

DIMENSIONS OF CONTAINERS FOR UNITS IN MODERATED ARRAYS

Container Designation	Material	Wall Thickness (cm)	Outside Dimensions (cm)	
			Base	Height
P^1	Plexiglas	0.64	12.9 x 12.9	12.1
P^2	Plexiglas	0.64	15.6 x 15.6	14.8
P^3	Plexiglas	1.27	17.9 x 17.9	17.2
P^4	Plexiglas	2.38	21.4 x 21.4	20.7
S^1	Iron*	0.66	14.1 diam.	13.2

*5-in. Schedule 40 iron pipe provided with end plates of thickness equal to the pipe wall.

and surrounded by a 15.2 cm thick paraffin reflector, was subcritical. The reactivity of the array increased as the thickness of the container walls separating the units was increased until the total thickness of the Plexiglas was 4.9 cm. Further increase reduced the reactivity. The detailed results of the experiments are shown in Fig. 4 where the reactivity of the array is expressed as a function of the Plexiglas thickness, including the walls of the P^3 boxes, separating the units.

The uranium density as a function of the total thickness of Plexiglas between adjacent units in the eight unit arrays surrounded by reflectors of various thickness are given in Fig. 5. The points shown at 3 and 7 cm on the thick paraffin reflector curve were obtained from Fig. 4. The addition of a 15.2 cm thick paraffin reflector to an unmoderated array reduced the critical density by a factor of about five; the insertion of a 4.8 cm thick Plexiglas moderator around the units of an otherwise unreflected array reduced the critical density by a factor of four. It is emphasized that this added moderator surrounded each unit and, consequently, introduced hydrogenous material into the reflector region. It may be observed, however, that simultaneous addition of a thick reflector and optimum moderator reduced the critical density to only 1/8 of that of the unreflected, unmoderated array. It is clear that the separate effects do not combine directly.

4.7. Mixed arrays

A brief but important exploration was conducted to determine the effect on array reactivity of varying the geometry or mass of a single unit in a critical array and of combining portions of different critical arrays. In one experiment the central unit of the critical array C_{27}^{3} [0; 6.363; 3.827; 0.96]

TABLE X

CRITICAL CONDITIONS FOR MODERATED ARRAYS OF 20.9 kg UNITS

Description of Array[a]	Paraffin Reflector Thickness (cm)	Surface Separation of Units[b] (cm)	Average Uranium Density in Array (g/cm^3)	Ratio of Array Height to $\sqrt{\text{Base Area}}$
$(C^2 \rightarrow P^1)_8$	0	4.082	5.810	0.95
	15.2	12.662	1.532	0.97
$(C^2 \rightarrow P^2)_8$	0	4.239	5.635	0.95
	1.3	5.875	4.170	0.96
	7.6	12.573	1.549	0.97
	15.2	12.929	1.482	0.97
$(C^2 \rightarrow P^3)_8$	0	6.619	3.670	0.96
	1.3	8.611	2.673	0.96
	15.2	14.503	1.226	0.97
$(C^2 \rightarrow P^4)_8$	0	10.239	2.110	0.97
	15.2	16.447	0.986	0.97
$(C^3 \rightarrow P^4)_{27}$	0	16.289	1.000	0.97
$(C^2 \rightarrow S^1)_8$	0	3.239	6.884	0.95
$(C^2 \rightarrow S^1 \rightarrow P^2)_8$	0	5.169	4.731	0.96

a. The first letter and superscript identify the average unit in the array described in Table III; the second letter and superscript identify the container (Table IX) in which each unit was centered; the subscript is the number of units in the array.

b. The error in the separation of the units in the unreflected arrays is ± 0.013 cm; in the reflected arrays it is ± 0.026 cm.

was replaced by a D^2 unit without a change in the lattice cell volume. The substitution of a unit having both a larger mass and a greater k_{eff} produced an array reactivity increase in excess of 1.5 $. In the second experiment, the central unit of the critical array B^2_{27} [0; 4.204; 5.185; 0.78] was replaced by a B^4 unit and the cell volume maintained. Although the uranium content of the B^4 unit was 124 g more than that of the B^2 unit, its k_{eff} was less and the replacement resulted in a decrease of about 5 cents in the array reactivity.

