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Criticality Studies with Plutonium Solutions

R. C. Lloyd, C. R. Richey, E. D. Clayton, and D. R. Skeen

Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington

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A series of criticality experiments were performed with plutonium (4.6% ^{240}Pu) nitrate solution in stainless steel spheres of 11.5-, 14-, and 15.2-in. diam. Reflectors of water, concrete, paraffin, and stainless steel were used; experiments were also performed on the 15.2-in. sphere unreflected. The spheres were made critical with plutonium concentrations varying from 24 to 435 g Pu/liter and molarity varying from 0.2 to 7.7. The minimum critical volumes for $Pu(NO_3)_4$ in water containing 4.6% ^{240}Pu were determined to be about 22 and 11 liters, respectively, for bare and reflected spheres at a concentration of 175 g Pu/liter. The effect of a 0.030-in. cadmium shell or a 4-in. air gap between the reflector and the vessel reduced the reflector worth to that of a nominal reflector (1-in. of water or less) for the concentrations of plutonium measured. Comparisons were made between experimental and theoretical results using multigroup diffusion theory.

INTRODUCTION

In processing reactor fuels, the establishment of nuclear safety limits requires criticality data on plutonium solutions, and as greater amounts of irradiated reactor fuels need processing, the criticality problems of safe storage and process facility design become more exacting. Some criticality experiments were performed with plutonium solutions in spherical and cylindrical geometries by Kruesi et al., in 1950-1951[1]. The recent experiments reported here were used to extend the previous criticality data on plutonium solutions in spherical geometry to higher concentrations of plutonium. These experiments, combined with those performed earlier, provide a comprehensive set of experimental data on which to base nuclear safety specifications for processing reactor fuels; the experiments also provide critical data against which computational methods of reactor physics may be checked.

MEASUREMENTS AND RESULTS

Criticality Data and Calculations

The critical assembly system[2] for these experiments was designed to be used with interchangeable vessels, which used the same control and safety rod system entering from the top, and the same fill and dump system on the bottom of the vessels.

Experiments were performed using thin stainless-steel spheres of 11.5-, 14-, and 15.2-in. diam the measured volumes were 12.95, 23.22, and 30.2 liters, respectively.

Properties of the fissile material which were studied were plutonium concentration and nitrate concentration. Other factors studied affecting criticality were containment material and reflector material. Factors not studied, such as temperature, ^{240}Pu content, and impurities, were kept as constant as possible. The temperature was maintained at 25°C, and impurities were kept low by periodic processing through ion exchange resin. The ^{240}Pu content of the Pu was 4.6%.

The Plutonium and nitrate concentrations were adjusted to make the spherical vessels critical in the "just-full" condition when possible. Data obtained from these measurements, and some data from measurements which were corrected to "just-full," are given in Table I.

Multigroup diffusion theory was used to compute criticality for a number of the $Pu(NO_3)_4$ solution spherical assemblies. The results of

[1]F. E. KRUESI et al., "Critical Mass Studies of Plutonium Solutions," HW-24514 DEL (May 19, 1952).

[2]W. A. REARDON et al., "Hazards Summary Report for the Hanford Plutonium Critical Mass Laboratory," HW-66266 (August 1, 1960).

