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# CRITICAL EXPERIMENTS WITH UNMODERATED PLUTONIUM OXIDE

REACTORS

S. R. BIERMAN and E. D. CLAYTON
*Battelle Memorial Institute, Pacific Northwest Laboratory*
*Richland, Washington 99352*

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*The results and analyses presented are from the latest series of experiments in a continuing program for determining the critical parameters of plutonium mixtures having concentrations typical of wet powders, precipitates, slurries, and polymers. Previous measurements in this program were made on 15 H/Pu fuel having $^{240}$Pu isotopic concentrations of 2.2 and 8.08 wt% and on 5 H/Pu fuel having a $^{240}$Pu isotopic concentration of 11.46 wt%. This latest series of experiments was conducted with fuel in which the $^{240}$Pu isotopic content has been increased to 18.35 wt% and the H/Pu atomic ratio decreased to essentially zero. The minimum critical slab thickness for a water-reflected homogeneous $PuO_2$-water system of this composition was determined to be 2.31 ± 0.06 cm as compared to only 1.15 ± 0.03 cm for Pu-water. Thus, having the plutonium in an oxide form at this degree of moderation results in an increase of about a factor of 2 in the critical thickness. For spherical geometry, the difference in critical mass between $PuO_2$ and Pu systems is about 2. Also, in the fast neutron spectrum of this fuel, the percent change in spherical critical mass per percent change in $^{240}$Pu content was determined to be 2.07 for the reflected case and 1.88 for the bare case. In general, results from these experiments indicate that the values for the critical sizes and masses of plutonium given in references such as TID-7028 should be increased for the highly concentrated systems.*

Part of the research effort at the Battelle-Northwest-operated Critical Mass Laboratory is concerned with determining the criticality parameters of plutonium mixtures over the entire range of neutron moderation. The cumulative results of this effort up through the early part of 1969, both experimentally and theoretically, were recently published.[1] The object of this paper is to present data from a later series of critical experiments in the undermoderated region of the neutron spectrum. In this region the plutonium mixtures are highly concentrated, typifying wet powders, precipitates, slurries, and polymers encountered in plutonium fuels fabrication and reprocessing. The experiments were carried out using our remote split-table machine[2] and essentially dry, unmoderated $PuO_2$ (0.04 H/Pu atomic ratio) containing 18.35 wt% $^{240}$Pu in the plutonium. The $PuO_2$ was in the form of 2- × 2- × $1\frac{1}{2}$-in. blocks, each compressed to a density of 5.762 ± 0.039 g Pu/cm$^3$ and encased, for contamination control, in a heat-shrinkable polyolefin plastic. A detailed description of the fuel is presented in Table I.

Experimental data were obtained from both bare and reflected parallelepipeds of $PuO_2$. The critical sizes of each of these assemblies are presented in Table II. The dimensions of each assembly have had experimentally determined corrections made to account for the effects of stacking voids and the cladding material used on each fuel block for contamination control. Thus, each critical assembly shown in Table II represents a solid mass of fuel at a density of 5.762 g Pu/cm$^3$. As observed in previous experiments[3] with $PuO_2$-polystyrene fuel having an H/Pu atomic ratio of 5, the critical mass of each of these assemblies shown in Table II was not affected by the stacking voids and cladding material. In these relatively fast systems, the fuel density reduction caused by stacking voids and cladding was offset by the increased neutron moderation caused by the cladding material.

