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Criticality of Plutonium Nitrate Solution Containing Soluble Gadolinium

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Experiments were performed to establish the effect of a soluble neutron absorber (gadolinium nitrate) on the criticality of plutonium nitrate solutions. The solutions contained plutonium at concentrations of ~116 g Pu/liter and at ~363 g Pu/liter. Measured quantities of gadolinium nitrate were mixed with these solutions to produce changes in critical solution height within a 24-in.-diam water-reflected cylinder. Gadolinium concentrations up to 20.25 g Gd/liter were used and the effect determined through the observed change in height. Monte Carlo calculations were used to compute the criticality factors (k_{eff}'s) for each of the measured critical configurations. The computed factors were below unity in each case (largest departure about 2% less than unity).

The gadolinium proved to be an effective neutron absorber. Its effectiveness decreased significantly, however, in the higher plutonium concentration and faster neutron spectrum. Although comparable values of k_∞ were computed (1.603 and 1.503) for the two plutonium concentrations in the experiments, the calculations show 2.4 g Gd/liter would be required to reduce k_∞ to unity in the first case, whereas about 72 g Gd/liter would have been required in the second (316 g Pu/liter solution).

Curves were prepared showing the computed quantities of gadolinium required to reduce k_∞ to unity as a function of plutonium concentration. Also included are computed critical radii for infinitely long cylinders of plutonium nitrate solution for several different gadolinium concentrations.

There was no evidence of chemical instability (or precipitation) of the gadolinium in the plutonium nitrate solution during the course of the experiments and over a 1-mo long test (a question of concern in using soluble poison for criticality control).

INTRODUCTION

Experimental criticality data are needed on soluble neutron absorbers (poisons) for an accurate determination of effectiveness relevant to their possible use for criticality control and prevention in the storage and processing of solutions bearing fissile material. A series of criticality experiments[1,2] was performed utilizing gadolinium as a soluble poison in plutonium nitrate solutions. The gadolinium was chosen because of its high neutron cross section, high solubility, and compatibility in the separation process. The experiments were oriented toward providing data on the effectiveness of gadolinium as a soluble absorber for use in nuclear criticality prevention; data on integral critical experiments also are provided against which calculational techniques and cross-section sets can be checked.

EXPERIMENTS AND CORRELATION OF DATA

Two series of criticality experiments were performed with plutonium nitrate solution homo-

[1] R. C. LLOYD and L. E. HANSEN, "The Effect of Soluble Nuclear Poisons on Criticality," Reactor Physics Quarterly Report, April, May, June 1971, BNWL-1522-3, Pacific Northwest Laboratory (1971).

[2] R. C. LLOYD, E. D. CLAYTON, and L. E. HANSEN, *Trans. Am. Nucl. Soc.*, **14**, 37 (1971).

geneously poisoned with gadolinium. Plutonium concentrations of 116 and 363 g Pu/liter with acid molarities of 1.85 and 4.1, respectively, were used. The ^{240}Pu content was 8.3 wt%. The experimental vessel in which the experiments were performed was a 61.03-cm i.d., stainless-steel cylinder, having a wall thickness of 0.079 cm. A second vessel enclosed the criticality vessel to provide for a water reflector having a minimum thickness of 20 cm on sides and bottom. The water reflector extended to the top of the experimental cylinder, h = 106.7 cm.

The gadolinium concentration in the first series was varied from 0 to 2.4 g Gd/liter in five equal steps while maintaining the plutonium concentration at 116 g Pu/liter. Figure 1 shows the measured critical height of the poisoned plutonium solution as a function of the gadolinium concentration. Calculated values of k_∞ have been made for these solutions (Fig. 2), which show k_∞ equal to unity at a gadolinium concentration of 4 g Gd/liter.

The second series of experiments involved gadolinium posioned solutions at a plutonium concentration of 363 g Pu/liter. For the more highly concentrated plutonium solution, the gadolinium concentration ranged from 4 to 20 g Gd/liter. Figure 3 shows the measured critical height as a function of the gadolinium concentration. At a gadolinium concentration of 20 g/liter, the critical mass of plutonium was 63.1 kg compared with about 21 kg in the absence of gadolinium.