Additional experiments used parts of critical eight-unit systems to illustrate the effect of multiple component replacement. In these experiments one half each of two different critical arrays were brought together along a common centre line until their cell boundaries coincided. The units of one of the critical arrays were right circular cylinders of aqueous uranyl nitrate solution contained in 0.64 cm thick Plexiglas vessels 20.32 cm

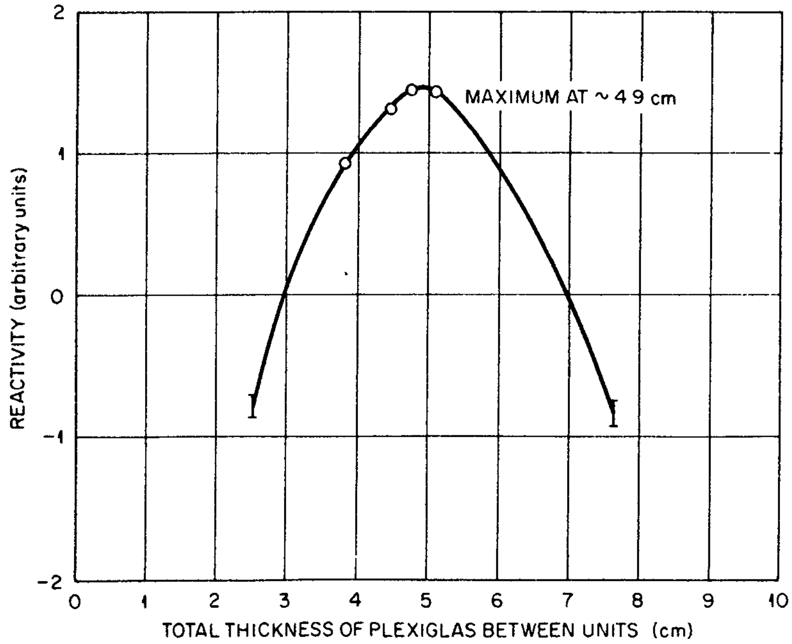

FIG. 4. The effect on reactivity of varying the thickness of Plexiglas between the 20.9 kg metal units of a paraffin-reflected eight-unit array.

o.d. and 19.05 cm in outside height. Each unit[4] contained 2.07 kg of uranium enriched to 92.6 wt.% ^{235}U at an H/^{235}U atomic ratio of 59.

Three assemblies of mixed units were attempted. In one, four C^2 units of the critical array C_8^2 [0; 2.248; 8.514; 0.95] were assembled with four D^1 units of the critical array D_8^1 [0; 3.542; 6.806; 1.18]. In another, four of the solution units at their critical spacing were mated with four units of the critical array of C_8^2 units; the composite array is shown in Fig. 6. In a third, four of the solution units were combined with one half the critical array C_8^6 [0; 1.466; 10.002; 1.77]. Each of the composite arrays was more than one dollar subcritical. The array of solution units and C^2 units was made critical by reducing the spacing between the C^2 units from 2.248 to 1.689 cm.

5. SERIES III: $U(92.6)O_2(NO_3)_2$ AQUEOUS SOLUTIONS

The units utilized in this series of experiments are described in Table XI for convenience. The volume of solution present in each of the units was carefully adjusted to 5.000 litres by weighing to ± 0.5 g. The ^{235}U content of the uranium was 92.6 wt.%.

The data describing regular three dimensional arrays are presented in Table XII. A majority of the experiments was performed with the F^1 units of solution having an H/ ^{235}U ratio of 59. A limited number of experiments were performed with more dilute solutions.

The behaviour of the arrays parallels that of individual critical units with respect to variations in concentration. Each of the critical arrays F_8^1

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[4] See section 5 for a complete description of critical conditions of arrays of these units.

