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DIXON CALLIHAN, Oak Ridge National Laboratory, Oak Ridge, Tennessee

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Criticality of Homogeneous Plutonium Oxide-Plastic Compacts at H:Pu=15*

C. R. Richey, J. D. White, E. D. Clayton and R. C. Lloyd

Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington

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Critical experiments were conducted with homogeneous mixtures of PuO₂-polystyrene (H:Pu atomic ratio of 15) containing both 2.2 and 8.0% Pu²⁴⁰. Criticality was determined for a series of Plexiglas reflected rectangular prisms ranging from near cubes, to long columns, and to thin slabs; bare arrays of near-cubic geometry were also studied. Critical thicknesses were 16.09 ± 0.41 and 5.99 ± 0.10 cm, respectively, for the bare and reflected infinite slabs of PuO₂-polystyrene containing 2.2% Pu²⁴⁰. Corresponding values for the 8.0% Pu²⁴⁰ mixtures were 18.48 ± 0.41 and 7.38 ± 0.09 cm. The infinite slab thicknesses for an equivalent Pu²³⁹-water mixture (H:Pu = 15, ρ = 1.62 g Pu/cm³) were 11.66 ± 0.30 and 4.38 ± 0.08 cm, respectively, for the bare and water-reflected slabs. Corresponding critical radii for infinitely long cylinders were 10.52 ± 0.16 and 6.54 ± 0.14 cm; radii for critical spheres were 13.81 ± 0.16 and 10.40 ± 0.17 cm.

INTRODUCTION

Under-moderated plutonium mixtures in the form of wet powders, precipitates, slurries, polymers and other highly concentrated plutonium mixtures constitute potential criticality hazards in chemical plants processing irradiated reactor fuels. The procedures for criticality control of highly concentrated plutonium mixtures are based primarily on the subcritical data of Goodwin and Schuske with heterogeneous cylindrical arrays of plutonium metal discs and Plexiglas¹. In their experiments the subcritical arrays did not exceed 85% of the critical mass predicted from the inverse neutron multiplication curves; therefore, some consistent uncertainties may exist in the results. This paper reports the results from critical experiments with homogeneous PuO₂-polystyrene compacts having a hydrogen-to-plutonium

atom ratio of 15. Two sets of compacts having 2.2 and 8.0% Pu²⁴⁰ were studied. A constant buckling conversion scheme was used to obtain critical dimensions for the basic geometrical shapes (e.g. spheres, infinite slabs and infinitely long cylinders.) The results are compared to those calculated using multigroup diffusion and transport (S₄) theory. Comparative data were also obtained on the reflector savings for 8 in. (20 cm) of water, Plexiglas, and 3wt%-U²³⁵-enriched UO₂(NO₃)₂ solution (≈ 374 g U/l).

DESCRIPTION OF EXPERIMENTS

Experimental Facility

The critical experiments were conducted in a large glove box within a heavily shielded cell at the Battelle-Northwest Critical Mass Laboratory². Critical arrays, in the form of rectangular prisms,

*This paper is based on work performed under USAEC Contracts AT(45-1)-1350 and AT(45-1)-1830. Permission to publish is gratefully acknowledged.

¹A. GOODWIN, Jr. and C. L. SCHUSKE, "Plexiglas- and Graphite-Moderated Plutonium Assemblies," *Reactor Sci. Tech.*, 15, 213, (October 1961).

²W. A. REARDON, E. D. CLAYTON, C. L. BROWN, R. H. MASTERSON, T. J. POWELL, C. R. RICHEY, R. B. SMITH and J. W. HEALY, "Hazards Summary Report for the Hanford Plutonium Critical Mass Laboratory," HW-66266, (August 1, 1960).

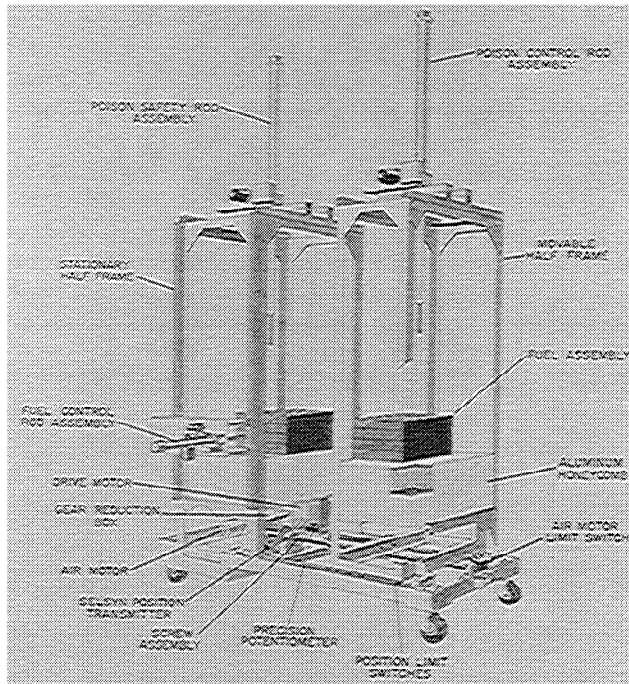


Fig. 1. Remote split-table machine.

were assembled by the Remote Split-Table Machine (RSTM) shown in Fig 1 (Ref. 3). Each table half, one stationary and the other movable, had a

