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CRITICALITY OF PLUTONIUM NITRATE SOLUTIONS IN SLAB GEOMETRY

CHEMICAL PROCESSING

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A series of criticality experiments was performed on plutonium nitrate solutions in slab geometry. The solutions contained plutonium at concentrations ranging between 58 and 412 g Pu/liter for material with three different isotopic contents: 4.6, 18.4, and 23.2 wt% ^{240}Pu. Acid molarities varied from 1.6 to 5.0.

The experiments were performed with a variable thickness slab-type vessel of 42-in. height and width, whose thickness could be adjusted throughout a range of 3 to 9 in. The experimental vessel was used with and without a water reflector and also with a 1-in.-thick Plexiglas reflector.

The critical experiment data from the finite slabs were corrected to yield values of critical thicknesses for one-dimensional infinite slabs, i.e., slabs of finite thickness but of infinite height and width. Analytical corrections, based on experimental data, were subsequently used to correct the critical infinite slab thicknesses for materials extraneous to the plutonium solutions, such as the effect of the stainless-steel vessel walls and room return neutrons. The analysis provided values for clean one-dimensional assemblies that were then used as an integral check of calculational methods using cross sections from the ENDF/B-II data file. The computed values of k_{eff} for these "clean assemblies" ranged between 0.988 and 1.040; the values increased somewhat with increasing concentration.

INTRODUCTION

As the volumes needed for storage and processing of fissile materials increase, slab geometry systems become of special interest, due to the large quantities that can be safely handled without the addition of poisons or materials that inhibit mixing. Despite the frequent use of slab storage tanks, however, criticality data for plutonium solutions in slab geometry are particularly lacking. The first experiments to be reported on plutonium nitrate solutions in slab geometry were performed in France.[1] The data presented herein provide information at different plutonium concentrations, ^{240}Pu isotopic contents, and excess nitrate molarities, supplementary to the French results. Of the three simplest (one-dimensional) geometries—the sphere, infinite cylinder, and infinite slab—the fewest data exist on the latter geometry for establishing nuclear criticality safety specifications.

EXPERIMENTAL DESCRIPTION

The slab assembly (Fig. 1) used in these experiments was of rather unique design in that its thickness could be adjusted over a range of 3 to 9 in. This range of adjustments was made possible by stainless-steel bellows fabricated around the periphery of the tank. The slab thickness change was accomplished by means of adjustment screws, located at the corners of the vessel, between the opposite sides of the slab; slab thickness was measured by means of a dial caliper. The height and width of the tank was 42 in., based on the average of the bellows variation. The stainless-steel sides (0.062 in.) were reinforced with an egg-crate-type structure that was effective in maintaining the side position to within ~0.010 in. on filling the tank. The assembly and its parts were of Type-304-L stainless steel. A reflected assembly was achieved by placing plastic in the egg crate or by attaching gasketed side plates and filling with water. The assembly was positioned in a large hood for contamination control. Experiments were performed at room temperature (23 ± 1°C).

Plutonium nitrate solutions of various acid molarities and plutonium concentrations with 4.6,[2] 18.4,[3] and 23.2 wt% ^{240}Pu were used in these measurements. Table I lists the isotopic content of plutonium in the various nitrate solutions.

For each condition of reflection on the assembly, several critical geometries were determined by changing the thickness of the slab and measuring the resultant critical height. Critical slab heights were determined by a least-squares extrapolation of volume/count rate versus height measurements, using the normal critical approach method.

MEASUREMENTS AND RESULTS

Experiments were performed using plutonium having three different isotopic compositions. These included plutonium nitrate solutions at 58 g Pu/liter, 4.6 wt% ^{240}Pu, with acid molarities of 2.3 and 5.0; a range of concentrations between 66 and 412 g Pu/liter, 18.4 wt% ^{240}Pu; and concentrations of 202 and 284 g Pu/liter, 23.2 wt% ^{240}Pu.

