Letters to the Editor

Calculations on Raschig-Ring-Poisoned Plutonium Solution Systems Using Hansen-Roach Cross Sections

In Ref. 1, Lloyd, Bierman, and Clayton reported results of Monte Carlo calculations on Raschig-ring-filled tanks containing plutonium nitrate solution. Using the KENO code[2] and Hansen-Roach[3] 16 group cross sections, they obtained a value of $k_{eff} = 0.886 \pm 0.006$ for an experimentally critical system of 391 g/liter solution and $\frac{1}{2}$ wt% boron rings. Lloyd, Bierman, and Clayton also report that the KENO code used with other cross-section sets gave better results (generally conservative), implying that Hansen-Roach cross sections are not suitable for such calculations.

Raschig-ring-poisoned plutonium solution systems are commonly used in process plants, and many facilities cannot afford to generate and evaluate new cross-section sets for each calculation. They must rely on "standard" sets such as the Hansen-Roach data and, therefore, it is important to know the range of applicability of such sets. As a preliminary to a series of calculations to provide data for determining plant criticality limits for Raschig-ring-filled tanks, I repeated the calculation for the 391 g/liter solution using the KENO-II code,[4] Hansen-Roach cross-section data, and an essentially identical geometry description, and found a k_{eff} of 1.0111 ± 0.0055. In a discussion with Bierman, I learned that the only difference in our calculations was that he had used infinite dilution plutonium cross sections for all solution concentrations, and I used finite dilution data.

The difference in calculational results for a very common plant problem seemed to justify a brief study of the choice of shielded cross sections. The effect of the selection of cross-section sets for the plutonium isotopes is shown for the 63 and 391 g/liter systems in Table I. The values for infinite dilution agree with those given in Ref. 1. The value of k_{eff} varies from 13% high to 12% low for the 391 g/liter solution, while the k_{eff} values for the 63 g/liter solution show no significant variation with choice of cross-section set. In the latter case, the more dilute system is more thermal, and fewer collisions occur in the resonance range of 0.1 to 550 eV (Hansen-Roach energy groups 9 through 15).

The results of k_{eff} calculations for all Hanford critical experiments using $\frac{1}{2}$ wt% boron rings are shown in Table II. The plutonium cross sections were chosen as follows: the macroscopic potential scattering cross section, Σ^P, was

TABLE I

Variation of k_{eff} with Cross-Section Set for a Raschig-Ring-Filled Tank Containing Plutonium Nitrate Solution

^{239}Pu Ratio[a]	$k_{eff} \pm \sigma$	
	63 g/liter Solution	391 g/liter Solution
20	0.9966 ± 0.0066	1.1269 ± 0.0077
100	---	1.0989 ± 0.0094
1×10^3	1.0099 ± 0.0072	1.0291 ± 0.0083
2×10^3	---	1.0122 ± 0.0078
4×10^3	---	0.9818 ± 0.0070
1×10^4	0.9995 ± 0.0057	0.9359 ± 0.0090
1×10^5	---	0.8992 ± 0.0071
∞	0.9903 ± 0.0060	0.8848 ± 0.0063

[a]The ratio of the potential scattering cross section of the solution to the ^{239}Pu number density. It was assumed that the corresponding ratio for ^{240}Pu was ten times that for ^{239}Pu (the actual value for 8.322% ^{240}Pu is 10.9 times the ^{239}Pu ratio).

TABLE II

Calculations of k_{eff} for Experimentally Critical Systems* of Plutonium Nitrate Solution in a Tank Filled with $\frac{1}{2}$ wt% Boron Raschig Rings

Solution Concentration (g/liter)	^{239}Pu Ratio[a]	^{240}Pu Ratio[a]	$k_{eff} \pm \sigma$[b]
63.0	1.1×10^4	1.2×10^5	1.0007 ± 0.0060
74.0	9.1×10^3	9.9×10^4	1.0129 ± 0.0063
110.0	5.9×10^3	6.4×10^4	0.9984 ± 0.0067
197.0	3.3×10^3	3.6×10^4	1.0126 ± 0.0072
271.0	2.3×10^3	2.5×10^4	1.0286 ± 0.0067
391.0	1.5×10^3	1.6×10^4	1.0111 ± 0.0055

*The critical data is taken from Ref. 1.