The calculated variation of k_∞ with the gadolinium content is given in Fig. 4. In this case, the results indicate that a gadolinium concentration of about 72 g Gd/liter would be required to reduce k_∞ to unity.

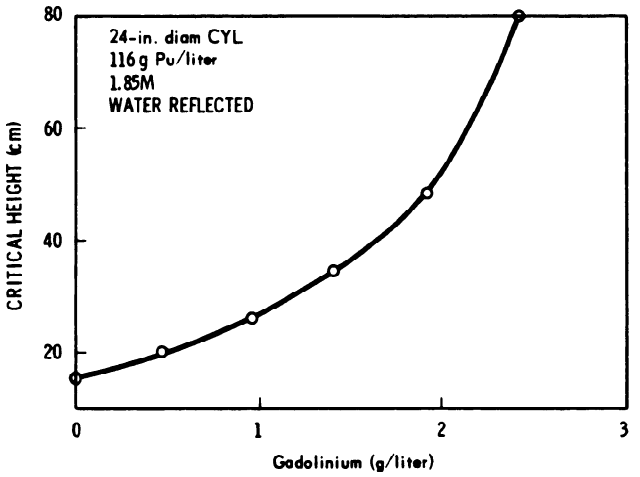

Fig. 1. Effect of gadolinium on the criticality of $Pu(NO_3)_4$.

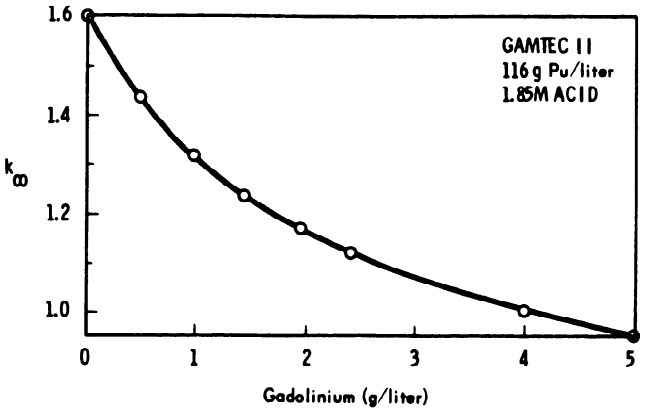

Fig. 2. Effect of gadolinium on k_∞ of $Pu(NO_3)_4$.

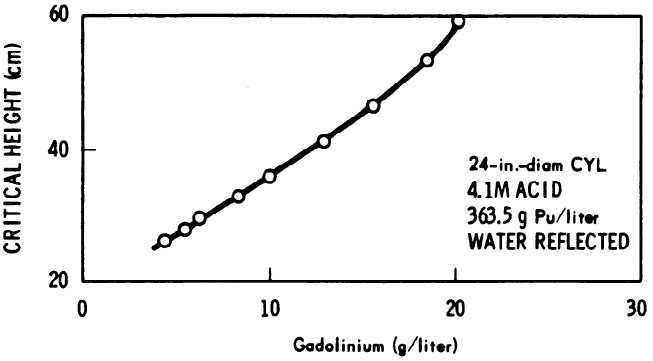

Fig. 3. Effect of gadolinium on the criticality of $Pu(NO_3)_4$.

Included in Table I are the critical values and calculated effective criticality factors for the experimental assemblies. KENO, a multigroup Monte Carlo code,[3] was used to calculate effective multiplication factors for the measured critical systems. The calculations were performed by processing 10 000 neutron histories. Multigroup cross-section data, 18 energy groups, used in these calculations were averaged by the GAM-TEC-II code,[4] the 17 fast group constants ($E >$ 0.683 eV) were averaged over a 66 group slowing down spectrum computed using the B_1 approximation to the Boltzmann equation. The thermal group data ($E <$ 0.683 eV) were averaged over a Wigner-Wilkins spectrum. The fine group cross-

[3]G. E. WHITESIDES and N. F. CROSS, "KENO—A Multigroup Monte Carlo Criticality Program," CTC-5, Union Carbide Corporation (1969).

