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CRITICAL THREE-DIMENSIONAL ARRAYS OF NEUTRON-INTERACTING UNITS

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ABSTRACT

As many as 125 five-liter units of concentrated aqueous uranyl nitrate solution were assembled in critical arrays. The solution, at a concentration of 415 g of uranium per liter, and having a specific gravity of 1.555, was contained in right circular cylinders of methacrylate plastic having a 0.64-cm-thick wall. The U^{235} content of the uranium was 92.6 wt%. The dependence of the number of units required for criticality as a function of the spacing has been determined. The critical number, N, as a function of spacing, within the range of these experiments, has been determined to be $N = N_0(\rho/\rho_0)^{-s}$, where N is the number of units in the critical array, ρ and ρ_0 are, respectively, the uranium density in the array and in the unit, and N_0 and s are constants depending upon the neutron reflector surrounding the array. For arrays with no reflector N_0 and s are 2.506 ± 0.014 and 1.928 ± 0.028. Experiments with 8 and 27 unit arrays at lower U^{235} concentration indicate that the value of s increases and that of N_0 decreases with decreasing concentration.

CRITICAL THREE-DIMENSIONAL ARRAYS OF NEUTRON-INTERACTING UNITS

INTRODUCTION

The experimental program at the Oak Ridge Critical Experiments Facility investigating the dimensions of critical arrays of individually subcritical units has been extended to include arrays of cylindrical volumes of aqueous solutions of enriched uranium having their height and diameter essentially equal. Although many data from experiments of this type with solutions have been previously reported,[1,2,3] they were derived, primarily, from critical arrays of identical cylinders of solution arranged so that their bases were coplanar. In some of these earlier experiments the height-to-diameter ratio of the individual units has been far from unity. In addition there have been critical experiments with slab-shaped volumes of solution[4] and with two-component arrays, one solid and the other liquid.[5] Although the data are generally and empirically applicable to nuclear safety problems, the earlier arrays are not amenable to calculational or even to broad empirical generalization owing to extremes in the geometry of the component units and to impossibilities of precise descriptions of their nuclear properties. The experiments reported here were designed to avoid some of these deficiencies. The units were compact, were regularly spaced in three dimensions in most instances, and were supported on a structure of low mass and of low neutron-absorbing materials. In addition to the number of units in the arrays and their spacing, the variables included the chemical concentration of the solution and the thickness of the hydrogenous reflector surrounding the array. Some of the data from this series of experiments have been reported previously.[6]

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1. D. Callihan, *et al.*, "Critical Mass Studies, Part IV," K-406 (Nov. 1949).
2. J. K. Fox, L. W. Gilley and D. Callihan, "Critical Mass Studies, Part IX, Aqueous U^{235} Solutions," ORNL-2367 (Feb. 1958).
3. L. W. Gilley, *et al.*, "Critical Arrays of Neutron Interacting Units," Neutron Phys. Div. Ann. Prog. Rept. Sept. 1, 1961, ORNL-3193, p. 159.
4. J. K. Fox and L. W. Gilley, "Critical Parameters of Aqueous Solution of U^{235}," Neutron Phys. Div. Ann. Prog. Rept. Sept 1, 1957, ORNL-2389, p. 77.
5. J. K. Fox and L. W. Gilley, "Critical Dimensions of Neutron-Interacting Slabs of Dissimilar Materials," ORNL-TM-494 (April 1963).
6. J. T. Thomas and J. K. Fox, "Critical Cubic Arrays of Neutron-Interacting Units of Aqueous Uranyl Nitrate Solution," Neutron Phys. Div. Ann. Prog. Rept. Sept. 1, 1962, ORNL-3360, p. 37.
   J. T. Thomas, TRANS. Amer. Nuc. Soc. 6, 169 (1963).

MATERIALS AND METHOD

Each subcritical unit was a right circular cylinder of aqueous uranyl nitrate solution contained in 0.64-cm-thick methacrylate plastic (Plexiglas, density 1.18 g/cm^3) 20.32 cm in outside diameter and 19.05 cm in outside height. The volume of each of the units was carefully adjusted to 5.000 liters by weighing to ±0.5 g. Solutions having concentrations of 415, 279 and 63.3 g of U per liter, having, respectively, a specific gravity of 1.555, 1.373 and 1.083, were used in these experiments. The corresponding U^{235} content of a five-liter volume was 1.921, 1.292 and 0.293 kg. The total nitrate in the solution corresponded to an N:U^{235} atomic ratio of 2.006. There were no other impurities in significant quantities. The U^{235} content of the uranium was 92.6 wt%.

