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Criticality of Plutonium-Uranium Mixtures Containing 5 to 8 wt% Plutonium

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Results are presented from a series of criticality experiments with homogeneous plutonium-uranium fuel mixtures with plutonium content in the range of 5 to 8 wt%. The H:(Pu+U) atomic ratios covered a range from 19.5 to a high of 80.7: the experiments provide data in the region of moderation where the minimum critical volume occurs for homogeneous plutonium-uranium systems with a plutonium content of 8 wt%. The minimum critical spherical radius for material containing 8 wt% Pu in PuO_2-UO_2-water mixtures was determined to be 14.98 cm.

The criticality factors were computed with neutron transport theory and Monte Carlo calculations, using the DTF-IV and the KENO-II codes with both ENDF/B-II and ENDF/B-III version cross sections. The calculated values were found to be within ±2% of unity.

INTRODUCTION

The experimental data which have been reported[1-4] on the criticality of homogeneous mixtures of plutonium and uranium describe materials of relatively high plutonium content. The results presented in this paper, which have appeared in preliminary reports,[5,6] are believed to be the only measurements with mixtures containing as low as 8 wt% plutonium. If reactor fuels of this or similar composition are to be processed safely and as economically as possible, experimental data are required to accurately establish the criticality safety limits involved in their handling.

The experiments reported on in this paper cover the plutonium content range between 5 and 8 wt%, and provide benchmark-type data for use in checking calculational techniques and cross-section sets used in establishing criticality safety limits. Experiments were performed with plutonium-uranium nitrate solutions and with plutonium-uranium oxide mixtures to provide, respectively, data for both well-moderated and undermoderated systems.

EXPERIMENT DESCRIPTION AND DATA

The experiments with plutonium-uranium nitrate solutions were performed in a water-reflected, cylindrical vessel with high ^{240}Pu content (~23 wt% ^{240}Pu) material in which the plutonium content was ~5 to 7 wt%. Figure 1 shows a schematic diagram of the experimental setup, which consists of a water-reflected cylindrical stainless-steel vessel. The experimental data are presented in Table I. The critical height was measured as a function of concentration change.

The first series of experiments was started

[1] J. H. CHALMERS, "Criticality Parameters for Mixtures of Plutonium Oxide, Uranium Oxide and Water," *Criticality Control of Fissile Material*, p. 3, International Atomic Energy Agency, Vienna (1966).

[2] S. R. BIERMAN and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **15**, 307 (1972).

[3] L. E. HANSEN, S. R. BIERMAN, and E. D. CLAYTON, "Criticality of Mixed PuO_2-UO_2 Systems," *Reactor Physics Quarterly Report*, BNWL-1150, p. 516, Battelle-Northwest Laboratory (1969).

[4] R. C. LLOYD and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **17**, 269 (1973).

[5] S. R. BIERMAN and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **15**, 805 (1972).

[6] R. C. LLOYD, E. D. CLAYTON, and S. R. BIERMAN, *Trans. Am. Nucl. Soc.*, **15**, 803 (1972).

TABLE I

Criticality of Plutonium-Uranium Nitrate Solution in Cylindrical Geometry

(Water-reflected, stainless-steel cylinder, i.d. = 61.028 cm; o.d. = 61.186 cm)

Plutonium Concentration[a] (g/liter)	Uranium Concentration[b] (g/liter)	Acid Molarity	Critical Height (cm)	Critical Volume (liter)	k_{eff} KENO ENDF/B-III
30.63	390.2	0.45	50.27	147.03	1.025 ± 0.006
29.00	394.5	0.44	54.66	159.89	1.014 ± 0.005
27.32	399.0	0.44	61.04	178.56	1.021 ± 0.006
25.71	403.3	0.37	70.49	206.20	1.019 ± 0.005
24.28	407.1	0.36	84.86	248.23	1.003 ± 0.005
23.66	408.7	0.36	94.56	276.62	1.008 ± 0.005
30.44	390.7	0.43	51.41	150.38	1.021 ± 0.006
29.13	373.8	0.47	52.29	152.98	1.017 ± 0.006
27.28	351.1	0.54	56.87	166.36	1.009 ± 0.006
25.76	331.3	0.56	60.99	178.39	1.006 ± 0.006
24.35	312.5	0.56	67.59	197.71	1.010 ± 0.005
23.04	296.7	0.54	76.96	225.13	1.006 ± 0.005
21.85	281.9	0.56	89.61	262.13	1.012 ± 0.004

