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Criticality of Plutonium Nitrate Solutions Containing Borated Raschig Rings

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Experimental criticality data on borated raschig rings in plutonium nitrate solutions are presented for use in establishing criticality safety limits and in verifying calculational methods for these type systems. The data cover the concentration range between 63- and 412-g Pu/liter for borosilicate-glass raschig rings containing 0.5 and 4.0 wt% boron, and stainless-steel raschig rings containing 1 wt% boron.

Criticality was possible in all three experimental vessels used (12-, 18-, and 24-in.-diam cylinders, 42-in. high) with no raschig rings. With rings randomly loaded in the vessels only the 24-in. cylinder could be made critical and then only when loaded with the 0.5 wt% borated rings. The minimum critical volume for this system, poisoned with 19.27 vol% borosilicate-glass rings containing 0.5 wt% boron, was determined to occur at about 300 g Pu/liter as compared to 175- to 200-g Pu/liter without the rings. The minimum critical mass occurred at ~110-g Pu/liter with the system poisoned, as compared with 30-g Pu/liter if the system had not been poisoned. Exponential measurements on the subcritical assemblies, loaded with 4 wt% borated rings displacing 18.78 vol% solution, indicated that negative bucklings existed for all plutonium nitrate solutions having concentrations below 391-g Pu/liter. Similar measurements on the subcritical assemblies, loaded with 1 wt% borated stainless-steel rings displacing 27 vol% solution, indicated that negative bucklings existed for all concentrations below 412-g Pu/liter.

Comparisons between the experimental data and the results of several calculational methods indicate that the validity of a particular calculational technique may be limited to a small concentration region. By treating the raschig rings as vertical parallel tubes displacing an equal volume of solution and using the Monte Carlo code KENO with GAMTEC-II cross sections averaged over the energy spectrum of the plutonium solution, k_{eff} values were calculated to within 2% of unity for the experimental critical assemblies presented in this paper. Other calculational methods and cross-section sets used resulted in values of k_{eff} departing from unity by as much as 12% low to 6% high, depending on the plutonium concentration. The various methods used are discussed in this paper.

INTRODUCTION

As enrichment requirements are increased and as power reactors go to plutonium-uranium fuel mixtures, fixed and soluble neutron absorbers are receiving more attention as a means of assuring criticality safety, particularly in those process areas for which the parameters affecting criticality are potentially subject to wide variations. Neutron absorbers are also being used to permit utilization of existing facilities for processing the higher enrichments being encountered.

A new standard on the use of raschig rings as neutron absorbers in solutions of fissile materials has recently been issued.[1] However, few data

[1]N16.4-1971, American National Standard, "Use of Borosilicate Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material," American National Standards Institute, Inc.

exist on the use of raschig rings in plutonium and uranium systems. In addition to the supporting document[2] upon which the new standard is based, experiments on uranium solutions have been published by Thomas et al.,[3] and recently some experimental work has been reported on plutonium solutions by Barbry et al.[4] To obtain additional data for use in establishing criticality safety guidelines and for checking calculational models, experiments have recently been performed at Hanford with stainless-steel raschig rings and with borosilicate-glass raschig rings in plutonium solution. The results of these experiments are presented in this paper, along with some calculational comparisons.

EXPERIMENTAL SYSTEM

Although the experiments covered a wide range of plutonium concentration, they could not cover all possible configurations and types of poison-loaded raschig rings. Consequently, the design of the experiments was oriented toward obtaining data for use in checking calculational techniques over the entire neutron energy spectrum normally encountered in plutonium-nitrate solutions, and to obtain relative data on the type of rings most widely used.