Paraffin (density 0.93 g/cm^3) and Plexiglas (a methacrylate plastic having a density of 1.18 g/cm^3) were used as hydrogenous reflectors surrounding the arrays and were spaced from the outer boundaries of an array a distance equal to one half the surface-to-surface separation of the units.

Criticality was achieved by filling some containers in the array by a remotely operated solution-handling system. In the larger arrays as many as five containers, located near the center of the lattice, were filled in this manner after having been assembled with the desired number of previously filled ones.

Figure 1 shows 125 units in an unreflected array which is nearly cubic both in its outline and in its lattice structure. The units are held in position on the frame of aluminum Unistrut by bolted lugs. The central location of the five control units, typical of the larger arrays, may be identified by the connecting polyethylene tubing. Figure 2 illustrates a typical Plexiglas-reflected array of 27 units with part of the reflector removed.

EXPERIMENTAL RESULTS

Although most of the results reported here describe arrays in which the solution concentration was 415 g of uranium per liter (corresponding to an H:U^{235} atomic ratio of 59) a few definitive experiments were performed with solutions containing 279 and 63.3 g of uranium per liter (H:U^{235} = 92 and 440, respectively).

Experiments with Solution Having an H:U^{235} Ratio of 59

Critical arrays were constructed of as many as 125 units (in a 5 x 5 x 5 lattice) unreflected and of as many as 27 units (in a 3 x 3 x 3 lattice) reflected. The surface-to-surface separation, equal in three dimensions, was measured in each array. The results are given in Table 1.

