REFERENCE 105

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Critical Experiments with Low-Moderated Homogeneous Mixtures of Plutonium and Uranium Oxides Containing 8, 15, and 30 wt% Plutonium

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The results from a series of criticality experiments with three different mixtures of oxides of plutonium and uranium are presented. The fuel mixtures consisted of ^{235}U-depleted uranium homogenized with ~8, 15, and 30 wt% plutonium and blended, homogeneously, with polystyrene to achieve H:(Pu + U) atomic ratios of ~7, 3, and 3, respectively. Critical sizes are given for rectangular parallelepipeds of each of the fuels fully reflected with a methacrylate plastic (Plexiglas). Critical sizes are also given for unreflected parallelepipeds of the 30-wt% plutonium-enriched fuel mixture. For the 30-wt% plutonium-enriched mixture, sufficient fuel was available to permit determining that the critical thickness of a fully reflected slab of this material, infinite in two dimensions, was 12.93 + 0.14 cm.

Comparisons were made between the critical assemblies and calculational results using ENDF/B-III cross sections and the KENO and DTF-IV computer codes. Wherever comparisons could be made, the DTF-IV and KENO results were within 1% of each other; however, some of the comparisons between calculations and experiments differed by 2 to 3% in k_{eff}.

INTRODUCTION

The initial series of criticality experiments, performed to provide data for the liquid-metal fast breeder reactor (LMFBR) fuel cycle outside reactors, were reported earlier.[1] These first experiments were conducted with mixed oxide fuels of ^{235}U-depleted uranium mixed with ~8, 15, and 30 wt% plutonium. Since these were the first experimental data applicable to fuel handling and processing outside reactors, the measurements were made at moderation ratios either near optimum with respect to mass or near optimum with respect to volume. The results from three additional series of criticality experiments with mixtures of plutonia and urania are presented in this paper. These latest fuel mixtures also consisted of ^{235}U-depleted uranium mixed with ~8, 15, and 30 wt% plutonium; however, these mixtures, having respective H:(Pu + U) atomic ratios of ~7, 3, and 3, are more undermoderated than those previously reported.

[1]S. R. BIERMAN, E. D. CLAYTON, and L. E. HANSEN, *Nucl. Sci. Eng.*, **50**, 115 (1973).

EXPERIMENTS

The experiments were performed using a remotely operated split-table machine. This device is shown in Fig. 1 with the table halves separated and the Plexiglas reflector partially removed to display the fuel core. Approach-to-critical neutron multiplication measurements were made on rectangular parallelepipeds of the 30-wt% plutonium fuel both bare and fully reflected with a methacrylate plastic (Plexiglas). However, measurements on the 8- and 15-wt% plutonium fuel mixtures were restricted to fully reflected assemblies by the prohibitively large volumes of fuel required for criticality.

Each of the fuels consisted of a homogeneous mixture of PuO_2-UO_2-polystyrene in the form of 5.09-cm square compacts having thicknesses of ~5.09 and 1.3 cm. Each compact was clad with #471 tape manufactured by Minnesota Mining and Manufacturing Company. A complete description is given in Table I for each of the three fuels, the cladding material, and the Plexiglas reflector used in the experiments. In all three fuel mixtures, the plutonium contained ~11.5 wt% ^{240}Pu, and the uranium was depleted to ~0.2 wt% ^{235}U.