The critical sizes shown in Table II also reflect corrections made for temperature changes during the course of the experiments. Because of the

TABLE I

Description of Fuel

| | |
|---|---|
| Dimension of compacts without cladding length × width × height | 5.13 × 5.13 × 3.81 ± 0.02 cm |
| Average dimension of stacked compacts with cladding length × width × height | 5.54 × 5.54 × 4.09 ± 0.04 cm |
| Average thickness of cladding (cm) | |
| Sides | 0.38 |
| Top | 0.13 |
| Bottom | 0.13 |
| Composition of cladding [atoms/(b cm)] | |
| H | $4.614 \times 10^{-2}$ |
| C | $2.906 \times 10^{-2}$ |
| O | $2.309 \times 10^{-2}$ |
| Composition of compacts [atoms/(b cm)] | |
| $^{238}Pu$ | $3.383 \times 10^{-5}$ |
| $^{239}Pu$ | $1.092 \times 10^{-2}$ |
| $^{240}Pu$ | $2.654 \times 10^{-3}$ |
| $^{241}Pu$ | $7.269 \times 10^{-4}$ |
| $^{242}Pu$ | $1.632 \times 10^{-4}$ |
| O | $3.094 \times 10^{-2}$ |
| H | $5.511 \times 10^{-4}$ |
| $PuO_2$ particle size (mm) | |
| Maximum | 0.0400 |
| Mean | 0.0074 |
| Minimum | 0.0025 |
| Plutonium density (g/cm$^3$) | 5.762 ± 0.039 |

relatively high plutonium density and $^{240}$Pu content, a high specific heat generation (about 32 W/liter or 5 W/kg) was obtained and resulted in temperature variations of as much as 30°C during the course of a single experiment. To obtain consistent sets of data, the count rates in each critical approach were corrected to 50°C by a technique developed by Lane and Perkins[4] at Aldermaston, United Kingdom, shown below

$$K = \frac{N}{T_1 - T_2}\left(\frac{1}{C_1} - \frac{1}{C_2}\right) ,$$

where

$N$ = number of fuel blocks

$T_1$ = temperature corresponding to count rate $C_1$

$T_2$ = temperature corresponding to count rate $C_2$

TABLE II

Experimental Data from $PuO_2$ Compacts at 5.762 g Pu/cm$^3$ 18.35 wt% $^{240}$Pu Isotopic Content 5.762 g Pu/cm$^3$, 0.04 H/Pu Atomic Ratio, 50°C

| | Critical Dimensions (cm) | | | Critical Mass (kg Pu) |
|---|---|---|---|---|
| Reflector | Length | Width | Height[a] | 18.35 wt% $^{240}$Pu |
| Plexiglas | 25.65 | 25.65 | 10.03 ± 0.01 | 38.3 ± 0.3 |
| Plexiglas | 25.65 | 30.78 | 8.98 ± 0.01 | 41.1 ± 0.3 |
| Plexiglas | 30.78 | 30.78 | 7.97 ± 0.03 | 43.8 ± 0.3 |
| Plexiglas | 30.78 | 41.05 | 6.86 ± 0.08 | 50.3 ± 0.7 |
| Plexiglas | 41.05 | 41.05 | 5.95 ± --- | 58.2 ± 0.3 |
| Bare | 30.78 | 30.78 | 20.90 ± 0.09 | 113.8 ± 0.9 |
| Bare | 30.78 | 30.78 | 21.12 ± 0.06 | 115.1 ± 0.8 |

[a]Critical height corrected for voids, cladding, and temperature effects. Bare assemblies also corrected for effects of structural supports.

$K$ = change in reciprocal count rate per degree change in temperature for $N$ fuel blocks.

Although there was an insufficient amount of fuel to achieve criticality unreflected, two bare critical assemblies were built using driver regions of $PuO_2$-polystyrene having an H/Pu atomic ratio of 5. (Experiments with this 5 H/Pu fuel have been previously reported.[3]) Each of the bare assemblies had the same cross-sectional dimensions but different amounts of driver fuel. Based on these measurements, a bare assembly of 5.762 g Pu/cm$^3$ of $PuO_2$, 30.78 × 30.78 × 20.99 ± 0.22 cm containing 114.6 ± 1.4 kg of plutonium would be critical. With our calculational techniques[5,6] we have been able to calculate this bare critical assembly to within 6 mk in $k_{eff}$. Consequently, it was felt that the extrapolation distance for the bare assembly could be calculated with a high degree of confidence. By using a calculated extrapolation distance of 3.57 cm for the bare assembly and the bare critical dimensions given above, the critical buckling for this essentially unmoderated $PuO_2$ material was determined to be 262.0 ± 1.7 m$^{-2}$.