The experiments were carried out in 42-in.-high, Type 304-L stainless-steel cylinders. Measurements were made at three different cylinder diameters—12, 18, and 24 in. The experimental vessels were fully reflected with water except directly above the solution. The cylindrical region above the solution in each measurement was made up of an air-raschig ring region directly above the solution, followed by an air region and then the stainless-steel top of the experimental vessel. The volume displacements of the rings were determined by measuring the water volume (using a volumetric flask) of the tank with and without the rings. The uncertainty in the volume displacements is <0.5%. A diagram of the experimental system is shown in Fig. 1. A photograph of a vessel filled with rings, prior to installation in the experimental system, is shown in Fig. 2.

The stainless-steel raschig rings were $\frac{1}{2}$-in. o.d., $\frac{1}{2}$ in. long, with a $\frac{1}{16}$-in.-thick wall, and contained 1±0.05 wt% natural boron. (The effectiveness of these types of rings is of special interest for use in sumps.) The glass rings were 1.5-in. o.d., 1.25-in. i.d., and 1.7 in. long. Measurements were made with two different boron concentrations in the glass rings. These were $\frac{1}{2}$±0.05 wt% and 4±0.1 wt% boron in the form of B_2O_3.

The plutonium concentration in the plutonium-nitrate solution was varied over a range between 63- to 412-g Pu/liter during the course of the measurements. Information on the various plutonium-nitrate solutions and raschig rings on

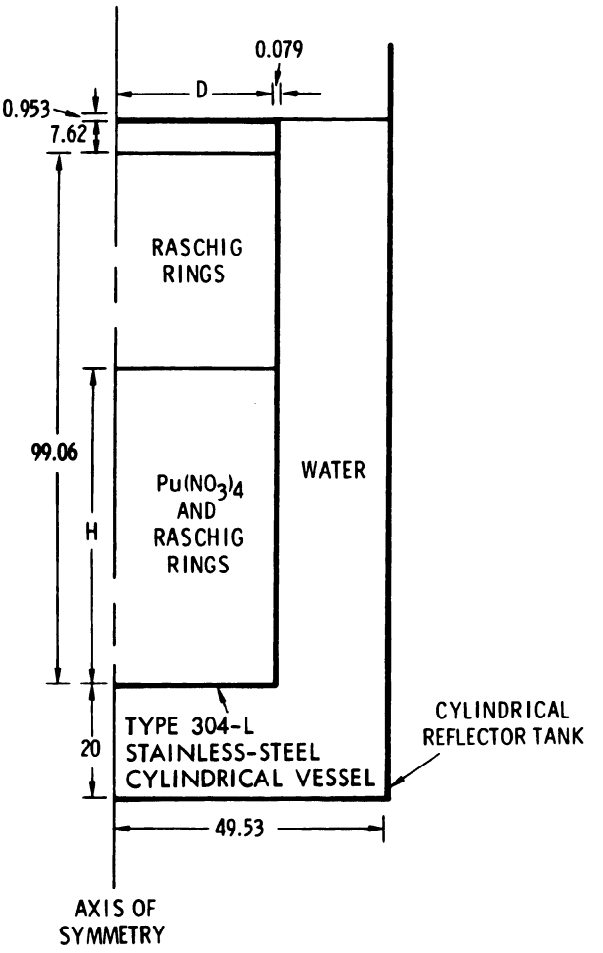

Fig. 1. Diagram of experimental system.

[2]J. T. THOMAS, J. K. FOX, and E. B. JOHNSON, "Critical Mass Studies. Part XIII. Borosilicate Glass Raschig Rings in Aqueous Uranyl Nitrate Solutions," ORNL-TM-499, Oak Ridge National Laboratory (1963).

[3]J. P. NICHOLS, C. L. SCHUSKE, and D. W. MAGNUSON, "Use of Borosilicate Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material," Y-CDC-8, Union Carbide Corporation, Y-12 Plant (1971).

[4]F. BARBRY, J. C. BOULY, R. CAISZERGUES, E. DEILGAT, M. HOUELLE, and P. LeCORCHE, "Etude Experimentale Et Theorique De L'Empoisonnement Heterogene De Solution De Matiere Fissile Par Des Tubes Ou Des Anneaux En Verre Au Borosilicate," Rapport CEA-#-3931, Centre d'Etudes Nucleaires de Saclay, Direction de la Protection et de la Surete Radiologiques, Service d'Etudes de Criticite (December 1969).