Several critical approach experiments were performed with each solution to determine the critical height as a function of the slab thickness. Using these determinations of critical dimensions

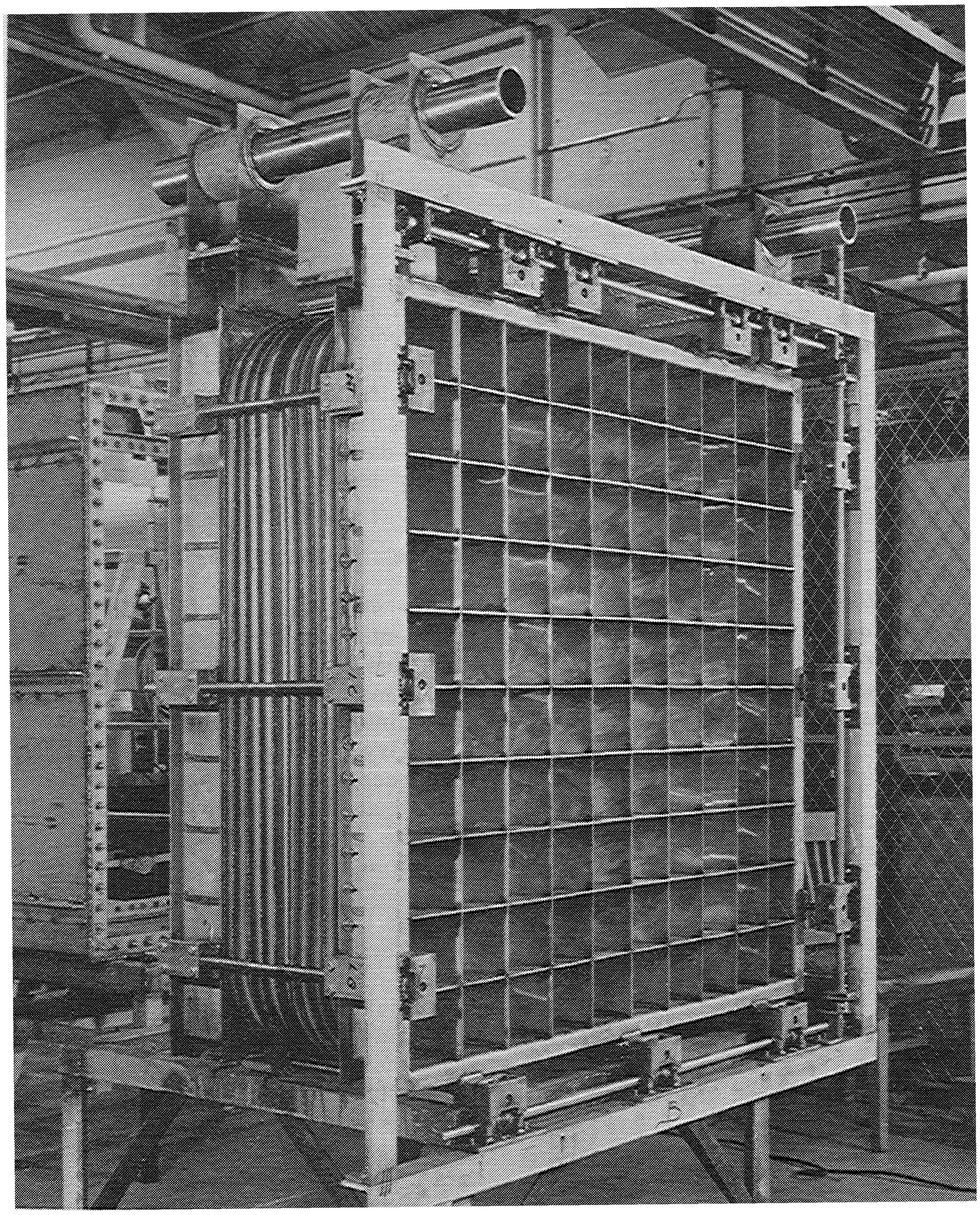

Fig. 1. Experimental slab tank assembly.

and by assuming constancy of buckling and extrapolation length, "best fit" values (i.e., minimum variation) were determined for the buckling and extrapolation length for each reflector condition. Table II gives values obtained for the buckling and extrapolation lengths of the various solutions, as well as critical infinite slab thicknesses derived for those same systems by buckling conversion from the experimental data. Although the material bucklings given may not be exact, their use for buckling conversion to infinite slab systems is well founded, due to the relatively large size of the experimental systems. The differences in critical thicknesses between a 42-in. slab and one of infinite dimensions are in the range of ~0.4 to 0.7 in. for the cases studied herein. Bucklings derived for the water-reflected systems are slightly greater (up to 8%) than the values derived for the same solutions when unreflected. The phenomenon is probably caused by the shift in the neutron energy spectrum associated with the water reflector. Figure 2 shows the variation in derived extrapolation lengths as a function of plutonium concentration for the 18.4 wt% ^{240}Pu solutions. These data are smoothly varying and increase as the plutonium concentration increases, thereby tending to confirm the self-consistency of the derived extrapolation lengths.