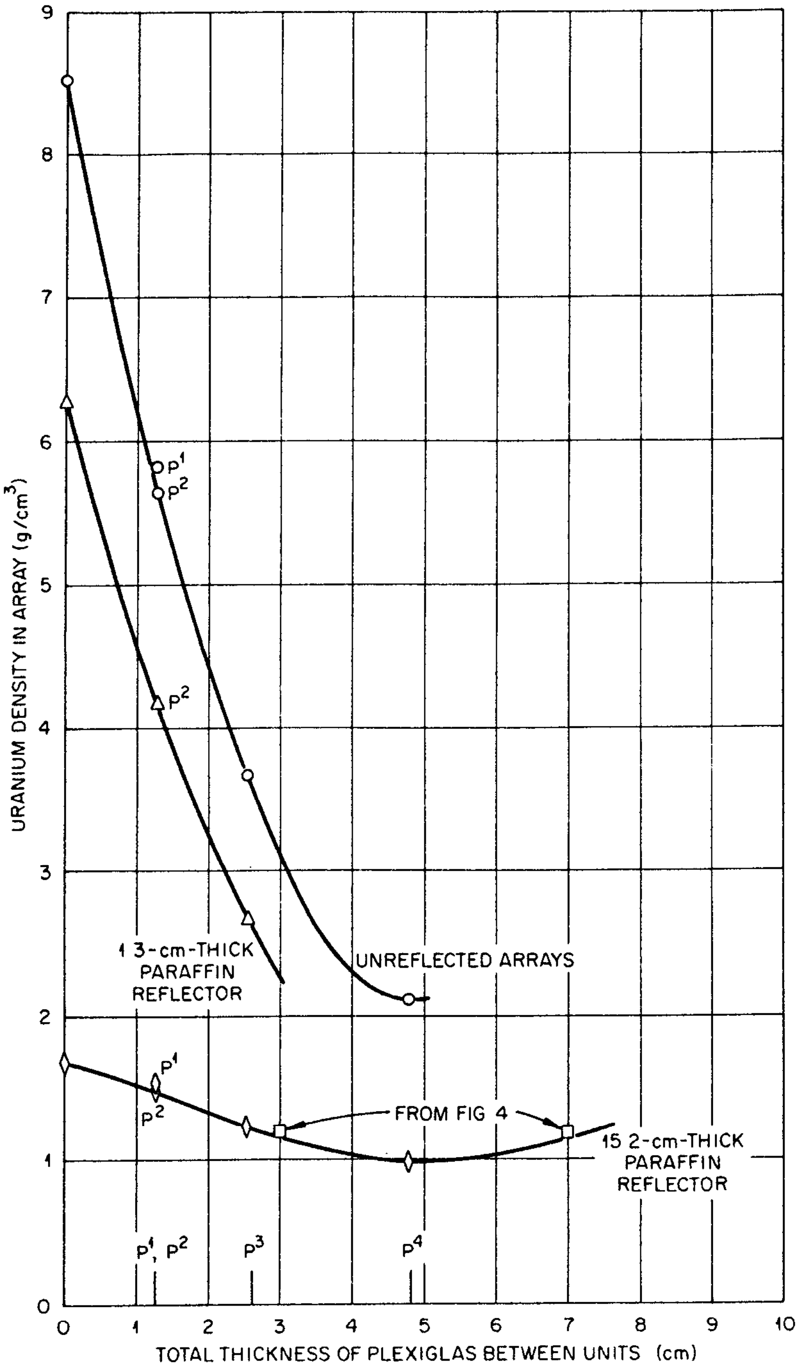

FIG. 5. The effect of Plexiglas as a moderator and paraffin as a reflector on the critical density of an eight-unit array of 20.9 kg units.

[0; 1.43; 0.214; 0.94] and F_8^2[0; 1.43; 0.144; 0.94] contained solutions of different uranium concentration although they had the same lattice volume, and the same total volume, within the uncertainty of the measured separation distance. The total mass present, however, differed by about 33%. The H/^{235}U ratio of the solution constituting these arrays was within a range including the concentration at which the minimum critical volume of an unreflected individual unit occurs. Within this range only slight variation in the critical volume is observed, although the variation of critical mass