Fig. 2. Experimental vessel loaded with glass raschig rings.

which measurements were made are given in Table I along with the associated experimental results. A detailed description of the rings is given in Table II and an isotopic analysis of the plutonium is given in Table III.

EXPERIMENTAL DATA

Criticality was possible, of course, in all three of the experimental vessels (12-, 18-, and 24-in.-diam cylinders, 42 in. high) when no neutron poisons were present. However, in the presence of neutron poisons, criticality was possible only in the 24-in.-diam cylinder containing the $\frac{1}{2}$ wt% borated glass rings. The plutonium-nitrate solution height required for criticality in this system is shown in Table I as a function of the plutonium concentration. A plot of these data, which were obtained by critical approach measurements, is shown in Fig. 3 to illustrate the sensitivity of the critical volume-to-concentration changes below about 100-g Pu/liter. Between about 200- and 400-g Pu/liter the critical volume is essentially constant for this poisoned system, with a minimum critical volume occurring at about 300-g Pu/liter. If the system had not contained the rings, the minimum critical volumes would have occurred between 175- and 200-g Pu/liter.[5,6] The relationship between the critical mass of this system and the plutonium concentration is shown in Fig. 4. The minimum critical mass occurs at about 110-g Pu/liter for this poisoned system as compared to 30-g Pu/liter[7] if the system had not been poisoned.

Exponential-type measurements were made on the 24- and 18-in.-diam cylinders containing 4 wt%

[5] R. C. LLOYD, C. R. RICHEY, E. D. CLAYTON, and D. R. SKEEN, *Nucl. Sci. Eng.*, **25**, 165 (1966).

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

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

TABLE I

Summation of Experiments

Plutonium Nitrate Solution					Raschig Rings			Experimental		
Pu[a] (g/liter)	Total Nitrate (g/liter)	Acid Molarity (H+)	Water[b] (g/liter)	Specific Gravity	Material[c]	Nominal Boron Content (wt%)	Solution Displaced (vol%)	Solution Radius (cm)	Solution Height[d] (cm)	Buckling (cm^{-2})
391	573.4	3.1	737.5	1.705	Glass	0.5	19.27	30.495	25.50	0.011135
271	415.1	2.4	824.5	1.513	Glass	0.5	19.27	30.495	25.07	0.011300
197	320.0	2.2	871.8	1.391	Glass	0.5	19.27	30.495	26.11	0.010912
110	300.0	2.9	860.1	1.273	Glass	0.5	19.27	30.495	35.66	0.008462
74	198.0	2.2	910.8	1.185	Glass	0.5	19.27	30.495	59.94	0.006024
63	180.0	2.0	924.0	1.169	Glass	0.5	19.27	30.495	93.98	0.004996
391	573.4	3.1	737.5	1.705	Glass	4.0	18.78	30.475	[e]	0.002
271	415.1	2.4	824.5	1.513	Glass	4.0	18.78	30.475	[e]	-0.0006
121	260.0	2.1	883.9	1.267	Glass	4.0	18.78	30.475	[e]	-0.006
275	480.0	3.4	779.6	1.538	Stainless	1.0	27.0	22.870	[e]	-0.011
412	602.0	3.5	721.5	1.739	steel	1.0	27.0	22.870	[e]	-0.006
275	480.0	3.4	779.6	1.538	None	0	0	15.263	28.804	0.017600

[a]Isotopic analysis given in Table III.
[b]Calculated by difference based on specific gravity.
[c]Chemical composition given in Table II.
[d]Critical height of solution.
[e]Vessel was completely filled and subcritical. Buckling values obtained from exponential measurements.