For water-reflected systems the slab vessel also achieved criticality when only partially filled. Consequently, since the vessel wall and reflector heights were constant, the extrapolation length for the top surface of the solution was determined such as to minimize the difference in buckling within each set of experiments through least-squares analysis.

The uncertainties in the critical slab thicknesses presented in Table II are estimated to be ±0.11 cm for a 95% confidence level. The infinite slab thicknesses obtained from buckling conversions of the experimental data are relatively insensitive to uncertainties in extrapolation lengths, as these errors are largely self-compensating in the conversion. The uncertainty in buckling conversion which results from errors in slab thickness and solution height, as well as the ex-

TABLE I

Isotopic Content of Plutonium in Nitrate Solutions

Isotope	wt% of Isotope		
	Solution 1 (4.6 wt% ^{240}Pu)	Solution 2[a] (18.4 wt% ^{240}Pu)	Solution 3[b] (23.2 wt% ^{240}Pu)
^{238}Pu	0.006	0.19	0.070
^{239}Pu	95.059	75.92	71.821
^{240}Pu	4.671	18.40	23.185
^{241}Pu	0.255	4.53	3.958
^{242}Pu	0.009	0.96	0.966

[a] ^{241}Am/Pu weight ratio = 0.0028.
[b] ^{241}Am/Pu weight ratio = 0.0021.

TABLE II

Measured Criticality Data for Plutonium Nitrate Solutions

^{240}Pu Content	Plutonium Concentration (g/liter)	Acid Molarity	Reflector	Material Buckling (m^{-2})	Effective Extrapolation Length (cm)	Critical Thickness of Infinite Slab of Solution Plus System[a] (cm)
4.6	58.0	2.3	Unreflected	182.3	4.3	14.7
4.6	58.0	2.3	Water	186.4	6.4	10.2
4.6	58.0	5.0	Unreflected	170.0	4.17	15.7
4.6	58.0	5.0	1-in. Plexiglas	170.5	5.64	12.8
4.6	58.0	5.0	Water	170.2	6.60	10.9
18.4	66.5	2.4	Unreflected	147.8	4.20	17.4
18.4	66.5	2.4	1-in. Plexiglas	148.4	5.68	14.4
18.4	66.5	2.4	Water	151.4	6.49	12.6
18.4	160.0	3.0	Unreflected	141.0	4.46	17.5
18.4	160.0	3.0	1-in. Plexiglas	136.0	6.30	14.3
18.4	160.0	3.0	Water	145.2	6.90	12.3
18.4	240.8	4.1	Unreflected	124.9	4.59	18.9
18.4	240.8	4.1	1-in. Plexiglas	122.3	6.46	15.5
18.4	240.8	4.1	Water	134.9	7.19	12.7
18.4	412.1	4.2	Water	114.6	7.69	14.0
23.2	283.8	2.2	Water	119.2	7.67	13.5
23.2	201.6	1.6	Unreflected	134.9	4.2	18.7
23.2	201.6	1.6	1-in. Plexiglas	135.1	5.8	15.4
23.2	201.6	1.6	Water	137.0	7.05	12.8

[a] Infinite thickness with vessel walls and supporting grid structure identical to that of experimental finite assembly.

trapolation length, is ±0.09. Independent of this is the uncertainty of chemical analysis. Individual uncertainties are as follows:

Slab thickness measurement	±0.01 cm
Determination of critical solution height	±0.040 cm
Chemical analysis	±0.060 cm
Uncertainty in extrapolation length	±0.3 cm .

The data presented in Table III contain additional uncertainties as a result of the corrections applied for the effects of tank walls, support structure, and room return. These corrections are summarized in Table IV. Considering the latter, the uncertainty in the critical thicknesses presented in Table III is ±0.2 cm.

It is interesting and, from a criticality safety viewpoint, important to note that the critical slab thickness is less for the 202 g Pu/liter solution than for the 284 g Pu/liter solution. The plutonium in both cases contained 23.2 wt% ^{240}Pu, and the lower concentration was produced by water dilution of the 284 g Pu/liter solution. Hence, the nitrogen-to-plutonium atomic ratio was held constant.