[a] ^{238}Pu = 0.07 wt%; ^{239}Pu = 73.00 wt%; ^{240}Pu = 22.80 wt%; ^{241}Pu = 3.22 wt%; ^{242}Pu = 0.91 wt%.
[b] ^{234}U = 0.01 wt%; ^{235}U = 0.66 wt%; ^{236}U = 0.01 wt%; ^{238}U = 99.32 wt%.

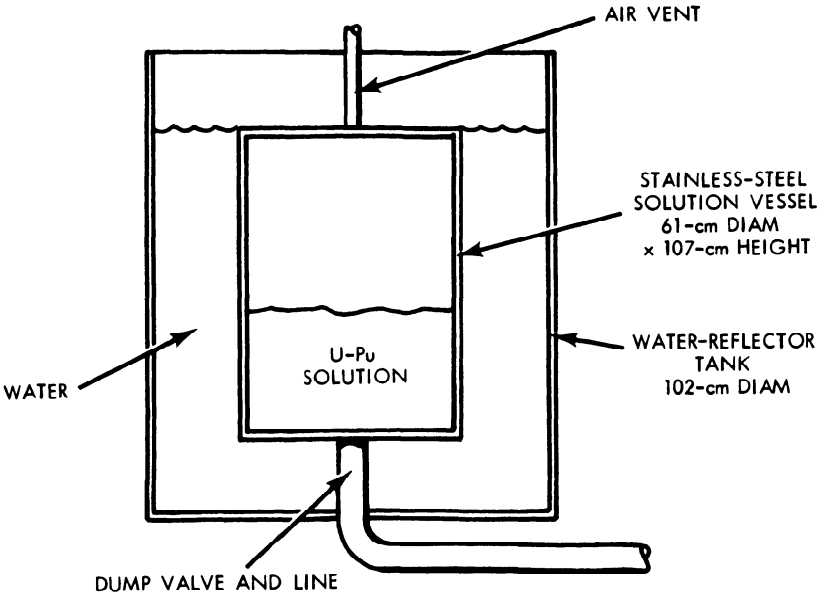

Fig. 1. Schematic diagram of experimental setup.

using solution with 30.63 g Pu/liter and 390.2 g U/liter, or ~7.3 wt% Pu. Uranium solution at 477 g U/liter was added in a series of dilution steps until criticality was achieved in a near-full cylinder with 23.66 g Pu/liter and 408.7 g U/liter, or ~5.5 wt% Pu.

The second series was again started at 30.44 g Pu/liter and 390.7 g U/liter, or ~7.2 wt% Pu, and diluted with low-molarity nitric acid until the vessel was critical in the near-full condition.

A comparison of the dilutions made with $UO_2(NO_3)_2$ and HNO_3 is shown in Fig. 2 where the critical height is plotted against plutonium concentration. The difference in the critical height is primarily due to the poison effect of the ^{238}U added in the dilution.

The experiments with the plutonium-uranium oxides were performed with homogeneous PuO_2-UO_2-polystyrene fuel compacts, 5.090 cm on a side and having thicknesses of 5.088 and 1.279 cm.