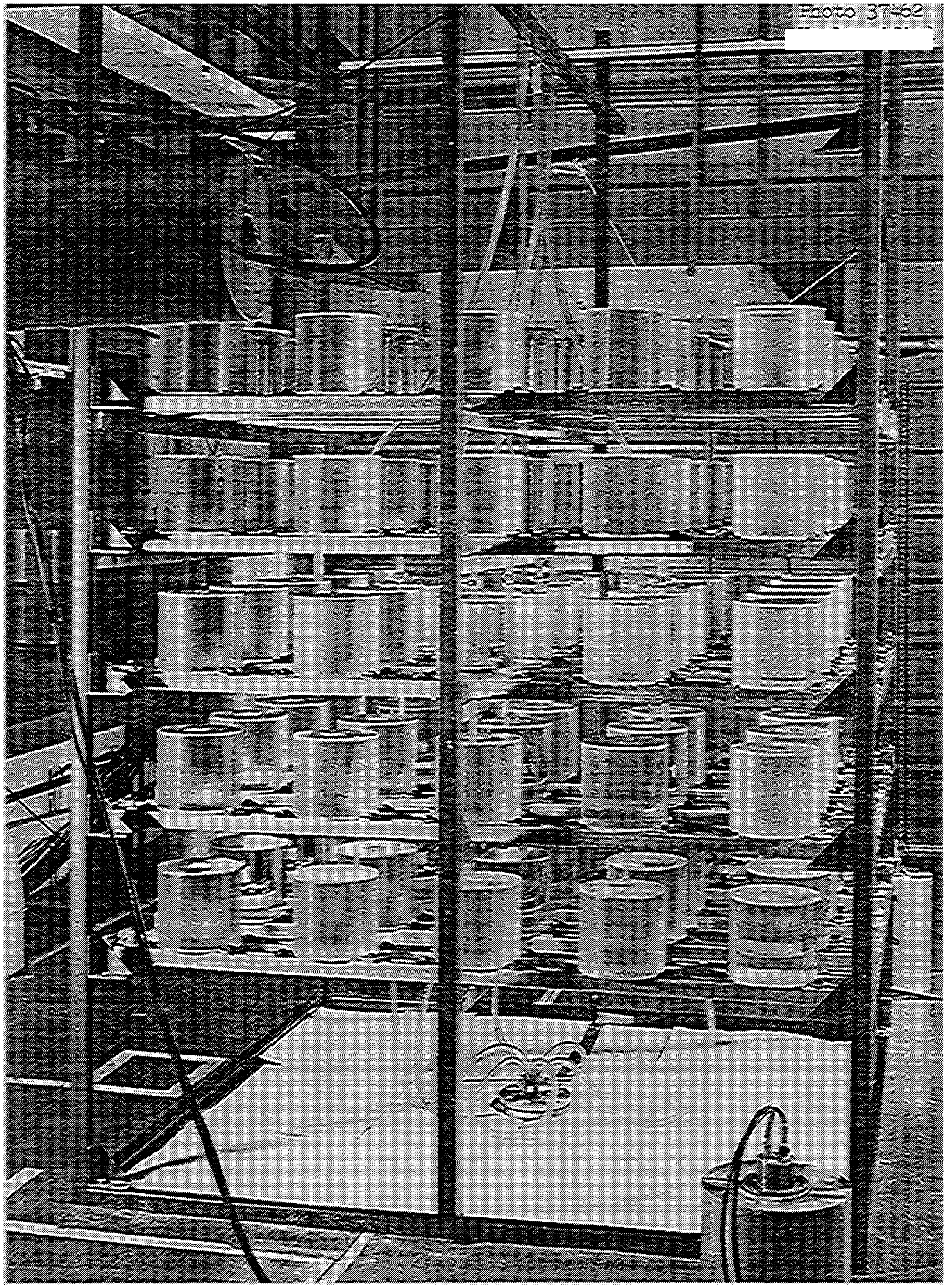

Fig. 1. View of a 125-Unit Unreflected Cubic Array.

