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CRITICAL EXPERIMENTS WITH HOMOGENEOUS PuO_2-POLYSTYRENE AT 50 H/Pu

REACTORS

KEYWORDS: *plutonium oxides, polystyrene, isotope-enriched materials, critical mass, plutonium-240, critical size, criticality*

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The results and analyses are presented from the latest series of experiments for determining the critical parameters of plutonium mixtures over the undermoderated region of the neutron energy spectrum. This latest series of experiments was conducted with PuO_2-polystyrene fuel having an atomic H/Pu ratio of 49.6 and a ^{240}Pu isotopic content of 18.53 wt%. In addition to providing basic criticality data, the experiments were designed to (a) establish the degree of correlation between the PuO_2-polystyrene and the plutonium nitrate solution fueled critical experiments performed at the Hanford Critical Mass Laboratory, (b) provide data for checking previously reported differences obtained by the neutron diffusion theory code HFN and the neutron transport theory code DTF-IV in calculating critical sizes of plutonium-water systems between about 500 and 1000 g Pu/liter, and (c) substantiate that high exposure, subcritical, fixed volume, plutonium-water systems can be made critical by dilution with water.

In contrast to the data previously obtained on this same PuO_2 material unmoderated, the density effect of having the plutonium in the oxide form was found to have essentially no effect on the reactivity at 49.6 H/Pu. At 49.6 H/Pu, the minimum critical slab thickness was determined to be 5.22 ± 0.07 cm for ^{239}Pu-H_2O as compared to 5.34 ± 0.07 cm for $^{239}PuO_2$-H_2O. However, the neutron poisoning effect of the ^{240}Pu at 49.6 H/Pu was at least five times that observed for the PuO_2 unmoderated. At this near-optimum concentration for maximum ^{240}Pu effects, the percent change in spherical critical mass per percent change in ^{240}Pu content was determined to be 12.1 for the reflected case and 10.2 for the bare case.

Based on the data derived from these experiments at 49.6 H/Pu, the HFN code was found to accurately calculate the plutonium-water systems at 521 g Pu/liter and should be used at this and lower concentrations in preference to the DTF-IV code for calculating critical parameters. As with the plutonium nitrate solutions at high concentrations, the DTF-IV neutron transport theory code calculations resulted in too small a critical size; however, the data obtained from the PuO_2-polystyrene experiments correlated well with those that have been obtained from plutonium nitrate solutions. The experiments also showed that, for high exposure plutonium-water systems, a given size vessel could be just critical ($k_{eff} = 1.0$) at three different concentrations below about 1000 g Pu/liter. Consequently, a critically safe vessel could be made critical by dilution alone if the ^{240}Pu content was high enough.

INTRODUCTION

Part of the research effort at the Battelle-Northwest operated Critical Mass Laboratory is concerned with determining the critical parameters of plutonium mixtures over the entire range of neutron moderation. To the extent possible, plutonium nitrate solutions have been used as the experimental fuel, since it offers the advantages of theoretical densities, easily varied fuel compositions, and easily varied geometries. However, the practical concentration limit for which plutonium nitrate solutions are stable is <500 g Pu/liter. Consequently, to obtain experimental data in that portion of the neutron energy spectrum characteristic of concentrations above 500 g Pu/liter, homogeneous mixtures of PuO_2-polystyrene have had to be used.

Experimental results have been reported for plutonium nitrate solutions[1,2] having concentrations between about 25 and 435 g Pu/liter and for PuO_2-polystyrene fueled assemblies[3-5] ranging in H/Pu atomic ratios corresponding to essentially unmoderated PuO_2 down to 1620 g Pu/liter. To obtain a closer tie-in of data between these solution- and solid-type experiments, a series of critical experiments was performed with a homogeneous mixture of PuO_2-polystyrene at an H/Pu atomic ratio corresponding to a theoretical density, plutonium-water concentration of 521 g Pu/liter (49.6 H/Pu). Results of these latest series of experiments are presented in this paper.

Data in this concentration range between 500 and 1000 g Pu/liter are also needed to resolve the differences[6] observed between diffusion theory and transport theory computations using the computer codes HFN[7] and DTF-IV.[8] The diffusion theory code, HFN, has been shown to be quite accurate for concentrations below 300 g Pu/liter, and indications are it is accurate up to about 500 g Pu/liter; however, it calculates too large a critical size above, at least, 1600 g Pu/liter. Conversely, the transport theory code, DTF-IV, has been shown to be quite accurate for concentrations above 1600 g Pu/liter but calculates a smaller critical size than HFN for concentrations between about 300 and 1600 g Pu/liter.