TABLE I

Description of Experimental Fuel and Reflector

	29.3-wt% Pu Fuel 2.8 H : (Pu + U) Atomic Ratio			15.0-wt% Pu Fuel 2.86 H : (Pu + U) Atomic Ratio			8 1-wt% Pu Fuel 7.3 H : (Pu + U) Atomic Ratio		
Fuel composition, 10^{24} atom/cm^3									
^{241}Am[a]	1.019×10^{-5}			2.765×10^{-5}			3.132×10^{-6}		
^{238}Pu	1.833×10^{-6}			5.737×10^{-7}			3.809×10^{-7}		
^{239}Pu	2.203×10^{-3}			1.118×10^{-3}			3.490×10^{-4}		
^{240}Pu	2.931×10^{-4}			1.478×10^{-4}			4.678×10^{-5}		
^{241}Pu[a]	4.934×10^{-5}			2.221×10^{-5}			7.745×10^{-6}		
^{242}Pu	5.636×10^{-6}			2.370×10^{-6}			1.116×10^{-6}		
^{235}U	9.401×10^{-6}			1.365×10^{-5}			9.116×10^{-6}		
^{238}U	6.172×10^{-3}			7.549×10^{-3}			4.606×10^{-3}		
O	1.869×10^{-2}			1.840×10^{-2}			1.178×10^{-2}		
C	2.666×10^{-2}			2.653×10^{-2}			3.567×10^{-2}		
H	2.417×10^{-2}			2.534×10^{-2}			3.680×10^{-2}		
Cladding composition, 10^{24} atom/cm^3									
H	4.489×10^{-2}			4.489×10^{-2}			4.489×10^{-2}		
C	3.111×10^{-2}			3.111×10^{-2}			3.111×10^{-2}		
Cl	7.240×10^{-4}			7.240×10^{-4}			7.240×10^{-3}		
Reflector composition, 10^{24} atom/cm^3									
H	5.666×10^{-2}			5.666×10^{-2}			5.666×10^{-2}		
C	3.510×10^{-2}			3.510×10^{-2}			3.510×10^{-2}		
O	1.428×10^{-2}			1.428×10^{-2}			1.428×10^{-2}		
PuO_2 particle size, μm									
95%	<20			<20			<20		
50%	<8			<5			<5		
5%	<2			<1			<1		
UO_2 particle size, μm									
95%	<40			<18			<18		
50%	<9			<5			<5		
5%	<2			<2			<2		
Polystyrene particle size, μm									
95%	<225			<225			<225		
50%	<150			<150			<150		
5%	<50			<50			<50		
Fuel Compact Sizes, cm	Unclad	Clad	Stacked[b]	Unclad	Clad	Stacked[b]	Unclad	Clad	Stacked[b]
Length	5.090 ± 0.005	5.114 ± 0.005	5.118 ± 0.005	5.090 ± 0.005	5.114 ± 0.005	5.129 ± 0.013	5.090 ± 0.005	5.114 ± 0.005	5.138 ± 0.089
Width[c]	5.083 ± 0.026	5.170 ± 0.026	5.174 ± 0.030	5.090 ± 0.005	5.114 ± 0.005	5.129 ± 0.013	5.090 ± 0.005	5.114 ± 0.005	5.138 ± 0.089
Heights	5.090 ± 0.005	5.114 ± 0.005	5.118 ± 0.005	5.082 ± 0.066	5.170 ± 0.067	5.173 ± 0.037	5.081 ± 0.003	5.168 ± 0.003	5.170 ± 0.060
	1.339 ± 0.026	1.363 ± 0.026	1.367 ± 0.030	1.265 ± 0.041	1.353 ± 0.044	1.395 ± 0.056	1.274 ± 0.008	1.361 ± 0.009	1.381 ± 0.064

[a]Concentration at time of experiments.
[b]Average dimension of space occupied by compacts.
[c]Full-size compacts were rotated 90 deg in the unreflected assemblies such that the width dimension shown here is the height.

