REFERENCE 106

R. C. LLOYD AND E. D. CLAYTON, "CRITICALITY OF PLUTONIUM-URANIUM NITRATE SOLUTIONS," NUCL. SCI. ENG. 60: 143-146 (1976).

NUCLEAR SCIENCE AND ENGINEERING®

THE RESEARCH JOURNAL OF THE AMERICAN NUCLEAR SOCIETY

VOLUME 60, NUMBER 2, JUNE 1976

NSENAO 60 (2) 113-194 (1976)

Indexed in "Engineering Index" and Abstracted in "Nuclear Science Abstracts"

NUCLEAR SCIENCE AND ENGINEERING is published monthly by the American Nuclear Society, Inc., with executive and business offices at 244 East Ogden Avenue, Hinsdale, Illinois 60521 — telephone 312/325-1991. The subscription rate is $50 per volume or $132 for three volumes per calendar year; single copies are $18 (special issues slightly higher). Microfiche subscriptions are also available at same rates. Address subscription order to the publisher. (Back issues of Volumes 1-17 are available from Academic Press, 111 Fifth Avenue, New York, New York 10003.) Second-class postage is paid at Hinsdale, Illinois, and at additional mailing offices. Printed in Danville, Illinois 61832, by Interstate Printers and Publishers, Inc. Inquiries about the distribution and delivery of NUCLEAR SCIENCE AND ENGINEERING and requests for changes of address should be directed to the American Nuclear Society. Allow 6 weeks for a change to become effective.

NUCLEAR SCIENCE AND ENGINEERING: 60, 143-146 (1976)

Criticality of Plutonium-Uranium Nitrate Solutions

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Received October 31, 1975
Revised January 13, 1976

A series of experiments was performed providing new criticality data on plutonium-uranium nitrate solutions in cylindrical and spherical geometry. For the experiments in cylindrical geometry, the plutonium content of the total uranium plus plutonium was ~30 wt%; whereas, in the case of the water-reflected spheres, measurements were performed with both 15 and 30 wt% plutonium. The uranium in the mixture was slightly depleted, containing 0.66 wt% ^{235}U. The plutonium concentration covered by these experiments ranged between 12.4 to 97.3 g Pu/ℓ (uranium plus plutonium concentrations between 30 to 310 g/ℓ). The ^{240}Pu content of the plutonium was 5.6 wt% in the first case and 4.7 wt% in the second. The experiments were analyzed using ENDF/B-III cross-section data, and criticality factors were computed in each case. Some comparative calculations also were made, showing the differences obtained with ENDF/B-II, ENDF/B-III, and GAMTEC cross sections. The KENO code, with ENDF/B-III cross sections, as well as the HFN code, provide conservative results on the criticality factors for these systems. The average value of the computed k_{eff} for the cylinders, using KENO, was 1.022, and for the spheres, 1.024 using HFN. Thus, using these methods and cross-section data, the computed critical masses and volumes would be expected to be smaller than those measured by ~2% in terms of k_{eff}.

INTRODUCTION

Criticality information is necessary for the design and operation of equipment and facilities for processing, fabrication, storage, and shipment of liquid-metal fast breeder reactor (LMFBR) fuel materials. Some experimental data were reported on criticality of homogeneous fuel materials containing both uranium and plutonium.[1-7] The objective of this series of experiments was to provide new criticality data on solutions in the 15 and 30% plutonium content range, adequate for use in assuring safe and economical criticality control throughout the LMFBR fuel recycle. The experimental data also provide useful benchmarks for data testing and analysis on integral criticality experiments for verification of the analytical techniques used in support of criticality calculations.

EXPERIMENT, DATA, AND ANALYSIS

Experiments were performed with plutonium-uranium nitrate solutions in water-reflected stainless-steel (Type 304L) cylindrical and spherical vessels. A schematic of the cylinder system is shown in Fig. 1. This vessel was 61.028-cm i.d. with a wall thickness of 0.079 cm, and was 107 cm in height. The water reflector surface was maintained at the top of the experimental vessel and was contained in a 102-cm-diam tank.

About 30 wt% of the plutonium-uranium of the nitrate solutions used in the cylindrical vessel was plutonium. The plutonium concentration varied from 12.4 to 97.3 g Pu/ℓ and the free nitric

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[2]S. R. BIERMAN and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **15**, 307 (1972).

