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CRITICAL EXPERIMENTS WITH HOMOGENEOUS PuO_2-POLYSTYRENE AT 5H/Pu

KEYWORDS: *fuels, plutonium oxides, polystyrene, compacting, criticality, mass, configuration, plutonium-240, enrichment, reprocessing, compacts*

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The results and analyses are presented from the latest series of experiments in a continuing program for determining the critical parameters of plutonium mixtures having concentrations that are typical of wet powders, precipitates, slurries, and polymers. The initial series of measurements in this program were made on 15 H/Pu fuel having ^{240}Pu isotopic concentrations of 2.2 and 8.08 wt%. The latest experiments were conducted with fuel having a ^{240}Pu isotopic concentration of 11.46 wt% and a H/Pu atom ratio of 5.

Generally, these results indicate that the published values for the critical sizes and masses of plutonium should be increased for the highly concentrated systems. Additional data are needed, however, to better establish the criticality curves in this region.

The 11.46 wt% ^{240}Pu isotopic concentration caused an increase of ~30% in the spherical critical mass of a bare ^{239}Pu-water system at 5 H/Pu. In the reflected system the increase was ~43%.

One of our research programs at the Battelle-Northwest Plutonium Critical Mass Laboratory is concerned with determining the critical parameters of plutonium mixtures having concentrations that are typical of wet powders, precipitates, slurries, and polymers. These types of highly concentrated plutonium mixtures are encountered in plutonium fuels fabrication processes and in the reprocessing of irradiated reactor fuels, and thus present a potential criticality hazard.

Currently, the procedures for criticality control of these types of mixtures are based, primarily, on subcritical experiments.[1] Our objective is to better define the criticality parameters of these types of mixtures by performing critical experiments over the Pu concentration range between 500 g/liter and 10 kg/liter. This corresponds to an H/Pu atom range from ~55 to 1. The effects of geometry and ^{240}Pu isotropic concentration on criticality will be determined during the course of these experiments.

The initial series of measurements[2,3] in this program were made on 15 H/Pu fuel having ^{240}Pu isotopic concentrations of 2.2 and 8.08 wt%. The object of this paper is to present data from our latest series of experiments, which were conducted with fuel having an 11.46 wt% ^{240}Pu isotopic concentration and an H/Pu atom ratio of 5. These experiments were carried out using our remote split-table machine (Fig. 1) and PuO_2-polystyrene fuel compacts, as described in Table I.

Experimental data were obtained from both bare and reflected rectangular parallelepipeds of the PuO_2-polystyrene fuel (Table II). The bare critical assembly was nearly a cube with a 35.84-cm square base and a critical height of 28.82 cm after corrections for external reflection and fuel-block cladding material. The critical mass of this assembly was 85.03 ± 0.2 kg. With our calculational techniques[4,5] we have been able to calculate this critical assembly to within 3 mk in k_{eff}.

The reflected critical assemblies ranged from a near cubic geometry of about 26 × 26 cm by 19 cm in height to a thin slab ~9-cm thick. Although corrections to these assemblies for stacking voids and cladding material do cause changes in the dimensions of these assemblies, the critical masses were found to remain essentially unchanged. The reason is that, in these relatively fast systems, the fuel density reduction caused by stacking voids and the cladding tape was offset by the increased neutron moderation caused by the cladding material.

TABLE I

Description of PuO_2-Polystyrene Compacts

Dimension of compacts without cladding (length × width × height), cm	5.12 × 5.12 × 3.81
Average dimension of stacked compacts with cladding (length × width × height), cm	5.21 × 5.16 × 3.88
Average thickness of cladding, cm	0.0254
Composition of cladding, atoms per barn-cm H	1.777×10^{-2}
C	1.184×10^{-2}
Cl	0.592×10^{-2}
Composition of compacts, atoms per barn-cm ^{239}Pu	4.986×10^{-3}
^{240}Pu	6.623×10^{-4}
^{241}Pu	1.382×10^{-4}
^{242}Pu	1.026×10^{-5}
O	1.159×10^{-2}
C	2.920×10^{-2}
H	2.920×10^{-2}
PuO_2 particle size, mm maximum	0.0250
mean	0.0088
minimum	0.0030
Plutonium density, g/cm^3	2.302

TABLE II

Experimental Data from PuO_2-Polystyrene Compacts at 2.302 gPu/cm^3 and 5H/Pu

Reflector	Critical Dimensions, cm Length	Width	Height	Critical Mass, kg of Pu Experimental	Corrected[a]
	11.46 wt% ^{240}Pu				
Plexiglas	25.88	25.88	19.04 ± 0.01	27.66 ± 0.01	27.66 ± 0.01
Plexiglas	31.24	30.96	14.77 ± 0.02	30.94 ± 0.04	30.94 ± 0.04
Plexiglas	41.66	41.28	11.03 ± 0.01	41.20 ± 0.03	41.20 ± 0.03
Plexiglas	52.07	51.60	9.38 ± 0.01	54.78 ± 0.02	54.78 ± 0.02
Bare	36.59	36.11	28.60 ± 0.05	82.88 ± 0.13	--
Bare[a]	35.84	35.84	28.82 ± 0.05	--	85.03 ± 0.2

[a]Data are corrected for voids and tape claddings; bare assemblies are corrected also for effects of structural supports.