TABLE II

Description of Raschig Rings

Ring Type	Glass	Glass	Steel
Dimensions, in.			
i.d.	1.25	1.25	0.4375
o.d.	1.50	1.50	0.50
Length	1.70	1.70	0.50
Boron, nominal wt%	0.5	4.0	1.0
Density, g/cm^3	2.51	2.26	7.84
Chemical composition, wt%			
SiO_2	91.55	80.29	---
B_2O_3	1.5	12.76	---
Al_2O_3	2.25	2.25	---
Fe_2O_3	0.05	0.05	---
Na_2O_3	3.50	3.50	---
K_2O	1.15	1.15	---
Fe	---	---	64.1
B	---	---	1.0
Mn	---	---	1.65
Si	---	---	0.64
Cr	---	---	18.3
Ni	---	---	14.1
C	---	---	0.03
P	---	---	0.01
S	---	---	0.01
Mo	---	---	0.11
Cu	---	---	0.05

TABLE III

Isotopic Analysis of Plutonium

Isotope	wt%
238	0.022
239	90.710
240	8.322
241	0.897
242	0.049

borated glass rings and 1 wt% borated stainless-steel rings to determine the bucklings of these subcritical systems. Buckling values for these systems are shown in Table I. In determining these bucklings, as well as those shown for the critical systems, an extrapolation distance of 7 cm was assumed for the water-reflected sides and bottom, and a 5-cm extrapolation distance was assumed for the partially reflected top. For the 4 wt% borated glass rings, only the 391-g Pu/liter concentration system had a positive buckling. Negative bucklings were determined for all lower concentrations and for both concentrations (275- and 412-g Pu/liter) poisoned with 1 wt% borated steel rings.

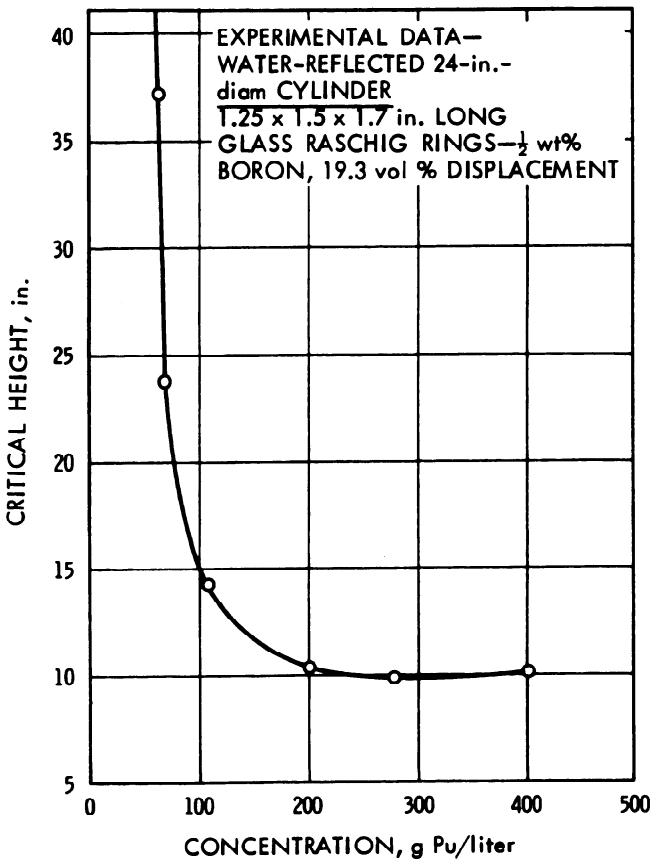

Fig. 3. Critical height as a function of concentration—24-in.-diam cylinder of plutonium nitrate solution containing ½ wt% borated glass raschig rings.