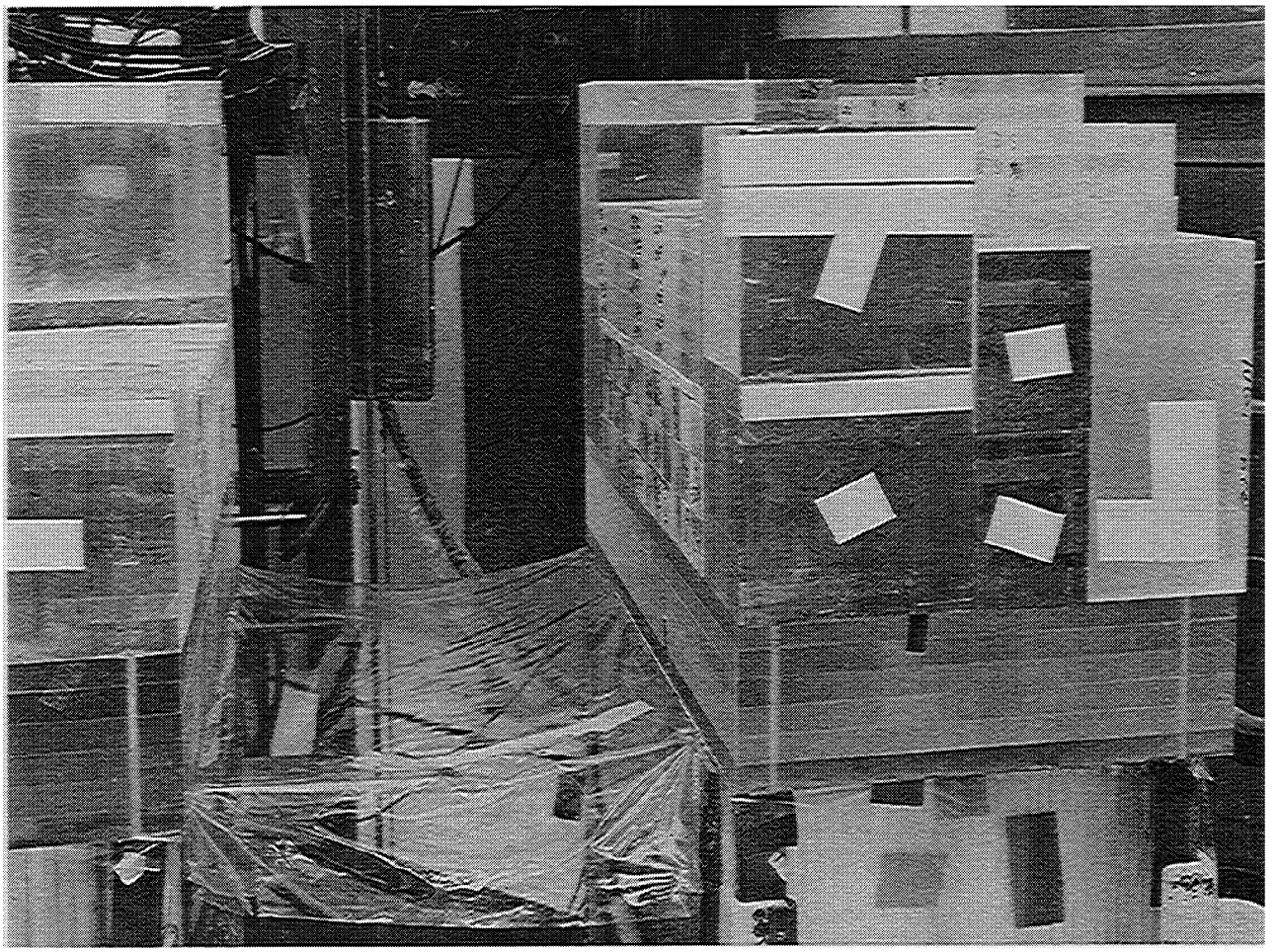

Fig. 1. The equipment and materials of the experiment.

EXPERIMENTAL DATA

The experimental critical assemblies, on which approach-to-critical measurements were made, are described in Tables II, III, and IV for 29.3-, 15.0-, and 8.1-wt% plutonium fuel mixtures, respectively. The approach-to-critical for each of these assemblies was made by incrementally loading fuel compacts to the top face of each assembly in a symmetrical manner with respect to the neutron flux. Since the neutron flux in these finite assemblies has a spatial dependency, the reactivity worth of a single fuel compact is dependent on its location in each assembly. However, since the top face of each assembly is rectangular and since the fuel compacts are of uniform composition, the neutron flux is symmetrical over this top face. By restricting the incremental loadings to ~1.3-cm-thick fuel compacts and fully loading either half of the top face, a spatially independent loading, smaller than one full layer of 5.09-cm-thick compacts, can be obtained. Consequently, the fractional layers given in Tables II, III, and IV should be treated as full layers of thinner fuel compacts having a thickness equal to the fractional layer times the thickness indicated for the compact.