[3]L. E. HANSEN, S. R. BIERMAN, and E. D. CLAYTON, "Criticality of Mixed PuO_2-UO_2 Systems," in *Reactor Physics Quarterly Report*, BNWL-1150, pp. 5.6-5.16, Pacific Northwest Laboratories (1969).

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[5]S. R. BIERMAN and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **15**, 805 (1972).

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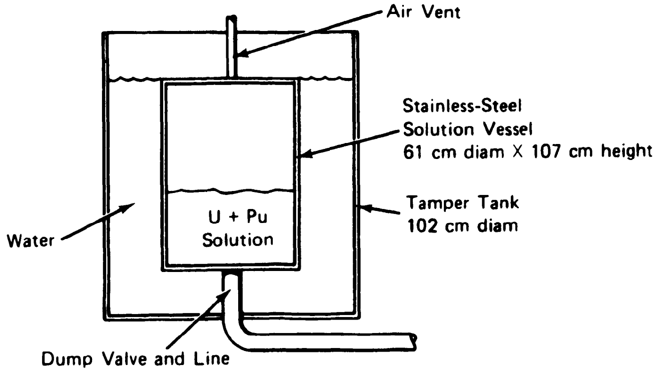

Fig. 1. Schematic of experimental set-up.

acid molarity changed from 1.3 to 4.8. The plutonium was in the tetravalent state. The ^{240}Pu content of the plutonium was 5.6%; the uranium contained 0.66% ^{235}U.

The critical approach method was used to obtain criticality, with the solution being pumped into the experimental vessel in incremental increases in height. The data obtained from the thirteen experiments performed are given in Table I. The critical heights were determined at various plutonium concentrations and acid molarities. The critical mass and volume are given for each measurement. These results can be seen more easily in Fig. 2, where the mass and volume are given as a function of plutonium concentration. In this plot, the minimum mass of ~6 kg occurs at a concentration of ~70 g (U+Pu)/ℓ. The volume curve near the minimum is very flat, but appears to reach a minimum at ~200 g (U+Pu)/ℓ.

The criticality factor was computed in each case. The k_{eff} values included in Table I were computed using ENDF/B-III cross-section data in an 18-group calculation with the KENO Monte Carlo code.[8] Epithermal energy groups were obtained via application of the ETOG (Ref. 9) code and the GAM portion of the EGGNIT code,[10] and the single-thermal group data via the FLANGE code[11] followed by the TEMPEST portion of the EGGNIT code. A thermal group energy cutoff

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[11] H. C. HONECK and D. R. FINCH, "FLANGE-II (VERSION 71-1): A Code to Process Thermal Neutron Data from an ENDF/B Tape," DP-1278 (ENDF-152), Savannah River Laboratory (1971).

TABLE I

Criticality of (U + Pu) Solution in Cylindrical Geometry

Plutonium Concentration[a,b] (g/ℓ)	Uranium Concentration (g/ℓ)	H^+	Specific Gravity	Critical Height (cm)	Critical Volume (ℓ)	Critical Mass		k_{eff} ENDF/B-III KENO
						kg (Pu + U)	kg Pu	
97.3	200.4	4.8	1.562	19.96	58.37	17.37	5.68	1.033 ± 0.008
87.9	184.3	4.4	1.512	19.48	57.00	15.52	5.01	1.035 ± 0.008
75.4	168.5	3.7	1.450	18.82	55.04	13.42	4.15	1.007 ± 0.008
63.1	147.7	2.9	1.389	18.72	54.76	11.54	3.46	1.026 ± 0.008
49.6	122.3	2.4	1.320	19.35	56.59	9.73	2.81	1.027 ± 0.008
39.9	96.7	2.2	1.257	20.32	59.44	8.12	2.37	1.016 ± 0.008
30.2	72.5	2.1	1.213	22.89	66.97	6.88	2.02	1.026 ± 0.008
25.3	60.9	2.0	1.182	25.12	73.48	6.33	1.85	1.019 ± 0.008
20.3	48.9	1.8	1.154	29.92	87.53	6.06	1.78	1.020 ± 0.006
17.2	41.5	1.7	1.132	36.04	105.44	6.19	1.81	1.023 ± 0.007
14.1	34.4	1.5	1.117	52.60	153.91	7.46	2.17	1.027 ± 0.006
13.3	32.2	1.4	1.110	64.01	187.24	8.52	2.49	1.007 ± 0.005
12.4	29.9	1.3	1.103	95.20	278.50	11.78	3.45	1.022 ± 0.005
								average k_{eff} = 1.022