TABLE I

Criticality of Plutonium Nitrate Solutions

Reflector	Sp. Gr.	Crit. Conc. (g Pu/liter)	Molarity	Total Nitrate (g NO_3/liter)	H_2O (g/liter)	H/Pu Atomic Ratio	Crit.[a] Mass (kg Pu)	Crit. k_{eff}
11.5-in. Stainless-Steel Sphere: 12.95 Liters (Vessel Wall Thickness 0.049 in.)								
Water	1.130	73	0.2	86	971	354	0.95	1.0034
	1.143	74.5	0.4	105	963	344	0.96	1.0023
	1.230	100	1.9	230	898	243	1.30	0.9971
	1.282	126	2.2	262	892	192	1.63	1.0057
	1.484	269	1.1	346	868	87	3.48	1.0039
	1.492	295[b]	0.8	303	893	81	3.82	1.0150
	1.680	435[b]	0.8	372	872	54	5.63	1.0124
Concrete: 11.25-in.	1.145	75	0.44	109	961	341	0.97	
	1.433	236	1.16	318	878	100	3.06	
14-in. Stainless-Steel Sphere: 23.22 Liters (Vessel Wall Thickness 0.044 in.)								
½-in. Paraffin[c]		53.6	2.2	190	930	470	1.24	
		62.9	2.8	238	912	396	1.46	
		63.8	3.5	278	886	382	1.48	
		89	4.0	338	847	263	2.07	
		96.7	4.2	354	841	241	2.25	
		97.4	4.5	377	833	238	2.26	
1-in. Paraffin[c]		53.6	4.3	323	855	443	1.24	
		85.1	6.4	482	763	256	1.98	
		97.6	6.1	473	765	223	2.27	
4-in. Concrete and 0.03-in. Cadmium[c]		40	1.1	110	946	634	0.93	
		50.9	4.6	336	834	457	1.18	
		232	6.7	650	689	86	5.39	
4-in. Concrete	1.093	32.8	1.1	118	949	770	0.76	
	1.143	35.5	2.9	216	889	684	0.82	
	1.253	45.2	5.62	410	792	495	1.05	
10-in. Concrete	1.085	29.6	1 07	118	936	848	0.69	
	1.209	36.6	4.39	310	858	651	0.85	
	1.285	43.4	6.37	445	790	518	1.01	
4-in. Air Gap; 6-in. Concrete	1.250	67.9	4.29	336	842	344	1.58	
	1.175	50.4	2.28	194	933	500	1.17	
4-in. Air Gap; 6-in. Concrete Hemisphere	1.193	75.0	2.10	209	907	328	1.74	
10-in. Concrete Hemisphere	1.146	46.4	2.05	175	923	538	1.08	
4-in. Concrete + 0.036-in. Stainless Steel	1.252	46.4	5.62	404	796	484	1.08	
4-in. Concrete + 0.72-in Stainless Steel	1.258	46.9	5.62	416	790	476	1.09	
Water	1.123	33.2	2.08	164	924	754	0.77	1.0006
	1.196	38.6	4.07	292	861	618	0.90	1.0056
	1.290	47.5	6.66	462	774	466	1.10	1.0059
Water and 0.030-in. Cadmium	1.126	46.9	1.4	136	942	540	1.09	
	1.238	69.0	3.9	313	852	341	1.60	
Water + 0.072-in. Stainless Steel	1.268	49.5	6.01	424	789	452	1.15	
15.2-in. Stainless-Steel Sphere: 30.2 Liters (Vessel Wall Thickness 0.048 in.)								
Unreflected[d]	1.081	39.0	0.4	64	978	668	1.18	1.0010
	1.429	172.3	4.9	486	766	125	5.20	0.9983
Stainless-Steel Shell 0.26-in. Thick	1.076	34.3	0.5	66	975	758	1.04	
Concrete Shell: Air Gap (6-in. Thick Concrete, 3.63-in. Air Gap)	1.071	29.0	0.6	64	977	900	0.88	
Water	1.060	24.4	0.5	58	977	1068	0.74	1.0004
	1.300	38.7	7.7	517	737	553	1.17	1.0078
Water and 0.08-in. Stainless Steel	1.060	25.2	0.5	60	974	1032	0.76	
Water Only[e]		23.6	0.5	55	979	1107	0.71	1.0038
Unreflected[e]		41.0	0.4	67	976	634	1.24	0.9991

[a] Includes 4.6 % ^{240}Pu; experimental values not corrected for vessel walls or neck supports, except where noted. Correction values are discussed in the text.

[b] Contains Pu(VI) and plutonium polymer, increasing the uncertainty to ± 5%.

[c] Values of nitrate corrected to full sphere from measurements close to full. Water calculated from nitric acid content and corrected by comparison to a known similar solution.

[d] These same solutions would be critical in spherical geometry at a volume of 31.1 liters with no stainless-steel walls or neck supports.

[e] Corrected for effect of vessel wall and neck supports.

these calculations are given in Table I; the values of k_{eff} should be unity to be in exact agreement with experiment.

The eighteen group constants utilized in the multigroup calculations were obtained from the GAMTEC-II computer code[3]. The 17 fast group constants were averaged over a 64-group spectrum from a B_1 approximation to the Boltzmann equation using a ^{239}Pu fission spectrum weighted source term. The thermal group constants were averaged over a Wigner-Wilkins spectrum utilizing cross sections normalized to the Sher data[4] for ^{239}Pu at 2200 m/sec. Resonance absorption by ^{240}Pu was treated by the Adler-Nordheim method[5]. The thermal group constants for the stainless steel shell were averaged over the Wigner-Wilkins spectrum for the respective $Pu(NO_3)_4$ solution, with the fast group constants averaged over a water slowing down spectrum.

The critical volumes for bare and water reflected spheres containing hypothetical ^{239}Pu-water mixtures, ranging in concentration from 15 to 250 g ^{239}Pu/liter were calculated from 18-group diffusion theory. The results are recorded in Table II.

Effect of Nitrate Concentration on Criticality

In order to determine the effect of the nitrate on critical mass, solutions with at least two different nitrate concentrations were measured in the same vessel. The nitrate was varied by dilution of concentrated solution with either high- or low-molarity nitric acid to obtain the desired solution.

The presence of NO_3 in the solution affects the critical mass primarily because of neutron capture in nitrogen, and displacement of hydrogen by the less moderating nitrogen. For overmoderated $Pu(NO_3)_4$ solutions, the critical mass increase is caused primarily by neutron capture in the nitrogen; whereas, for undermoderated solutions, the loss of hydrogen moderation is predominant. This is readily seen in Fig. 1 where the critical concentration of plutonium is plotted as a function of nitrate density for the water reflected 11.5-, 14.0-, and 15.2-in. spheres. The critical mass rapidly increases as the NO_3 density increases in undermoderated solutions with the 11.5-in. sphere. For critical solutions more optimally moderated in the 14- and 15.2-in. spheres, the increase in critical mass is considerably smaller. The computed critical concentrations for zero density NO_3 are 65.6, 28.8, and 23.4 g Pu/liter for the 11.5-, 14.0-, and 15.2-in. water reflected spheres, respectively.