³C. R. RICHEY, E. D. CLAYTON, R. H. ODEGAARDEN, J. D. WHITE and W. A. REARDON, "Hazards Summary Report for the Hanford Plutonium Critical Mass Laboratory, Supplement No. 1: The Remote Split-Table Machine," HW-66266 SUP 1 REV, (October 1963).

steel framework supporting the horizontally operated safety- and control-rod drives. Together, the table halves formed a surface 30-in. (76 cm) wide and 42- (24- and 18-) in. (107- (61- and 46-) cm) long, 20 in. (51 cm) above the floor level. A 12-in.-thick, (30-cm) low-density aluminum honeycomb material ($\rho = 0.037 \text{ g/cm}^3$) covered both table halves. Each fuel assembly was clamped to a $\frac{1}{8}$ -in.-thick (0.159 cm) aluminum plate, which rested on the honeycomb base. The safety and control rods displaced either fuel or reflector materials; therefore each assembly was relatively free of voids. The reflector displacement rods were free of extraneous neutron absorbers. While using the fuel displacement rods, the fuel was encased in steel jackets and inserted into a steel sleeve traversing the array.

PuO₂-Polystyrene Compacts

The PuO₂-polystyrene compacts were fabricated by pressing 100-mesh polystyrene and PuO₂ into a 2-in. (5 cm) cubic mold. The advantages of polystyrene as the matrix material are its excellent molding properties at high temperatures and pressures and its stability under alpha irradiation from the radioactive plutonium.

Each compact was spray painted with about a 1-mil-thick (0.025 mm) coat of aluminum paint and covered with rubberized plastic approximately 10-mils (0.25 mm) thick. By cutting a number of 2-in. cubes into 1-in.- and 0.5-in.-thick (1.3-cm) blocks (Fig 2) it was possible to vary the array in increments of 0.5 in. The PuO₂ was filtered

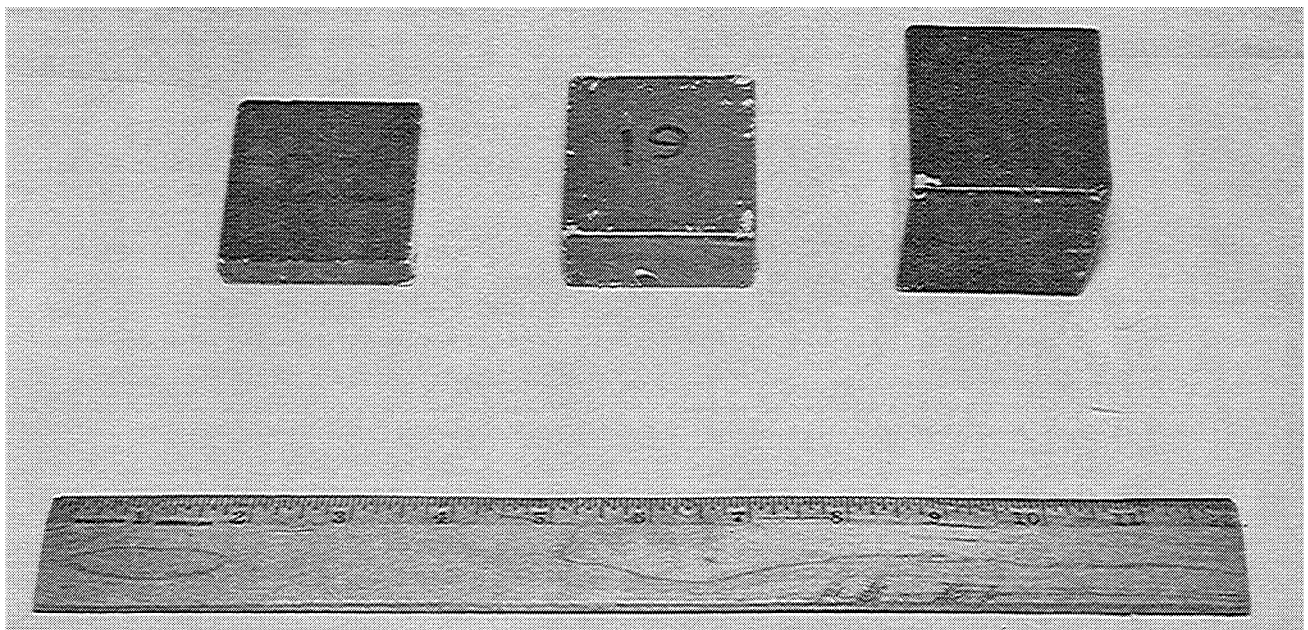


Fig. 2. PuO₂-polystyrene compacts.

through a 100-mesh screen setting a 5.8-mil (0.15-mm) upper limit on particle size. Approximately 35 and 77% of the PuO_2 particles in the 2.2 and 8.0% Pu^{240} materials, respectively, were smaller than 1.7 mils (0.04 mm). Recorded in Table I are the dimensions and masses corresponding to the cubic compacts. The values given, except for the thickness of the plastic coating, are averages over 100% of inventory. The quoted uncertainties define the range into which two-thirds of the compacts fall. Isotopic content of the plutonium is given in Table II.