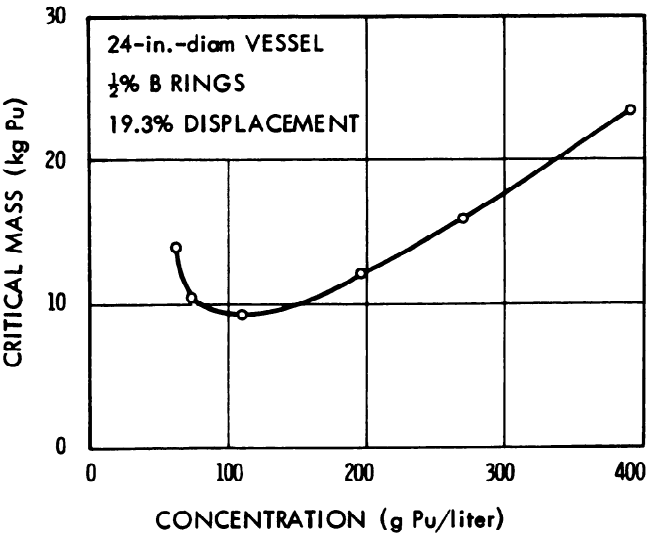

Fig. 4. Critical mass as a function of concentration—24-in.-diam cylinder of plutonium nitrate solution containing ½ wt% borated glass raschig rings.

For nonpoisoned systems, the buckling is essentially constant between 275- and 415-g Pu/liter. However, in these highly poisoned systems, the buckling would be expected to increase with the concentration. Since the boron in the rings is basically a thermal-neutron poison, fewer neutrons are absorbed in the rings as the neutron spectrum becomes faster. Consequently, the rings become less effective as an absorber and the buckling must increase.

THEORY EXPERIMENT CORRELATION

The presence of a fixed, randomly oriented thermal-neutron absorber in an experimental system complicates considerably any attempt to reproduce that system with calculations. To determine what geometrical model best simulates these complex systems and which calculational techniques can be expected to best reproduce the experimental data, k_{eff} values were calculated, by several different techniques, for different models of the critical systems containing ½ wt% borated glass rings. The results of these calculations are summarized in Table IV.

The simplest approach of homogenizing the rings and the plutonium solution on an atom basis results in nonconservative values for establishing criticality safety limits. Using GAMTEC-II cross-section data,[8] both diffusion theory and KENO-I[9] Monte Carlo can calculate the critical condition for unpoisoned plutonium nitrate solutions quite accurately (an example at 275-g Pu/liter is shown in Table IV). However, both tend to overestimate the critical size of the poisoned systems shown in Table IV. The calculated values appear to improve as the plutonium concentration increases, that is, as the neutron spectrum hardens. Apparently, homogenizing the experimental assemblies on an atom basis results in too much additional boron (which is essentially a thermal-neutron absorber) being made available for thermal-neutron capture in the calculations. Treating the boron as essentially a soluble poison in this manner would not have much effect on k_{eff} calculated for the faster systems, whereas its

[8] 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 Laboratories (1965).

[9] G. E. WHITESIDES and N. F. CROSS, "KENO—A Multigroup Monte Carlo Program," CTC-5, Union Carbide Corporation (1969). This program incorporates 16-group neutron cross sections reported by Hansen and Roach in "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos Scientific Laboratory (1961).

TABLE IV

Calculated Critical Assemblies of $Pu(NO_3)_4$ Solution Containing Borated Raschig Rings

Solution Concentration g Pu/liter	Raschig Rings	Homogeneous Model		Equal-Volume Vertical-Tube Model				
		Diffusion Theory[a]	KENO[a,b]	KENO[a,c,d]	KENO[a,c,e]	KENO[c,f]	GEM IV[b,c,g]	GEM IV[g,h]
275	None	0.994	1.001 ± 0.009	1.001 ± 0.009	1.001 ± 0.009	---	---	---
391	[i]	0.990	0.996 ± 0.010	1.025 ± 0.005	1.012 ± 0.006	0.886 ± 0.006	1.063 ± 0.019	1.038 ± 0.017
271	[i]	0.998	0.981 ± 0.008	---	---	---	---	1.055 ± 0.026[j]
197	[i]	1.000	0.983 ± 0.008	1.034 ± 0.007	1.001 ± 0.006	0.951 ± 0.006	---	1.057 ± 0.023[j]
110	[i]	0.971	0.954 ± 0.008	---	---	---	---	1.020 ± 0.018
74	[i]	0.937	0.942 ± 0.007	1.130 ± 0.006	---	---	---	1.000 ± 0.013[j]
63	[i]	0.934	0.929 ± 0.005	1.140 ± 0.005	0.978 ± 0.004	0.998 ± 0.005	0.996 ± 0.012	0.979 ± 0.008[j]