The measured values are shown in Fig. 2, where total nitrate is plotted against H:Pu atom ratio. For a constant volume, it is seen that the plots form straight lines, making possible the extrapolation across zero-molarity and zero-nitrate lines, providing values of interest for nuclear safety minimums and for theoretical comparisons.

The extrapolation to zero nitrate gives 60 ± 3, 29.5 ± 1, and 24.0 ± 1 g Pu/liter for $Pu(NO_3)_4$ containing 4.6% ^{240}Pu in the 11.5-, 14.0-, and 15.2-in. spheres, respectively. These values compare reasonably well with those calculated and shown in Fig. 1. Interpolation between volume lines provides an estimation of the critical values for intermediate volumes. By interpolation, the

[3]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 (March 1965).

[4]R. SHER and J. FELBERBAUM, "Least-Squares Analysis of 2200 m/sec Parameters of ^{233}U, ^{235}U, and ^{239}Pu," Neutron Cross Section Evaluation Group, BNL-722 (June, 1962).

[5]F. T. ADLER, G. W. HINMAN, and L. W. NORDHEIM, "The Quantitative Evaluation of Resonance Integrals," *Proc. 2nd. UN Intern. Conf. Peaceful Uses At. Energy*, **16**, 1988 (1958).

TABLE II

Criticality of ^{239}Pu-Water Mixtures in Spherical Geometry

Pu, g/liter	Bare Spheres		Water-Reflected Spheres	
	Crit. Volume liters	Crit. Mass kg of ^{239}Pu	Crit. Volume liters	Crit. Mass kg of ^{239}Pu
15	78.998	1.185	52.502	0.788
20	49.029	0.981	30.122	0.602
30	31.443	0.943	17.685	0.531
35	27.878	0.976	15.235	0.533
40	25.537	1.021	13.639	0.546
50	22.580	1.129	11.662	0.583
75	19.415	1.456	9.525	0.714
100	18.114	1.811	8.646	0.865
250	16.173	4.043	7.364	1.841

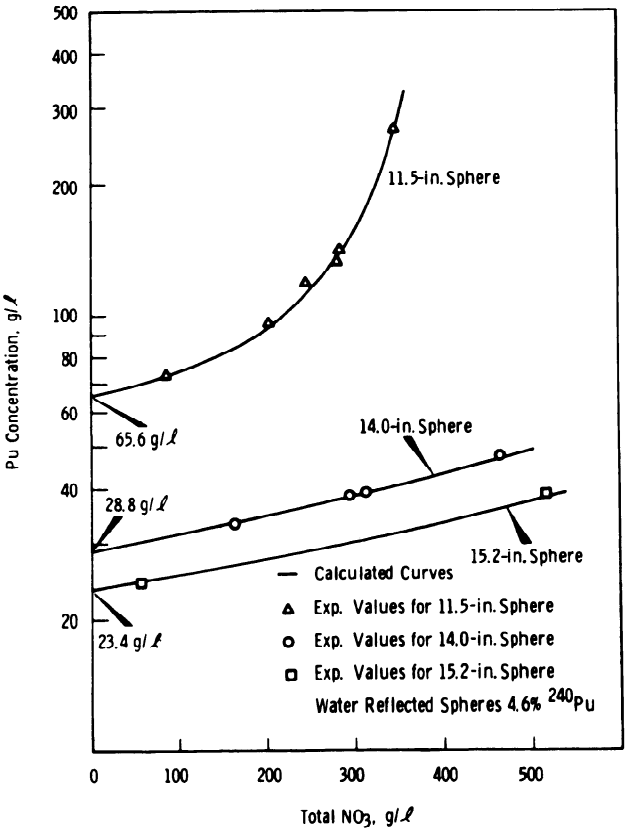

Fig. 1. Effects of nitrate on the critical concentration of plutonium.

volume of the tangent line to the zero-molarity curve can be estimated, which determines the minimum volume and optimum concentration that can be made critical for solution in the form of $Pu(NO_3)_4$ (i.e., a larger or smaller concentration results in a larger volume for criticality).

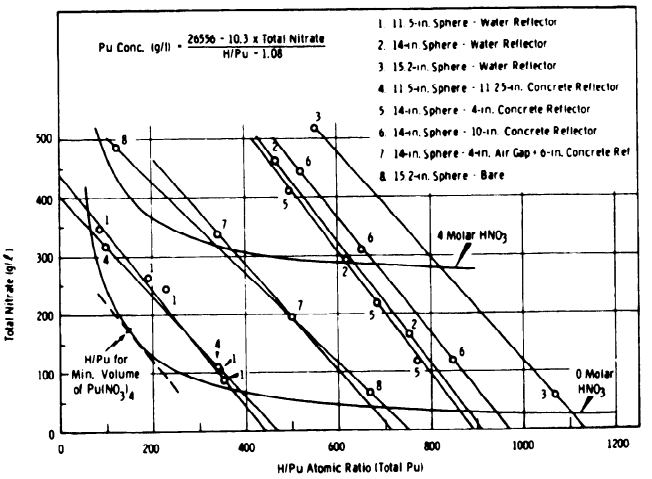

Fig. 2. Criticality of plutonium nitrate solutions. (Plutonium contains 4.6% ^{240}Pu)

The minimum volume of about 11 liters at a concentration of 175 ± 25 g Pu/liter is for $Pu(NO_3)_4$ solution containing 4.6% ^{240}Pu in a water-reflected sphere. Similarly, for bare spheres, though extrapolations are longer, a minimum critical volume of about 22 liters was estimated at zero molarity and for 4.6% ^{240}Pu.