TABLE I
Properties of PuO_2 -Polystyrene Compacts

	2.2% Pu^{240}	8.0% Pu^{240}
Dimension of Cubic Compacts (Including Plastic Coating)	2.036 ± 0.012 in. (5.171 ± 0.030 cm)	2.016 ± 0.010 in. (5.121 ± 0.025 cm)
Dimension of Bare Cubic Compacts (Including ≈ 1 mil Aluminum Coating)	2.014 ± 0.012 in. (5.116 ± 0.030 cm)	2.000 ± 0.010 in. (5.080 ± 0.025 cm)
Thickness of Plastic Coating	0.011 ± 0.003 in. (0.028 ± 0.008 cm)	0.008 ± 0.002 in. (0.020 ± 0.005 cm)
Plutonium per Cubic Compact, g	150.1 ± 2.0	137.2 ± 1.2
Polystyrene per Cubic Compact, g	123.3 ± 1.9	111.7 ± 1.0
Plutonium Density in Bare Cubic Compacts, g/cm^3	1.12 ± 0.02	1.05 ± 0.02

TABLE II
Isotopic Content of Plutonium

Isotope	Percent of Isotope Present	
	2.2% Pu^{240}	8.0% Pu^{240}
Pu^{238}	< 0.01	< 0.01
Pu^{239}	97.69 ± 0.04	91.16 ± 0.07
Pu^{240}	2.20 ± 0.04	8.06 ± 0.07
Pu^{241}	0.11 ± 0.02	0.75 ± 0.01
Pu^{242}	< 0.01	0.034 ± 0.001

Experimental Method

Source-neutron multiplication measurements served as a guide for succeeding fuel additions during the assembly of the critical arrays. Spontaneous fission, primarily from Pu^{240} , supplied the required neutron source. Three proportional neutron counters with scaling circuits provided the quantitative measurements of the source-neutron multiplication. Although each experimental array was made critical, criticality was usually achieved with a partial or incomplete layer of compacts and with the control rod partially inserted. A least-squares analysis of the inverse neutron multiplication data was therefore used to obtain the critical dimensions. The data utilized in the analysis was obtained with greater than 80% of the critical mass assembled.

EXPERIMENTAL RESULTS

Criticality with 2.2 and 8.0% Pu^{240} compacts was obtained in rectangular prisms ranging from near cubes, to long columns, to thin slabs. These arrays were enclosed on all surfaces by 6 in. (15 cm) of Plexiglas ($\rho = 1.2 \text{ g}/\text{cm}^3$). It is shown in the section on 'Reflector Savings Measurements' that this is equivalent to an infinitely thick Plexiglas reflector. The safety and control rods utilized were the reflector displacement type, hence the fuel core was relatively free of extraneous materials. Except for partial reflection by the Plexiglas control and safety rods, two otherwise-bare near-cubic arrays of 2.2 and 8.0% Pu^{240} compacts were made critical. Because of an insufficient number of compacts, the 8.0% Pu^{240} array was made critical with a 1-in. layer of 2.2% Pu^{240} compacts on its top surface. A least-squares analysis of the source-neutron multiplication data with both reflecting rods withdrawn provided the critical size for the completely bare arrays. Approximately 98% of the bare critical masses was assembled when the rod-reflected arrays became critical. The perturbation due to the layer of 2.2% Pu^{240} compacts on top of the 8.0% Pu^{240} array was evaluated by an 18-group diffusion-theory calculation; the correction increased the critical height by 1.3%.

Recorded in Table III are the critical dimensions for each array. Quoted errors are standard deviations from the least-squares analysis and do not reflect any uncertainties in individual source-neutron multiplication measurements. Also given are results corrected to 'clean arrays' free from extraneous material. The basis for the corrections are as follows:

The $\frac{1}{16}$ -in.-thick aluminum plate and low-density honeycomb base, on which the bare arrays rested,

TABLE III
Criticality Data for PuO₂-Polystyrene Compacts, H:Pu = 15

Reflector	Critical Dimensions, cm			Critical Mass, kg of Pu	
	Length	Width	Height	Exptl Data	Corr Data ^a
2.2% Pu ²⁴⁰ Compacts					
Plexiglas	51.69	46.13	9.04 ± 0.06	23.39 ± 0.17	24.14 ± 0.18
Plexiglas	41.35	38.46	10.34 ± 0.05	17.84 ± 0.09	18.42 ± 0.09
Plexiglas	31.01	31.01	13.13 ± 0.02	13.70 ± 0.02	14.14 ± 0.02
Plexiglas	25.86	25.86	16.43 ± 0.06	11.92 ± 0.04	12.30 ± 0.04
Plexiglas	23.27	23.27	19.79 ± 0.05	11.63 ± 0.03	12.01 ± 0.03
Plexiglas	20.68	20.68	24.87 ± 0.06	11.54 ± 0.03	11.91 ± 0.03
Plexiglas	15.52	18.08	50.04 ± 0.13	15.23 ± 0.04	15.72 ± 0.04
Bare	31.01	31.01	31.87 ± 0.06	33.25 ± 0.06	
Bare ^a	30.68	30.68	32.84 ± 0.52		34.62 ± 0.55
8.0% Pu ²⁴⁰ Compacts					
Plexiglas	51.31	68.25	10.36 ± 0.03	37.19 ± 0.11	38.09 ± 0.11
Plexiglas	35.92	35.92	15.42 ± 0.03	20.39 ± 0.04	20.89 ± 0.04
Plexiglas	30.78	30.78	18.56 ± 0.02	18.07 ± 0.02	18.46 ± 0.02
Plexiglas	25.65	25.65	25.03 ± 0.05	16.88 ± 0.03	17.29 ± 0.03
Plexiglas	25.65	25.65	25.13 ± 0.03	16.95 ± 0.02	17.36 ± 0.02
Plexiglas	20.52	20.52	49.15 ± 0.01	21.22 ± 0.01	21.73 ± 0.01
Bare	35.92	35.92	35.54 ± 0.02	46.93 ± 0.03	
Bare ^a	35.63	35.63	36.04 ± 0.60		48.04 ± 0.80