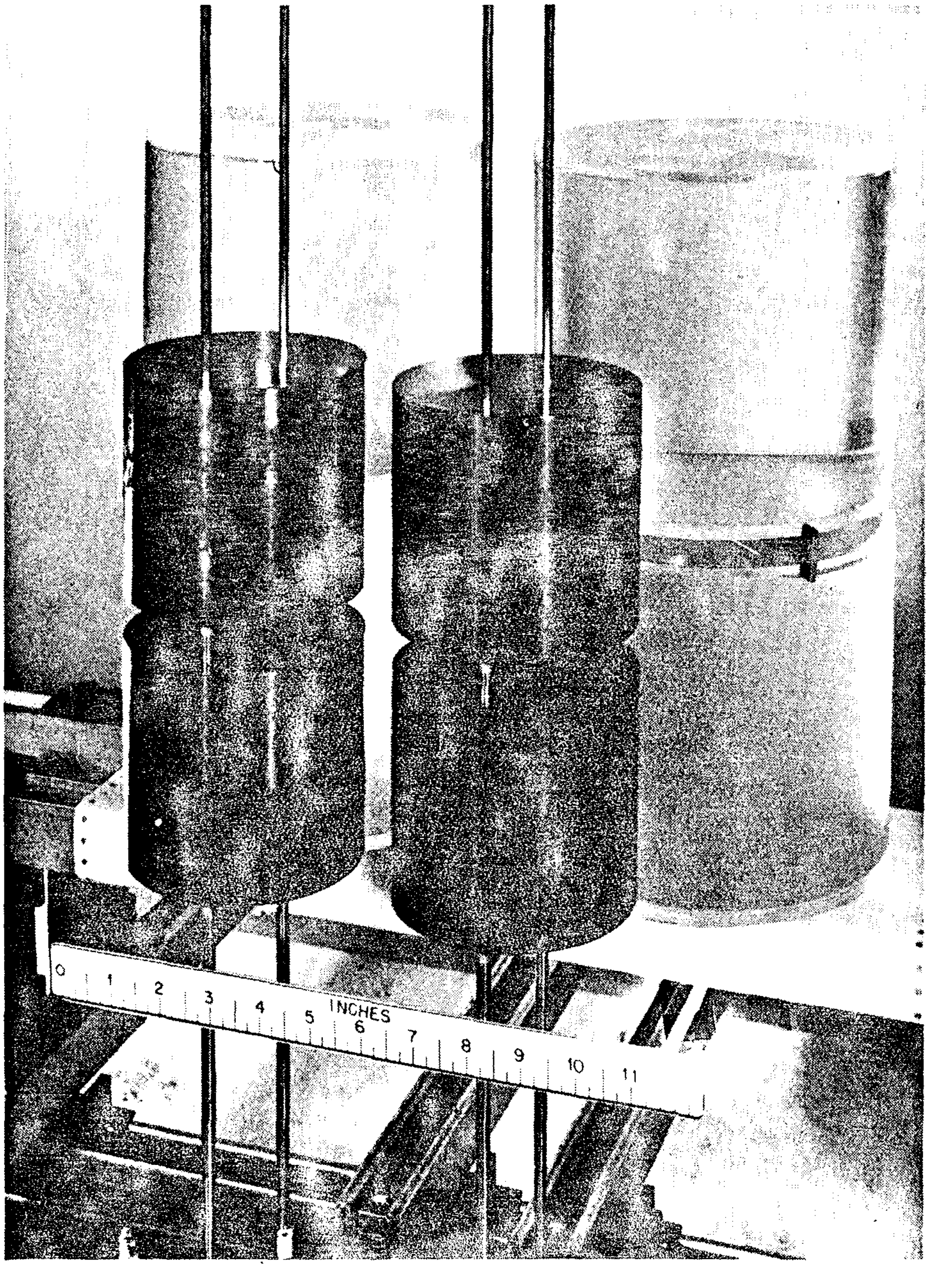

FIG. 6. Composite array of four 20.9 kg U(93.2) metal units and four 2.1 kg U(92.6) units of $UO_2(NO_3)_2$ solution.

is comparable to the difference observed in the arrays. The specific reactivity of an F^2 unit in arrays is, nevertheless, smaller than that of an F^1 unit. This observation was verified by comparing the critical array $F^{1,2}_{27}$ [0; 6.41; 0.107; 0.95] to F^{1}_{27}[0; 6.48; 0.114; 0.95] where it is shown that a decrease in spacing was required when five of the F^1 units in the latter array were replaced by F^2 units.

The specific reactivity of a unit in an array is further reduced by decreasing its uranium concentration as shown by the eight and twenty-seven unit arrays of F^3 units.

5.1. Some planar arrays

Several other experiments were performed with two dimensional arrays of solution at a concentration of 415 g(U)/l. Nineteen units arranged in a

TABLE XI

DESCRIPTION OF FIVE LITRE UNITS CONSTITUTING ARRAYS

Containers: 0.64-cm-thick Plexiglas cylinders 20.32 cm o.d. and 19.05 cm outside height[a]

Unit Designation	Aqueous Solution			
	Concentration g(U)/litre	Specific Gravity	Atomic Ratio $H/^{235}U$	Uranium Mass, kg
F^1	415	1.555	59	2.074
F^2	279	1.373	92	1.395
F^3	63.3	1.083	440	0.316

a. Content of each unit was 5.000 ± (3 x 10^{-4}) litres determined by weighing to ± 0.5 g.

single tier with their centres in a triangular pattern were critical, unreflected, at a unit surface separation of 1.35 cm. It was observed that 16 units in a single tier, in contact, arranged in a square pattern, and nonreflecting were subcritical with an apparent source neutron multiplication of approximately 6. Four units in a single tier, square pattern, with a surface separation of 3.94 cm were critical when surrounded by a 15.2 cm thick paraffin reflector at the cell boundaries.

6. SERIES IV: $U(4.9)O_2F_2$ AQUEOUS SOLUTION[5]

The experiments reported in this section were performed with uranyl fluoride solution in which the ^{235}U content of the uranium was 4.9 wt.%. The concentration, as in the series with U(92.6) solutions, was chosen to be as near that estimated to result in a minimum critical volume for an unreflected individual unit as solubility permits. Aluminium cylinders 24.1 cm i.d. by 152.4 cm in height, having a wall and a bottom thickness of 0.32 and 0.64 cm, respectively, were used as solution containers. The effect of unit variation on the criticality of arrays was studied by filling the cylinders to various depths. A description of the average units used in the arrays assembled is given in Table XIII. The physical properties of the polyethylene, used as a reflector, and of the Plexiglas, used as a moderator, are given in section 2.2.

The critical conditions for unreflected two dimensional, or planar, arrays are given in Table XIV. Although only planar arrays were assembled, significant variation of unit h/d ratio, of array pattern and of array shape

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[5] This series of experiments was conducted by Johnson [4,5].