[4]L. L. CARTER, C. R. RICHEY, and C. E. HUGHEY, "GAMTEC-II: A Code for Generating Consistent Multigroup Constants Utilized in Diffusion and Transport Theory Calculations," BNWL-35, Pacific Northwest Laboratory (1965).

TABLE I

Criticality of Gd_2O_3-Poisoned Plutonium Solutions*

Concentration		Critical Values			GAMTEC-KENO Computed k_{eff}
Plutonium[a] (g Pu/liter)	Gadolinium (g Gd/liter)	Height (cm)	Volume (liter)	Mass (kg Pu)	
116	0.00	15.44	45.17	5.240	0.993 ± 0.009[b]
116	0.48	20.16	58.98	6.842	0.980 ± 0.008
116	0.96	25.98	75.99	8.815	0.980 ± 0.006
116	1.42	34.48	100.85	11.698	0.978 ± 0.006
116	1.92	48.28	141.24	16.383	0.976 ± 0.005
116	2.38	80.37	235.10	27.272	0.983 ± 0.005
363	4.40	26.52	77.56	28.192	0.977 ± 0.006
363	5.28	28.07	82.08	29.838	0.986 ± 0.006
363	6.28	29.62	86.65	31.497	0.997 ± 0.007
363	8.21	32.84	96.10	34.932	0.984 ± 0.006
363	9.88	36.04	105.40	38.311	0.992 ± 0.006
363	12.58	41.02	120.00	43.608	0.986 ± 0.006
363	15.55	46.76	136.76	49.714	0.985 ± 0.006
363	18.4	53.70	157.05	57.089	0.977 ± 0.005
363	20.25	59.36	173.63	63.113	0.985 ± 0.004

*61.03-cm i.d. water-reflected, cylindrical, stainless-steel vessel.

[a]Chemical analysis - 116 g Pu/liter, 1.85 M acid, 238.9 g NO_3/liter total, sp. gr. 1.2552
- 363 g Pu/liter, 4.1 M acid, 634.1 g NO_3/liter total, sp. gr. 1.7010.
Isotopic analysis (wt) - ^{238}Pu = 0.004, ^{239}Pu = 90.677, ^{240}Pu = 8.379
- ^{241}Pu = 0.851, ^{242}Pu = 0.049.

[b]Statistical deviation from Monte Carlo calculation.

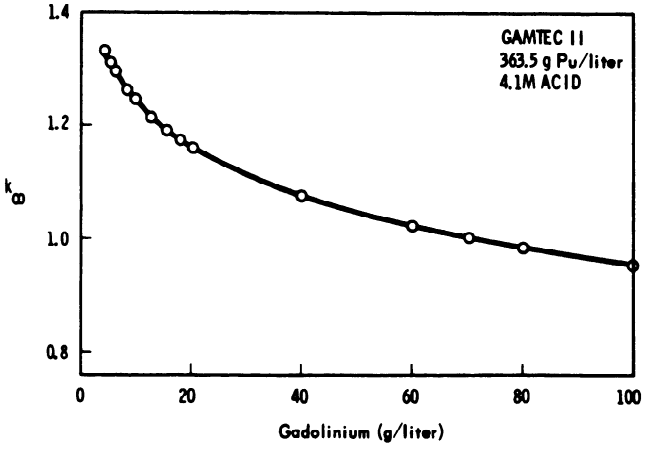

Fig. 4. Effect of gadolinium on k_∞ of $Pu(NO_3)_4$.

section data on the GAMTEC library were obtained from the GAM-I and TEMPEST libraries[5,6] for all isotopes of interest here except ^{239}Pu and gadolinium. The ^{239}Pu cross-section data used were normalized to the 0.0253-eV values obtained from a 1962 least-squares analysis by Sher and Felberbaum.[7] The gadolinium fine-group data were obtained from the ENDF/B library in a suitable fine-energy group structure by the FLANGE-II code[8] and the ETOG code[9] for the thermal and fast energy ranges, respectively. The material identification number of the data used is 1030.

As can be seen in Table I, the computed, criticality factors are within 2% of the experimental values (unity), and are nonconservative from a criticality safety standpoint.

[5]G. D. JOANOU and J. S. DUDEK, "GAM-I: A Consistent P_1 Multigroup Code for the Calculation of Fast Neutron Spectra and Multigroup Constants," GA-1850, General Atomic (1961).