The reflected critical assemblies ranged over a series of slabs from about 10 to 6 cm in thickness to permit determining the critical thickness of a slab of this fuel, infinite in two dimensions. The critical dimensions of each of these reflected assemblies are shown in Table II. The critical height of each of these assemblies is plotted in Fig. 1 as an inverse function of its core cross-sectional area. A linear least-squares fit extrapolation of this data to an inverse cross-sectional area of zero yields a critical thickness of 3.34 ± 0.10 cm for a slab of this essentially unmoderated

$PuO_2$ material infinite in two dimensions. A corresponding extrapolation distance of 8.20 ± 0.22 cm was obtained for the reflected infinite slab by equating the critical dimensions of each reflected assembly to the critical buckling of 262.0 ± 1.7 $m^{-2}$ determined for this material from the bare assembly. The extrapolation distances thus obtained for each reflected assembly are also shown in Fig. 1 as an inverse function of the respective assembly core cross-sectional area.

For establishing criticality limits and for checking nuclear constants and calculational techniques, these slab data need to be expressed in equivalent spherical, cylindrical, and cubic geometries. Consequently, the experimental data were converted at the critical buckling of 262.0 ± 1.7 $m^{-2}$ to obtain critical sizes for these geometries, both bare and fully reflected. These derived critical sizes are shown in Table III. The standard deviations on the calculated extrapolation distances are estimated to be ±0.2 cm. The remaining standard deviations shown were obtained by propagation of errors. Also shown in Table III are critical sizes for Pu-water and $PuO_2$-water at their theoretical density corresponding to an 0.04 H/Pu atomic ratio. These were obtained by making density corrections to the 5.762 g Pu/cm$^3$ $PuO_2$ data.

Values are also shown in Table III which have been corrected to zero $^{240}$Pu content to show the relatively small effect that $^{240}$Pu has on the critical size of this highly concentrated system. In this high neutron energy system the percent change in spherical critical mass per percent change in $^{240}$Pu content is only 2.07 for the re-

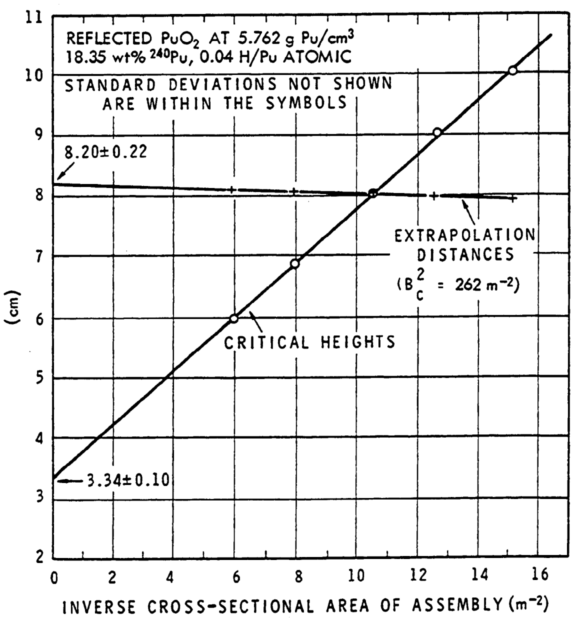


Fig. 1. Extrapolation distances and measured critical height.