THEORY-EXPERIMENT CORRELATIONS

The HFN multigroup diffusion theory code,[4] previously used to predict with good accuracy the criticality of plutonium solutions in spheres,[5] also was used to compute criticality for the bare and water-reflected slabs.

Calculations, normalized to experimental data, were used to determine the effect of the stainless-steel tank walls, support structure, and room return neutrons on the measured critical slab thicknesses. The necessary corrections were made in the manner discussed in a previous paper.[6] The magnitudes of the corrections applied to the experimental slab thicknesses are given in Table IV.

Table III gives the unreflected and water-reflected infinite slab thicknesses corrected for

TABLE III

Criticality of Infinite Slabs of Plutonium Nitrate Solution
(Experimental results and calculations with ENDF/B-II cross-section data)

Description of $Pu(NO_3)_4$ Solution	H/Pu (atom ratio)	Total NO_3 (g/liter)	Type Reflector	Critical Thickness of Infinite Slab as Derived from Experiments (cm)	Computed Criticality Factor, Multigroup Diffusion Theory-HFN Code	Computed Critical Thickness (cm)
58 g Pu/liter, 4.6 wt% ^{240}Pu, 2.34 M HNO_3	425	203	Unreflected	16.91	1.014	16.60
			Water	9.13	0.999	9.15
58 g Pu/liter, 4.6 wt% ^{240}Pu, 5.00 M HNO_3	383	397	Unreflected	17.99	1.000	18.00
			Water	9.78	0.988	10.08
66.5 g Pu/liter, 18.4 wt% ^{240}Pu, 2.36 M HNO_3	367	219	Unreflected	19.79	1.020	19.17
			Water	11.38	1.002	11.31
160 g Pu/liter, 18.4 wt% ^{240}Pu, 3.04 M HNO_3	144	360	Unreflected	20.05	1.027	19.17
			Water	11.21	1.022	10.49
202 g Pu/liter, 23.2 wt% ^{240}Pu, 1.60 M HNO_3	115	313	Unreflected	21.19	1.022	20.34
			Water	11.70	1.008	11.41
241 g Pu/liter, 18.4 wt% ^{240}Pu, 4.14 M HNO_3	89	495	Unreflected	21.48	1.031	20.34
			Water	11.64	1.014	11.15
284 g Pu/liter, 23.2 wt% ^{240}Pu, 2.23 M HNO_3	78	436	Unreflected	---	---	21.40
			Water	12.43	1.007	12.13
412 g Pu/liter, 18.4 wt% ^{240}Pu, 4.16 M HNO_3	46	664	Unreflected	---	---	21.90
			Water	13.01	1.040	12.23

materials extraneous to the $Pu(NO_3)_4$ solution and reactivities computed for these slabs. Computed critical slab thicknesses are also given. All calculations were performed using ENDF/B-II cross-section data averaged over 18 broad energy groups by the GAMTEC-II code.[7] The ENDF/B cross sections were obtained from the ENDF/B-II point energy library data through use of the FLANGE-II code[8] to process the thermal data, and the ETOG code[9] to process the epithermal data for broad group averaging in GAMTEC-II. All of the isotopes of plutonium, including the small quantity of ^{241}Am, were accounted for in these calculations. The ^{241}Am results from the beta decay of the 13-h half-life ^{241}Pu. Under some circumstances, the buildup of ^{241}Am can cause significant changes in the value of k_{eff}. Calculations show that ^{241}Am can, by itself, be made critical with sufficient quantity in an essentially unmoderated state (71.4 kg for a steel-reflected metal sphere).

For the two high ^{240}Pu content solutions used in these experiments, the computed effect of the ^{241}Am was found to be small, amounting to a decrease in k_{eff} of from ~0.1 to 0.3%. The effect is sensitive to both ^{241}Am concentration and the hydrogen/plutonium ratio of the solution.