It turns out that the critical height in the reflected systems is a linear inverse function of the core cross-sectional area. Thus, by correcting these dimensions for cladding material and stacking voids and plotting as in Fig. 2, the critical thickness of 5.88 ± 0.21 cm for an infinite slab of this material can be obtained by a least-squares fit extrapolation to $1/A = 0$.

For establishing criticality limits and for checking nuclear constants and calculational techniques, these slab data need to be expressed as equivalent spherical, cylindrical, and cubic geometries. Since our calculational techniques reproduced the bare critical assembly of PuO_2-polystyrene to within 3 mk, it was felt that the extrapolation distance for the bare assembly could be calculated with a high degree of confidence. By using the calculated extrapolation distance of 2.98 cm for the bare assembly and the corrected measured critical dimensions given in Table II, the critical buckling for the 5H/Pu plutonium-oxide-polystyrene material was determined as 194.5 ± 4/m^2. By equating the critical dimensions of each reflected assembly to this critical buckling, extrapolation distances were obtained for each

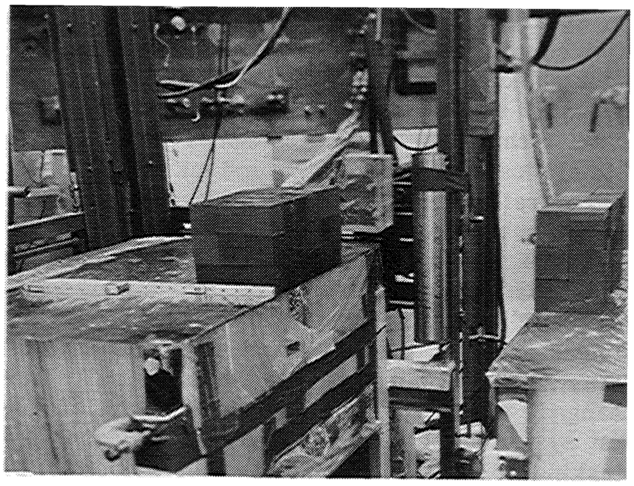

Fig. 1. Remote split-table machine.

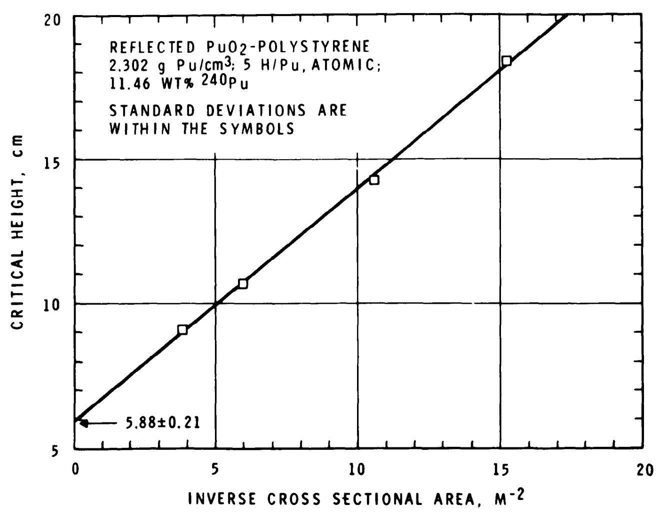

Fig. 2. Measured critical height.

assembly. These, and the critical heights corresponding to each assembly's horizontal buckling, are shown in Fig. 3. A linear least-squares fit extrapolation of these points to zero buckling results in an extrapolation distance of 8.27 cm and a corresponding critical thickness of 5.98 cm for the reflected infinite slab. This is within 2% of the measured critical thickness of 5.88 cm. Also, the extrapolation distance is only 0.05 cm less than that obtained by equating the measured critical thickness to a buckling of $194.5/m^2$. Consequently, the experimental data were converted at this constant buckling to obtain critical sizes for spherical, cylindrical, and cubic geometries. These derived critical sizes are shown in Table III for both the bare and reflected systems. The standard deviations on the calculated extrapolation distances are estimated to be ± 0.2 cm. The remaining standard deviations were obtained by propagation of errors.