TABLE III

Critical Dimensions in Spherical, Cylindrical, Cubical, and Slab Geometries at H/Pu Atomic Ratio of 0.04

| Geometry | $PuO_2$-Water 5.76 g Pu/cm$^3$ 18.35 wt% $^{240}$Pu[a] λ cm | $PuO_2$-Water 5.76 g Pu/cm$^3$ 18.35 wt% $^{240}$Pu[a] $X^b$ cm | $^{239}$Pu-Water 19.16 g Pu/cm$^3$ 0.0 wt% $^{240}$Pu $X^b$ cm | $^{239}PuO_2$-Water 9.96 g Pu/cm$^3$ 0.0 wt% $^{240}$Pu $X^b$ cm | Pu-Water 19.16 g Pu/cm$^3$ 18.35 wt% $^{240}$Pu[a] $X^b$ cm | $PuO_2$-Water 9.96 g Pu/cm$^3$ 18.35 wt% $^{240}$Pu[a] $X^b$ cm |
|---|---|---|---|---|---|---|
| | Bare Assemblies | | | | | |
| Infinite slab | 3.62 ± 0.2 | 12.17 ± 0.28 | 3.71 ± 0.09 | 6.38 ± 0.14 | 4.13 ± 0.00 | 7.07 ± 0.16 |
| Sphere | 3.60 ± 0.2 | 15.81 ± 0.20 | 5.20 ± 0.07 | 8.41 ± 0.11 | 5.74 ± 0.07 | 9.22 ± 0.12 |
| Infinite cylinder | 3.58 ± 0.2 | 11.28 ± 0.20 | 3.60 ± 0.06 | 5.96 ± 0.11 | 3.99 ± 0.07 | 6.57 ± 0.12 |
| Cube | 3.57 ± 0.2 | 26.48 ± 0.28 | --- | --- | --- | --- |
| | Reflected Assemblies[c] | | | | | |
| Infinite Slab | 8.20 ± 0.22 | 3.34 ± 0.10 | 0.82 ± 0.02 | 1.60 ± 0.04 | 1.15 ± 0.03 | 2.31 ± 0.06 |
| Sphere | 8.17 ± 0.38 | 11.24 ± 0.38 | 4.22 ± 0.14 | 6.57 ± 0.22 | 4.70 ± 0.16 | 7.39 ± 0.25 |
| Infinite cylinder | 8.17 ± 0.38 | 6.69 ± 0.31 | 2.38 ± 0.11 | 3.86 ± 0.17 | 2.72 ± 0.13 | 4.48 ± 0.21 |
| Cube | 8.13 ± 0.38 | 17.36 ± 0.54 | --- | --- | --- | --- |

[a]Isotopic concentration.
[b]Critical thickness of slab or cube, critical radius of sphere or cylinder.
[c]5.762 $PuO_2$-water reflected with Plexiglas, other systems reflected with water.

flected case and 1.88 for the bare case, as compared to values of about 10 and 12 for a more thermalized system. A calculated neutron energy spectrum for the $PuO_2$ fuel used in the experiments is shown in Table IV.

As with previously published data[2,3] for 15 and 5 H/Pu material, these data for essentially unmoderated systems are compared in Figs. 2 through 5 with data[7] from TID-7028 for slab, cylindrical, and spherical geometries. In all three

TABLE IV

Calculated 18-Group Flux for $PuO_2$

5.762 g Pu/cm$^3$, 0.04 H/Pu Atomic Ratio
18.35 wt% $^{240}$Pu Isotopic Content

| Group | Lower Energy | Relative Flux |
|---|---|---|
| 1 | 7.79 MeV | 0.0036 |
| 2 | 6.07 | 0.0103 |
| 3 | 4.72 | 0.0247 |
| 4 | 3.68 | 0.0398 |
| 5 | 2.87 | 0.0621 |
| 6 | 1.74 | 0.1596 |
| 7 | 1.35 | 0.0847 |
| 8 | 183.00 keV | 0.4789 |
| 9 | 24.80 | 0.1260 |
| 10 | 3.36 | 0.0094 |
| 11 | 454.00 eV | 0.0007 |
| 12 | 101.00 | |
| 13 | 37.30 | |
| 14 | 13.70 | |
| 15 | 5.04 | 0.0002 |
| 16 | 1.86 | |
| 17 | 0.683 | |
| 18 | 0 | |