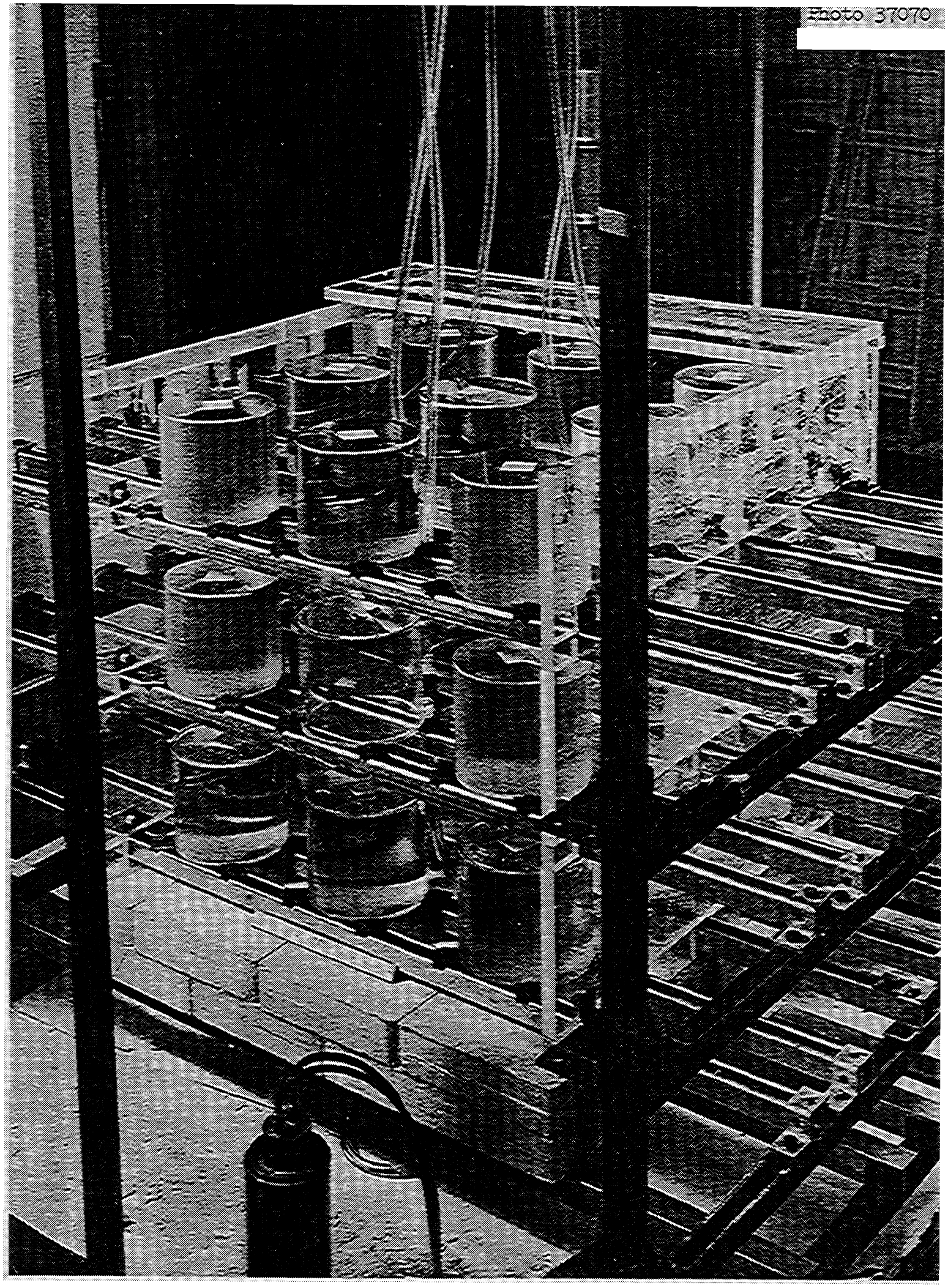

Fig. 2. View of a 27-Unit Cubic Array with 2.54-cm-thick Plexiglas Reflector on Five Sides and a 15.24-cm-thick Paraffin Base.

Table 1. Surface-to-Surface Separation of Units in Critical Three Dimensional Arrays with Reflectors of Various Thicknesses

Units: Five liters of $U(92.6)O_2(NO_3)_2$ solution having a concentration of 415 g of uranium per liter and a specific gravity of 1.555 contained in 0.64-cm-thick Plexiglas cylinders 20.32 cm OD and 19.05 cm in outside height.

Number of Units in Array	Reflector: Material	Reflector: Thickness (cm)	Surface-to-Surface Separation of Units[a] (cm)	Average Uranium Density in Array (g/cc)
8	None		1.43	0.214
8	Paraffin	1.27	3.28	0.167
8	"	3.81	6.91	0.108
8	"	7.62	8.48	0.091
8	"	15.24	8.99	0.087
8	Plexiglas	1.27	3.00	0.173
27	None		6.48	0.113
27	Paraffin	1.27	9.02	0.086
27	"	3.81	13.69	0.055
27	"	15.24	16.53[b]	0.043
27	Plexiglas	1.27	8.76	0.088
64	None		10.67	0.072
125	None		14.40	0.052

a. The uncertainty in the separation is ±0.13 cm.
b. The separation was 16.91 cm when one face of the cube was reflected by Plexiglas 15.24 cm thick.

Throughout another series of experiments the reflector on the bottom of 8- and 27-unit arrays was a 15.24-cm-thick slab of paraffin and the reflector on the remaining five sides was varied. These experiments simulate a condition where stored fissile material rests on a thick concrete floor. The results are given in Table 2.

Several other experiments were performed with two-dimensional arrays of solution at a concentration of 415 g of uranium per liter. Nineteen units arranged in a single tier with their centers in a triangular pattern were critical, unreflected, at a surface-to-surface separation of 1.35 cm. It was observed that 16 units in a single tier, in contact, arranged in a square pattern, and unreflected were subcritical with an apparent source neutron multiplication of approximately 6. Four units in a single tier, square pattern, with a surface-to-surface separation of 3.94 cm were critical when surrounded by a 15.24-cm-thick paraffin reflector.

Although in all the experiments reported in the tables the spacing between the Plexiglas containers was established by an aluminum structure, a few measurements were made with a light wooden frame as the spacer. The effect of the additional hydrogenous material increased the critical spacing up to 6% over the range examined. Had the support been wooden shelving the increase in the spacing may have been as much as 20%.

An indication of the neutron spectral change occurring in the neutron flux within these arrays upon the addition of a reflector was obtained by observing the corresponding change in the cadmium fraction. A BF_3 counter, 0.635-cm-OD by 2.54 cm long, was placed, with and without a cadmium cover, midway between the central and an adjacent unit in each of two arrays. The cadmium fraction in a 15.24-cm-thick paraffin-reflected 27-unit array was 0.96 while that in an unreflected 125-unit array was 0.86.

Experiments with Solutions at Other Uranium Concentrations

The results of a number of experiments performed with 8- and 27-unit arrays of solution more dilute than 415 g of uranium per liter are given in Table 3. From earlier work[2] it is known that the $H:U^{235}$ atomic ratio corresponding to a minimum critical volume for unreflected individual systems is in the range $60 < H:U^{235} < 90$; therefore only small differences in spacing are expected over this range.