[6]R. H. SHUDDE and J. DYER, "TEMPEST-II: A Neutron Thermalization Code NAA Program Description," TID-18284, Atomics International (1962).

[7]R. SHER and J. FELBERBAUM, "Least Squares Analysis of 2200 m/sec Parameters of U^{233}, U^{235}, and Pu^{239}," BNL-722, Brookhaven National Laboratory (1962).

[8]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).

[9]D. E. KUSNER, R. A. DANNELS, and S. KELLMAN, "ETOG-I: A FORTRAN-IV Program to Process Data from the ENDF/B File to the MUFT, GAM, and ANISN Formats," WCAP-3845-1 (ENDF-114), Westinghouse Electric Corporation (1969).

CALCULATIONS FOR NUCLEAR SAFETY GUIDELINES

Calculations were made to determine the quantity of gadolinium required as an additive to various plutonium solutions to result in $k_\infty = 1$ in one case and to 0.96 in the second. The solutions were assumed to be made up from plutonium containing 100% ^{239}Pu and to contain no excess acid. The results are presented in Fig. 5. Note the reduction in the effectiveness of gadolinium as a neutron absorber at higher concentrations, as the spectrum hardens.

Since the computed criticality factors were nonconservative by up to about 2%, i.e., the critical configurations were calculated as being subcritical by up to this amount, the curves in Fig. 5 have been adjusted accordingly to provide correct estimates at $k_\infty = 1.0$, and also at $k_\infty = 0.96$. The two curves show the amount of gadolinium required to reduce k_∞ to unity and by 4% below unity. The curves also serve to illustrate the quantity of gadolinium that might be required to specify a certain level of subcriticality, the latter type of knowledge being of principal interest in criticality prevention.

The relative effectiveness of gadolinium, cadmium, and boron poisons are shown in Fig. 6. The cadmium and boron values come from curves in ARH-600.[10] Boron appears to be better at higher concentrations; however, due to the low solubility of boron compounds, poisoning ($k_\infty < 1$) of solutions above 100 g Pu/liter becomes academic.

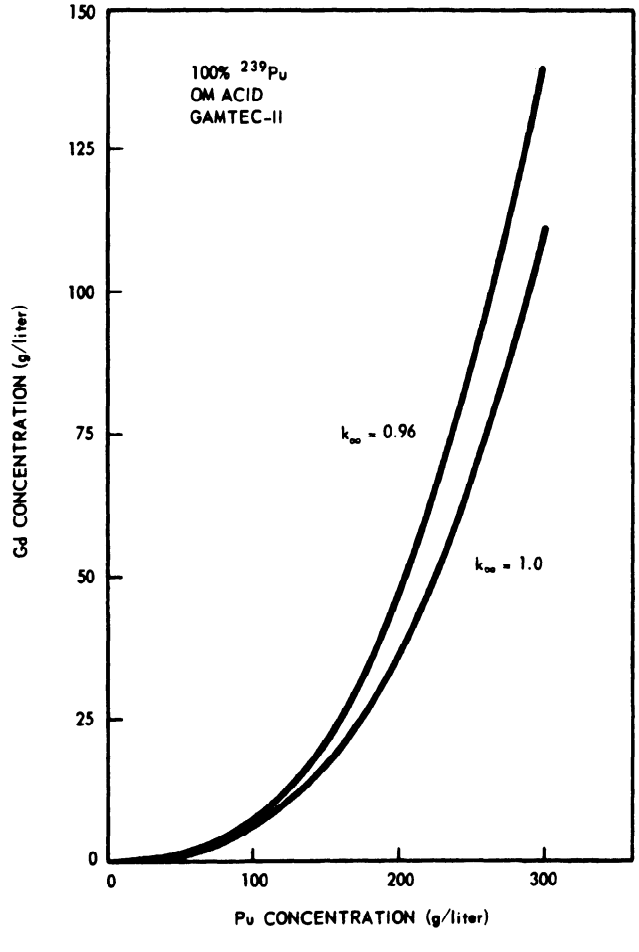

Fig. 5. Gadolinium required as an additive in plutonium solution to result in $k_\infty = 1$.