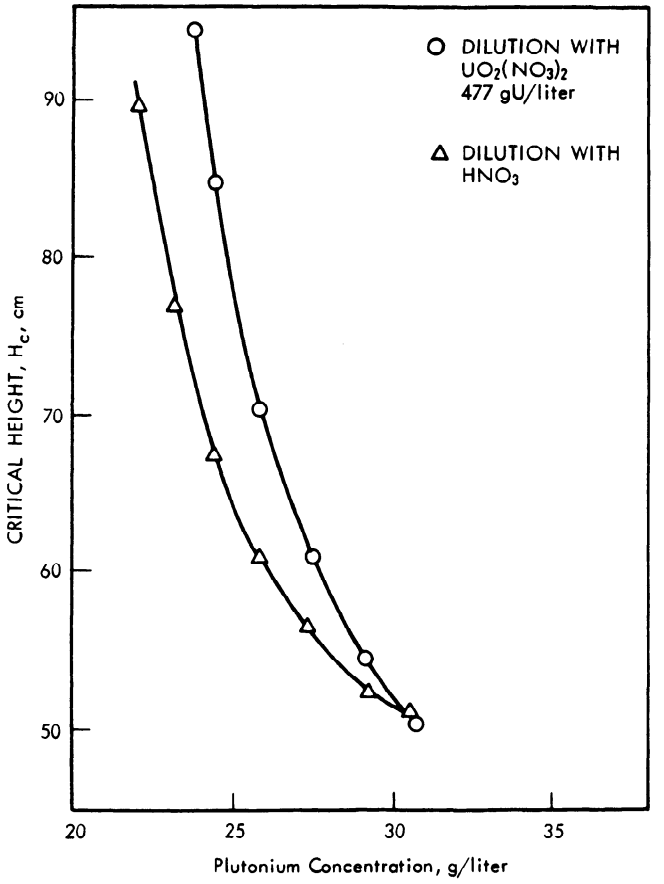

Fig. 2. Effect of dilution.

The PuO_2-UO_2 mixture contained 7.6 wt% PuO_2, and the oxide-polystyrene mixture had an atomic H:(Pu+U) of 19.5. A complete description of the compacts, including cladding, composition, and size, is given in Table II.

The critical assemblies consisted of rectangular parallelepipeds of fuel fully reflected with 15 cm of Plexiglas. A portion of one experimental assembly is shown in Fig. 3 with the remotely operated split-table, used in the experiments, fully open and with part of the reflector material removed. The critical heights for six such assemblies, having different cross-sectional dimensions, were determined. The results of these measurements are shown in Table III, along with the interpolated size for a Plexiglas-reflected cube of this fuel.

In Table III, the experimental assemblies are defined both in terms of clad fuel compacts and as corrected critical dimensions of fuel only. The correction for cladding material and stacking voids was determined experimentally, as shown in Fig. 4, and amounted to a 12.7% reduction in the mass of each critical assembly.

The approach to critical for each of the assemblies shown in Table III was made by incrementally loading fuel elements 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 depends on its location in each assembly. However, since the top face of each assembly is rectangular and the fuel compacts are of uniform composition, the neutron flux is symmetrical over this top face. By restricting the incremental loadings to fuel compacts ~1.27 cm in thickness and fully loading either half of the top face, a spatially independent loading, smaller than one

TABLE II

Description of Experimental Fuel and Reflector Materials for Experiments with Oxides

Cladding material per compact (g)	3.175
Cladding density (g/cm^3)	1.12
Composition of cladding (at./b-cm)	
H	4.489×10^{-2}
C	3.110×10^{-2}
Cl	0.724×10^{-2}
Composition of reflector	
H	5.712×10^{-2}
C	3.570×10^{-2}
O	1.428×10^{-2}
Composition of fuel compacts (at./b-cm)	
^{241}Am[a]	3.766×10^{-6}
^{238}Pu	9.535×10^{-7}
^{239}Pu	1.092×10^{-4}
^{240}Pu	3.688×10^{-5}
^{241}Pu	8.945×10^{-6}
^{242}Pu	4.689×10^{-6}
O	4.348×10^{-3}
H	4.155×10^{-2}
C	4.287×10^{-2}
^{235}U	2.897×10^{-6}
^{238}U	1.968×10^{-3}
PuO_2-UO_2 particle size (μm)	
90%	<20
50%	<5
10%	<0.8
Polystyrene particle size (μm)	
95%	<225
50%	<150
5%	<50
Uranium density (g/cm^3)	0.779 ± 0.006
Plutonium density (g/cm^3)	0.064 ± 0.001
Fuel compact density (g/cm^3)	1.884 ± 0.015