^a Data corrected for plastic coating. An additional correction for the base material was applied to the bare arrays.

were evaluated by placing a duplicate arrangement on the top surfaces of the arrays. The corresponding corrections increased the critical height of the 2.2 and 8.0% Pu²⁴⁰ bare arrays by 0.18 ± 0.04 and 0.16 ± 0.03 in. (0.46 ± 0.10 and 0.41 ± 0.08 cm), respectively.

To evaluate the effect of the plastic coating on each compact, its thickness was varied by inserting rubberized plastic shims between each compact. Measurements were made for both the bare and reflected near-cubic arrays with the results being summarized in Table IV. Fuel-removal control and safety rods were used in the bare 2.2% Pu²⁴⁰ measurements, thus accounting for the differences in critical dimension quoted in Table III and Table IV. Adding the shims increased the horizontal dimensions, and the critical height was observed to vary inversely as the plastic thickness. The critical volumes, however, behaved differently; as the shim thickness increased, the critical volume decreased for the bare arrays and increased for the reflected arrays. The plastic coating affects criticality primarily in two ways: 1) it decreases the effective plutonium density, and 2) it increases the neutron moderation in the fuel core. While a decrease in density will increase the critical volume, the direction of the moderator effect depends on whether the array is under- or over-moderated. As expected, the moderating effect predominates in the more under-moderated

bare arrays; in the less under-moderated reflected arrays the density effect is slightly predominate. Since the volume corrections for the near-cubic reflected arrays were small (< 1%), the critical dimensions for the reflected arrays were not altered on correcting to the clean arrays. Corrections were applied, however, to the critical mass values for differences in the effective plutonium density.

ANALYSIS OF EXPERIMENTS

Constant Buckling Conversion

The critical bucklings for the 2.2 and 8.0% Pu²⁴⁰ bare arrays were calculated by the usual one-group expression

$$B_c^2 = \frac{\pi^2}{(a + 2\lambda)^2} + \frac{\pi^2}{(b + 2\lambda)^2} + \frac{\pi^2}{(c + 2\lambda)^2}, \quad (1)$$

where a , b , and c are the critical dimensions of the arrays. The extrapolation length λ is defined by

$$\lambda = \frac{1}{2} (T_e - T_c), \quad (2)$$

where T_e and T_c are critical heights computed from an 18-group diffusion-theory calculation using two different exterior boundary conditions; T_e was computed with the group fluxes vanishing at the exterior boundary (albedo = -1.0), and T_c was

TABLE IV
Criticality as a Function of Plastic Thickness

Thickness of Plastic Coating ($\times 10^{-3}$ cm)	Critical Dimensions, cm			Critical Volume, Liters	Critical Mass, kg of Pu
	Length	Width	Height		
	2.2% Pu ²⁴⁰ Bare Array				
99	31.88	31.88	31.19 \pm 0.15	31.70 \pm 0.15	31.70 \pm 0.16
64	31.45	31.45	31.98 \pm 0.08	31.63 \pm 0.08	32.94 \pm 0.08
28	31.01	31.01	33.66 \pm 0.07	32.37 \pm 0.07	35.15 \pm 0.08
0	30.68	30.68	34.49 \pm 0.51	32.46 \pm 0.48	36.35 \pm 0.54
2.2% Pu ²⁴⁰ Reflected Array					
97	23.88	23.88	19.13 \pm 0.18	10.91 \pm 0.10	10.91 \pm 0.10
61	23.57	23.57	19.61 \pm 0.20	10.89 \pm 0.11	11.35 \pm 0.11
28	23.27	23.27	19.79 \pm 0.15	10.72 \pm 0.08	11.64 \pm 0.09
0	23.01	23.01	20.12 \pm 0.18	10.65 \pm 0.10	11.93 \pm 0.11
8.0% Pu ²⁴⁰ Bare Array					
94	36.95	36.95	33.20 \pm 0.01	45.33 \pm 0.01	42.30 \pm 0.01
46	36.27	36.27	34.10 \pm 0.02	44.86 \pm 0.03	44.27 \pm 0.03
20	35.92	35.92	35.45 \pm 0.06	45.74 \pm 0.08	45.48 \pm 0.08
0	35.63	35.63	35.88 \pm 0.59	45.55 \pm 0.75	47.83 \pm 0.79
8.0% Pu ²⁴⁰ Reflected Array					
81	26.26	26.26	23.41 \pm 0.02	16.14 \pm 0.01	15.29 \pm 0.01
53	25.98	25.98	24.31 \pm 0.10	16.41 \pm 0.07	16.05 \pm 0.07
20	25.65	25.65	25.04 \pm 0.03	16.47 \pm 0.02	16.74 \pm 0.02
0	25.45	25.45	25.40 \pm 0.28	16.45 \pm 0.18	17.27 \pm 0.18

computed using the usual boundary condition that the group fluxes vanish at the extrapolated boundary (albedo = 3.15×10^{-2}). The computed values of λ for the 2.2 and 8.0% Pu²⁴⁰ bare arrays are, respectively, 2.55 and 2.76 cm, which result in critical bucklings of 222.7 ± 1.9 and $173.3 \pm 1.3 \text{ m}^{-2}$.

