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The Criticality of High Burnup Plutonium

R. C. Lloyd and E. D. Clayton

Battelle-Pacific Northwest Laboratories, Experiments and Operations Section
Richland, Washington 99352

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A series of criticality experiments have been completed with plutonium nitrate solutions made up from extremely high burnup fuel (^{239}Pu isotopic concentration $<\frac{1}{2}$ total Pu). The measurements were performed on a large, 61-cm-diam, water-reflected, cylindrical vessel. The critical experiment data were analyzed by means of the KENO Monte Carlo code utilizing both ENDF/B-II and -III cross sections; the computed criticality factors were in the range of 1.6 to 1.9% above unity. The effects of the various heavy element isotopes on criticality were analyzed. The results show the importance of properly accounting for, and treating, the effects of each of the various isotopes in computing criticality. Even the presence of as little as 1% of ^{241}Am can cause a change in the reactivity of the solutions used in these experiments by ~1%.

INTRODUCTION

Plutonium isotopes other than ^{239}Pu are invariably present in all plutonium. The presence of these isotopes can significantly affect the criticality control limits established for ^{239}Pu. The effect on criticality of the even-neutron nuclides such as ^{240}Pu and ^{242}Pu is to cause the critical dimensions and masses for the isotopic mixture to be greater than those for ^{239}Pu alone. However, the effects are strongly dependent on the neutron spectrum and any moderating diluent, such as water, that may be present. The highly fissile odd-neutron nuclide ^{241}Pu will cause significant reductions in the critical dimensions and masses below those of ^{239}Pu.

The minimum critical mass for ^{241}Pu has been calculated to be only 260 g for a concentration of 32 g/liter in water, and the limiting critical concentration for this nuclide in water is only ~5 g/liter.[1]

A series of experiments has been completed with plutonium nitrate solution made up from extremely high burnup fuel (^{239}Pu isotopic concentration <1/2 total Pu). The experiments were performed to establish the combined effects of the various heavy element isotopes on criticality. The data are provided for use in criticality safety guidance. The comparison of calculated criticality with measurements provides an integral check on the cross sections and computational methods used.

EXPERIMENTAL PROCEDURE

The experiments were performed with a 61.028-cm-i.d., water-reflected, cylindrical vessel (Fig. 1). The vessel was constructed out of

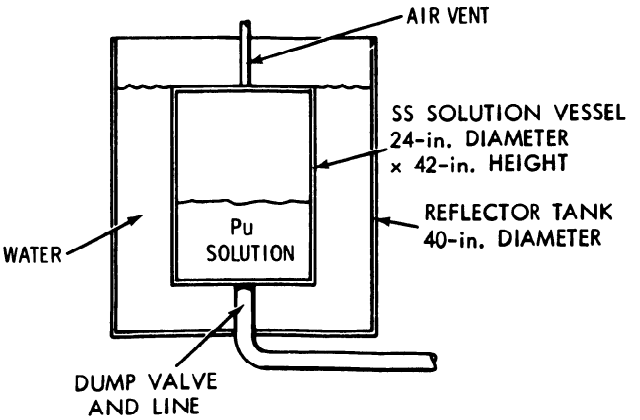

Fig. 1. Schematic of experimental setup.

[1]E. D. CLAYTON and S. R. BIERMAN, *Actinides Rev.*, 1, 408 (1971).

Type-304-L stainless steel with a wall thickness of 0.079 cm. Plutonium concentrations were in the range of ~40 to 140 g Pu/liter with nitric acid molarities being from ~1.5 to 5. The plutonium was in the tetravalent state. The isotopic concentration of the plutonium, as measured on August 11, 1971, was ^{238}Pu - 0.2 wt%, ^{239}Pu - 41.4 wt%, ^{240}Pu - 42.9 wt%, ^{241}Pu - 10.8 wt%, and ^{242}Pu - 4.7 wt%. A nonnegligible amount of ^{241}Am was present to the extent of being 1.08 wt% of the total Pu. Gadolinium contamination came from tanks that contained solutions used previously in poisoned experiments.

DATA CORRELATION AND ANALYSIS

Nine data points were obtained with the critical heights and masses determined for various plutonium concentrations and nitric acid molarities in the plutonium nitrate solutions. Figure 2 shows the variation in the measured critical height as a function of plutonium concentration.

It is evident that, for a solution system such as this, the critical mass, as well as the critical volume, displays a minimum with concentration change in the large cylindrical vessel. The effect, in this case, is somewhat enhanced due to the fact that a low nitrate water mixture was used for dilution of the solution starting at ~5*M* nitric. Thus, the acid molarity and nitrate concentration are decreased more, relative to the initial point, than if 5*M* nitric acid had been used. The criticality connotation is that the addition of water to any solution containing plutonium at more than

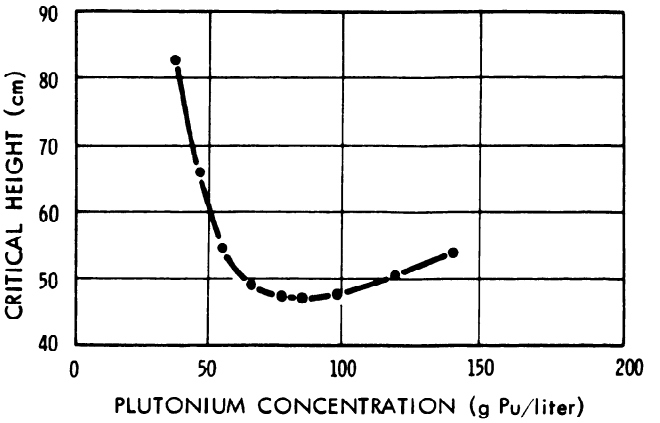

Fig. 2. Criticality of high burnup plutonium solutions.