Dimensions of fuel compacts (cm):

	Length	Width	Thicknesses
unclad compacts	5.090 ± 0.005	5.090 ± 0.005	5.088 ± 0.018 1.279 ± 0.040
clad compacts	5.120 ± 0.005	5.120 ± 0.005	5.148 ± 0.018 1.339 ± 0.040
clad compacts + voids	5.153 ± 0.010	5.153 ± 0.010	5.172 ± 0.025 1.422 ± 0.043

[a]Isotopic analysis made on March 23, 1972; experiments performed March 1972.

TABLE III

Experimental Criticality Data for PuO_2-UO_2-Polystyrene Fuel Mixtures

[843 g (Pu + U)/liter at 7.6 wt% Pu; atomic ratios: H/(Pu + U) = 19.5, H/Pu = 259; ^{240}Pu content of Pu = 23 wt%; ^{235}U content of U = 0.151 wt%]

Plexiglas-Reflected Experimental Assemblies				Critical Dimensions (cm)		
Fuel Length[a] (5.09 cm)	Fuel Width[a] (5.09 cm)	Fuel Height[b] (5.088 cm)	Fuel Height[b] (1.279 cm)	Length	Width	Height[c]
12	13	6	3.434 ± 0.012	61.08 ± 0.06	66.17 ± 0.07	30.49 ± 0.15
12	12	7	0.632 ± 0.004	61.08 ± 0.06	61.08 ± 0.06	31.80 ± 0.16
13	13	6	2.317 ± 0.020	66.17 ± 0.07	66.17 ± 0.07	29.24 ± 0.16
12	10	8	0.068 ± 0.008	61.08 ± 0.06	50.90 ± 0.05	35.61 ± 0.18
12	11	7	1.920 ± 0.016	61.08 ± 0.06	55.99 ± 0.06	33.24 ± 0.16
10	10	9	0.703 ± 0.028	50.90 ± 0.05	50.90 ± 0.05	40.76 ± 0.20
--	--	-	---	47.27 ± 0.20	47.27 ± 0.20	47.27 ± 0.20[d]

[a]Number of 5.09-cm fuel compacts.

[b]Number of 5.088- and 1.279-cm-thick fuel compacts. Fractional layers should be treated as full layers having the indicated fractional thickness.

[c]Experimentally determined corrections have been made to account for the reactivity effects of the cladding material and the stacking voids.

[d]Cube dimensions obtained by interpolation between critical assemblies.

TABLE IV

Criticality Data for PuO_2-UO_2-Polystyrene Fuel Mixtures

[843 g (Pu + U)/liter at 7.6 wt% Pu; atomic ratios: H/(Pu + U) = 19.5, H/Pu = 259; ^{240}Pu content of Pu = 23 wt%; ^{235}U content of U = 0.151 wt%]