Equating B_c^2 to the corresponding bare-array value, extrapolation lengths for the reflected arrays are obtained upon substituting the respective critical dimensions into Eq. (1); the resulting values of λ are plotted in Fig. 3 as a function of horizontal buckling B_H^2 . The errors indicated in Fig. 3 are standard deviations reflecting the respective uncertainty in the measured critical height. A least-squares analysis of the data yields extrapolation lengths of 7.53 ± 0.03 and 8.24 ± 0.05 cm, respectively, for the 2.2 and 8.0% Pu²⁴⁰ reflected infinite slabs. Corresponding values calculated from Eq. (2) are 7.51 and 8.22 cm, respectively. The measured critical heights for reflected rectangular prisms are plotted in Fig. 4 as a function of B_H^2 .

Critical dimensions for bare and Plexiglas reflected spheres, cubes, infinite slabs, and infinitely long cylinders from a constant buckling conversion are given in Table V. The values of λ utilized in the buckling conversions were calculated from Eq. (2) except for the reflected cubes, which were

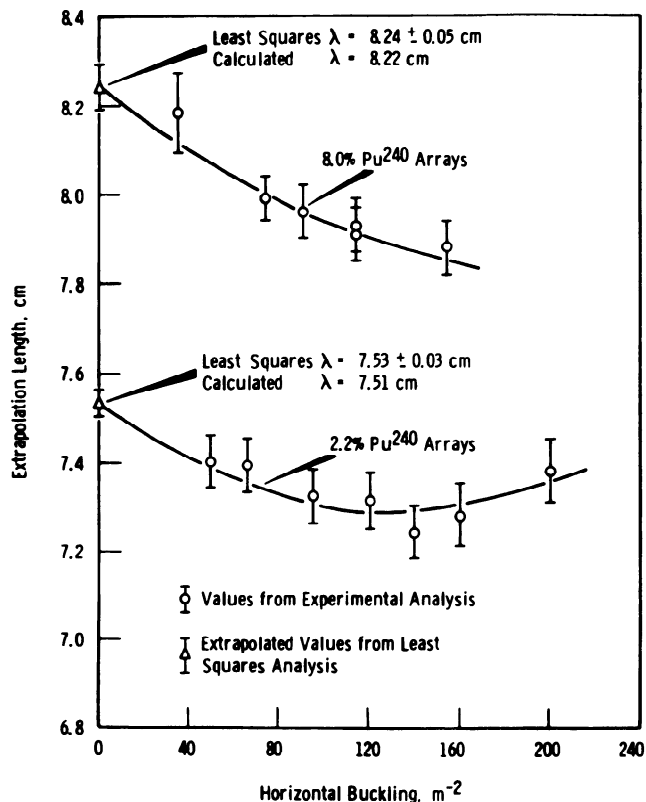


Fig. 3. Extrapolation length as a function of horizontal buckling for PuO₂-polystyrene compacts.

TABLE V
Critical Dimensions for PuO₂-Polystyrene in Spherical, Cylindrical, Cubic, and Slab Geometry, H:Pu = 15

	Bare Arrays		Plexiglas-Reflected Arrays	
	λ , cm	X_c , ^a cm	λ , cm	X_c , ^a cm
	2.2% Pu²⁴⁰, $B_c^2 = 222.7 \pm 1.9 \text{ m}^{-2}$			
Infinite Slab	2.48 ± 0.20	16.09 ± 0.41	7.53 ± 0.03	5.99 ± 0.10
Sphere	2.47 ± 0.20	18.58 ± 0.22	7.51 ± 0.20	13.54 ± 0.22
Infinite Cylinder	2.47 ± 0.20	13.64 ± 0.21	7.52 ± 0.20	8.59 ± 0.21
Cube	2.55 ± 0.20	31.36 ± 0.29	7.28 ± 0.06	21.90 ± 0.19
8.0% Pu²⁴⁰, $B_c^2 = 173.3 \pm 1.3 \text{ m}^{-2}$				
Infinite Slab	2.69 ± 0.20	18.48 ± 0.41	8.24 ± 0.05	7.38 ± 0.09
Sphere	2.69 ± 0.20	21.17 ± 0.21	8.22 ± 0.20	15.64 ± 0.21
Infinite Cylinder	2.68 ± 0.20	15.59 ± 0.21	8.23 ± 0.20	10.04 ± 0.21
Cube	2.76 ± 0.20	35.82 ± 0.26	7.82 ± 0.06	25.70 ± 0.17

^a X_c denotes the critical dimensions, i.e. thickness of slab, radii of sphere and cylinder.