Calculations have also indicated that, for plutonium systems having a ^{240}Pu isotopic content greater than 5 wt%, there are three different plutonium concentrations below about 1000 g Pu/liter for which k_{eff} is unity[6] in the same sized vessel. That is, for plutonium-water systems having a ^{240}Pu isotopic content greater than 5 wt%, a subcritical plutonium concentration exists which can be made critical by dilution alone. Critical experiments with plutonium nitrate solutions[9] having a ^{240}Pu isotopic content of 23.2 wt% have tended to substantiate this phenomenon in that a smaller critical slab thickness was required for 202 g Pu/liter solution than for 284 g Pu/liter solution. However, in this concentration range the critical size is not very sensitive to concentration changes. To provide additional data on this aspect of the effects of ^{240}Pu on criticality, as well as to provide data on the neutron poisoning effects of ^{240}Pu, the experimental fuel was fabricated using PuO_2 having a relatively high ^{240}Pu content.

EXPERIMENTS AND DATA REDUCTION

The experiments were carried out using our remote split-table machine[3] and PuO_2-polystyrene in the form of 2- × 2- × 2-in. and 2- × 2- × $\frac{1}{2}$-in. blocks, each compressed to a density of 367 ± 4 g Pu/liter and encased, for contamination control, in 6-mil-thick tape (MM&M #471). The fuel blocks were fabricated with PuO_2 having a ^{240}Pu isotopic content of 18.35 ± 0.22 wt%. A detailed description of the fuel is presented in Table I. The reflected assemblies were fully reflected with 15 cm of Plexiglas and its composition is also given in Table I.

Experimental data were obtained at room temperature from both bare and Plexiglas-reflected parallelepipeds. The critical sizes of each of these assemblies are presented in Table II. Replacement type experiments were performed to experimentally determine the corrections required 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 49.6 ± 0.7 H/Pu fuel at a density of 367 ± 4 g Pu/liter.

TABLE I

Description of Fuel

Dimension of 2-in. cubes without cladding, L×W×H, cm	5.09 × 5.09 × 5.09 ± 0.01
Average dimension of stacked 2-in. cubes with cladding, L×W×H, cm	5.19 × 5.19 × 5.16 ± 0.04
Average thickness of cladding, cm	0.018
Composition of cladding, atoms/(b cm)	
H	4.489×10^{-2}
C	3.113×10^{-2}
Cl	0.724×10^{-2}
Composition of Plexiglas, atoms/(b cm)	
H	5.712×10^{-2}
C	3.570×10^{-2}
O	1.428×10^{-2}
Composition of compacts, atoms/(b cm)[a]	
^{238}Pu	2.125×10^{-6}
^{239}Pu	6.952×10^{-4}
^{240}Pu	1.689×10^{-4}
^{241}Pu	3.952×10^{-5}
^{242}Pu	1.045×10^{-5}
^{241}Am	7.345×10^{-6}
O	2.082×10^{-3}
H	4.575×10^{-2}
C	4.505×10^{-2}
PuO_2 particle size, mm	
Maximum	0.0070
Mean	0.0012
Minimum	0.0003
Plutonium density, g/cm³	0.367 ± 0.004

[a] Americium-241 content determined 5-27-70. Reflected experimental data obtained by 6-6-70. Bare experimental data obtained 9-9-70.

TABLE II

Experimental Data from PuO_2-Polystyrene Compacts Plutonium at 18.35 wt% ^{240}Pu, 49.6 ± 0.7 H/Pu Atomic, 367 g Pu/liter

Reflector	Critical Dimensions (cm)			Critical Mass
	Length	Width	Height[a]	kg of Pu
Piexiglas	61.08	61.08	16.35 ± 0.06	22.4 ± 0.3
Plexiglas	50.90	61.08	17.48 ± 0.05	19.9 ± 0.2
Plexiglas	50.90	50.90	18.68 ± 0.08	17.7 ± 0.2
Plexiglas	50.90	45.81	19.69 ± 0.07	16.8 ± 0.2
Plexiglas	40.72	45.81	22.06 ± 0.06	15.1 ± 0.2
Plexiglas	40.72	40.72	23.58 ± 0.10	14.3 ± 0.2
Plexiglas	40.72	30.54	29.64 ± 0.12	13.5 ± 0.2
Bare	46.08 ± 0.50	40.96 ± 0.44	49.12 ± 0.50	34.0 ± 0.3

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

Although there was an insufficient amount of fuel to achieve criticality unreflected, a bare critical assembly was built using a driver region of PuO_2-polystyrene having an H/Pu atomic ratio of 5. Experiments with this 5 H/Pu fuel have been previously reported.[4] The worth of the 5 H/Pu driver region was calculated in terms of an equivalent height of the 49.6 H/Pu fuel region. Based on this corrected measurement, a bare assembly 46.08 × 40.96 × 49.12 ± 0.050 cm containing 34.0 ± 0.3 kg of plutonium, would be critical. By using a calculated extrapolation distance of 2.72 ± 0.2 cm for the bare assembly and the bare critical dimensions given above, the critical buckling for this 49.6 H/Pu, 367 g Pu/liter, PuO_2-polystyrene material was determined to be 116.2 ± 1.0 m^{-2}.