Reflector	Experimental Critical Dimensions[a] (cm)			KENO, k_{eff}	
	Length	Width	Height	ENDF/B-II	ENDF/B-III
Plexiglas	61.08 ± 0.06	66.17 ± 0.07	30.49 ± 0.15	0.971 ± 0.005	---
Plexiglas	61.08 ± 0.06	61.08 ± 0.06	31.80 ± 0.16	0.987 ± 0.005	0.987 ± 0.005
Plexiglas	66.17 ± 0.07	66.17 ± 0.07	29.24 ± 0.16	0.990 ± 0.007	---
Plexiglas	61.08 ± 0.06	50.90 ± 0.05	35.61 ± 0.18	0.978 ± 0.006	---
Plexiglas	61.08 ± 0.06	55.99 ± 0.06	33.24 ± 0.16	0.986 ± 0.005	---
Plexiglas	50.90 ± 0.05	50.90 ± 0.05	40.76 ± 0.20	0.983 ± 0.007	0.974 ± 0.007
Average k_{eff}	---	---	---	0.983	---
Plexiglas	∞	∞	20.0[b]	0.983 ± 0.007	0.981 ± 0.007

[a]Experimentally determined corrections have been made to account for the reactivity effects of the cladding material and the stacking voids.

[b]Thickness corresponding to average calculated "critical k_{eff}" of 0.983.

full layer, can be obtained. Consequently, the fractional layers given in Table III should be treated as full layers of thinner fuel compacts having a thickness equal to the fractional layer times the full-sized compact.

DATA ANALYSIS AND COMPARISON OF EXPERIMENTS WITH CALCULATIONS

Effective neutron multiplication factors were calculated for each of the experimental assemblies as indicated in Tables I and IV. The calculations were made with the Monte Carlo code KENO (Ref. 7), using 18-group GAMTEC-II (Ref. 8)

[7]G. E. WHITESIDES and N. F. CROSS, "KENO—A Multigroup Monte Carlo Criticality Program," CTC-5, Oak Ridge Computer Technology Center (1969).

[8]D. E. KUSNER, R. A. DANNELS, and S. KELLMAN, "ETOG-1: 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 Corp. (1969).