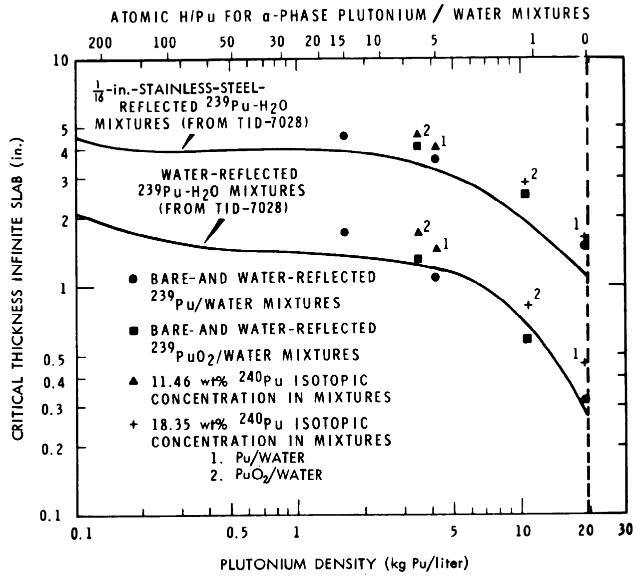


Fig. 2. Data derived from critical slabs of $PuO_2$-polystyrene atomic H/Pu for $\alpha$-phase plutonium/water mixtures.

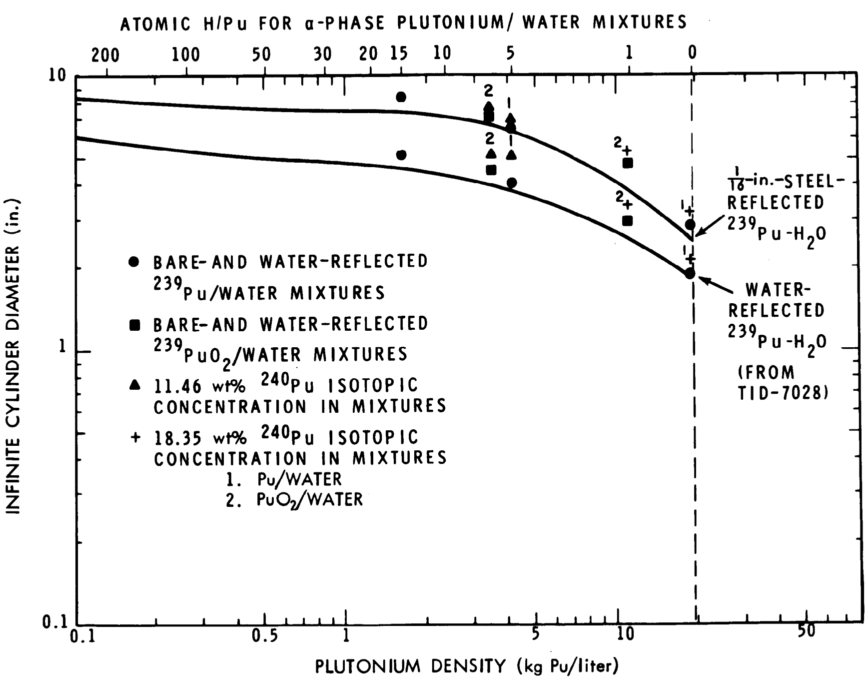


Fig. 3. Data derived from critical slabs of $PuO_2$-polystyrene atomic H/Pu for $\alpha$-phase plutonium/water mixtures.