Effect of ^{240}Pu

The plutonium utilized in the critical experiments contained 4.6% ^{240}Pu. The ^{240}Pu is basically a neutron absorber in $Pu(NO_3)_4$ solutions, thus increasing the critical mass. Its effect on criticality for the 14.0-in. water-reflected sphere is illustrated in Fig. 3. The experimental data recorded for 0.54, 3.17, and 4.05% ^{240}Pu are from experiments of Kruesi et al.[1]. The calculated curves were computed from 18-group diffusion theory. Resonance self-shielding effects were treated by the Adler-Nordheim method within the GAMTEC-II code.

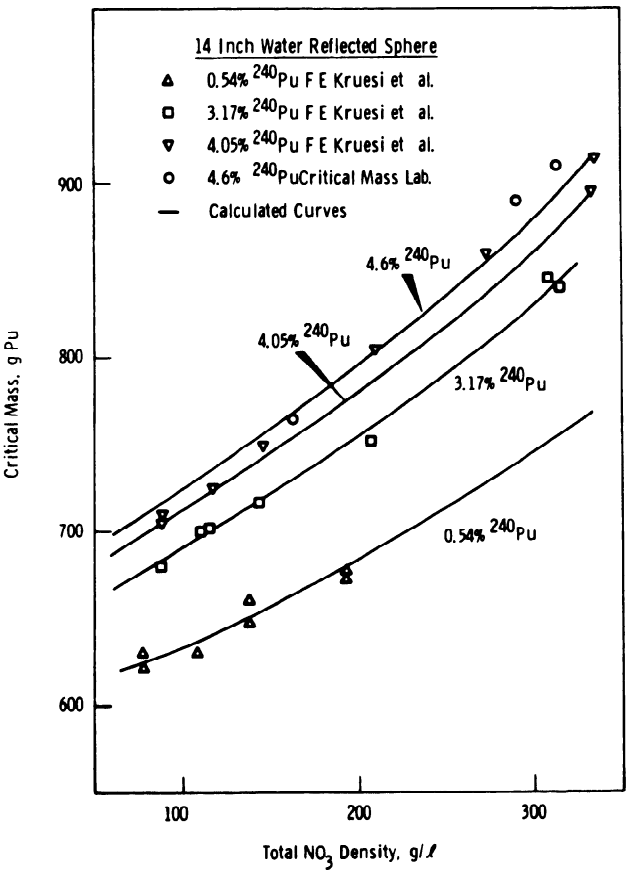

Fig. 3. Effects of ^{240}Pu on critical mass.

Reflector Conditions

Types of Reflectors

Several different reflector materials were used. A tamper tank about 40-in. diam was mounted on the system, providing an effectively infinite water reflector for the assembly vessels. Shells of paraffin were made in hemispherical molds to give $\frac{1}{2}$-in. and 1-in. reflector thicknesses for the 14-in. sphere. The concrete reflector was made in the form of two spherical shells, each consisting of orange-peel type sections for convenience in handling. The outer shell was 6 in. thick with 22-in. i.d. The inner shell was made to fit the vessel and had an o.d. of 22 in. The weight of the 10-in. concrete reflector for the 14-in.-diam sphere was about 1600 lb. Variations of concrete reflector used in experiments are shown in Fig. 4. A chemical analysis of the concrete is given in Table III.

The most difficult quantity to determine in the concrete is the water content. The lower value listed in Table III was determined by heating the sample to 700°C and measuring the electric current conducted by the water vapor driven off.

TABLE III

Results of the Concrete Analysis

(Density of the Concrete = 2.35 g/cm^3)			
Compound	wt %	Compound	wt %
MgO	2.17	K_2O	1.45
Al_2O_3	12.08	MnO_2	0.17
CaO	10.34	SiO_2	54.9
Fe_2O_3	7.57	H_2O[a]	2.19
Na_2O	< 0.25	H_2O[b]	11.06

[a] H_2O liberated at ignition temperatures up to 700°C.

[b] Quantity of H_2O required for 100% material balance.

Since any water remaining in the sample would not be recorded, it is expected that the true figure for the water content would be higher than that reported. This, then, would explain a portion of the material which could not be accounted for, since a material balance of only 91% was obtained.