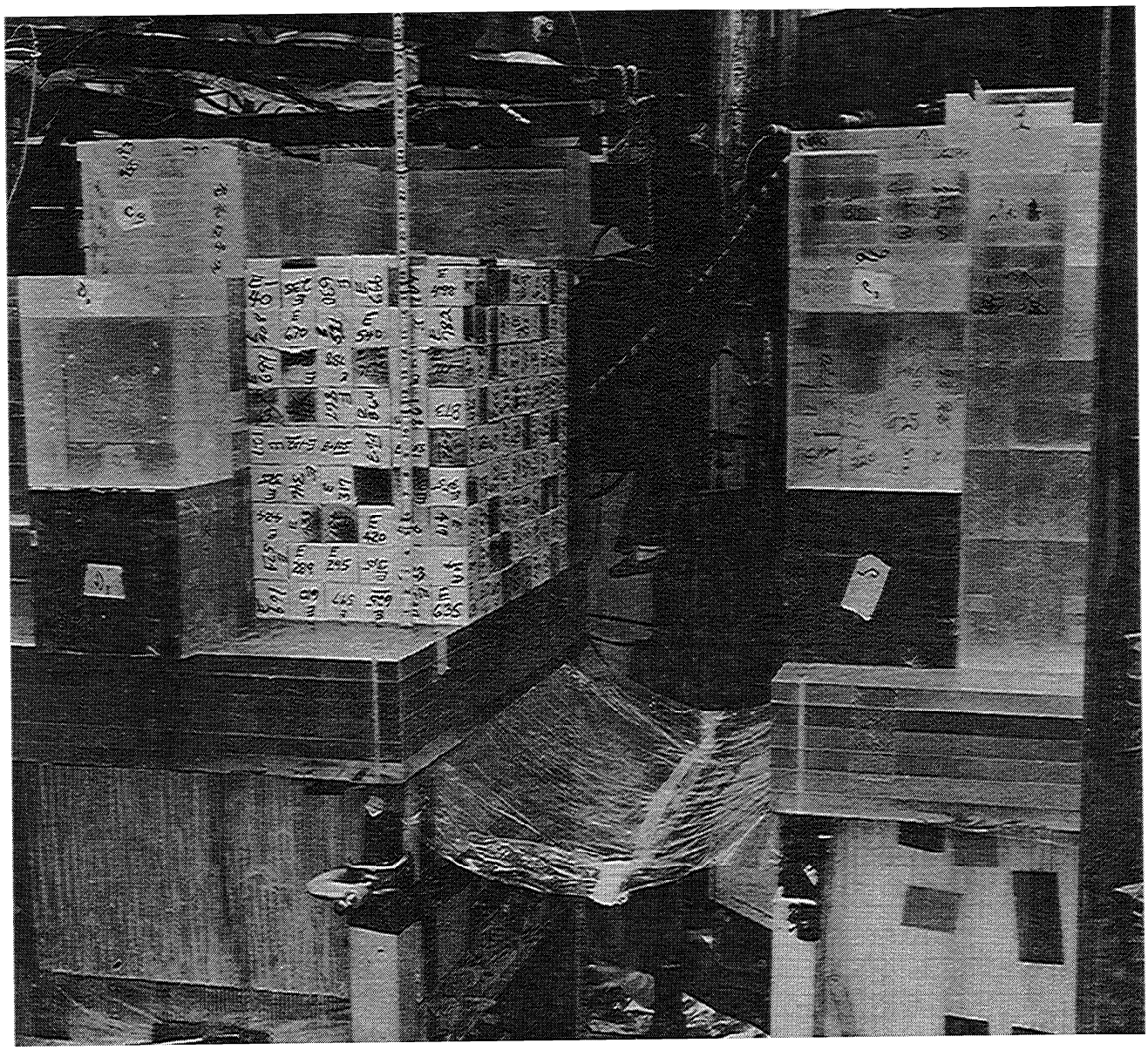

Fig. 3. Experimental assembly with part of the reflector removed.

averaged cross sections that had been processed from the ENDF/B-II and the ENDF/B-III with the FLANGE-II (Ref. 9) and ETOG-II (Ref. 10) codes.

Calculated k_{eff} values for the nitrate solution systems are shown in Table I and indicate that the calculations have a positive bias of ~1.3%. In contrast, the calculated k_{eff} values shown in Table IV for the PuO_2-UO_2-polystyrene compacts indicate that the calculations are biased low by ~1.7%. As shown in Table IV, the low bias for this more concentrated fuel is obtained with either the ENDF/B-II or the ENDF/B-III data.

The critical size for a cube of the PuO_2-UO_2-polystyrene fuel was determined to be 47.27 ± 0.2 cm on a side by interpolation, as shown in Fig. 5, between two critical assemblies. However, a linear extrapolation to obtain the critical thickness for a slab of this fuel infinite in two dimensions was prevented by a lack of fuel. A linear extrapo-

[9]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).

[10]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, Battelle-Northwest Laboratory (1965).