An approach-to-critical curve for one of the experimental assemblies is shown in Fig. 2 and is typical of the measurement data obtained on each of the other finite assemblies described in Tables II, III, and IV. (As can be seen in Fig. 2, there is no discernable difference between the "full-layer" fuel loading of 1.3-cm-thick compacts and the "half-layer" fuel loading data.)

To provide a more simplified geometry for use in calculations, the reactivity worth of the ~1.3-cm-thick fuel compacts with respect to the full-sized fuel compacts was determined for each fuel mixture by replacement measurements. The critical height in terms of the full-sized compacts is presented in Tables II, III, and IV, for each of the experimental assemblies, as the "corrected height." Except for the mixture containing 29.3

TABLE II

Critical Assembly Configurations and Calculated k_{eff} for 29.3 wt% Pu, 2.8 H: (Pu + U) PuO_2-UO_2-Polystyrene Fuel Compacts

(Temperature at core center was 60 ± 5°C for the unreflected assemblies and 55 ± 5°C for the reflected assemblies. Room temperature was 20 ± 2°C.)

Critical Number of Fuel Compacts						
Reflector	Length[a] (5.090 cm)[b]	Width (5.083 cm)[b]	Height (5.090 cm)[b]	Height (1.339 cm)[b]	Corrected Height[c] (5.090 cm)[b]	KENO k_{eff}
None	10	10	8	3.768 ± 0.008	8.888 ± 0.019	1.025 ± 0.008
None	10	11	8	1.510 ± 0.002	8.252 ± 0.004	1.024 ± 0.005
Plexiglas	7	7	7	0.646 ± 0.001	7.060 ± 0.010	1.002 ± 0.005
Plexiglas	8	8	5	2.548 ± 0.004	5.615 ± 0.008	1.000 ± 0.004
Plexiglas	9	9	4	4.192 ± 0.011	4.997 ± 0.013	0.984 ± 0.007
Plexiglas	10	10	4	1.300 ± 0.002	4.316 ± 0.007	---
Plexiglas	10	10	4	1.297 ± 0.003	4.308 ± 0.009	1.000 ± 0.006
Plexiglas	12	10	4	0.038 ± 0.001	3.948 ± 0.006	0.987 ± 0.004
Plexiglas	12	12	3	2.791 ± 0.004	3.668 ± 0.005	0.990 ± 0.004
Plexiglas	12	13	3	2.407 ± 0.005	3.578 ± 0.006	0.986 ± 0.005
Plexiglas	14	13	3	2.068 ± 0.007	3.487 ± 0.009	1.000 ± 0.006
Plexiglas	∞	∞	-	---	2.540 ± 0.028[d]	0.993 ± 0.006

[a]Note that the assembly dimensions are expressed in number of blocks or compacts.

[b]Length, width, and thickness of unclad fuel blocks. Interchange width and height dimensions for the two unreflected assemblies.

[c]The 1.339-cm-thick fuel expressed as 5.090- × 5.090- × 5.083-cm fuel to account for its measured reactivity worth in terms of the full-size fuel. (Voids and cladding effects were observed to be negligible in the full-size fuel.)

[d]Obtained from linear extrapolation of data from the finite assemblies.

TABLE III

Critical Assembly Configurations and Calculated k_{eff} for 15.0 wt% Pu, 2.86 H: (Pu + U), PuO_2-UO_2-Polystyrene Fuel Compacts

(Temperature at core center was 50 ± 2°C; room temperature was 20 ± 2°C.)

Critical Number of Fuel Compacts						
Reflector	Length (5.090 cm)[a]	Width (5.090 cm)[a]	Height (5.082 cm)[a]	Height (1.265 cm)[a]	Corrected Height[b] (5.082 cm)[a]	KENO k_{eff}
Plexiglas	10	10	9	6.651 ± 0.016	10.749 ± 0.031	0.994 ± 0.002[c]

[a]Length, width, or thickness of unclad fuel.