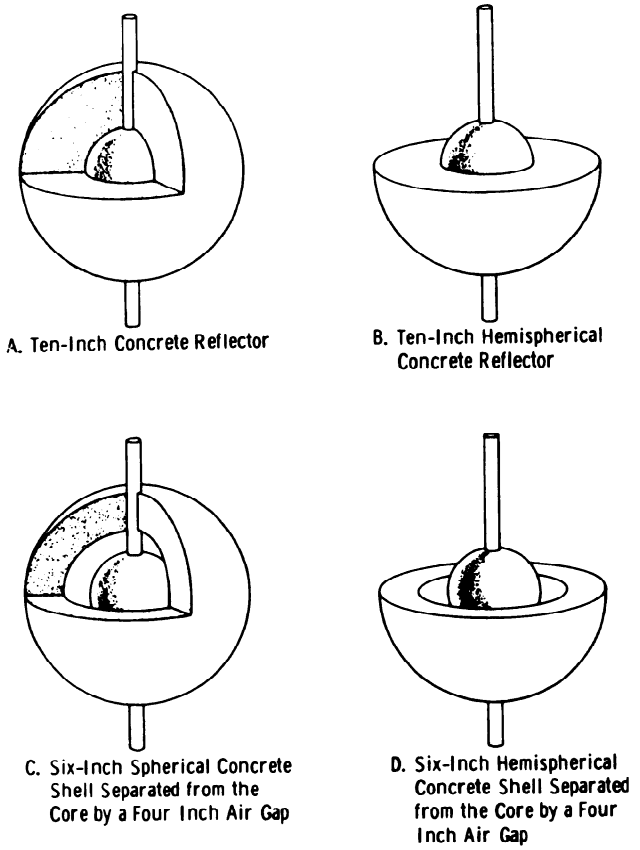

Fig. 4. Concrete reflector variations of the 14-in.-diam sphere.

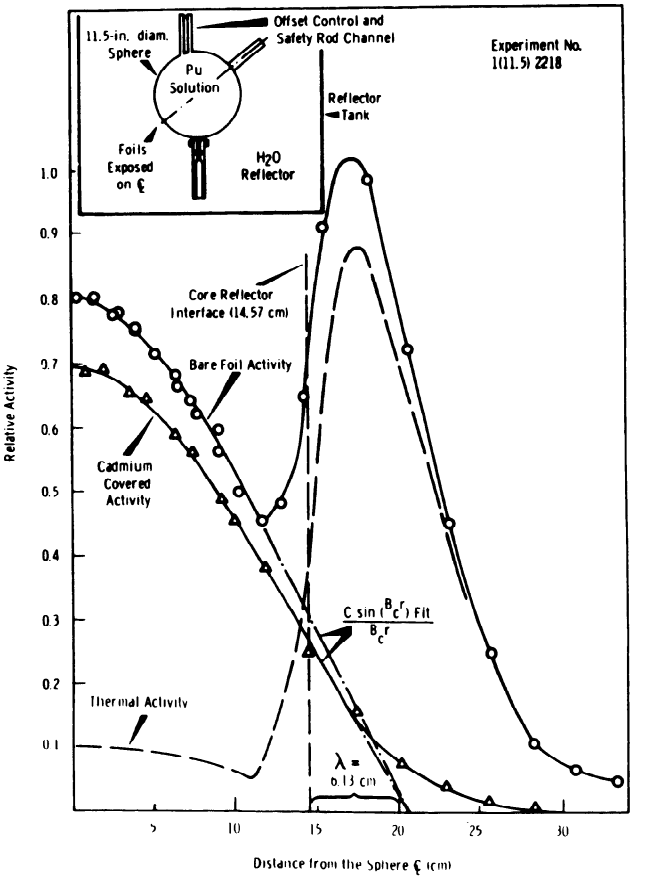

Fig. 5. Gold foil traverse in 11.5-in.-diam water-reflected sphere.

Measurements made in the 14-in.-diam sphere show that a 10-in.-thick layer of concrete is a better reflector than water; the critical concentration is about 10-12% less than when the sphere is water reflected. The 4-in. concrete reflector is not quite as good as a water reflector. Measurements made in the 11.5-in.-diam sphere show that the 11.25-in. concrete reflector was slightly less effective than the water reflector, although the two were nearly equivalent. These measurements suggest a slight, expected, dependence of the concrete reflector worth on the spectrum within the critical assembly core.

Since the albedo for concrete is higher than that of water, the concrete returns more neutrons to the core, but their worth is less since the neutrons have higher energies than if reflected from water. Although fewer neutrons are returned from the water reflector, those which are returned have a more thermal spectrum and a higher worth, since the slow neutrons are more readily captured in the fissile core material; the worth is also related to the H:X ratio of the core. Thus, although the number of neutrons being returned to the core is different in the two cases, the effective reflector savings of water and concrete are nearly equivalent. The flux shapes are illustrated in Figs. 5 and 6.