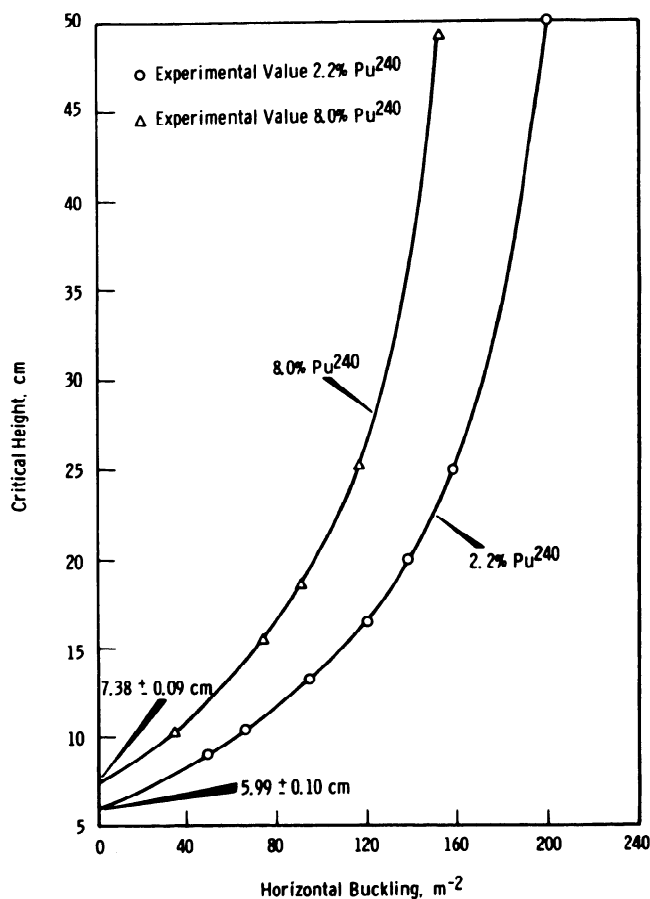


Fig. 4. Critical height as a function of horizontal buckling for PuO₂-polystyrene compacts.

taken from the data plotted in Fig. 3. Quoted errors on the critical dimensions are standard deviations reflecting the uncertainties in the measured critical heights and extrapolation lengths.

The uncertainty in the calculated extrapolation lengths is estimated to be ± 0.2 cm.

Multigroup Analysis

The critical dimensions of PuO₂-polystyrene in the form of bare and Plexiglas reflected spheres, infinite slabs, and infinitely long cylinders were computed using multigroup diffusion and S_4 approximations to the transport equation^{4,5}. Corresponding calculations were made for bare and water-reflected mixtures of Pu²³⁹-water ($\rho = 1.62 \text{ g Pu}^{239}/\text{cm}^3$) and PuO₂-water ($\rho = 1.50 \text{ g Pu}^{239}/\text{cm}^3$) at a H:Pu atomic ratio of 15. The results are summarized in Table VI. The S_4 calculations in general agree better with experiment than do the diffusion theory results. Some difficulty in treating the Pu²⁴⁰ capture is evident from the difference in computed and experimental values for the 8.0% Pu²⁴⁰ arrays.

The multigroup constants for both the S_4 and diffusion-theory calculations were obtained from the GAMTEC-2 computer code⁶. The 17-fast-group constants ($E > 0.683 \text{ eV}$) were averaged over a 64-group slowing-down spectrum from a B_1 approximation of the Boltzmann equation using a Pu²³⁹ fission-spectrum-weighted source term. The one-thermal-group constants were averaged over

⁴J. R. LILLEY, "Computer Code HFN-Multi-Group Multi-Region Neutron Diffusion Theory in One Space Dimension," HW-71545, (November 17, 1961).

⁵G. B. CARLSON, W. J. WORLTON, G. GUBER and M. SHAPIRO, "DTF Users Manual," UNC Phys/Math-3321, Vol. 1.

⁶L. L. CARTER, C. R. RICHEY and C. E. HUGHEY, "GAMTEC-2: A Code for Generating Consistent Multi-Group Constants Utilized in Diffusion and Transport Theory Calculations," BNWL-35, (March 1965).

TABLE VI
Calculated Criticality for PuO₂-Polystyrene, Pu²³⁹-Water, and Pu²³⁹O₂-Water
Mixtures in Various Geometrical Shapes, H:Pu = 15

	PuO ₂ -Polystyrene, Pu ²⁴⁰ = 2.2%			PuO ₂ -Polystyrene, Pu ²⁴⁰ = 8.0%			Pu ²³⁹ -Water, Pu ²⁴⁰ = 0.0		Pu ²³⁹ O ₂ -Water, Pu ²⁴⁰ = 0.0	
	Exptl, ^a cm	P ₁ Calc, cm	S ₄ Calc, cm	Exptl, ^a cm	P ₁ Calc, cm	S ₄ Calc, cm	P ₁ Calc, cm	S ₄ Calc, cm	P ₁ Calc, cm	S ₄ Calc, cm
	Bare Arrays									
Inf Slab Thickness	16.09 ± 0.41	16.23	16.00	18.48 ± 0.41	18.98	18.87	11.90	11.59	12.42	12.14
Radius of Sphere	18.58 ± 0.22	18.71	18.31	21.17 ± 0.21	21.69	21.48	14.19	13.61	14.70	14.17
Radius of Inf Cyl	13.64 ± 0.21	13.75	13.39	15.59 ± 0.21	15.97	15.73	10.33	9.86	10.73	10.28
Reflected Arrays ^b										
Inf Slab Thickness	5.99 ± 0.10	6.17	6.21	7.38 ± 0.09	7.98	8.02	4.54	4.49	4.84	4.81
Radius of Sphere	13.54 ± 0.22	13.67	13.46	15.64 ± 0.21	15.97	15.90	10.82	10.34	11.23	10.78
Radius of Inf Cyl	8.59 ± 0.19	8.70	8.53	10.04 ± 0.21	10.33	10.23	6.82	6.49	7.12	6.81

^aValues derived from experimental data.