Correlation of Data for the Solution at an $H:U^{235}$ of 59

Assuming that the quantity of uranium in a critical array varies inversely with the average uranium density to some power, s, the number of units in an array with a specified reflector may be expressed as

Table 2. Surface-to-Surface Separation of Units in Critical Three-Dimensional Arrays with Reflectors of Various Thicknesses on Five Sides and a 15.24-cm-thick Paraffin Reflector on the Bottom

Units: Five liters of $U(92.6)O_2(NO_3)_2$ solution having a concentration of 415 g of uranium per liter and a specific gravity of 1.555 contained in 0.64-cm-thick Plexiglas cylinders 20.32 cm OD and 19.05 cm in outside height.

Reflector		Surface-to-Surface Separation of Units[a] (cm)	Average Uranium Density in Array (g/cc)
Material	Thickness (cm)		
		8-Unit Arrays	
Paraffin	1.27	3.86	0.155
	3.81	7.26	0.103
	7.62	8.71	0.088
	15.24	8.99	0.087
Plexiglas	1.27	3.61	0.160
	2.54	5.41	0.128
	4.45	7.39	0.102
	6.35	8.64	0.089
	11.43	9.53	0.082
	15.24	9.60	0.081
		27-Unit Arrays	
Paraffin	1.27	9.88	0.079
	3.81	14.27	0.052
	7.62	15.85	0.044
	15.24	16.53	0.043
Plexiglas	1.27	9.58	0.081
	2.54	11.94	0.064

a. The uncertainty in the separation is ±0.13 cm.

Table 3. Surface-to-Surface Separation of Units of U^{235} Solutions of Various Concentrations in Critical Three-Dimensional Arrays

Units: Five liters of $U(92.6)O_2(NO_3)_2$ solution contained in 0.64-cm-thick Plexiglas cylinders 20.32 cm OD and 19.05 cm in outside height.

Number of Units in Array	$H:U^{235}$ Atomic Ratio	Uranium Density in the Solution (g/l)	Reflector		Surface-to-Surface Separation of Units[a] (cm)	Average Uranium Density in Array (g/cc)
			Material	Thickness (cm)		
8	92	279	Paraffin[b]	11.43	8.71	0.060
8	92	279	None		1.43	0.144
27	92	279	None		6.40	0.077
8	440	63.3	None		0[c]	0.040
27	440	63.3	None		2.41	0.029
27	d	d	None		6.41	0.107

a. The uncertainty in the values of the separation is ±0.13 cm.
b. Array was reflected on bottom by 15.24-cm-thick paraffin.
c. Array subcritical, k_{eff} ~0.6.
d. Five control units in center tier at $H:U^{235}$ = 92 and remaining 22 units at $H:U^{235}$ = 59.

$$N = N_o \, (\rho/\rho_o)^{-s} \quad , \qquad (1)$$

where the constant N_o is defined by the limit as ρ, the uranium density within the array, approaches ρ_o, the uranium density within a unit. The data of Table 1 are shown in this manner in Fig. 3. In calculating the ratio ρ/ρ_o the effect of the container was considered by increasing the unit volume by an amount equal to the volume reflector savings that would occur by placing a 0.64-cm-thick Plexiglas reflector around a bare critical sphere of the same solution. This correction, for units of optimum cylindrical geometry, was evaluated as 320 cc of solution. It should be noted that the determination of the exponent, s, does not depend upon this correction. The uncertainty in the constants s and N_o of Eq. (1) arises primarily from the error in the critical spacing of the units, the uncertainty in the mass present being much smaller. The experimental values of s and N_o, for the arrays described in Table 1, assuming Eq. (1), are presented in the following table.

Reflector			
Material	Thickness (cm)	s	N_o
None	0	1.928 ± 0.028	2.506 ± 0.014
Paraffin	1.27	1.831 ± 0.118	1.699 ± 0.316
	3.87	1.797 ± 0.098	0.794 ± 0.125
	15.24	1.721 ± 0.083	0.606 ± 0.084
Plexiglas	1.27	1.804 ± 0.116	1.842 ± 0.341

A comparison of the effect of reflectors of various thicknesses about these critical arrays of solution with the effect of reflectors on individual critical units is made in Fig. 4. Data are given for two arrays of different spacing, for a single unit of $U(93)O_2F_2$ solution at an $H:U^{235}$ of 74, and for a sphere of U(93) metal. The data for the single unit of solution and that for the metal sphere are reported in Reference 7. Although the addition of an effectively infinite hydrogenous reflector to a single unit reduces the critical mass by a factor of only approximately 2, a similar reflector about an array of units of solution reduces its content to about 1/5 the unreflected mass at the same unit spacing.