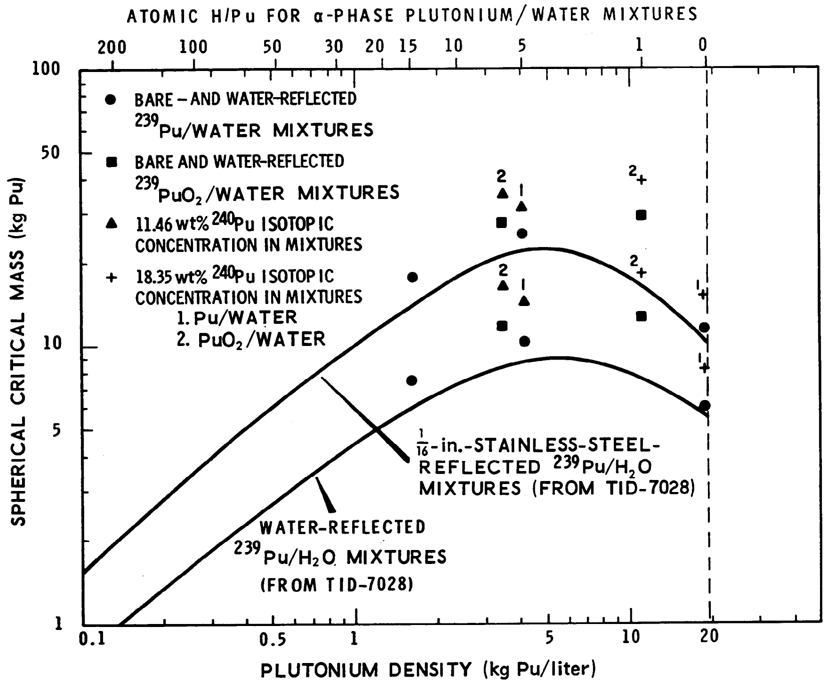


Fig. 4. Data derived from critical slabs of $PuO_2$-polystyrene atomic H/Pu for α-phase plutonium/water mixtures.

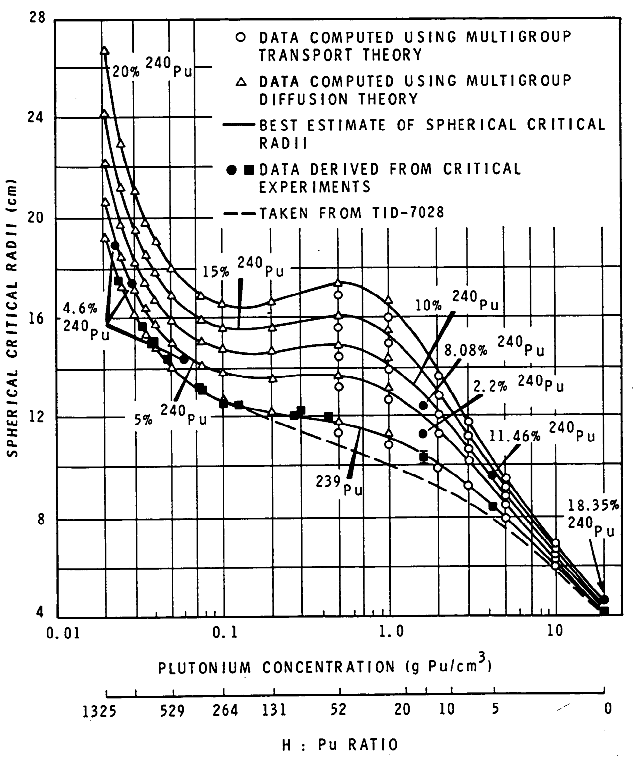


Fig. 5. Water-reflected spherical critical radii of Pu(metal)/water mixtures.

geometries the critical sizes derived from the experiments are slightly larger than those shown in TID-7028. Calculated values for the spherical critical mass and radii obtained by using GAMTEC-II 18-group constants[5] with the transport theory code[6] DTF-IV also are shown in Fig. 5.

It was anticipated that these experiments with essentially unmoderated $PuO_2$ would either verify or disprove that a reflected cube could have a smaller critical mass than a reflected sphere. The reflected critical mass of a cube of this essentially unmoderated $PuO_2$ was observed to be less than that for a reflected sphere of the same material. However, as with previous observations with 5 H/Pu and 15 H/Pu,[2,3] this phenomenon was not so pronounced that it could not be accounted for by inaccuracies in the measurements.

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

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