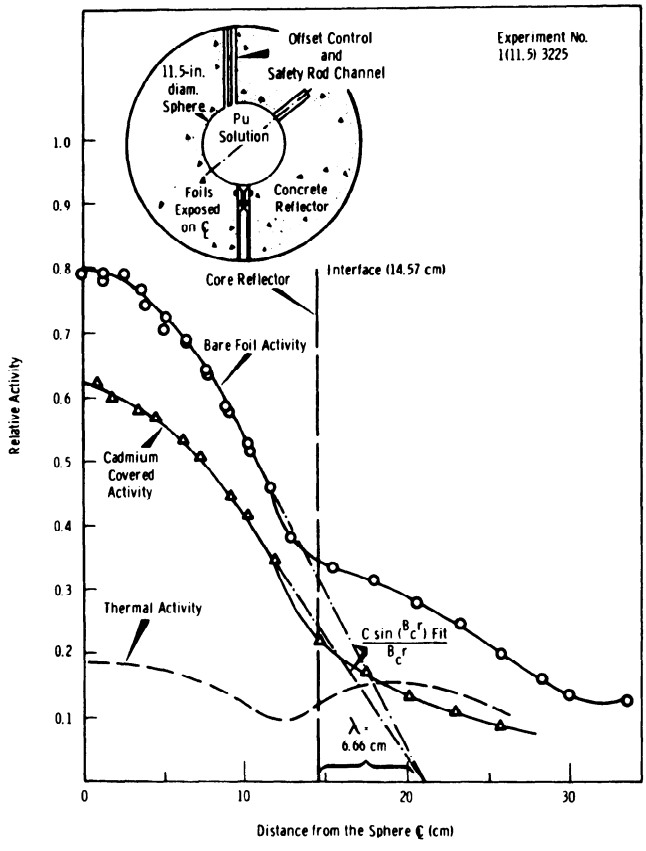

Fig. 6. Gold foil traverse in 11.5-in.-diam concrete-reflected sphere.

Stainless-Steel Shell Effect

The stainless steel of the vessel wall was evaluated by making criticality measurements with additional shells added to the vessel. The data from these measurements permitted an extrapolation to zero steel thickness. The effects on criticality, as measured in terms of volume, are given in Table IV. Also reported are the shell worths as computed from 18-group diffusion theory calculations for water-reflected measurements. The vessel walls were 0.049, 0.044, and 0.048 in. for 11.5-, 14.0-, and 15.2-in.-diam spheres, respectively.

Although the effect will not be linear for a thick reflector of stainless steel, over a small variation

TABLE IV

Effect of Stainless-Steel Wall for Water-Reflected Spheres

Sphere Diam. (in.)	Reflector	Concentration (g Pu/liter)	Acid Molarity	Total Nitrate (g/liter)	Volume Effect of Stainless Steel (mℓ/mil)		% Differ. in Volume Using Calc. Value
					Exp.	Calc.	
15.2	Water	26.0	0.53	60	8.8	22.1	1.76
14.0	Water	34.4	2.08	154	12.2	18.2	1.03
14.0	Water	40.4	4.13	297	10.6	17.1	1.12
14.0	Water	49.2	5.96	492	9.0	14.9	1.02
11.5	Water	119.0	2.02	250	4.6	6.4	0.25
11.5	Water	275.0	0.76	290	3.4	2.6	0.56
14.0	Concrete	46.9	5.6	416	4.7	---	---
14.0	Concrete	32.7	1.2	118	6.4	---	---
15.2	Bare	42.4	0.4	67	-6.8	---	---
14.0	Bare	105.2	2.4	261	-13.9	---	---

in thickness (up to 150 mil), the effect is nearly linear. Table IV indicates that the volume worth of the stainless steel increases quite rapidly at lower concentrations.

Measurements were also made in the bare and water-reflected 15.2-in.-diam sphere where the concentration was adjusted to accomplish criticality with the sphere "just-full" as shown in Table I.

The effects of the vessel neck and support were evaluated by using a neck mockup. For the reflected assembly, the effect was very small. The corrected critical volume for a sphere completely covered with one inch of paraffin would be about 0.6% larger. The corrected critical volume for full concrete and water reflectors would be ~0.4% less. Measurements with the neck mockup on the 15.2-in. bare sphere showed the critical volume for the clean sphere would be about 0.60 liter larger, or about 2%.

Cadmium at Vessel-Reflector Interface

A 0.030-in.-thick cadmium shell was used at the reflector-vessel interface in several experiments. The effect of this shell on the extrapolated critical volumes is shown in Table V.

As shown in Table V, the cadmium shell caused an increase in critical volume (or mass) of about 10-12% over a wide range of concentrations when the sphere was reflected. From data taken with the 4-in. concrete reflector, the percent volume increase varies inversely with concentration.

Two measurements in the 14-in.-diam sphere with the vessel covered with 0.030-in. Cd and backed by water were made to determine the concentration for criticality in the full sphere as shown in Table I.

The critical concentration in the 14-in. sphere, when wrapped with cadmium and water reflected, corresponds closely to that of a 12.2-in.-diam water-reflected sphere, or 15.2-in.-diam bare sphere as determined from interpolation of data from "just-full" spheres. From comparisons with data in which paraffin reflectors of $\frac{1}{2}$ and 1 in. were used, the cadmium is seen to reduce the reflector savings of the water to less than that from a 1-in. paraffin reflector. For dilute solutions, the composite Cd-H_2O reflector is equivalent to a paraffin reflector about 0.7-in. thick.

There results are of particular interest in nuclear safety, since under the concentration range (30-70 g Pu/liter), the data show that a vessel wrapped with cadmium would only be nominally reflected (reflector equivalent to 1 in. or less of water). The "safe" dimension of a cadmium-covered tank could then be based on the condition of a nominal reflector, rather than on the more restrictive fully reflected condition with its consequent smaller size. There was no measurable effect on criticality by placing of cadmium shield on bare sphere, which indicates the presence of some neutron reflection from the hood walls.