The reflected critical assemblies ranged over a series of slabs from about 30 to 16 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 least-squares fit extrapolation of these data to an inverse cross-sectional area of zero yields a critical thickness of 10.87 ± 0.14 cm for a slab of this PuO_2-polystyrene material infinite in two dimensions. A corresponding extrapolation distance of 9.04 ± 0.10 cm was obtained for the reflected infinite slab by equating the critical dimensions of each reflected assembly to the critical buckling of 116.2 ± 1.0 m^{-2} determined for this material for the bare assembly. The extrapolation distances thus obtained for each reflected assembly are also shown in Fig. 1 as an inverse function of the core cross-sectional area of the respective assembly.

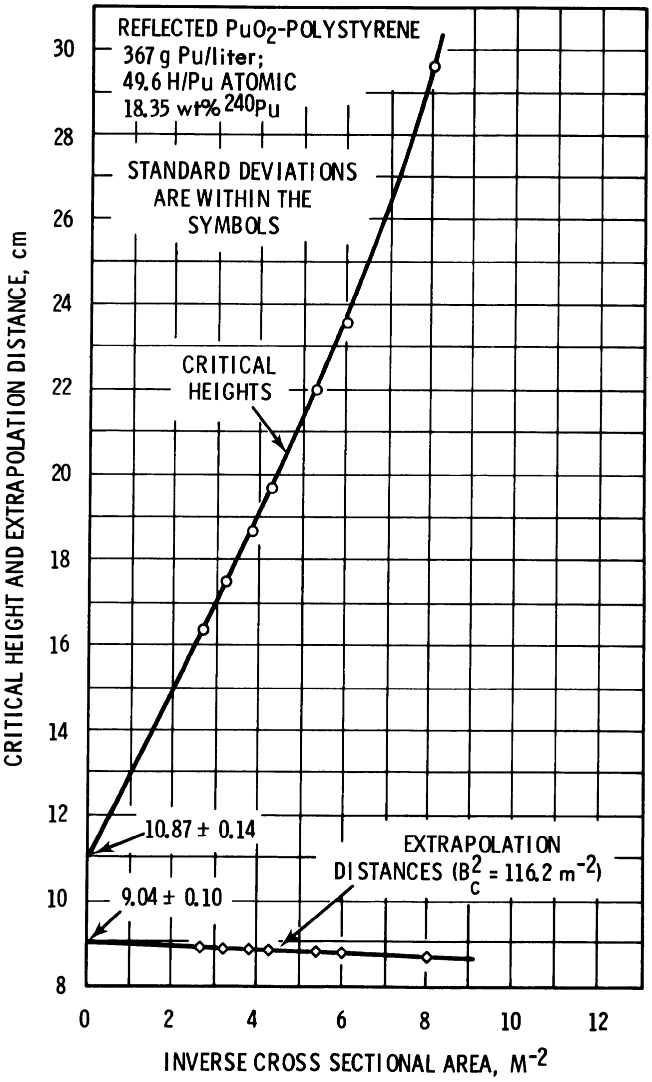

Fig. 1. Extrapolation distances and measured critical heights.

DATA ANALYSIS

For establishing criticality limits and for checking nuclear constants and calculational techniques, these experimental slab data have been expressed in equivalent spherical, cylindrical, and rectangular geometries. The experimental data were converted at the critical buckling of 116.2 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 plutonium-water and PuO^2-H_2O at theoretical densities corresponding to a 49.6

TABLE III

Critical Dimensions in Spherical, Cylindrical, Cubic, and Slab Geometries at 49.6 H/Pu, Atomic Ratio