[a] 18-group averaged GAMTEC-II cross sections using the original GAMTEC-II library data. Collapsed to two groups for diffusion theory calculations.

[b] The uncertainty assigned to each of the calculated values of k_{eff} is the standard deviation determined by the statistics of the particular Monte Carlo calculation. In the KENO code 20 000 neutron histories were traced; in the GEM IV, 2000 histories.

[c] Buckling-converted rectangular geometry with tubes on square pitch.

[d] Raschig ring cross sections averaged over the neutron energy spectrum characteristic of raschig ring material.

[e] Raschig ring cross sections averaged over the neutron energy spectrum characteristic of the respective plutonium solution.

[f] 16-group averaged Hansen-Roach cross sections.

[g] GEM-IV cross sections.

[h] Cylindrical geometry of experiments with tubes on triangular pitch.

[i] 1.25- × 1.50- × 1.70-in. raschig rings containing $\frac{1}{2}$wt% boron.

[j] A normal distribution of mR values not attained. Indicated computer time prohibitively long.

effectiveness would be considerably increased in the more thermal systems.

Treating the randomly oriented rings as equal volume, vertical, parallel tubes in a square array and using Hansen and Roach cross sections in the KENO calculations result in an acceptable k_{eff} of 0.998 ± 0.005 for the 63-g Pu/liter critical assembly. However, as the plutonium concentration is increased to 391-g Pu/liter, the KENO calculated k_{eff}, using Hansen-Roach cross sections, decreases to 0.886 ± 0.006 for the critical assembly. These same calculations using GAMTEC-II cross sections, averaged over the energy spectrum of the neutrons in the rings and over the energy spectrum of the neutrons in the plutonium nitrate solution, respectively, result in overly conservative values of k_{eff} for each of the critical assemblies. However, at the more important higher concentrations (with respect to utilization of raschig rings to gain increased processing capacity) the k_{eff} values are only 2 to 3% high. Averaging the neutron cross sections of the material comprising the rings over the energy spectrum of the respective solutions results in KENO-calculated k_{eff} values closer to unity for each of the critical assemblies; however, the value for the more thermalized 63-g Pu/liter system is about 2% low.

Monte Carlo calculations using the computer code GEM-IV[10] were made on the low concentration 63-g Pu/liter system and on the 391-g Pu/liter system for comparison with the KENO calculations. As in the heteorgeneous KENO calculations, the raschig rings were treated as a square array of vertical parallel tubes displacing the same volume of solution as the rings. As can be seen in Table IV, about a 7% spread was obtained between the GEM-IV k_{eff} values for the 63-g/liter and 391-g/liter critical assemblies. The GEM-IV values tend to be slightly higher than the KENO-calculated values. However, it should be pointed out that the GEM-IV cross-section library data were used in the GEM calculations, whereas the GAMTEC-II cross sections, as previously mentioned, were used in the KENO calculations. Consequently, the comparison is not particularly valid with respect to calculational techniques; however, it does provide a comparison that might be useful in the application of these Monte Carlo codes to plant systems.