[b]The 1.265-cm-thick fuel expressed as 5.082-cm-thick fuel to account for its measured reactivity worth in terms of the 5.082-cm fuel. (Other measured corrections for cladding and void effects are not available.)

[c]Determined from ten KENO calculations in which the random number sequence was started at a different place each time.

wt% plutonium, this corrected height does not include corrections for the cladding material or stacking voids.

The reactivity effect of the stacking voids and the cladding material was determined experimentally to be negligible for the 29.3-wt% plutonium fuel mixture. The negative reactivity effects, caused by the stacking voids and the reduced fuel density because of the cladding being present, were compensated for by the positive worth of the additional hydrogen and carbon introduced into each of these undermoderated assemblies by the cladding material. No attempt was made to measure the void and cladding effects in the 15.0- and 8.1-wt% plutonium fuel mixtures because of their size and the limited amount of fuel available.

TABLE IV

Critical Assembly Configurations and Calculated k_{eff} for 8.1 wt% Pu, 7.3 H:(Pu + U) PuO_2-UO_2-Polystyrene Fuel Compacts

(Temperature at core center was 47 ± 4°C; room temperature was 20 ± 2°C.)

Critical Number of Fuel Compacts						
Reflector	Length (5.090 cm)[a]	Width (5.090 cm)[a]	Height (5.081 cm)[a]	Height (1.274 cm)[a]	Corrected Height[b] (5.081 cm)[a]	KENO k_{eff}[c]
Plexiglas	10	9	9	1.135 ± 0.001	9.274 ± 0.003	1.025 ± 0.005
Plexiglas	10	11	7	3.239 ± 0.001	7.780 ± 0.002	1.027 ± 0.004
Plexiglas	12	12	6	2.526 ± 0.003	6.609 ± 0.007	1.021 ± 0.006
Plexiglas	14	13	5	3.733 ± 0.005	5.919 ± 0.008	1.021 ± 0.003

[a]Length, width, or thickness of unclad fuel.

[b]The 1.274-cm-thick fuel expressed as 5.081-cm-thick fuel to account for its measured reactivity worth in terms of the 5.081-cm fuel. (Other measured corrections for cladding and void effects are not available.)

[c]The 18-energy-group EGGNIT-averaged cross sections using FLANGE-ETOG processed ENDF/B-III data.

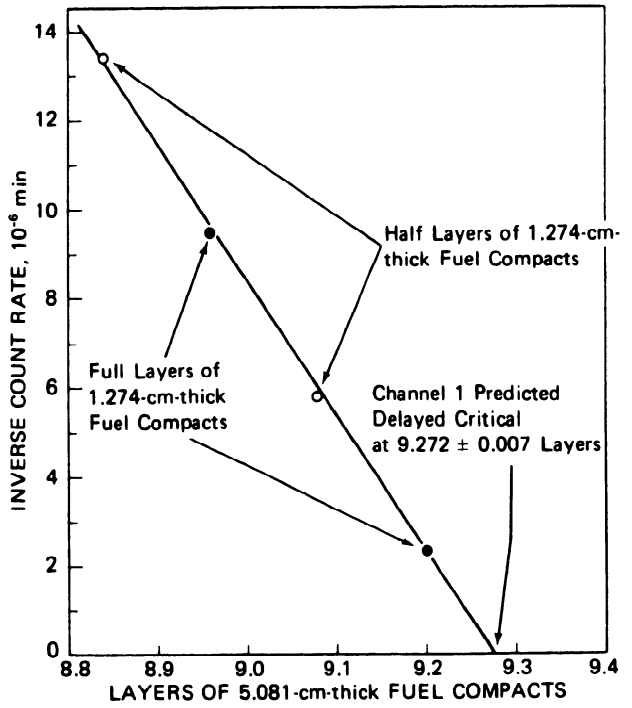

Fig. 2. Typical critical approach measurement with counting channel 1.

Also, the voids and cladding material can be adequately described in a Monte Carlo-type calculation.