	PuO_2-Polystyrene 0.367 g Pu/cm^3 18.35 wt% ^{240}Pu[a]		^{239}Pu-H_2O 0.521 g Pu/cm^3 0.0 wt% ^{240}Pu	$^{239}PuO_2$-H_2O 0.508 g Pu/cm^3 0.0 wt% ^{240}Pu	Pu-H_2O 0.521 g Pu/cm^3 18.35 wt% ^{240}Pu	PuO_2-H_2O 0.508 g Pu/cm^3 18.35 wt% ^{240}Pu
Geometry	λ (cm)	X[b] (cm)	X[b] (cm)	X[b] (cm)	X[b] (cm)	X[b] (cm)
Bare Assemblies						
Infinite slab	2.65 ± 0.2	23.85 ± 0.29	13.40 ± 0.16	13.60 ± 0.16	19.55 ± 0.24	19.85 ± 0.24
Sphere	2.65 ± 0.2	26.50 ± 0.21	15.49 ± 0.12	15.69 ± 0.12	22.02 ± 0.17	22.30 ± 0.17
Infinite cylinder	2.64 ± 0.2	19.67 ± 0.21	11.32 ± 0.12	11.48 ± 0.12	16.26 ± 0.17	16.50 ± 0.18
Cube	---	45.04 ± 0.30	---	---	---	---
Reflected Assemblies[c]						
Infinite slab	9.04 ± 0.10	10.87 ± 0.14	5.22 ± 0.07	5.34 ± 0.07	9.43 ± 0.12	9.64 ± 0.12
Sphere	9.12 ± 0.10	20.03 ± 0.12	11.65 ± 0.07	11.81 ± 0.07	17.19 ± 0.10	17.45 ± 0.10
Infinite cylinder	9.08 ± 0.10	13.23 ± 0.12	7.39 ± 0.07	7.51 ± 0.07	11.39 ± 0.10	11.58 ± 0.10
Cube	---	32.08 ± 0.17	---	---	---	---

[a] Isotopic concentration.

[b] Critical thickness of slab or cube, critical radius of sphere or cylinder.

[c] PuO_2-polystyrene reflected with Plexiglas, other systems reflected with water.

H/Pu atomic ratio. These were obtained by making transport theory corrections to the 367 g Pu/liter PuO_2-polystyrene data for density and the presence of carbon.

Also shown in Table III are values which have been corrected to zero ^{240}Pu content to show the large effect that ^{240}Pu has on the critical size at this near-optimum concentration for neutron poisoning with ^{240}Pu. In this relatively thermalized neutron energy system, the percent change in the spherical critical mass of total plutonium per percent change in ^{240}Pu content is 12.1 for the reflected case and 10.2 for the bare case, as compared to values of about 2 for an unmoderated plutonium-water system containing up to 20 wt% ^{240}Pu. A GAMTEC-II[10] calculated neutron energy spectrum, using the original GAMTEC-II cross-sections library, is shown in Table IV for the PuO_2-polystyrene fuel used in the experiments.

TABLE IV

Calculated 18-Group Neutron Flux for PuO_2-Polystyrene

367 g Pu/liter, 49.6 H/Pu Atomic Ratio
18.35 wt% ^{240}Pu Isotopic Content

Group	Lower Energy	Relative Flux
1	7.79 MeV	0.0028
2	6.07	0.0083
3	4.72	0.0184
4	3.68	0.0293
5	2.87	0.0401
6	1.74	0.1094
7	1.35	0.0545
8	183.00 keV	0.2738
9	24.80	0.1152
10	3.36	0.0799
11	454.00 eV	0.0731
12	101.00	0.0512
13	37.30	0.0292
14	13.70	0.0280
15	5.04	0.0245
16	1.86	0.0242
17	0.683	0.0152
18	0	0.0229

For consistency with previously reported experimental results and data presentation on plutonium systems, the original GAMTEC-II cross-section data developed[10] at the Hanford Critical Mass Laboratory were used in analyzing and presenting these current data. Currently, a thorough testing and evaluation of the ENDF/B cross-section data is in progress[11] with these and other previously reported critical experiments. Preliminary k_{eff} values, however, based on ENDF/B-II cross-section data for the bare and the Plexiglas-reflected critical infinite slabs of PuO_2-polystyrene at 49.6 H/Pu are shown in Table V as an initial comparison between the original GAMTEC-II cross sections (CML) and the ENDF/B-II cross sections. In each case, 18-group GAMTEC-II averaged cross sections were used with the diffusion theory code, HFN,[7] and the transport theory code, DTF-IV.[8] The ENDF/B-II cross sections were obtained from the ENDF/B-II point energy library data by using the FLANGE code to process the thermal data and the ETOG code to process the epithermal data for broad group averaging in GAMTEC-II. As can be seen in the table, calculated k_{eff} values using these cross-section data were consistently higher than comparable values obtained from the calculations with the CML cross sections. Both sets of calculations indicate, also, that the bare infinite slab thickness of 23.85 cm may be slightly conservative with respect to criticality safety.

As with previously published data[3-5] from undermoderated PuO_2-polystyrene experiments, these current data are shown graphically in Figs. 2 through 5 along with the previous data. In all three geometries the critical sizes for ^{239}Pu-H_2O derived from the current experiments are slightly larger than those shown in TID-7028[12] and are in general agreement with the previously reported data derived from undermoderated PuO_2-polystyrene experiments.