TABLE XII

CRITICAL CONDITIONS FOR REGULAR THREE DIMENSIONAL ARRAYS OF $U(92.6)O_2(NO_3)_2$-FIVE LITRE SOLUTION UNITS WITH VARIOUS PARAFFIN REFLECTORS

Array Description	Paraffin Reflector Thickness (cm)	Surface Separation of units[b] (cm)	Average Uranium Density in Array (g/cm^3)	Ratio of Array Height to $\sqrt{\text{Base Area}}$
F_8^1 (2x2x2)	0	1.43	0.214	0.94
	1.3	3.28	0.167	0.95
	3.8	6.91	0.108	0.95
	7.6	8.48	0.091	0.96
	15.2	8.99	0.087	0.96
F_{27}^1 (3x3x3)	0	6.48	0.114	0.95
	1.3	9.02	0.086	0.96
	15.2	16.53[c]	0.043	0.96
F_{64}^1 (4x4x4)	0	10.67	0.072	0.96
F_{125}^1 (5x5x5)	0	14.40	0.052	0.96
F_8^2 (2x2x2)	0	1.43	0.144	0.94
	11.4[d]	8.71	0.060	0.96
F_{27}^2 (3x3x3)	0	6.40	0.077	0.95
F_8^3 (2x2x2)	0	0.0[e]	0.040	0.94
F_{27}^3 (3x3x3)	0	2.41	0.029	0.95
$F_{27}^{1,2}$ (3x3x3)[f]	0	6.41	0.107	0.95

a. The letter and the superscript identify the average unit in the array described in Table XI; the subscript is the number of units in the array; the numbers in parentheses are the horizontal and vertical dimensions, respectively, of the array expressed in number of units.

b. The uncertainty in the values of the separation is ± 0.13 cm.

c. The separation was 16.91 cm where one face of the array was reflected by Plexiglas 15.2-cm-thick.

d. The array was reflected on the bottom by 15.2-cm-thick paraffin.

e. Array subcritical $k_{eff} \sim 0.6$.

f. Five control units in centre tier are F^2 units and remaining 22 units are F^1.

are presented. Figure 7 is a photograph of the critical array L_{19}^3[0; 14.75; 0.276; 0.98].

The container dimensions prohibited construction of arrays completely surrounded by a reflector at the cell boundaries. In one experiment, however,

TABLE XIII

DESCRIPTION OF UNITS CONSTITUTING ARRAYS WITH $U(4.9)O_2F_2$ SOLUTIONS

Aluminium containers: 24.1 cm i.d.; 0.32 cm lateral wall thickness;
0.64-cm-thick bottom; 152.4 cm high
Solution: $U(4.9)O_2F_2$; sp.gr. 2.0201; 901.38 g of U per litre
$H/^{235}U$ = 496.6

Unit Designation	Solution Height (cm)	h/d Ratio	Volume of Solution (litres)	Uranium Mass (kg)
L^1	61.0	2.53	27.826	25.082
L^2	122.0	5.06	55.652	50.164
L^3	142.2	5.90	64.867	58.470

a 15.2 cm thick polyethylene reflector was placed, at the cell boundaries, on the lateral surfaces of a 3 × 3 × 1 square array. The unit surface separation was 11.94 cm and criticality was achieved when the solution depth in three control cylinders, constituting a centre row, was 132.6 cm. The remaining six cylinders had a solution depth of 142.2 cm, the L^3 unit of Table XIII.

In another experiment, the thickness of Plexiglas for optimum array moderation of these units was determined. An array of nine L^3 units, in a square pattern, spaced at 15.46 cm with a 15.2 cm thick polyethylene reflector on the four lateral array boundaries was subcritical in the absence of a moderator. Variations in the thicknesses of Plexiglas centred between the units produced critical arrays with different solution heights in the three control cylinders. The results are shown in Fig. 8 where the control cylinder solution depths, normalized to the depth with 1.3 cm thickness, are given as a function of the Plexiglas thickness between adjacent units. The broad minimal portion of the curve shows that 1.5 cm thick Plexiglas produces optimum moderation of the array of these units.

7. CALCULATIVE METHODS

No review of the various methods of computing the criticality of arrays is required to reveal that those methods limited to the neutronics of individual subcritical units and relying on geometric proportioning of leakage neutrons are inadequate to cope with reflection, moderation and other array modifications. For this reason the application of Monte Carlo codes, or methods treating the neutronics of system criticality rather than of subcritical units, are preferred. Two Monte Carlo codes and an analytic code have been applied to a representative group of experiments from the series in this paper.

TABLE XIV

CRITICAL CONDITIONS FOR UNREFLECTED PLANAR ARRAYS OF $U(4.9)O_2F_2$ SOLUTION UNITS

Array Description[a]	Array Pattern	Unit Surface Separation[b] (cm)	Average Uranium Density in Array (g/cm^3)	Ratio, Array Height to $\sqrt{\text{Base Area}}$
L_7^1	Tri.	1.89	0.649	0.96
L_9^1	Sq.	1.32	0.593	0.80
L_{16}^1	Sq.	3.62	0.492	0.57
L_{25}^1	Sq.	5.24	0.421	0.44
L_5^2	Tri.	1.37	0.689	2.27
L_7^2	Tri.	4.44	0.538	1.76
L_{19}^2	Tri.	13.23	0.297	0.88
L_9^2	Sq.	4.85	0.452	1.43
L_{16}^2	Sq.	9.16	0.333	0.97
L_{25}^2	Sq.	12.42	0.270	0.72
L_5^3	Tri.	1.47	0.684	2.63
L_7^3	Tri.	5.11	0.514	2.00
L_{19}^3	Tri.	14.75	0.276	0.98
L_9^3	Sq.	5.61	0.429	1.62
L_{16}^3	Sq.	10.44	0.310	1.08
L_{25}^3	Sq.	14.12	0.248	0.80

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[a] The letter and superscript identify the average unit in the array described in Table XIII. The subscript is the number of units in the array.