DATA CORRELATION AND ANALYSIS

Neutron multiplication constants (k_{eff}) were calculated for each of the critical assemblies listed in Tables II, III, and IV and having the respective material compositions given in Table I. The ^{241}Pu-^{241}Am concentrations, listed in Table I, were determined at the time of the respective set of experiments and changed less than the reproducibility of the sample analyses during the course of each set of experiments.

Because of the geometries involved, the calculations were performed primarily with the Monte Carlo computer code KENO (Ref. 2). However, where possible, supportive P_1 neutron transport theory calculations were also performed using the DTF-IV computer code[3] with four orders of quadrature. Both the KENO and DTF-IV calculations were made using 18-energy-group EGGNIT-averaged[4] cross sections that had been processed from the ENDF/B-III with the FLANGE (Ref. 5) and ETOG (Ref. 6) codes. Because of the relatively small mean particle diameters (<10 μm) obtained in the fuels, particle size was not considered in

[2]G. E. WHITESIDES, "KENO–A Multigroup Monte Carlo Criticality Program," CTC-5, Computing Technology Center, Union Carbide Corporation (1969).

[3]K. D. LATHROP, "DTF-IV–A Fortran IV Program for Solving the Multigroup Transport Equation with Anisotropic Scattering," LA-3373, Los Alamos Scientific Laboratory (1965).

[4]C. R. RICHEY, "EGGNIT: A Multigroup Cross Section Code," BNWL-1203, Battelle Pacific Northwest Laboratory (1969).

[5]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).

[6]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).

the EGGNIT calculations. However, based on previous calculations[7] for systems containing plutonium only, these particle sizes are not expected to appreciably affect the calculated k_{eff}. In all of the KENO calculations, 150 generations of 300 neutrons each were used, and the average k_{eff} reported for each calculation was obtained from at least 40 of these generations in succession. In the DTF-IV calculations, mesh points uniformly spaced every $\frac{1}{2}$ cm or less were used. Transverse buckling for the three-dimensional systems was accounted for by making DB^2 calculational corrections, which are indigenous to the DTF-IV code and take into consideration P_1 scattering, to the absorption cross sections.

To obtain a correct source distribution in each of the KENO calculations, initial generations were successively deleted in calculating the average k_{eff} for the KENO run. However, although the average k_{eff} tends not to be affected monotonically as additional generations are deleted, the statistics associated with the calculation allow for a considerable amount of variation between k_{eff} averages. Even with a source distribution characteristic of the material and geometry, the k_{eff} average will fluctuate randomly as the number of generations, over which the average is made, is decreased. To consistently select an average k_{eff} value from each KENO run, a technique recently employed[8] in evaluating the statistical fitting of time-dependent neutron decay phenomena was used. This technique makes use of the standard deviation obtained in the calculation and the random fluctuation of the average k_{eff} within the standard deviation as successive generations are ignored in the averaging process. Paraphrasing Ref. 8, the generation to begin the averaging process over is taken to be the one for which the upper confidence limit opens upward and the lower confidence limit opens downward, providing that averaging over later generations results in k_{eff} averages above and below that obtained with the chosen generation.

For the nine fully reflected critical assemblies of 29.3-wt% plutonium fuel shown in Table II, the average k_{eff} calculated by KENO was 0.994, with a spread of 1.6% between the highest and lowest values. Even though the random number sequence was started at the same place for each of these calculations, the variations in geometry resulted in different sequencing for each case. Consequently, a spread of from ~1.5 to 2% in the calculated values should be expected based on the standard deviations observed on the individual calculations. For the two unreflected critical assemblies shown in Table II, an average k_{eff} of 1.024 was obtained from single KENO calculations on each assembly.

Neutron transport theory calculations with DTF-IV were made, for comparison with the KENO results, on the two unreflected assemblies of 29.3-wt% plutonium fuel, and on the fully reflected slab of this fuel infinite in two dimensions. A k_{eff} of 1.012 was calculated with DTF-IV for each of the two unreflected critical assemblies as compared with the KENO values of 1.024 ± 0.005 and 1.025 ± 0.008. For the critical infinite slab, a DTF-IV k_{eff} of 1.000 was obtained, as compared with the 0.993 ± 0.006 obtained from a single KENO calculation.