The critical data for plutonium-polystyrene at the Pu density of 2.302 g/cm^3 have been corrected for density and for the effects of the more symmetrical scatterer carbon to obtain critical sizes for Pu-water and PuO_2-water systems at their theoretical densities corresponding to an atom ratio of 5 H/Pu. The values shown in Table III have also been corrected to zero ^{240}Pu content. To obtain a better perspective of these data, a comparison of spherical critical mass is made in Fig. 4 with data from TID-7028 and with calculated values obtained by using GAMTEC-II[5] 18-group constants with the transport theory code DTF IV.[4] Since systems in this Pu concentration range are more apt to occur as PuO_2 mixtures, derived data for theoretical density PuO_2-water at 5 H/Pu are also shown. Also shown in Fig. 4 is the poisoning effect of the ^{240}Pu and Pu-water from previously published data[2,3] on 15 H/Pu material.

In this intermediate neutron energy system, the presence of 11.46 wt% ^{240}Pu causes an increase in the critical mass of ~30% in the bare case and 43% in the reflected. However, caution should be exercised in utilizing this increase when establishing criticality safety limits, since it can be easily nullified by fuel lumping in an actual plant-type situation.

In a reflected cubic assembly whose neutron flux has a large thermal component, core leakage and the importance of neutron thermalization in the reflector is not as great as in a relatively fast-neutron energy system. Consequently, the

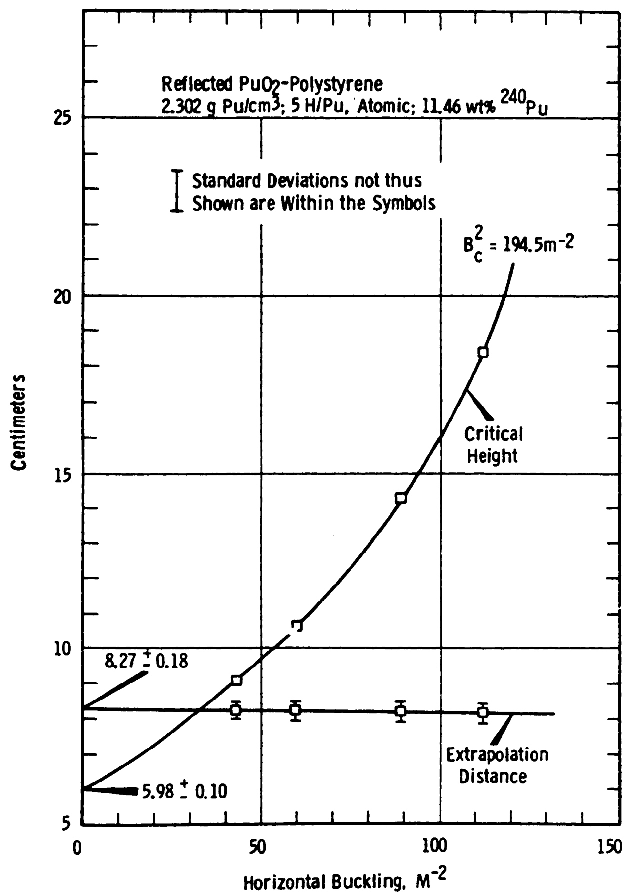

Fig. 3. Critical height and extrapolation distances as a function of horizontal buckling.

TABLE III

Critical Dimensions in Spherical, Cylindrical, Cubic and Slab Geometries at 5 H/Pu Atomic Ratio

Geometry	PuO_2-Polystyrene 2.302 g Pu/cm³ 11.46 wt% ^{240}Pu[a] λ cm	PuO_2-Polystyrene 2.302 g Pu/cm³ 11.46 wt% ^{240}Pu[a] X^b cm	^{239}Pu-water, 4.158 g Pu/cm³ 0.0 wt% ^{240}Pu X^b cm	$^{239}PuO_2$-Water, 3.464 g Pu/cm³, 0.0 wt% ^{240}Pu X^b cm
	Bare Assemblies			
Infinite slab	2.85 ± 0.2	16.83 ± 0.31	9.30 ± 0.17	10.60 ± 0.20
Sphere	2.84 ± 0.2	19.68 ± 0.23	11.29 ± 0.13	12.56 ± 0.14
Infinite cylinder	2.84 ± 0.2	14.40 ± 0.22	8.15 ± 0.12	9.11 ± 0.14
Cube	2.98 ± 0.2	33.05 ± 0.35	--	--
	Reflected Assemblies[c]			
Infinite slab	8.27 ± 0.18	5.88 ± 0.21	2.71 ± 0.05	3.24 ± 0.06
Sphere	8.37 ± 0.35	14.15 ± 0.37	8.39 ± 0.13	9.34 ± 0.14
Infinite cylinder	8.31 ± 0.35	8.93 ± 0.36	5.12 ± 0.21	5.77 ± 0.23
Cube	8.31 ± 0.35	22.39 ± 0.54	--	--