7. H. C. Paxton, J. T. Thomas and A. D. Callihan, "Critical Dimensions of Systems Containing U^{235}, Pu^{239}, and U^{233}," TID-7028 (1963).

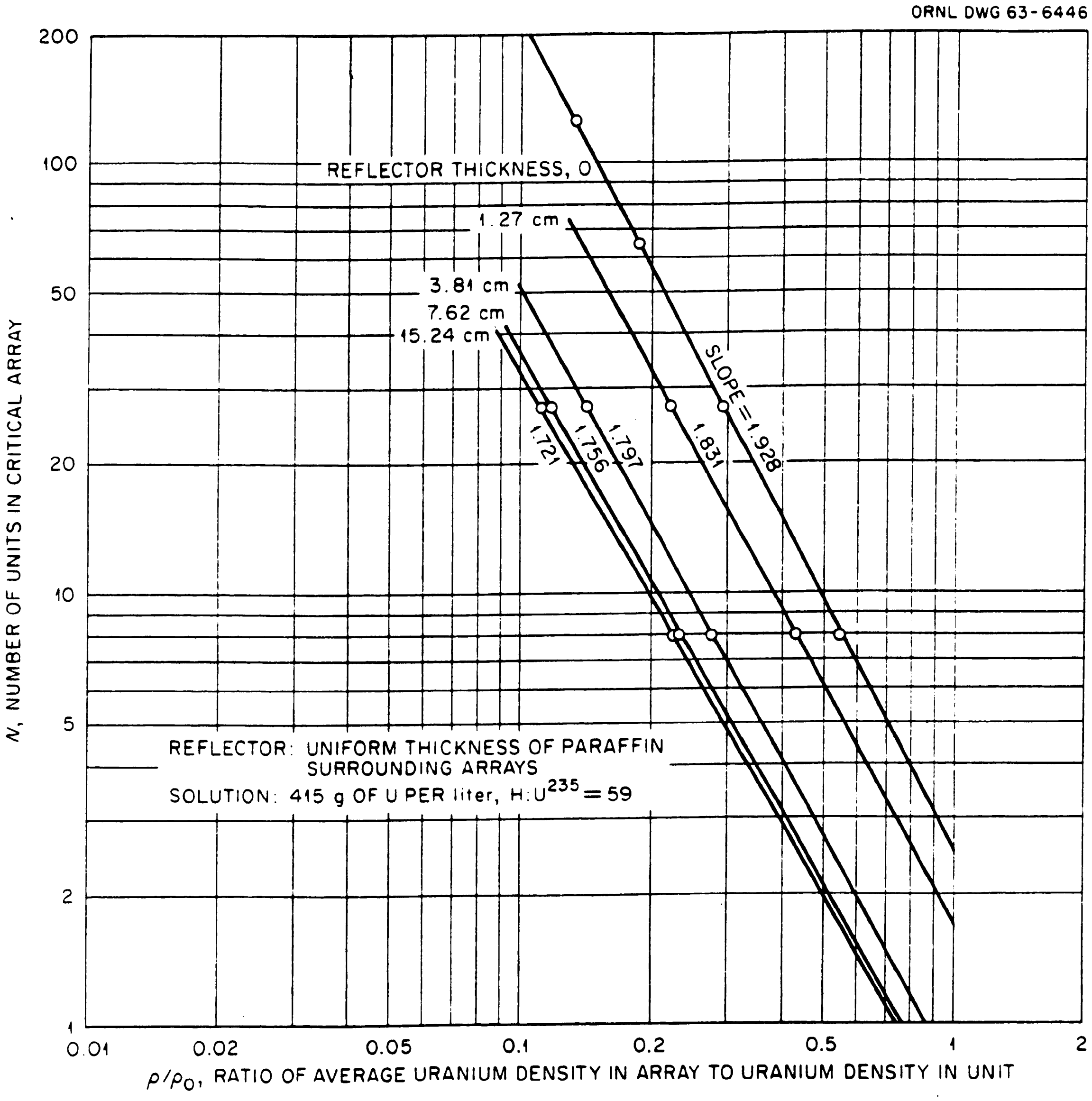

Fig. 3. **Mass-Density Representation of Unreflected and Reflected Critical Three-Dimensional Arrays of $U(92.6)O_2(NO_3)_2$ Solution in Five-Liter Units.**