Since only one critical assembly of 15-wt% plutonium fuel was available against which to compare calculations, several KENO calculations were made with different starting places in the random number sequence. The results of these calculations are shown graphically in Fig. 3. If it is considered, for all practical purposes, that a KENO-computed average k_{eff} lies within four standard deviations (4σ) of the true calculated average k_{eff} (i.e., the objective k_{eff}), then the true average k_{eff} that would ultimately be obtained from many KENO calculations on this critical assembly would lie in the k_{eff} range between 0.984 and 1.003, as indicated in Fig. 3. Consequently, based on this limited number of calculations and the cross sections used, the best average calculated value of k_{eff} for the 15-wt% plutonium critical assembly is 0.994 ± 0.002.

The results from single KENO runs on each of the four critical assemblies of 8.1-wt% plutonium fuel are shown in Table IV. All four of these were 2 to 3% high in k_{eff}, with the average k_{eff} being

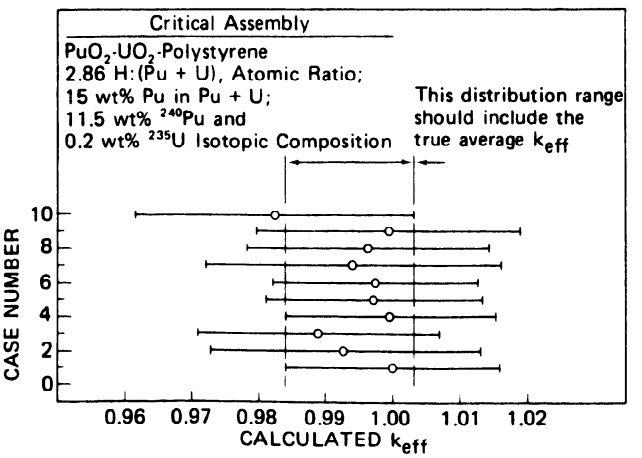

Fig. 3. Distribution of KENO-calculated average k_{eff} and 4σ limit ranges as a function of the random number sequence.

[7]L. E. HANSEN, E. D. CLAYTON, R. C. LLOYD, S. R. BIERMAN, and R. D. JOHNSON, *Nucl. Appl.*, **6**, 371 (1969).

[8]W. PFEIFFER, J. R. BROWN, and A. C. MARSHALL, *Nucl. Technol.*, **27**, 352 (1975).

1.024. Although additional KENO runs on these four assemblies may result in a lower overall average k_{eff} it would probably lie well above unity. Previous calculations on similarly fueled assemblies, having a relatively thermal neutron energy spectrum as does this 8.1-wt% plutonium fuel (91 H:fissile atomic ratio), have also resulted in k_{eff} values 2 to 3% above unity.[1]

SUMMATION

With two exceptions, the data covered in this paper, along with the data previously published[1,9] provide sufficient experimental data for validating calculations on undermoderated LMFBR-type homogeneous plutonium-uranium mixtures containing up to 30 wt% plutonium. Criticality data are still needed on dry or moist oxide mixtures and on mixtures having 25 to 40 wt% plutonium in the plutonium.

In hydrogenous materials, mixed (Pu + U) oxides containing up to ~30 wt% plutonium in the (Pu + U) exhibit minimum critical volumes at concentrations below that of the dry oxide (see curves in Ref. 1). With the data currently available, these minimums, and the curvature in the critical volume curves, are adequately defined for 8- and 15-wt% plutonium fuel mixtures. Plutonium-uranium mixtures containing 30 wt% plutonium exhibit an essentially constant critical volume at concentrations, in hydrogenous material, above ~500 kg/m^3. Two experimental points, one at an H/(Pu + U) corresponding to 550 kg oxide/m^3 and one corresponding to 5477 kg/m^3, are now available for establishing this constant volume region of the 30-wt% plutonium fuel mixture.

ACKNOWLEDGMENT

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[9]R. C. LLOYD, S. R. BIERMAN, and E. D. CLAYTON, *Nucl. Sci. Eng.*, **55**, 51 (1974).