[a] Isotopic composition:

^{239}Pu-93.964 wt%	^{238}U-99.324 wt%
^{240}Pu-5.585 wt%	^{236}U-0.012 wt%
^{241}Pu-0.352 wt%	^{235}U-0.659 wt%
^{242}Pu-0.072 wt%	^{234}U-0.005 wt%
^{238}Pu-0.027 wt%	

[b] Chemical impurities:

$$\frac{Fe}{Pu} = 1.67 \times 10^{-2} \text{ by weight}$$

$$\frac{Gd}{Pu} = 2.493 \times 10^{-4} \text{ by weight}$$

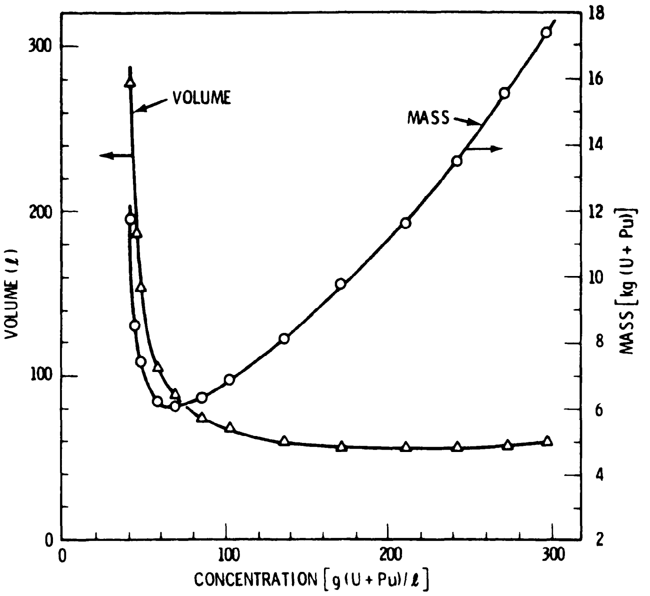

Fig. 2. Criticality of plutonium-uranium solution in cylindrical geometry.

of 0.683 eV was used in these calculations. Ten thousand neutron histories were used in the KENO calculations, resulting in a standard deviation of ±0.008. The computed criticality factors were found to be slightly above unity, with the average value of k_{eff} being 1.022.

The experiments in spherical geometry utilized low-^{240}Pu-content (~4.7 ^{240}Pu) material with the plutonium concentration in the uranium being ~30 wt% of the total uranium and plutonium in the first

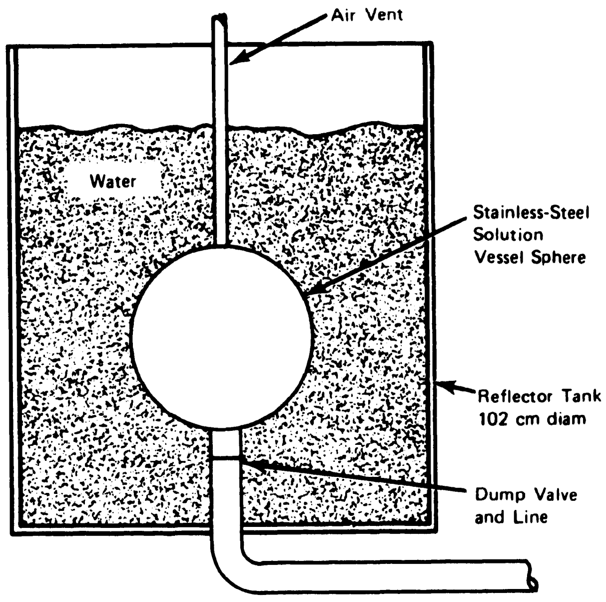

Fig. 3. Schematic of experimental set-up.