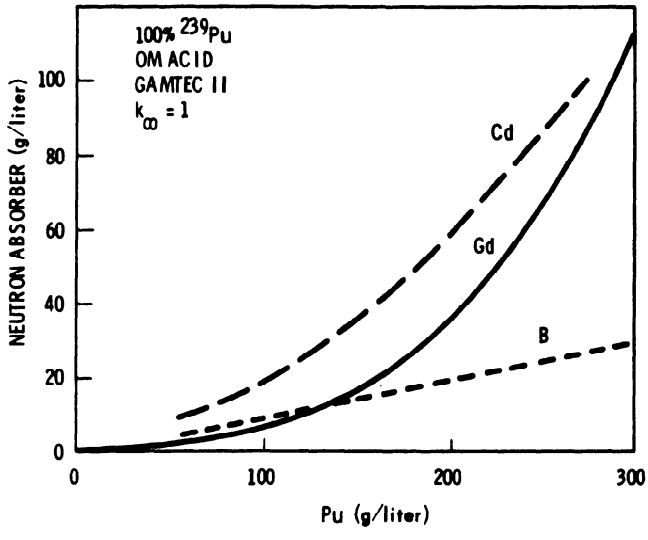

Fig. 6. Neutron absorbers required as an additive in plutonium solution to result in $k_\infty = 1$.

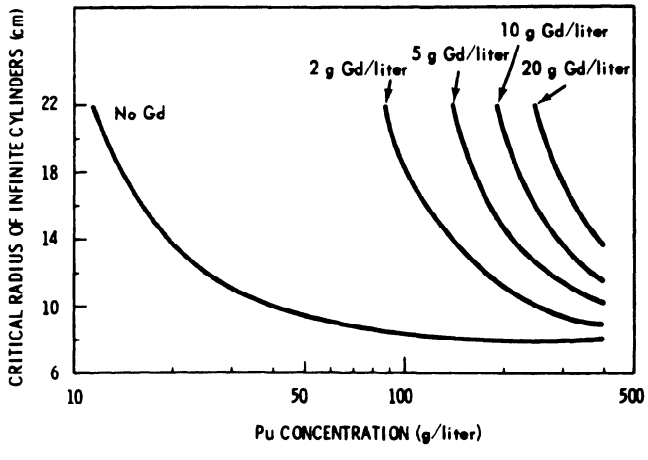

Fig. 7. Criticality of $Pu(NO_3)_4$ containing $Gd(NO_3)_3$ in infinitely long cylinders.

[10] R. D. CARTER, G. R. KIEL, and K. R. RIDGWAY, *Criticality Handbook, Volume II*, ARH-600, Atlantic Richfield Hanford Company (1969).

Calculated critical radii of infinite cylinders over a range of plutonium concentrations up to 400 g Pu/liter for several additions of gadolinium up to 20 g Gd/liter are shown in Fig. 7. These were calculated for water-reflected systems of ^{239}Pu containing no excess nitric acid and neglecting the stainless-steel walls of the container, which would give more conservative values than would be found in practice. Critical radii of cylinders of solutions containing gadolinium are compared to the radii of solutions having no gadolinium present.[11] Calculations were made using GAMTEC-II to generate 18-group cross sections. The HFN diffusion theory code[12] was used to calculate critical radii of the infinite cylinders.

[11]C. R. RICHEY, *Nucl. Sci. Eng.*, **31**, 32 (1968).

[12]J. R. LILLEY, "Computer Code HFN—Multigroup, Multiregion Neutron Diffusion Theory in One Space Dimension," HW-71545, General Electric Company (1961).

CONCLUSIONS

Gadolinium was found to be a very effective poison for solutions in the lower concentration ranges, but rapidly decreases in its effectiveness at higher plutonium concentrations as the neutron spectrum hardens. There was no evidence of instability (or precipitation) of the gadolinium in the plutonium nitrate solution during the course of the experiments and over a 1-mo-long test.

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, were found to be within 2% of unity, indicating a bias on the low side, or a nonconservative result; i.e., the critical systems were calculated as being slightly subcritical.

ACKNOWLEDGMENT

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