[b] The uncertainty in the separation values is ± 0.08 cm.

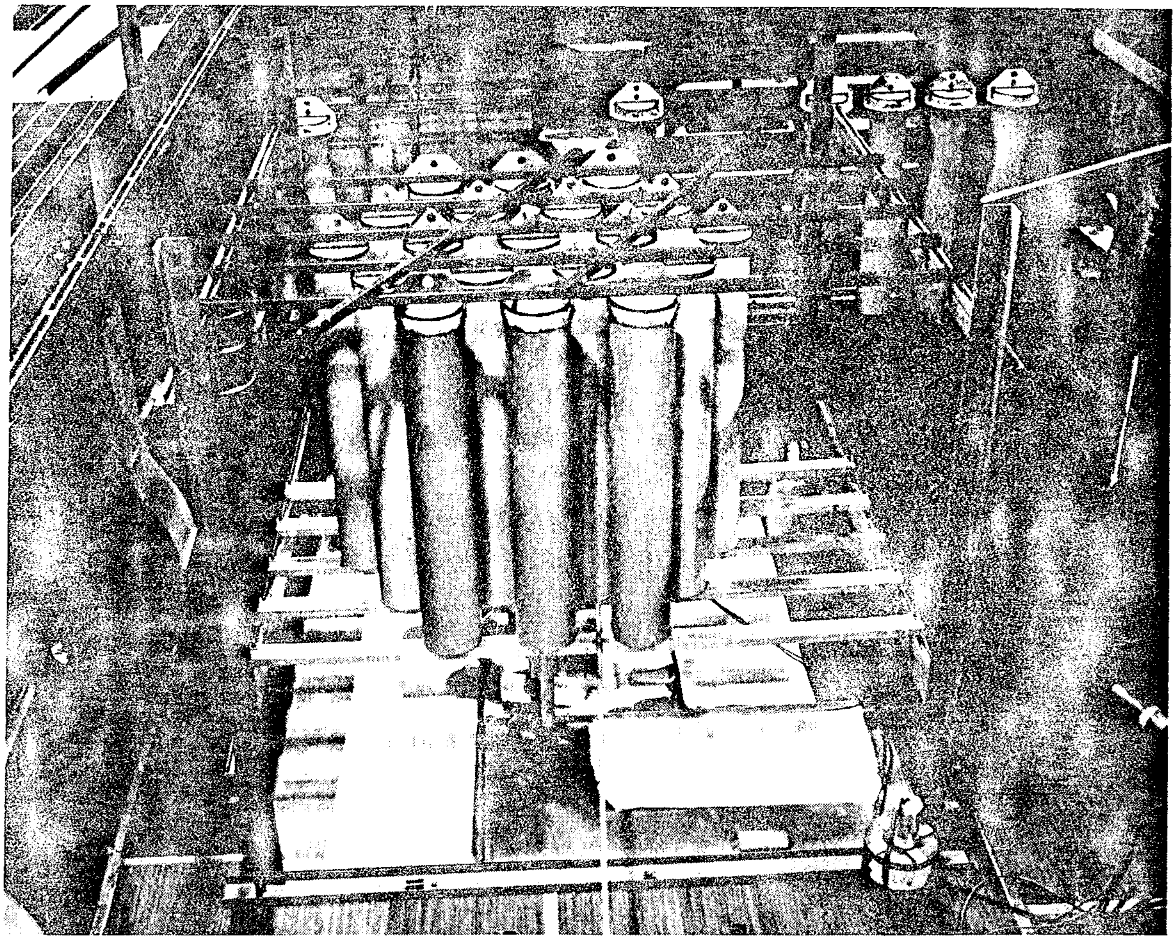

FIG. 7. A view of the unreflected nineteen-unit array of $U(5)O_2F_2$ solution in a triangular pattern. Each unit contains 58.5 kg of uranium.

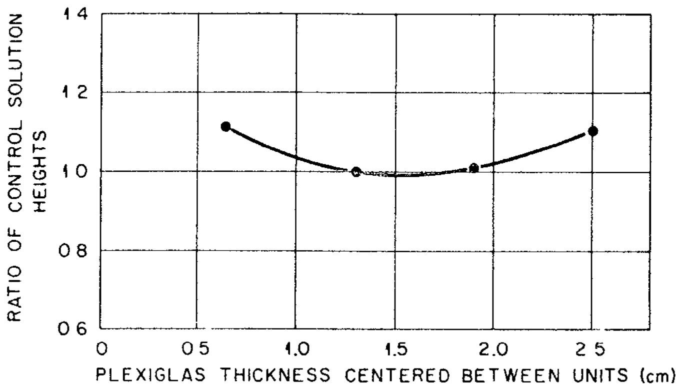

FIG. 8. Effect on critical solution height in three control cylinders of varying the thickness of Plexiglas between 58.5 kg uranium solution units of a polyethylene-reflected nine-unit array.