In Fig. 5, previously reported spherical critical

TABLE V

Comparison Between HFN and DTF-IV Calculations at 49.6 H/Pu Atomic Ratio

Critical Assembly				k_{eff}			
				DTF-IV		HFN	
Core	Reflector	Geometry	Slab Thickness	CML[a]	ENDF/B[b]	CML[a]	ENDF/B[b]
PuO_2-polystyrene at 367 g Pu/liter; 18.35 wt% ^{240}Pu in Pu	none	infinite slab	23.85 ± 0.29	1.028	1.049	1.014	1.041
	full Plexiglas	infinite slab	10.87 ± 0.14	0.998	0.009	0.998	1.004

[a] 18-group GAMTEC-II averaged cross sections using the original GAMTEC-II library.[10]

[b] FLANGE-ETOG-ENDF/B-II 18-group GAMTEC-II averaged cross sections.

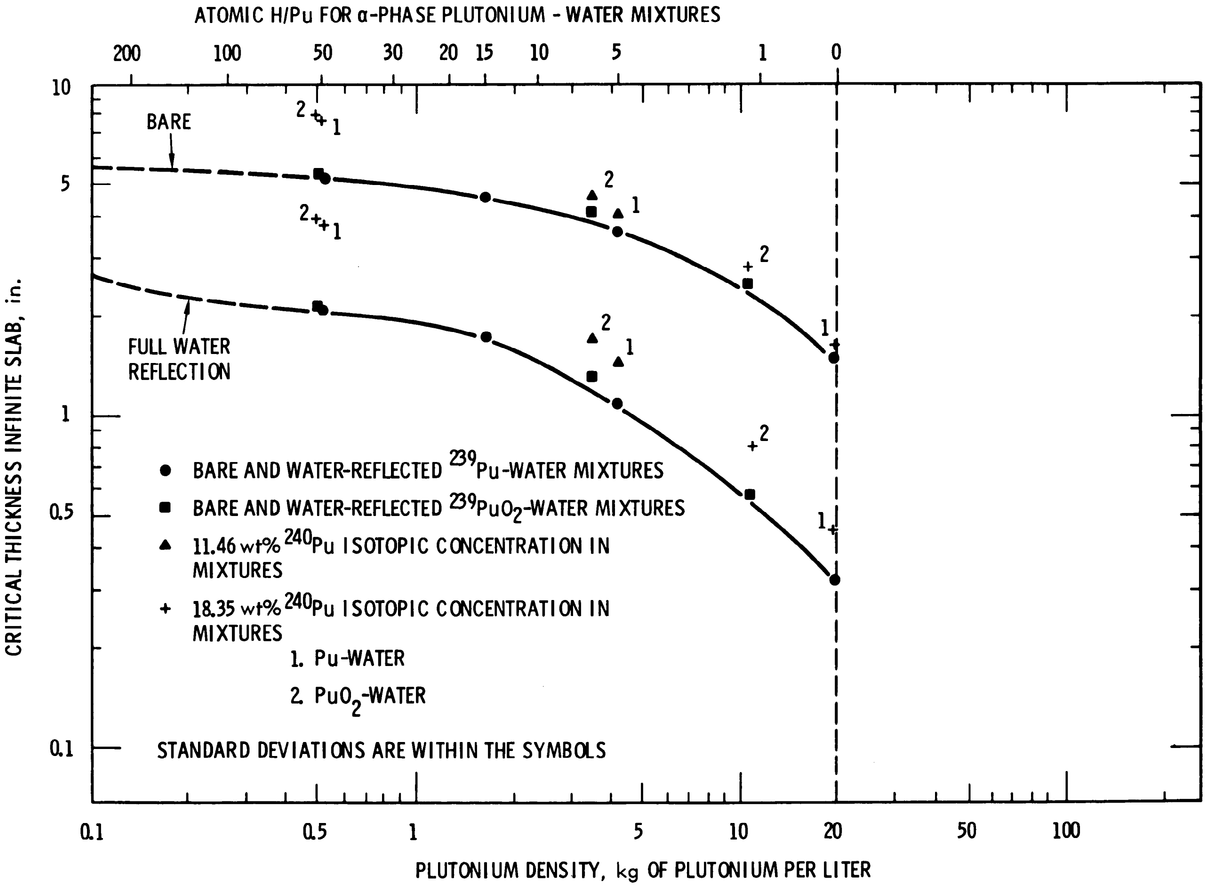

Fig. 2. Data derived from critical slabs of PuO_2-polystyrene.