~80 g Pu/liter will cause a reduction in critical volume and may also cause a reduction in critical mass. For ^{239}Pu-water solutions, the critical mass will display a minimum with concentration, but the critical volume continues to decrease monotonically as the plutonium concentration is increased.

The data from the criticality experiments (conducted from August 9 to 19, 1971) with the plutonium have been reduced, yielding critical volumes for the plutonium nitrate solutions used in the 61.028-i.d., water-reflected, cylindrical vessel. The results are given in Table I.

Values of k_{eff} were computed for the critical assemblies by means of the 18-group cross sec-

TABLE I

Criticality of High Burnup Plutonium Solutions

Pu Concentration[a,b] (g/liter)	Nitric Acid Molarity	Critical Height in 24-in.-diam Cylinder (cm)	Critical Volume (liter)	Critical Mass (kg)	Computed k_{eff} with the KENO Code[c]	
					ENDF/B-II	ENDF/B-III
140.0	5.02	54.70	158.3	22.16	1.027 ± 0.008	1.033 ± 0.007
116.0	4.14	50.55	147.9	17.16	1.020 ± 0.006	1.023 ± 0.007
99.3	3.67	48.26	141.2	14.02	1.012 ± 0.007	1.018 ± 0.006
85.5	3.12	47.00	137.5	11.76	1.020 ± 0.005	1.018 ± 0.006
75.6	2.74	47.29	138.3	10.46	1.006 ± 0.007	1.020 ± 0.007
65.1	2.41	49.12	143.7	9.35	1.014 ± 0.007	1.013 ± 0.005
56.3	2.05	52.83	154.5	8.69	1.022 ± 0.007	1.019 ± 0.006
46.8	1.70	63.47	185.7	8.69	1.014 ± 0.005	1.011 ± 0.006
40.6	1.46	80.92	236.7	9.61	1.007 ± 0.005	1.016 ± 0.005
					1.016	1.019

[a]Isotopic Composition - wt%: ^{238}Pu = 0.2%; ^{239}Pu = 41.4%; ^{240}Pu = 42.9%; ^{241}Pu = 10.8%; ^{242}Pu = 4.7%.
[b]^{241}Am = 1.08% Pu; Gd = 0.0089% Pu.
[c]The water content of the solutions was determined from the equation:

$$H_2O\ (g/liter) = 1000 - 0.3618\ Pu\ (g/liter) - 33.05\ (molarity\ nitric\ acid).$$

tions and the KENO Monte Carlo code.[2] Briefly, the 18-group averaged cross-section data were obtained from the ENDF/B-II and -III libraries by using the FLANGE code[3] to process the thermal data and the ETOG code[4] to process the epithermal data for broad-group averaging in the GAMTEC-II code.[5] The calculated values of k_{eff} are presented in Table I. The checks against the integral critical experiments display a somewhat positive bias, with the average computed k_{eff} being ~2% above unity where ENDF/B-III cross-section data are used.

A general concern in using plutonium experimental data as a benchmark to evaluate computational methods has been the proper accounting for the effect of the various isotopes. It is known that, under some circumstances, the quantity of ^{241}Am present can cause significant changes in the value of k_{eff}. (The ^{241}Am results from the beta decay of the 13-yr half-life ^{241}Pu.) 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).[1]

The computed effect for the 1.08 wt% ^{241}Am is to lower the value of k_{eff} from 0.08 to 1.2%, as shown in Table II. The effects of the other isotopes have been examined for the solution containing 85.5 g Pu/liter. The results are presented in Table III.

TABLE II

Effect of Americium-241

Pu Concentration (g/liter)	δK for 1.08% ^{241}Am
40.6	-0.008
85.5	-0.010
140.0	-0.012

TABLE III

Effects of Plutonium Isotopes

Isotope (wt%)	δK
^{238}Pu (0.2)	-0.001
^{242}Pu (4.7)	-0.009
^{240}Pu (42.9)	-0.371
^{241}Pu (10.8)	+0.103
^{241}Pu replaced by ^{239}Pu	-0.036

In the case of ^{241}Pu, the change in k_{eff} is that which would result from adding 10.8 wt% of this isotope to the solution originally containing no ^{241}Pu, so as to result in the final concentration of 85.5 g Pu/liter. It may be more appropriate to consider how much more reactive the ^{241}Pu would be relative to a like addition of ^{239}Pu. This is shown in the last entry in Table III. If the 10.8 wt% ^{241}Pu were replaced with a like amount of ^{239}Pu, k_{eff} would be less by ~3.6%; that is, treating the ^{241}Pu as equivalent to ^{239}Pu would cause a significant error of nearly 4% in k_{eff} which would be nonconservative (k_{eff} calculated ~4% too small).

CONCLUSIONS

For accurate estimates of criticality, it is important that each isotope be properly accounted for, and treated, in the calculations on high burnup fuels. The effect of the various isotopes depends not only on their relative concentrations but also on the degree of moderation in the system. The results show that treating ^{241}Pu as equivalent to ^{239}Pu in the material of these experiments would result in underestimating k_{eff} by ~4%. Even the presence of small quantities of ^{241}Am can cause significant changes in reactivity, or be the cause of significant errors in the reactivity.

The criticality factors (k_{eff}'s) computed by means of the KENO code using the 18-group cross sections generated by the GAMTEC-II code and ENDF/B-III cross sections were found to be within 2% of unity, indicating a bias on the high side, or a conservative result; that is, the critical systems were calculated as being slightly supercritical. The latter should not be construed to imply similar results of accuracy in calculations on more highly concentrated solutions (undermoderated systems) where the effect of the ^{240}Pu (42.9 wt% in this case) would be much more pronounced in its effect on reactivity than those reported in this paper.

ACKNOWLEDGMENT

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