TABLE II

Criticality of (U + Pu) Solution in Spherical Geometry

Approximate Percent Plutonium	Sphere Wall Thickness (cm)	Plutonium[a] Concentration (g/ℓ)	Uranium[a] Concentration (g/ℓ)	Gadolinium Concentration (g/ℓ)	Acid Molarity	Specific Gravity	Critical Radius (cm)	Critical Volume (ℓ)	Critical Mass kg (Pu + U)	Critical Mass kg Pu	k_{eff} ENDF/B-III HFN
30	0.112	70.93	157.1	0.051	3.12	1.429	17.87	23.90	5.45	1.69	1.022
30	0.122	35.05	75.7	0.025	1.49	1.215	19.30	30.14	3.34	1.06	1.018
15	0.122	45.6	264.9	0.005	2.1	1.491	19.31	30.18	9.37	1.38	1.031

[a]Isotopic composition, wt%:

Plutonium	*Uranium*
^{238}Pu 0.01	^{235}U 0.66
^{239}Pu 95.09	^{236}U 0.01
^{240}Pu 4.66	^{238}U 99.32
^{241}Pu 0.22	
^{242}Pu 0.01	

TABLE III

Effect of Gadolinium on the Critical Spherical Radius

Experiment	Measured R_c (cm)	Gadolinium (g/ℓ)	k_{eff} [a]	Calculated R_c with no Gadolinium (cm)
001R	17.87	0.051	1.022	17.14
007	19.30	0.025	1.018	18.36
011	19.31	0.005	1.031	19.26

[a]Assuming no gadolinium, the k_{eff} values would be 1.047, 1.045, and 1.032, respectively.

two experiments and ~15 wt% in the latter one. Figure 3 shows a schematic of the experimental set-up. The spheres were water-reflected with 18 cm of water above the sphere. The vent was 5.7-cm-i.d. tubing with a 0.3-cm-thick wall. Spheres of 35.7 and 38.6 cm diam were used in the experiments. These diameters were determined from accurate volume measurement. The sphericity of the vessels was very good; the maximum variation in the measured diameter was 0.1 cm. Table II gives the experimental data and the calculated k_{eff} values attained with ENDF/B-III cross sections. The 18-group multigroup constants were obtained using the GAMTEC-II code. The k_{eff}'s were calculated using the HFN code. These values also showed a code-positive bias with an average of ~2.4% above unity.

Experiments performed in the system before the present set of measurements involved the use of gadolinium poison. Some of the gadolinium from the lines and tanks was picked up during transfer of these solutions, even though considerable rinsing was done before beginning the experiments. The effect of this gadolinium contaminant is shown in Table III. The critical radii were calculated assuming the same k_{eff} as calculated but removing the gadolinium. Although the nuclear densities of the gadolinium were very small, its effect on k_{eff} was appreciable. Calculating k_{eff} disregarding the gadolinum gave k_{eff} as 1.047, 1.045, and 1.032, respectively, It is very important to include the effect of small quantities of such contaminants with large neutron cross sections.

In the case of the spheres, comparative calculations also were made showing the differences obtained using GAMTEC, ENDF/B-II, and ENDF/B-III cross-section data. The values are given in Table IV. For these experiments, the average k_{eff} is ~1.3, 1.6, and 2.4% high, respectively, with GAMTEC, ENDF/B-II, and ENDF/B-III cross-section data.

The results of these calculational correlations indicate we were able to calculate k_{eff} for the critical experimental systems with the (U+Pu) solutions to within ~2% on the conservative side of criticality. However, note that no improvement was obtained in using the ENDF/B-III cross-section data, which gave, in fact, the poorest agreement in the sphere experiments.

TABLE IV

Comparison of Cross-Section Sets

(k_{eff} from HFN Code)

Experiment Number	GAMTEC	ENDF/B-II	ENDF/B-III
001R	1.011	1.015	1.022
007	1.007	1.010	1.018
011	1.020	1.024	1.031
Average	1.013	1.016	1.024

CONCLUSION

The experimental data cover a wide range of (U+Pu) concentrations (30 to 310 g/ℓ) and bracket the minimum critical mass and minimum critical volume. Using these data as calculational benchmarks, the results can be extended to areas of specific interest or need.

The KENO code, with ENDF/B-III cross-section data, as well as the HFN code, provides conservative results on the criticality factors for these systems; that is, the calculated values of k_{eff} are greater than the experimental ones. The average value of the computed k_{eff} is 1.022 for the cylinders and 1.024 for the spheres. Thus, the calculated critical masses and volumes would generally be smaller than those measured (by ~2% in terms of k_{eff}).

ACKNOWLEDGMENT

This paper is based on work performed under Contract No. E(45-1)-1830 between the U.S. Energy Research and Development Administration and Battelle Memorial Institute.