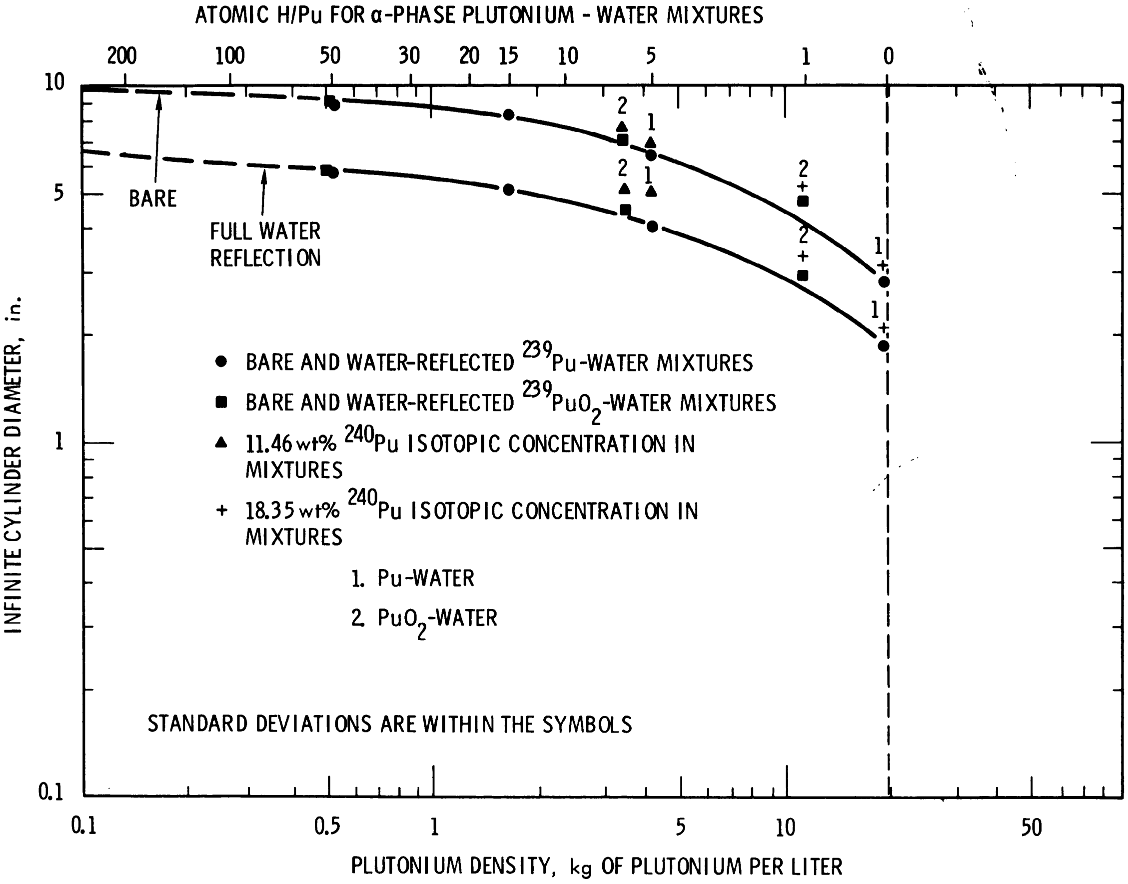

Fig. 3. Data derived from critical slabs of PuO_2-polystyrene.

radii obtained from plotonium nitrate solutions are also included. Also shown in Fig. 5 are calculated spherical critical radii obtained by using GAMTEC-II[10] 18-group CML cross sections with the transport theory code, DTF-IV,[8] and the diffusion theory code, HFN.[7] Previous studies[13] indicate that little improvement can be gained in the transport theory calculated values for these type systems by using higher than first order anisotropic scattering or fourth order of quadrature. Consequently, the S_4 approximation with P_1 scattering was used in the DTF-IV calculations. As can be seen in Fig. 5, the spherical critical radius for 521 g Pu/liter ^{239}Pu-H_2O, derived from the 49.6 H/Pu PuO_2-polystyrene experiments, is in good agreement with that previously obtained[2] from a plutonium nitrate solution experiment at 435 g Pu/liter. (The water-reflected spherical critical radius for ^{239}Pu-H_2O at 435 g Pu/liter is 12.00 cm, and at 521 g Pu/liter it is 11.65 cm.) Also, it can be seen in Fig. 5 that, at 521 g Pu/liter, the diffusion theory code, HFN, yields a more correct calculated value of the critical size than does the transport theory code, DTF-IV.