The effect of 18.4 wt% ^{240}Pu on critical slab thickness has been demonstrated in Fig. 3 where

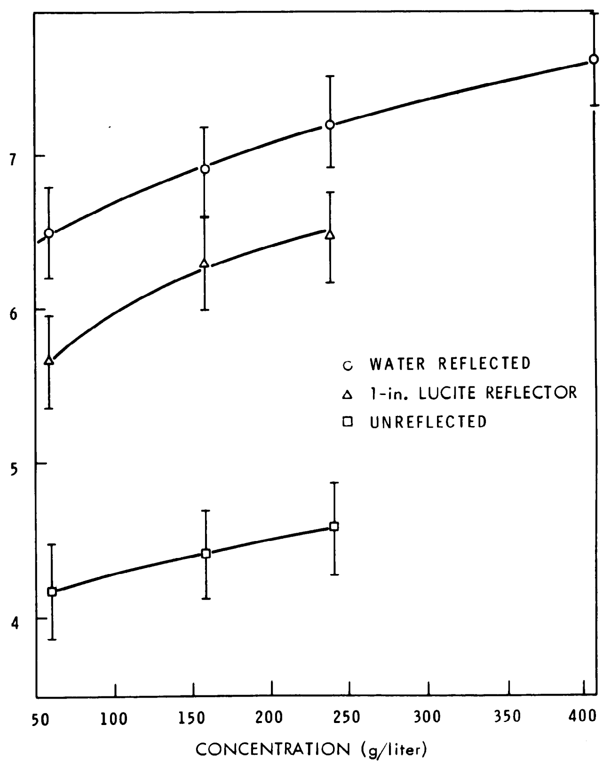

Fig. 2. Extrapolation length—slab tank—18.4% ^{240}Pu.

TABLE IV

$Pu(NO_3)_4$ Solution Slab Tank Corrections

	2.34 M HNO_3 58 g Pu/liter 4.6% ^{240}Pu (cm)	5.00 M HNO_3 58 g Pu/liter 4.6% ^{240}Pu (cm)	2.36 M HNO_3 66.5 g Pu/liter 18.4% ^{240}Pu (cm)	3.04 M HNO_3 160 g Pu/liter 18.4% ^{240}Pu (cm)	4.14 M HNO_3 240.8 g Pu/liter 18.4% ^{240}Pu (cm)	4.16 M HNO_3 412.1 g Pu/liter 18.4% ^{240}Pu (cm)	1.60 M HNO_3 202 g Pu/liter 23.2% ^{240}Pu (cm)	2.23 M HNO_3 284 g Pu/liter 23.2% ^{240}Pu (cm)
Tank walls, reflected	-0.732	-0.757	-0.793	-0.611	-0.530	-0.414	-0.584	-0.512
Tank walls, unreflected	+0.326	+0.338	+0.338	+0.357	+0.365	---	+0.359	---
Support structure, reflected	-0.335	-0.360	-0.375	-0.451	-0.498	-0.549	-0.476	-0.512
Support structure, unreflected	+0.832	+0.866	+0.865	+0.926	+0.955	---	+0.941	---
Room return (13%), unreflected	+1.05	+1.09	+1.15	+1.23	+1.23	---	+1.24	---

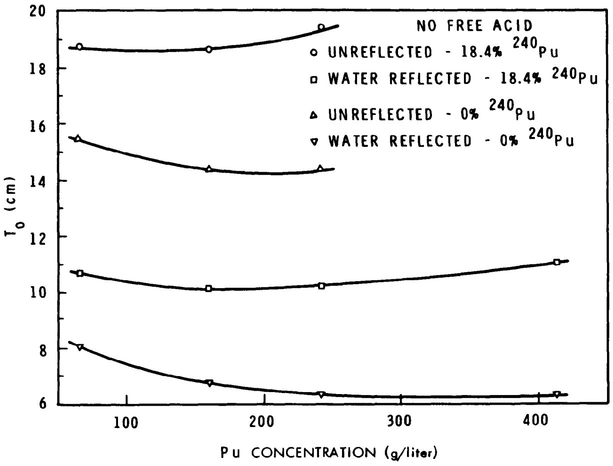

Fig. 3. Effect of ^{240}Pu on critical slab thickness of $Pu(NO_3)_4$ solution.

computed critical thicknesses are given for various concentrations of plutonium in plutonium nitrate solutions. These values were calculated for systems with no extraneous material and for solutions having no free acid. It was previously pointed out that the measured critical slab thickness was smaller for a concentration of 202 than at 284 g Pu/liter. For the plutonium nitrate solutions, the critical slab thickness will exhibit a minimum with plutonium concentration; the concentration at which this minimum critical slab thickness occurs depends on both the nitrate concentration and the ^{240}Pu content of the plutonium. In the concentration range near this minimum, 150 to 200 g Pu/liter, it is estimated that the critical slab thickness would be doubled if the plutonium contained 25 to 30 wt% ^{240}Pu.

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