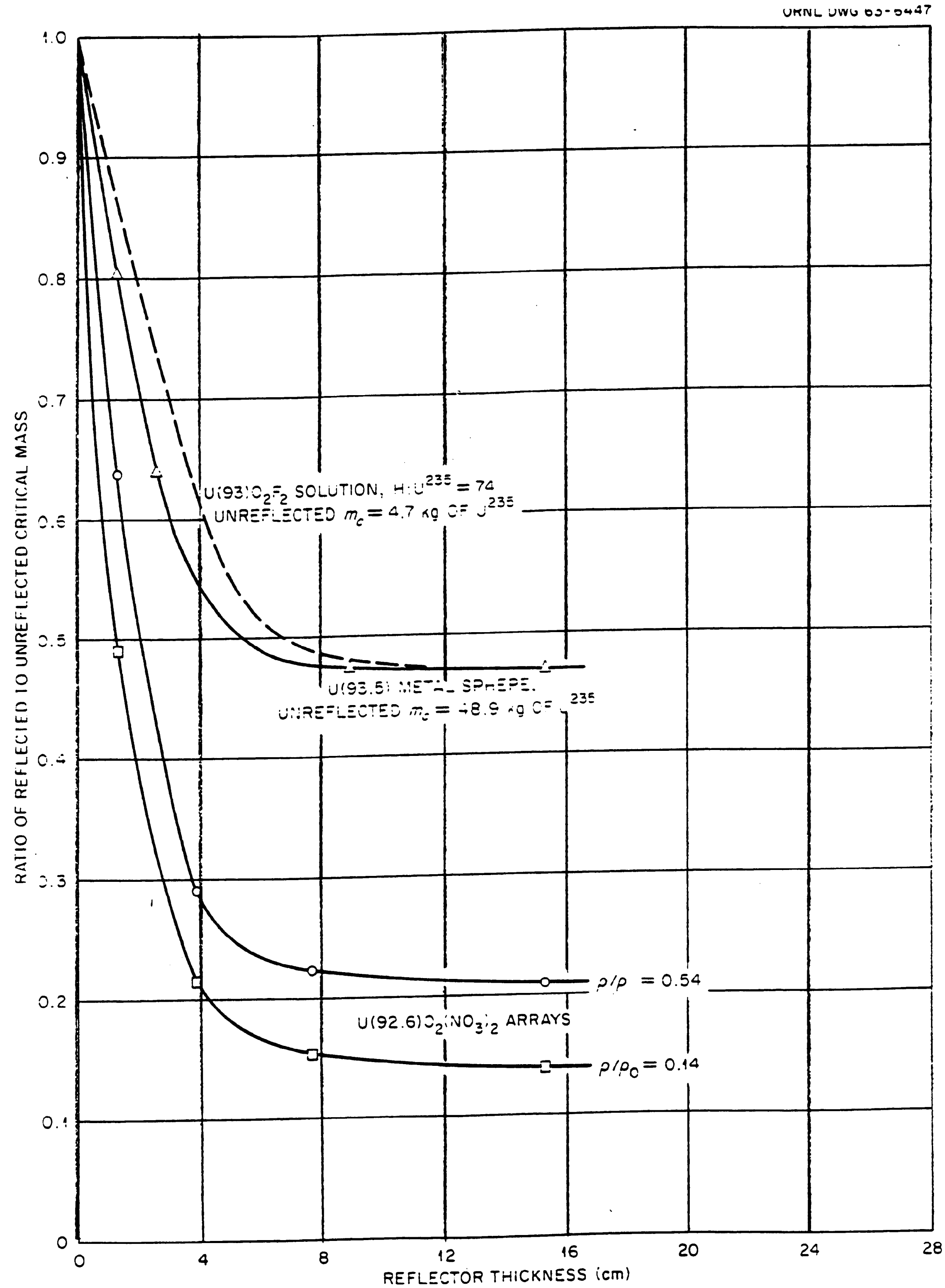

Fig. 4. Fractional Reduction of Critical Mass for Various Reflector Thicknesses Surrounding Individual Systems and Three-Dimensional Arrays of $U(92.6)O_2(NO_3)_2$ Solution Units.

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