The GEM-IV code can accommodate triangular pitched tubes in cylindrical geometry. Since this more nearly reproduces the actual experimental assemblies than does the buckling-converted square arrays necessitated in the KENO calculations, k_{eff} values were calculated for each of the critical assemblies shown in Table IV by treating the raschig rings as triangular-pitched parallel vertical tubes displacing the same volume of solution as the rings. This treatment of the experimental assemblies appears to result in a

[10] P. J. HEMMINGS, "The GEM Code," AHSB (S) R 105, United Kingdom Atomic Energy Authority (1967). Neutron cross sections were supplied with the GEM-IV code.

reasonably close grouping of the calculated k_{eff} values about unity for the critical systems. However, as indicated in Table IV, the computer time required to obtain the necessary normal distribution in the GEM-IV parameter MR (a measure of k_{eff}) for several of the critical systems prevented obtaining reliable k_{eff} values for these systems. Even if the computer time were expended, it does not appear that the GEM calculations, with their inherently long running times, offer any advantage over the KENO calculations using GAMTEC-II cross-section data with both the ring and the plutonium-solution cross sections averaged over the neutron energy spectrum of the respective solutions.

Treating the randomly oriented raschig rings as a uniform array of mutually perpendicular rings was not extensively investigated. However, recent studies reported by Thomas and Rückert[11] on these experimental systems have shown a decrease of about 1% in k_∞ by going from tubes to rings. In studies on the Saclay experiments, Barbry et al.[4] also obtained a decrease in k of about 1% by treating the rings as mutually perpendicular instead of vertical parallel tubes. Thomas and Rückert also reported that no trend was observed with plutonium concentration by going to rings.

To evaluate the actual poisoning effect of the rings, KENO calculations were made with the rings treated as vertical parallel tubular voids in a square array displacing the same volume of solution as the rings. The calculations were made for the 63-g/liter, the 197-g/liter, and the 391-g/liter experimental critical assemblies to provide a comparison over the entire concentration range covered by the experiments. The k_{eff} values calculated for these three systems are shown in Table V. Also shown in Table V are the comparable k_{eff} values calculated by KENO for the actual poisoned critical assemblies at these three concentrations. In each case 18-group GAMTEC-II cross-section data, averaged over the neutron energy spectrum of the respective plutonium solution, were used.

As expected, the reactivity effect is due primarily to neutron absorption in the low, 63-g Pu/liter, concentration solution. In this thermalized system, the poisoning effect accounts for about 85% of the total 30% decrease in k_{eff} caused by the introduction of the raschig rings into the solution. Conversely, the calculations indicate that neutron poisoning is nil in the high, 391-g Pu/liter, concentration solution. The small reactivity effects

TABLE V

Relative Poisoning and Density Effects of Raschig Rings on the Reactivity of $Pu(NO_3)_4$ Solutions

Borated Glass Raschig Rings Containing $\frac{1}{2}$ wt% Boron			
Solution Concentration g Pu/liter	KENO Calculated k_{eff}[a]		
	Critical Assembly[b]	Raschig Rings as Void Tubes	Raschig Rings Removed[c]
391	1.012 ± 0.006	1.010 ± 0.006	1.033 ± 0.006
197	1.001 ± 0.006	1.076 ± 0.006	1.119 ± 0.006
63	0.978 ± 0.004	1.330 ± 0.004	1.393 ± 0.005

[a]18-group averaged GAMTEC-II cross sections using the original GAMTEC library data (Ref. 8).

[b]Raschig rings treated as parallel vertical tubes displacing the same volume of solution as the raschig rings.

[c]Critical height of solution decreased by 19.27%, the percent solution displaced by the raschig rings.

(1 to 2% on k_{eff}) observed in this relatively undermoderated system appear to be entirely due to the density reduction caused by the rings displacing solution. What poisoning effects are present are masked by either the statistics in the calculations or the use of tubular voids in the calculations. The use of the vertical parallel voids in the calculations probably results in a small amount of neutron leakage that does not actually exist.