^bPu²³⁹-water and Pu²³⁹O₂-water mixtures reflected by water; PuO₂-polystyrene compacts reflected by Plexiglas.

a Wigner-Wilkins spectrum using Sher-normalized Pu²³⁹ cross sections⁷. Resonance absorption by Pu²⁴⁰ was treated by the Adler-Nordheim method⁸. In the S₄ calculations, the angular distribution of elastically scattered neutrons was defined through the P₁ term in the Legendre expansion.

To convert the PuO₂-polystyrene data to Pu²³⁹-water and Pu²³⁹O₂-water mixtures at an equivalent hydrogen-to-plutonium ratio requires more than a simple density correction. Polystyrene, (C₈H₈CHCH₂)_n, contains carbon in a one-to-one correspondence to hydrogen, whereas the hydrogen in water is in a two-to-one correspondence to oxygen. The presence of the additional carbon in polystyrene significantly affects the neutron leakage and consequently the critical mass. Since elastic scattering of neutrons by hydrogen is strongly forward in direction, displacement of hydrogen by the more symmetric scatterer, carbon, tends to reduce neutron leakage. This effect is particularly important at higher neutron energies (E > 0.01 MeV), where the cross sections for carbon are competitive to those for hydrogen.

The critical dimensions for bare and water-

reflected spheres, infinite slabs, and infinitely long cylinders of Pu²³⁹-water (H:Pu atomic ratio = 15) were obtained by

$$X(\text{Pu}^{239}\text{-H}_2\text{O}) = X_{\text{exptl}}(\text{PuO}_2\text{-(C}_8\text{H}_8)_n) \times \frac{X_{\text{calc}}(\text{Pu}^{239}\text{-H}_2\text{O})}{X_{\text{calc}}(\text{PuO}_2\text{-(C}_8\text{H}_8)_n)}$$

where $X_{\text{exptl}}(\text{PuO}_2\text{-(C}_8\text{H}_8)_n)$ is the experimentally derived critical dimension for the 2.2% Pu²⁴⁰ compacts and $X_{\text{calc}}(\text{Pu}^{239}\text{-H}_2\text{O})$ and $X_{\text{calc}}(\text{PuO}_2\text{-(C}_8\text{H}_8)_n)$ are corresponding values from multi-group S₄ calculations given in Table VI. The resulting infinite slab thicknesses are 11.66 ± 0.30 and 4.36 ± 0.12 cm for the bare and water-reflected slabs, respectively. Corresponding critical radii for infinitely long cylinders are 10.52 ± 0.16 and 6.54 ± 0.14 cm; radii for critical spheres are 13.81 ± 0.16 and 10.40 ± 0.17 cm.

Critical thicknesses computed from multigroup S₄ theory for infinite slabs of Pu²³⁹-water mixtures having 0.1 to 10.0 g Pu²³⁹/cm³ are recorded in Fig. 5. Note that the critical slab thicknesses derived from the critical measurements agree with the calculated values; whereas, the data evaluated from the subcritical experiments of Goodwin and Schuske consistently underestimate the critical thicknesses^{1,9}.

⁷R. SHER and J. FELBERBAUM, "Least Squares Analysis of 2200 m/sec Parameters of U²³⁵, U²³⁸, and Pu²³⁹," "Neutron Cross Section Evaluation Group," BNL 722, (June 1962).

⁸F. T. ADLER, G. W. HINMAN and L. W. NORDHEIM, "The Quantitative Evaluation of Resonance Integrals," *Proc. 2nd Intern. Conf. Peaceful Uses Atomic Energy*, 16, P/1988 (1958).

⁹H. C. PAXTON, "Correlation of Experiment and Theoretical Criticality Data, Comparative Reliability, Safety Factors for Criticality Control," LAMS-2537, (March 1961).

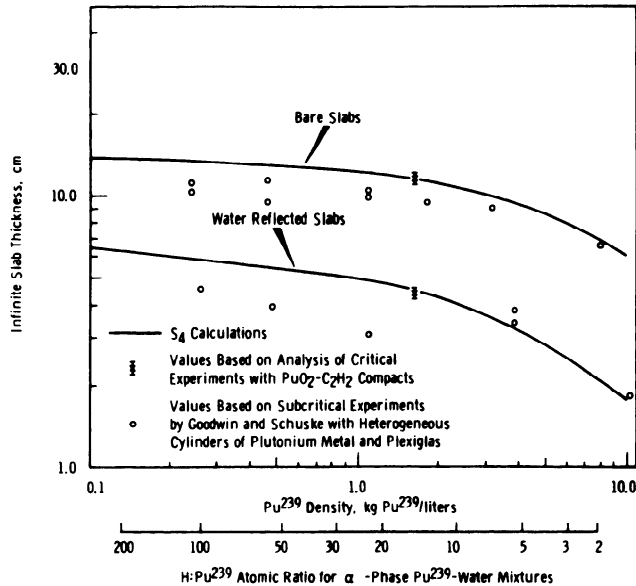


Fig. 5. Critical infinite slab thickness vs concentration for Pu^{239} -water mixtures.