One of the Monte Carlo codes is GEM[6], a neutronics code written for the IBM-7090 computer. The GEM programme input is a simple description of the material, unit, cell and reflector. The system is divided in two by

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[6] Private communication from Roskell and Hemmings of the Authority Health and Safety Branch, U.K.A.E.A.; see also Ref. [10].

a neutronically important surface separating a 'core' from a 'reflector' which may not necessarily correspond to the true core-reflector boundary. In the programme neutron tracking is done by stages, where a stage begins with the passage of a preselected number of neutrons into the core and terminates when all of the descendants of those neutrons re-enter the core. The calculation provides the ratio of the neutron population at the boundary at the end of a stage to the population at the beginning. This ratio, together with neutron accountings made at the boundaries during tracking, provides estimates of k_{eff} and other properties such as spectra and fluxes.

The second code is the O5R, a general purpose Monte Carlo neutron transport programme written for the IBM-7090 and the CDC-1604A computers [6]. Unlike GEM, the geometric input here is complex, requiring the specification of all surfaces in the array. The programme calculates the fission distribution from a batch of a preselected number of neutrons with a specified distribution in space. The resulting distribution is then assigned to the succeeding batch of neutrons and a new distribution calculated. This procedure is repeated for the desired number of batches. The multiplication factor is obtained by calculating the ratio of the number of neutrons produced in each batch to the number of source neutrons, i.e. k_{eff} for each generation and then averaging it over all the batches. In this method a matrix of probabilities that a neutron in one region will cause a fission in another region is used to determine when the effects of the assumed initial source distribution have disappeared. Only subsequent batches are used in computing the multiplication factor. The output of O5R is, in addition to the k_{eff}, a history of all the collisions from which the spectra, fluxes, and other measurable quantities may be obtained. In the application of the programme to the experiments considered here, a simpler treatment utilizing monoenergetic neutrons and assuming isotropic scattering has been used. Mihalczo [9] has shown that when only the multiplication factors are to be calculated this treatment is reliable for a single material in unreflected, complicated geometry.

The third code is an analytic programme prepared by Clark [7] for an IBM-704 computer and has been described as a practical method for computing neutron interaction in groups of fissionable units. The approximations made to simplify the calculations sufficiently characterize it and are the following. A one-group spatial distribution of neutrons is assumed to satisfy the wave equation. The unit and cell are replaced by a sphere and by a cube, respectively, of the same volumes, retaining thereby the uranium density of the experimental array. In the calculation of reflected arrays the reflector is assigned the actual dimensions of the experiment and an albedo. The value of the albedo is that of an infinite slab, having the thickness of the reflector used, on an infinite slab core of fissionable material with the same composition as the units. The emitted and incident neutron currents in the array are treated as though they were uniform over the entire surface of each unit. The angular distribution of emitted neutrons is assumed proportional to the cosine of the angle between the direction of emission and the normal to the emitting surface element. The extent of the array considered in the calculation is limited to those units, which, either by complete or partial shadowing, intercept all emergent neutrons. A boundary condition employed is that the incoming neutron current for an unreflected isolated unit is zero in order to express the total transport cross-section in terms of an extrapolation distance which is consistent with the unreflected critical size and material buckling of an individual unit having the same composition as a unit in the array. The programme computes the maximum eigenvalue of a set of homogeneous equations for the neutron currents as a function of the spacing and of the reflector albedo.

A display of typical results from the application of these codes to the experiments of these series is presented in Table XV. The group of experiments represents a wide variation in both unit and array properties.

8. CONCLUSIONS

Regular three-dimensional arrays may be characterized as low density individual critical systems. The similarity is apparent when the data are expressed graphically as total mass versus the average uranium density in the array. When the data are examined in this manner it is immediately evident that the effect of a reflector on an array depends strongly on the energy of the leakage neutrons. Although this observation is not surprising, the magnitude of the factors by which the reflector reduces the critical number of units and the range of these factors is important to the handling of fissile materials. The factors observed in these experiments were about 13 for the metal units, 7 for the U(93) solution units and less than 3 for the U(4.9) solution units. This enhanced reflector effect, compared to that occurring for individual critical units, can be associated with the relatively high neutron leakage through the area between the units in an array.

The effect of partial reflection by a thick reflector, on the other hand, is relatively small, and appears to not violate the usual factors of safety.

Unit shape has less effect on array reactivity than does array shape. Large changes in the reactivity of unreflected arrays may be accomplished by altering the unit shape, but these reactivity changes are greatly diminished by the addition of a reflector.