Although the neutron poisoning effects of 18.35 wt% ^{240}Pu are given in Table III, its effect on ^{239}Pu-H_2O at 521 g Pu/liter, and the indication that three concentrations exist for which a given size vessel is just critical ($k_{eff} = 1$), is better illustrated in Fig. 5. As can be seen in Fig. 5, the critical radius for this system is slightly less than that calculated by the diffusion theory code HFN for this same system containing 20 wt% ^{240}Pu. However, the critical radius, at 521 g Pu/liter, is considerably larger than some of the HFN-calculated critical radii at lower concentrations. This substantiates the observations made by Lloyd[9] with plutonium nitrate solutions at concentrations between 200 and 300 g Pu/liter. It also confirms that the critical size of a plutonium-water system can experience a minimum at a concentration other than that of the pure metal. Consequently, in setting criticality limits the credibility of diluting, as well as concentrating, in a given vessel must be established whenever the neutron poisoning effect of ^{240}Pu is considered, and if, as is generally the case, the plutonium concentration is below about 1000 g/liter.

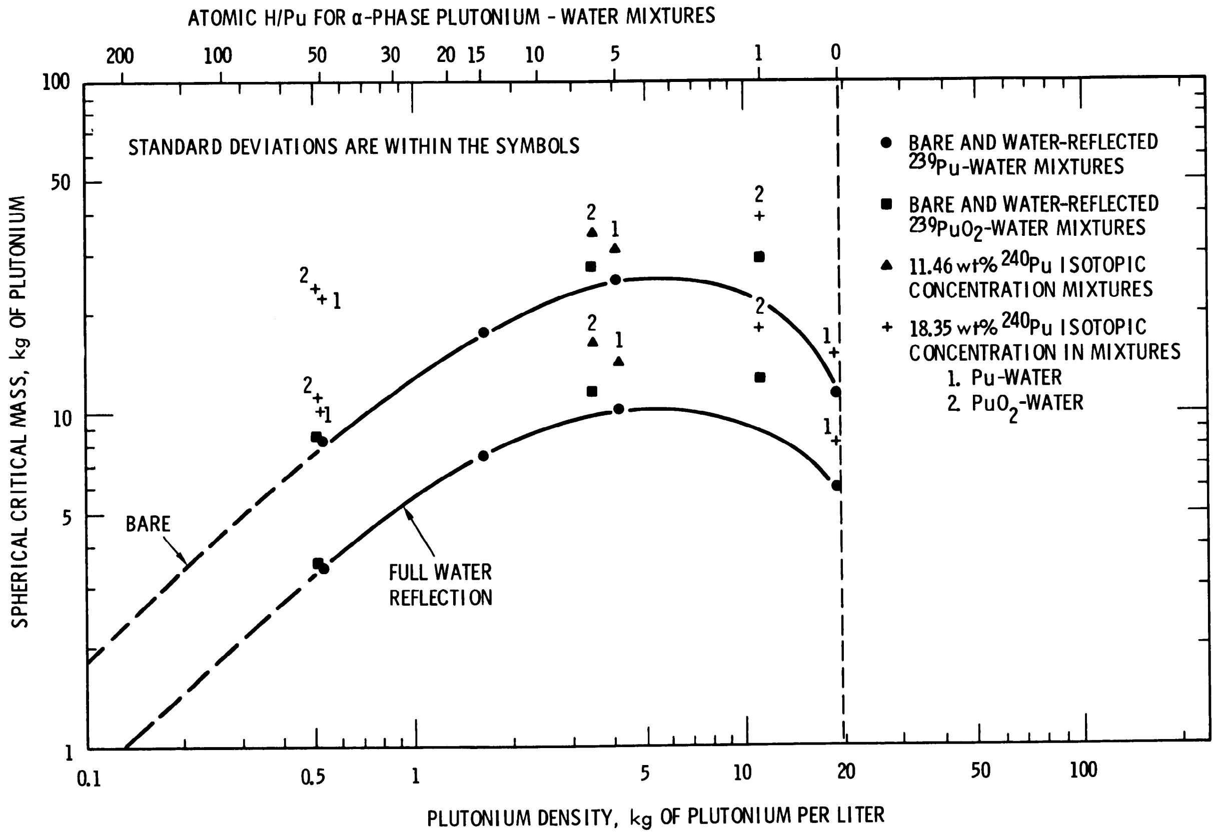

Fig. 4. Data derived from critical slabs of PuO$_2$-polystyrene.

To further evaluate the phenomenon in which a subcritical vessel of fixed volume could become critical by dilution with water, further data at 1000 g Pu/liter would be useful with plutonium containing about 20 wt% ^{240}Pu. Experiments reported by Lloyd[9] provide data for the slightly overmoderated case, and the experiments reported in this paper provide data at the concentration for which ^{240}Pu effects are essentially a maximum.