Air Gaps at Vessel-Reflector Interface

Experiments were performed to obtain data with concrete separated from the vessel by an air gap for use in evaluating nuclear safety of in-plant equipment proximate to reflectors. Measurements in the 14-in.-diam sphere show that the effect of an air gap between the concrete reflector and the core on the critical concentration varies with plutonium concentration and total nitrate. Extending the results to zero nitrate (Pu-H_2O mixture), the critical concentration is increased by about 40% as a result of the air gap, whereas at 300 g NO^3/liter, the concentration for criticality is increased by about 80%. For this vessel, with the concrete and 4-in. air gap, the results show equivalence to a paraffin reflector of about 1-in. thickness. Also, the 4-in. air gap is almost as effective in increasing critical concentration as removal of the 10-in. concrete reflector from one half of the sphere. Measurement in the 15.2-in. sphere with a (3-5/8)-in. air gap gave a reduction in concentration (and mass) over a bare sphere of about 35%. This compares to a reduction of about 60% for full

TABLE V

Effect of 0.030-Inch Cadmium Shell on Critical Volume

Sphere Diam (in.)	Reflector	Pu Concentration (g/liter)	Critical Volume Increase (%)
14	4 in. of concrete	231	10.0
14	4 in. of concrete	51	10.6
14	4 in. of concrete	33	12.3
14	1 in. of paraffin	127	11.4
15.2	Bare	42.5	0

water reflection. These experiments show that an air gap between a reflector and storage container can appreciably affect criticality.

Neutron Flux Measurements—Extrapolation Length and Buckling

The 11.5- and 15.2-in.-diam spheres were provided with re-entrant tubes traversing their centers in which foils could be irradiated for neutron flux measurements. Gold foils of 5-mil thickness and 0.25-in. diam were used in the irradiations; cadmium covers were 21 mil thick. The stainless steel re-entrant tube had an o.d. of 0.572 in. with a wall thickness of 0.062 in. The foils were placed perpendicular to the centerline of this tube and separated by aluminum spacers.

Gold foil irradiations were made in full spheres with both the control and safety rods withdrawn. On occasion, criticality was obtained with some solution in the neck of the sphere, but the effect of this perturbation on the measured flux was not observable; the measured distribution was symmetric with respect to the sphere center. Typical plots of the foil activity versus sphere radius are shown in Figs. 5-8 for various reflector conditions measured.

The gold cadmium ratio was essentially constant to within an inch of the boundary in all irradiations. A least-squares analysis of the gold foil activity to the function, $A_0 \sin B_c r / B_c r$, in the region of constant cadmium ratio provided values for the extrapolation lengths (λ_{eff}) and critical bucklings (B_c^2). The critical bucklings and extrapolation lengths are quoted in Table VI.

The foil activities in the reflector regions (Figs. 5-8) clearly indicate the differences in thermalization properties of concrete and water reflectors. The peak of the thermal flux which occurs about 1 in. from the core boundary in the water-reflected systems is essentially absent in the concrete curves. The small variation in the neutron flux across the air gap is clearly shown in Fig. 8.

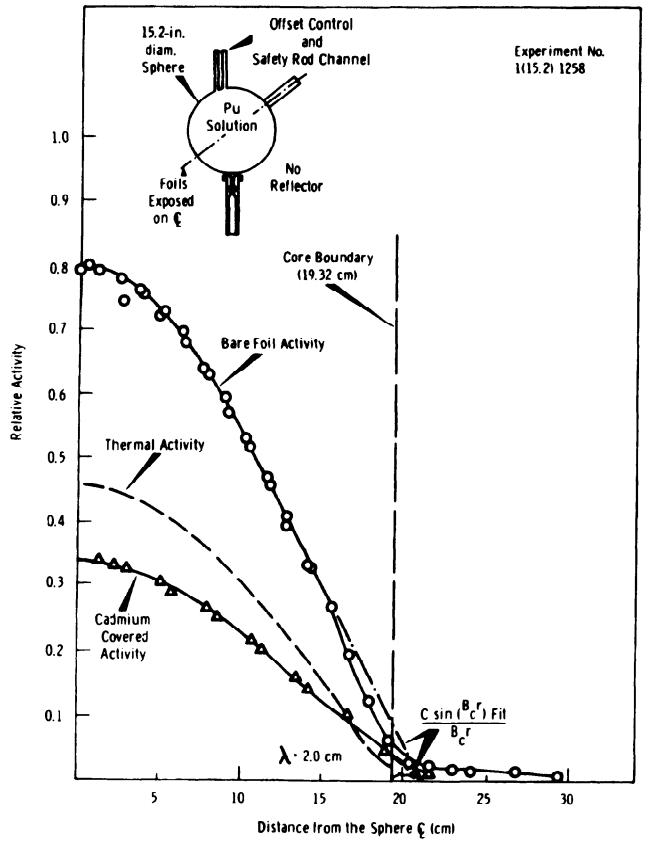

Fig. 7. Gold foil traverse in 15.2-in.-diam bare sphere.