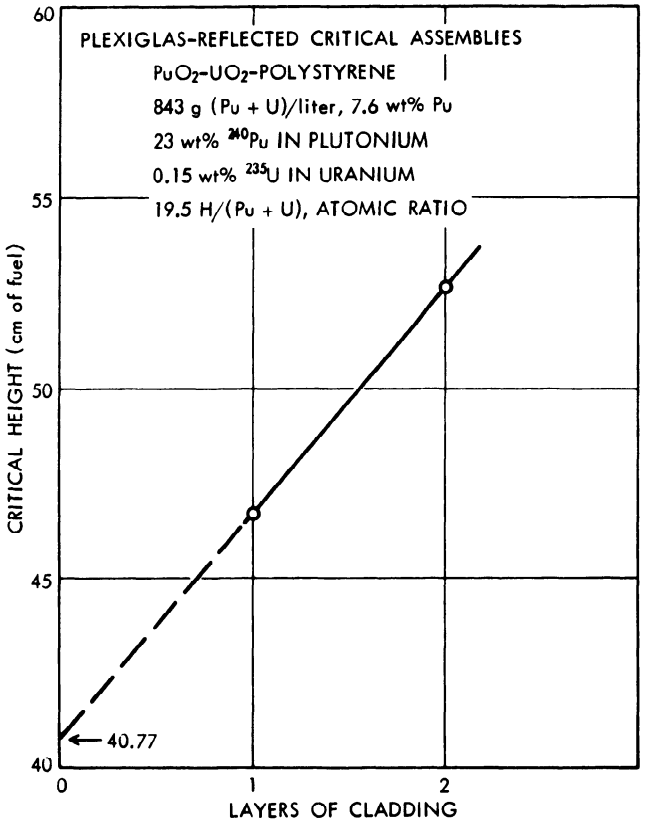

Fig. 4. Combined effect of voids and fuel cladding on critical height of reflected assemblies.

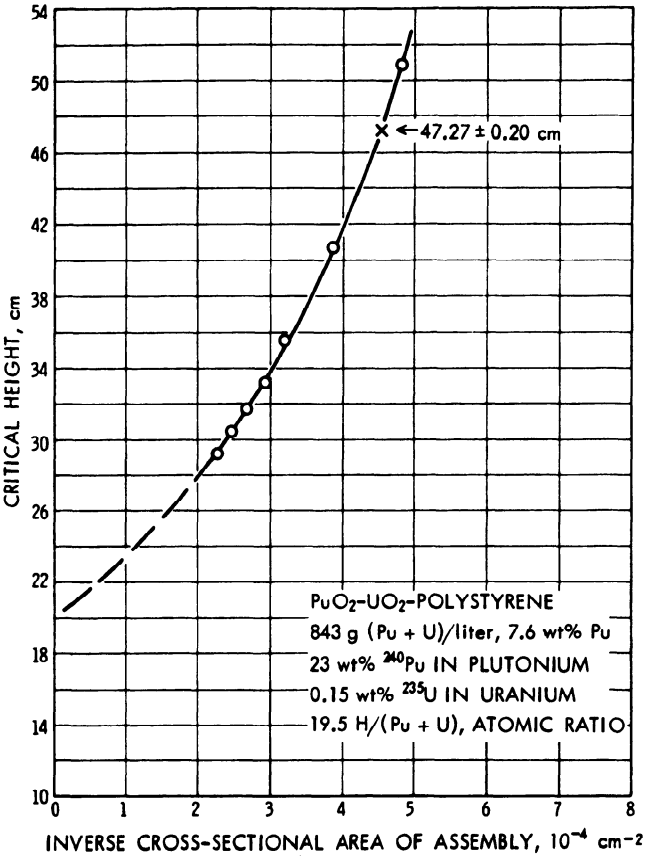

Fig. 5. Measured critical height as a function of the cross section of the assembly.