The observed effect of array moderation by hydrogenous materials is due to a combination of neutron energy degradation, neutron scattering and leakage, and neutron absorption. An upper limit of the factors by which the critical number of units in an unmoderated array is reduced is that observed for the metal systems. The factor was about 4 in the absence of a reflector and about 2 with a reflector.

The data from the mixed arrays appear to be a demonstration of the contrapositive of the result derived by Thomas and Scriven [11] for pairs of dissimilar containers of fissile materials in air. In a mixed critical array consisting of equal portions of two other regular critical arrays, each composed of identical units, the separation of adjacent unlike units is less than the geometric mean of the separations of the like units — a result in keeping with the concept of arrays as individual low density systems.

The good agreement between the results of experiments and of calculations suggests that calculative techniques may have reached a stage where the reliability of their results is suitable for application to safety evaluation problems.

TABLE XV

COMPARISON OF THREE CODES FOR CALCULATION OF MULTIPLICATION FACTOR FOR SOME CRITICAL SYSTEMS

Array Description	GEM	05R[a]	H. K. Clark [7,8]
A^{6}_{64} [0; 3.952; 4.693; 0.61]	1.005	0.971	--
A^{2}_{27} [0; 2.007; 7.767; 0.55]	1.027	1.004	--
B^{1}_{8} [0; 0.902; 11.374; 0.73]	1.012	0.975	--
B^{3}_{8} [0; 0.229; 11.497; 0.85]	1.038	1.010	--
B^{2}_{27} [15.2; 4.204; 5.185; 0.78]	1.013	--	--
C^{2}_{8} [0; 2.248; 8.514; 0.95]	1.019	--	0.959
C^{2}_{8} [15.2; 11.986; 1.669; 0.97]	1.017	--	1.102
C^{3}_{27} [0; 6.363; 3.827; 0.96]	1.023	0.995	0.987
C^{3}_{27} [15.2; 19.147; 0.744; 0.98]	1.027	--	1.079
C^{4}_{16} [0; 1.516; 10.059; 0.24][b]	1.018	0.993	--
C^{4}_{16} [0; 3.891; 6.027; 0.67][c]	1.023	0.997	--
$(C^{2}\rightarrow S^{1})_{8}$ [0; 3.239; 6.884; 0.95]	1.029	--	--
$(C^{2}\rightarrow S^{1}\rightarrow P^{2})_{8}$ [0; 5.169; 4.731; 0.96]	1.031	--	--
D^{2}_{27} [0; 8.494; 2.980; 1.10]	1.017	--	0.991
D^{2}_{27} [3.8; 19.606; 0.817; 1.06]	1.014	--	0.972
F^{1}_{8} [0; 1.43; 0.214; 0.94]	1.006	--	--
F^{1}_{8} [15.2; 8.99; 0.087; 0.96]	1.021	--	1.051
F^{1}_{64} [0; 10.67; 0.072; 0.96]	--	--	1.051
L^{3}_{16} [0; 10.44; 0.310; 1.08]	1.001	--	--
L^{3}_{9} [15.2; 11.94; 0.282; 1.40][d]	1.001	--	--

a. Program utilized monoenergetic neutrons and isotropic scattering.

b. Units were arranged as (4x4x1).

c. Units were arranged as (2x4x2).

d. The reflector is on the 4 lateral surfaces of the array and the solution height in the three control cylinders, constituting a centre row, is 132.6 cm.

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DISCUSSION

R. CAIZERGUES: How many initial neutrons are assumed for these calculations?

J.T. THOMAS: In the Monte Carlo calculations with the O5R code we use typical batches of 400 neutrons, the history of each batch being completed before the next is introduced. This process can be repeated as often as desired, but usually 30 to 40 batches are adequate.

J.-M. MOREAU: In Table XV, where you give the results of calculations for various configurations, the variations from unity seem rather large (almost 4%). Do you really get unity for spherical critical units of the same material — in other words are these discrepancies due essentially to the geometry? I am rather surprised, because in the experimental cases we have calculated, the discrepancies have never exceeded the statistical error.

J.T. THOMAS: I omitted to mention the statistical limits on the values given. The number of neutrons tracked in the GEM calculations average 5000 per problem; the confidence interval for these values is accordingly not very good. However, the requirements of nuclear safety did not seem to justify further expenditure to improve the results in all the cases mentioned.

I might say that, with the O5R code, accuracy to within ± 1% has been achieved many times with only 12 000 neutron histories. I am not acquainted with the accuracy of Mr. Clark's results. To answer your question more specifically, however, the discrepancies are clearly not related to geometry.

W. SCHÜLLER: Given the large amount of experimental data now available on interacting or rays, do you consider it reasonable to maintain the safety factor of 5 applied in transport regulations — particularly in view of the fact that data for large numbers of units must be obtained by extrapolation?

J.T. THOMAS: Personally I feel that the factor of 5 ought to be reviewed. In any case it should be made clear that this factor is not necessarily what nuclear safety specialists would recommend; rather, it seems to reflect an opinion held by the transport specialists of several countries.