CONCLUSIONS

The results from the critical experiments reported in this paper provide a correlating point between the plutonium critical experiments performed with plutonium nitrate solution and those performed with PuO$_2$-polystyrene compacts. A good correlation was found to exist between the two types of experiments. It was also observed that, due, apparently, to compensating errors, the diffusion theory code HFN calculates the critical condition better than the transport theory code DTF-IV at concentrations of 521 g Pu/liter and less.

In contrast to data previously obtained with unmoderated PuO$_2$, the density effect of having the plutonium in the oxide form had essentially no effect on the reactivity at 49.6 H/Pu, whereas the neutron poisoning effect of the 18.35 wt% ^{240}Pu was at least five times that observed for the unmoderated PuO$_2$. It appears, also, from the experiments that, for high exposure plutonium-water systems, a given size vessel could be just critical (k_{eff} = 1.0) at three different concentrations below about 1000 g Pu/liter. That is, a critically safe vessel could be made critical by dilution alone if the ^{240}Pu content were high enough. The experiments also imply that the ENDF/B-II cross-section data result in conservative values with respect to nuclear criticality safety; however, considerably more testing against experimental data is needed to establish conclusions of this kind.

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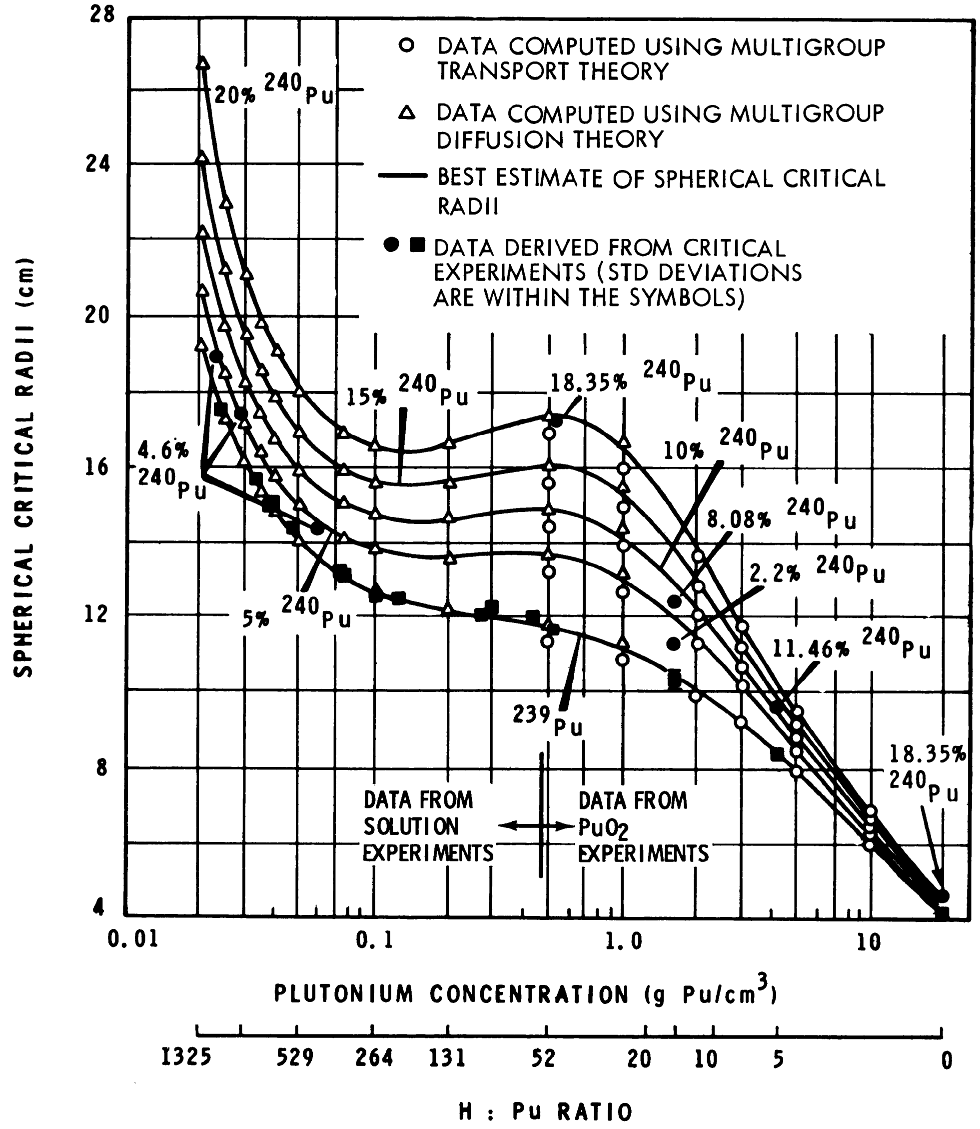

Fig. 5. Water-reflected spherical critical radii of plutonium(metal)-water mixtures.

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