As the boron content of the rings is increased, neutron absorption becomes the predominant mechanism by which the reactivity is decreased over the entire solution concentration range. This is illustrated in Table I where it is shown that the geometric buckling for the critical 391-g Pu/liter solution increases from 0.002 cm^{-2} to about 0.011 cm^{-2} when the boron content of the rings is decreased from 4 to 0.5 wt%. Although some variation in the volume of solution displaced by the two different rings was unavoidable in the respective experimental assemblies (18.78 and 19.27 vol%), this reduction in reactivity is also indicated by the calculations. For the 391-g Pu/liter solution, a KENO-calculated k_{eff} value of 0.819 ± 0.005 was obtained with 4 wt% boron in the rings as compared to the 1.012 ± 0.007 value obtained with 0.5 wt% boron. This 19% reduction in k_{eff} is due to the added neutron absorption in boron.

CONCLUSIONS

With no raschig rings, criticality is possible in all three experimental vessels used (12-, 18-, and 24-in.-diam cylinders, 42 in. high). With rings randomly loaded in the vessels, only the 24-in.-diam cylinder could be made critical, and then only with the 0.5 wt% borated rings. The exponential measurements on the subcritical assemblies

[11]W. THOMAS and N. T. RÜCKERT, "Criticality Calculations of the Poison Effect of Boron Raschig Rings in Plutonium Nitrate Solutions," MRR-96, Technische Universitat München (1971).

loaded with the 4 wt% borated glass rings and the 1 wt% borated steel rings showed that negative bucklings exist for these systems at concentrations below 391-g Pu/liter and 412-g Pu/liter, respectively.

The processing advantages gained by using raschig rings for criticality prevention are indicated by the results from the critical assemblies containing 0.5 wt% borated rings. The minimum critical volume for unpoisoned plutonium nitrate solution normally occurs between about 175- to 200-g Pu/liter. With the system poisoned with 0.5 wt% borated glass raschig rings displacing 19.27 vol% solution, the concentration region in which the minimum critical volume was found to occur shifted upward to about 300 Pu/liter. Even with the 19.27% loss in volume caused by the rings, the vessel capacity increased at least from 175-g Pu/liter of vessel to 242-g Pu/liter of vessel for the minimum volume condition.

At low boron concentrations, density reduction, caused by the rings displacing solution, is essentially the only means by which the reactivity of highly concentrated solutions is decreased. As the plutonium concentration decreases and the neutron spectrum becomes more thermalized, neutron absorption becomes the primary mechanism decreasing the reactivity. At boron concentrations of about 4 wt% in the rings, both calculated values and experimental data show the density effects on the reactivity to be small in comparison to neutron absorption.

A comparison of geometrical models simulating the randomly oriented raschig rings and calculational methods indicates that the experimental data can be adequately reproduced. Using the KENO Monte Carlo code with GAMTEC-II cross-section data averaged over the neutron energy spectrum of the solution and simulating the rings with vertical parallel tubes displacing an equal volume of solution result in calculated k_{eff} values for the experimental critical assemblies that are within 2% of unity. This calculational approach results in slightly conservative values, with respect to criticality safety, at plutonium concentrations above 200-g Pu/liter. At lower concentrations the calculated critical condition is slightly nonconservative for establishing criticality safety limits. Conservative values of k_{eff} were obtained over the entire concentration range between 63- and 391-g Pu/liter with this same geometrical model and KENO by averaging the neutron cross sections of the ring material over the energy spectrum characteristic of the rings.

It appears that the ability to calculate the reactivity of a raschig ring poisoned system is more sensitive to the plutonium concentration than it is for unpoisoned systems. KENO calculations using Hansen-Roach cross sections reproduced the critical condition at 63-g Pu/liter but are 12% low on k_{eff} at 391-g Pu/liter. The opposite results were obtained with diffusion theory using GAMTEC-II cross sections and homogenizing the solution and raschig rings. About an 8% variation in k_{eff} was obtained using the Monte Carlo code GEM-IV. Consequently, the validity of a calculational method should be checked in the concentration range in which it is to be used.

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