[a]Isotopic concentration.
[b]Critical thickness of slab or cube and critical radius of sphere or cylinder.
[c]Polystyrene assemblies reflected with Plexiglas; water systems reflected with water.

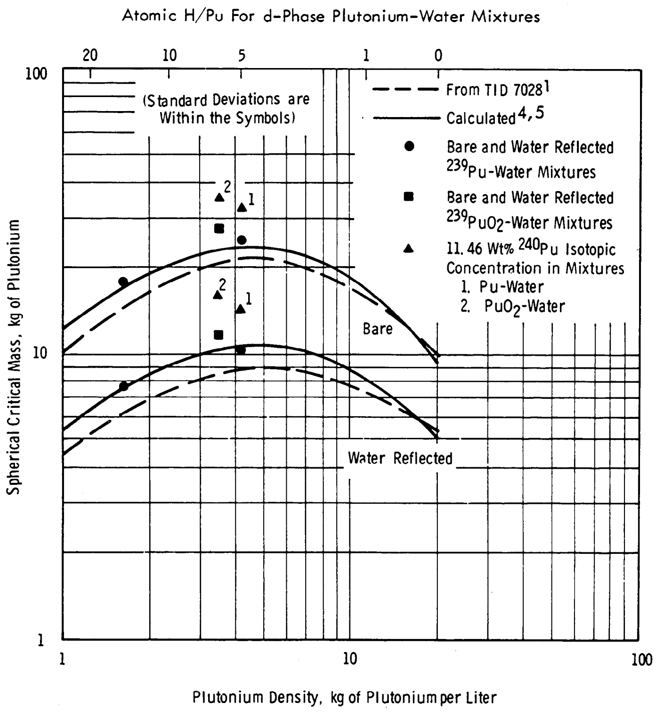

Fig. 4. Data derived from critical slabs of PuO_2-polystyrene.

corners of a cube do not make a significant contribution to the reactivity of the assembly, and, in fact, a sphere is the optimum geometry, since it has the lower surface- (and thus lower leakage-) to-mass ratio. However, in relatively fast systems, such as these 5 H/Pu assemblies, core leakage is greater, and neutrons that are thermalized in the reflector and returned to the core are worth considerably more in the vicinity of the corners. This could result in a reflected cube having a smaller critical mass than a reflected sphere of the same material. For the 5 H/Pu material used in these experiments, and in the previous experiments conducted with 15 H/Pu material, the reflected critical mass of a cube has been observed to be less than that for a reflected sphere of the same material. However, this phenomenon has never been so pronounced that it could not be accounted for by the inaccuracies in the measurements. It is anticipated that currently planned experiments with unmoderated PuO_2 will either verify or disprove the existence of this phenomenon.

The results from the experiments to date indicate that the values for the critical sizes and masses of plutonium, given in such references as TID-7028, should be increased over the intermediate neutron energy range. However, additional data are needed at H/Pu ratios around 30 and 55 and in the vicinity of Pu metal for a better definition of the variation in critical mass with concentration and ^{240}Pu content. Experiments that will provide these data are planned.

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REFERENCES

1. H. C. PAXTON, J. T. THOMAS, DIXON CALLIHAN, and E. B. JOHNSON, "Critical Dimensions of Systems Containing U^{235}, Pu^{239}, and U^{233}," TID-7028, U.S. Dept. of Commerce (June 1964).

2. C. R. RICHEY, J. D. WHITE, R. C. LLOYD, and E. D. CLAYTON, "Homogeneous Critical Experiments With Plutonium/Plastic Mixtures in the Intermediate Energy Range," *Trans.Amer. Nucl. Soc.*, **7**, 81 (1964).

3. C. R. RICHEY, J. D. WHITE, E. D. CLAYTON, and R. C. LLOYD, "Criticality of Homogeneous Plutonium Oxide-Plastic Compacts at H:Pu = 15," *Nucl. Sci. Eng.*, **23**, 150 (1965).

4. B. G. CARLSON, W. J. WORLTON, G. GUBER, and M. SHAPIRO, *DTF Users Manual I & II*, United Nuclear Corporation, White Plains, New York (1963).

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