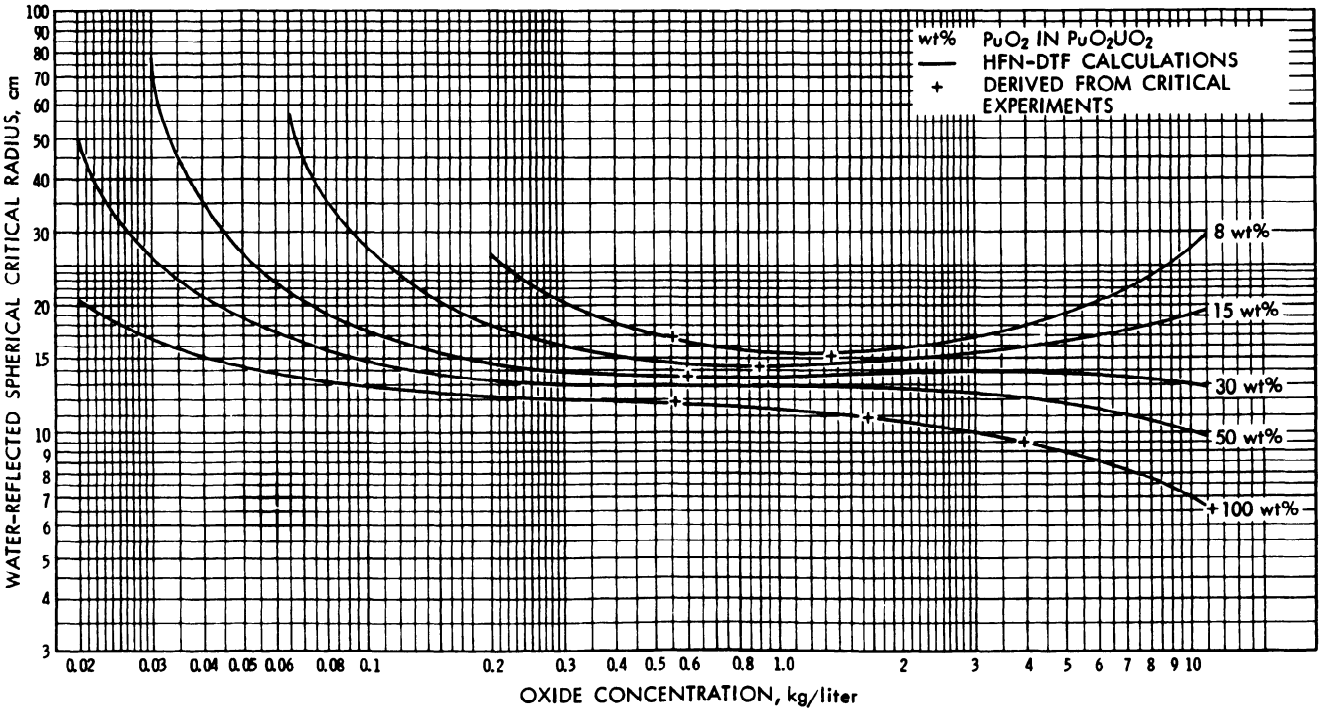

Fig. 6. Water-reflected spherical critical radii, $^{239}PuO_2$-U (0.71)O_2-H_2O.

lation of the three or four thinner critical assemblies results in a critical infinite slab thickness of ~15 cm, which calculation indicate to be ~10% low in k_{eff}. A more accurate value for the critical thickness of an infinite slab is 20 cm and was arrived at by searching for the slab thickness having a k_{eff} equal to the average k_{eff} of 0.983 within the accuracy obtained in the KENO computations on the finite systems. At 20 cm and using the same cross sections, transport theory calculations with DTF-IV (Ref. 11) yield a k_{eff} of 0.990, which is within one standard deviation of the KENO value.

To reduce the PuO_2-UO_2-polystyrene data to a theoretical oxide density system, DTF-IV computations were made at a "critical k_{eff}" of 0.990. The derived critical radius of a 14.98-cm fully reflected sphere of 8 wt% plutonium-enriched natural uranium oxide-water, at the theoretical concentration corresponding to an H:(Pu+U) ratio of 19.5, is compared in Fig. 6 with previously calculated values for theoretical density oxide-water systems. If the 23 wt% ^{240}Pu and associated 5.6 wt% ^{241}Pu that were present in the experimental fuel are included in the oxide-water system, the critical radius is increased by about a third, or to 21.67 cm.

[11]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).

CONCLUSIONS

These criticality experiments provide a data base for a wide range of plutonium-uranium concentrations within the range of 5 to 8 wt% Pu. These data can be used to establish more accurate criticality safety limits for fuels with the above plutonium concentrations. The critical spherical radius near optimum moderation for 8 wt% enriched PuO_2-UO_2-water systems was derived to be 14.98 cm. Using ENDF/B-III cross sections, it is shown that k_{eff} can be computed over a wide range of concentrations of uranium and plutonium to within ±2%.

ACKNOWLEDGMENT

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