[a]The ratio of the potential scattering cross section of the solution to the ^{239}Pu or ^{240}Pu number density.

[b]All calculations were done using the KENO-II code with Hansen-Roach 16 group cross sections.

[1]R. C. LLOYD, S. R. BIERMAN, and E. D. CLAYTON, *Nucl. Sci. Eng.*, **50**, 127 (1973).

[2]G. E. WHITESIDES and N. F. CROSS, "KENO, A Multigroup Monte Carlo Criticality Program," CTC-5, Computing Technology Center, Nuclear Division, Oak Ridge, Tennessee (Sep. 1969).

[3]GORDON E. HANSEN and WILLIAM H. ROACH, "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos Scientific Laboratory (Dec. 1960).

[4]G. E. WHITESIDES, Personal Communication (Dec. 1971).

calculated as

$$\Sigma^P = 22.0\,x\,[H] + 10.0\,x\,[N] + 3.8\,x\,[O] ,$$

where the variable in brackets denotes the number density of nuclide x in at./b-cm. The cross-section set for each plutonium isotope is chosen according to the value of the ratio of Σ^P to the plutonium isotope number density. Interpolation was used when the calculated ratio fell between two tabulated values. The average value of k_{eff} for the data shown in Table II is 1.0107 ± 0.0108, with no apparent drift in k_{eff} as a function of solution concentration.

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Comments on "Calculations on Raschig-Ring-Poisoned Plutonium Solution Systems Using Hansen-Roach Cross Sections"

We were pleased to note the good results obtained by Dickinson[1] in calculating criticality for some of our plutonium nitrate Raschig-ring-filled vessels[2] utilizing Hansen-Roach cross sections. Use of the formula provided for macroscopic potential scattering cross sections, with interpolation when the calculated ratio falls between two tabulated values, provides very good results and attests to the quality of the experiments.

In the data analysis, our purposes were not so much directed toward evaluation of cross-section sets *per se*, but rather to examine a few methods of treating the Raschig rings. The method used to mock up the random array of Raschig rings for the purposes of carrying out the calculations is to be emphasized and is of principal concern. In our treatment, good results can be obtained with the Monte Carlo calculations by treating the Raschig rings as parallel vertical tubes displacing the same volume of solution as the rings with the neutron cross sections of the rings being averaged over the neutron energy spectrum characteristic of the plutonium solution. Going to greater complexity, such as splitting the parallel tubes into short sections of length equal to the Raschig rings, with gaps between each, did not improve the results. This latter information was not included in the original paper.

TABLE I

Plutonium Concentration (g Pu/liter)	k_{eff} Using KENO-II[a,b,c]	
	ENDF/B-III	GAMTEC-Library
391	1.022 ± 0.006	1.012 ± 0.006
197	1.032 ± 0.006	1.001 ± 0.006
63	0.990 ± 0.006	0.978 ± 0.005

[a]ENDF/B-III data were processed using the FLANGE-ETOG codes.

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

[c]Equal-volume vertical-tube model.

One of the principal purposes of the paper was to present experimental data for use in validation of cross sections and calculational methods. Because of the apparent sensitivity of poisoned systems to plutonium concentration, the validity of the calculational method and cross sections to be used should always be checked in the concentration range that is of interest. Since the original paper was presented, some additional calculational correlations, which may be of further interest, have been made using evaluated nuclear data file cross sections (ENDF/B-III version) with 18 energy groups (Table I). Comparison is made with values presented in the original paper.

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[1]D. DICKINSON, *Nucl. Sci. Eng.*, **54**, 367 (1974).

[2]R. C. LLOYD, S. R. BIERMAN, and E. D. CLAYTON, *Nucl. Sci. Eng.*, **50**, 127 (1973).