ANALYSIS OF EXPERIMENTAL ERROR

The critical mass is determined from the measured critical volume and knowledge of the plutonium concentration determined by chemical analysis of a sample of the solution. The volume of the solution in the vessel is measured as a function of the height of solution prior to criticality measurements, so that error in volume becomes dependent on error in solution height measurement. The solution height is determined

TABLE VI

Measured Extrapolation Lengths and Critical Bucklings

Sphere Diam (in.)	Reflector	Pu Conc. (g/liter)	H/Pu	Au Cd Ratio	λ_{eff} (cm)	Buckling (cm^{-2})
11.5	Water	269	83	1.15	6.13 ± 0.20	0.02303 ± 0.00045
11.5	Concrete	236	96	1.27	6.66 ± 0.15	0.02190 ± 0.00031
11.5	Concrete	75	337	1.87	5.87 ± 0.16	0.02363 ± 0.00037
11.5	Water	76	332	1.96	5.55 ± 0.26	0.02439 ± 0.00063
15.2	Bare	39	662	2.34	2.00 ± 0.07	0.02182 ± 0.00015
15.2	Concrete with $(3\frac{5}{8})$-in. Air Gap	29	891	3.43	3.78 ± 0.14	0.01850 ± 0.00023
15.2	Water	24	1062	3.74	4.90 ± 0.22	0.01683 ± 0.00031
15.2	Water	39	563	2.76	5.79 ± 0.15	0.01565 ± 0.00019

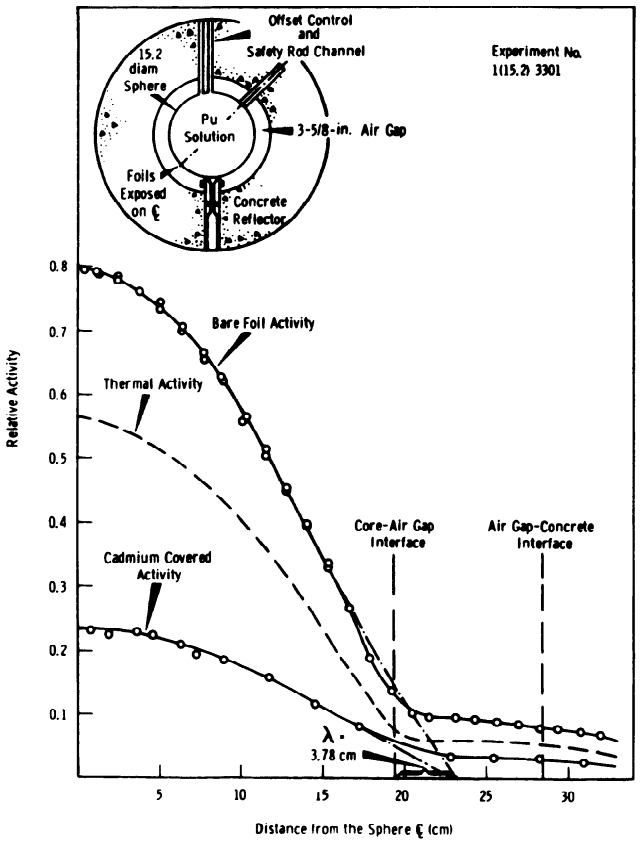

Fig. 8. Gold foil traverse in a 15.2-in.-diam concrete-reflected sphere with air cap.

by a servomanometer system with a claimed accuracy of 0.001 to 0.003 in. The height is also checked by a sight tube. The volume when the sphere is full becomes quite insensitive to the height measurement. Successive measurements of criticality using the same solution have shown differences of the order of 0.020 liters, which is probably within the uncertainly of the volume measurement.

A difference of up to 0.04 liters was found upon comparing the extrapolated critical volumes from inverse multiplication curves to the critical volumes for the same solutions made critical and and corrected for control rods.

It is not expected that uncertainty in critical volume determination would be greater than ± ¼%.

Chemical analyses of the solution samples are carried out by the Chemical Laboratory and quoted standard deviations are as follows:

Pu concentration	± ½%
H^+ concentration	± 1%
Specific gravity	± 0.4%
Total nitrate	± 1½%
H_2O concentration	± 5% .

Ranges of about 3% in the concentration have been observed on multiple analyses of the same solutions. Multiple analyses are made on critical solutions to keep the uncertainty reasonably small. It is believed that the uncertainty of the concentration is about ± 1%. Under present measurement techniques, it is believed that the major source of error of the critical mass is in the determination of the critical plutonium concentration which would lead to uncertainty in the critical mass of about ± 1%.

Since the water analysis is the most difficult of the chemical analyses to accurately measure, the water content was computed by measurement of other components of the solution and specific gravity and by assuming a material balance.

SUMMARY

Criticality measurements were made with plutonium nitrate solutions in stainless-steel spheres of 11.5-, 14-, and 15.2-in. diam. The plutonium contained 4.6% ^{240}Pu. Various reflector materials were used, including water, concrete, paraffin, and stainless steel; the 15.2-in. vessel was also used nonreflected. Calculation using multigroup diffusion theory showed agreement with the experimental criticality data.

A minimum volume for water reflected and nonreflected spheres containing $Pu(NO_3)_4$ (4.6% ^{240}Pu) was determined from extrapolation of experimental data to be 11 and 22 liters, respectively, and occurred at about 175 g Pu/liter (i.e., a larger or smaller concentration would result in larger volume for critical). Measurements indicated concrete was a slightly better reflector than water at lower concentrations of Pu but slightly less at higher concentration, indicating some dependence on neutron spectrum. The extrapolation length and buckling were determined from the neutron flux measurements in the assemblies. The placement of 0.030-in. cadmium or a 4-in. void between the reflector and vessel reduced the reflector to the equivalent or less than a nominal reflector for the Pu concentration studied.

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