REFLECTOR SAVINGS MEASUREMENTS

The reflector savings of various thicknesses of Plexiglas located on two opposite surfaces of an otherwise bare rectangular prism of 2.2% Pu^{240} compacts was measured. The cross-sectional dimensions of the rectangular prism were 12.21×12.21 in. (31.01×31.01 cm). Since two opposite surfaces were reflected, the reflector savings δ is defined as one-half the decrease in critical length due to the reflector. The results plotted in Fig. 6 show that δ for 6-in.-thick (15-cm) Plexiglas is approximately that for infinitely thick Plexiglas. A 30-mil-thick (0.76-mm) cadmium sheet at the interface of the fuel core and 8-in.-thick (20-cm) Plexiglas reflector reduced δ from 4.10 ± 0.03 to 1.51 ± 0.07 cm, a difference of 2.59 ± 0.08 cm.

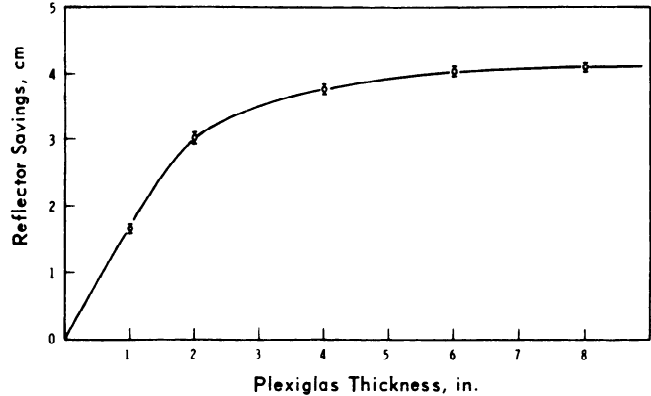


Fig. 6. Reflector savings as a function of plexiglas thickness.

Thus, the 30-mil cadmium resulted in a δ equivalent to 0.9 ± 0.1 -in.-thick (2.3 ± 0.3 -cm) Plexiglas.

Recorded in Table VII are comparative data on the reflector savings of 8 in. of water, Plexiglas, and a 3wt% U^{235} -enriched $\text{UO}_2(\text{NO}_3)_2$ solution (≈ 374 g U/l) at one end of the rectangular prism. The water and $\text{UO}_2(\text{NO}_3)_2$ solutions were contained in an 8-in.-thick rectangular tank 12×12 in. (30×30 cm); the tank was fabricated from 61-mil-thick (1.5-mm) stainless steel. To simulate the steel tank when testing the Plexiglas reflector, one measurement was made with a 61-mil-thick stainless-steel plate inserted at the fuel-Plexiglas interface.

Although the density of hydrogen in Plexiglas is less than in water, Plexiglas was found to have the larger reflector savings, i.e. $\delta(\text{C}_5\text{H}_8\text{O}_2) - \delta(\text{H}_2\text{O}) = 0.61 \pm 0.10$ cm. This is principally due to the increased amounts of carbon and oxygen in Plexiglas. Neutrons undergoing scattering with either carbon or oxygen have a better chance of being reflected to the fuel core than if scattered by hydrogen with its strongly forward-scattering component.

TABLE VII
Reflector Savings for Eight Inches of Water, Plexiglas,
and 3Wt%-Enriched Uranyl Nitrate

Reflector	Interface Material	Critical Length, cm	Reflector Savings cm
None	---	32.41 ± 0.05	---
Water	61-mil (1.5-mm) Stainless Steel	29.67 ± 0.05	2.74 ± 0.07
$\text{UO}_2(\text{NO}_3)_2$	61-mil (1.5-mm) Stainless Steel	27.63 ± 0.08	4.78 ± 0.09
Plexiglas	61-mil (1.5-mm) Stainless Steel	29.06 ± 0.05	3.35 ± 0.07
Plexiglas	None	28.75 ± 0.05	3.66 ± 0.07

SUMMARY

Criticality data from experiments with PuO_2 -polystyrene mixtures (H:Pu atomic ratio = 15) are given and applied in an analysis to obtain critical dimensions of basic geometrical shapes. Thus, the infinite slab thicknesses for bare and Plexiglas-reflected 2.2% Pu^{240} compacts are 16.09 ± 0.41 and 6.03 ± 0.17 cm, respectively; corresponding values for the 8.0% Pu^{240} compacts are 18.47 ± 0.40 and 7.41 ± 0.18 cm. Converting the experimental data to an equivalent Pu^{239} -water mixture (H:Pu = 15, Pu density = 1.62 g/cm^3) resulted in infinite slab thicknesses of 11.66 ± 0.30 and 4.36 ± 0.12 cm, respectively, for the bare and water-reflected slabs. Corresponding critical radii for infinitely long cylinders are 10.52 ± 0.16 and 6.54 ± 0.14 cm; radii for critical spheres are 13.81 ± 0.16 and 10.40 ± 0.17 cm. These results are in consistent agreement with those computed from multigroup S_4 theory. Corresponding data evaluated from the

subcritical experiments of Goodwin and Schuske give consistently smaller slab thicknesses, thus indicating a need for additional critical experiments at other H:Pu atomic ratios ranging from 1 to 300.

The reflector savings due to 8 in. of Plexiglas was found to be larger than an equal thickness of water. Placing a 30-mil-thick cadmium sheet at the fuel core-Plexiglas interface reduced the effectively infinite Plexiglas reflector to one of 0.9 ± 0.10 in. (2.3 ± 0.3 cm).

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