BASIS FOR EXTENDING LIMITS IN ANSI STANDARD FOR MIXED OXIDES TO HETEROGENEOUS SYSTEMS

CRITICALITY SAFETY

TECHNICAL NOTE

EUGENE D. CLAYTON *Pacific Northwest Laboratory*
Energy Systems Department, P.O. Box 999, Richland, Washington 99352

HUGH K. CLARK *E. I. DuPont de Nemours and Company*
Savannah River Laboratory, Theoretical Physics Division
Aiken, South Carolina 29801

GORDON WALKER *U.K. Atomic Energy Authority*
Safety and Reliability Directorate
Chemical Plants, Laboratories and Criticality Group
Wigshaw Lane, Culcheth, Warrington, Lancashire WA3 4NE, England

RICHARD A. LIBBY *Pacific Northwest Laboratory*
Energy Systems Department, P.O. Box 999, Richland, Washington 99352

Received January 27, 1986
Accepted for Publication June 2, 1986

Subcommittee 8 of the Standards Committee of the American Nuclear Society is revising the Standard for Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors to include limits on heterogeneous systems. In connection with this effort, a number of criticality calculations were completed for mixed-oxide ($PuO_2 + UO_2$) fuel pins in water. The concentration of PuO_2 in the UO_2 (natural uranium) covered the range from 3.0 to 34 wt%. The isotopic makeup of the plutonium was also varied, up to 25 wt% ^{240}Pu and 15 wt% ^{241}Pu.

A search was made on fuel pin diameters and water-to-fuel volume ratios to obtain minimum critical dimensions and masses for a given fuel composition. Calculations made independently by several different members of the Work Group are compiled and compared, together with the proposed subcritical control limits for the Standard. Some difficulties were encountered with calculations pertaining to 30% PuO_2 at ^{240}Pu concentrations at water-to-fuel volume ratios and fuel pin diameters outside the area covered by any critical experiment. For this reason, dimensional limits on heterogeneous systems are not being proposed at this time for the Standard with 30% PuO_2 at a ^{240}Pu content of 25%.

In general, for a given fuel composition of mixed oxides, a heterogeneous arrangement of fuel pins of optimum diameter in water results in substantially smaller minimum critical dimensions than are obtainable for an aqueous homogeneous plutonium-uranium fuel mixture.

INTRODUCTION

Subcommittee 8 of the Standards Committee of the American Nuclear Society is revising the Standard for Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors[1] to include subcritical limits for heterogeneous systems. In connection with this effort, a number of calculations have been completed for heterogeneous systems of mixed-oxide (MOX) fuel pins in water.[2] Because the range of oxide and ^{240}Pu concentrations covered by the Standard and the pin diameters where minima occur lies somewhat outside the area of critical experiments that have been done, these calculations were later supplemented by calculations done by other members of the Work Group. All the calculations are compiled and compared here, together with proposed limiting values. In addition, some problems encountered when the concentration of PuO_2 in MOX is 30% (the upper end of the range in the Standard) are discussed.

DISCUSSION

The original calculations[2] were done by Libby who used the code EGGNIT-II (Ref. 3) to generate 17-group macroscopic cross sections in the epithermal neutron energies for a cell of fuel rod material and water. The cross sections for the single thermal group were calculated either with the code BRT-I (THERMOS) (Ref. 4) or using the TEMPEST portion of EGGNIT. These codes used cross-section data from the ENDF/B-III and -IV libraries, which were processed by FLANGE (Ref. 5)-ETOG (Ref. 6). The macroscopic cross sections were used in HFN (Ref. 7), a one-dimensional diffusion theory code. Each HFN calculation was run as a

critical search on the major dimension: the slab width, cylinder diameter, or spherical radius. The HFN calculations were made to determine the minimum critical dimension of water-reflected systems as a function of rod size and pitch. Correlations of this method of calculation with some 42 critical experiments with lattices of PuO_2-UO_2 rods in water showed it to have positive bias, with k_{eff} calculated for the experiments ranging from ~1.02 to 1.05 (Ref. 2). The bias, however, was not incorporated in the calculation of critical and subcritical dimensions.

Later, Clark, of the Savannah River Laboratory, utilized the GLASS code[8] for performing cell calculations for lattices of rods to obtain flux and volume-weighted cross sections for an equivalent homogeneous medium. An 84-group structure was used, and cell-averaged cross sections were collapsed either to a 16-group structure as close as possible to the Hansen-Roach structure,[9] with P_1 scattering and with upscatter removed, or to a 2-group structure, with cross sections adjusted for diffusion theory.[10,11] The 84-group cross sections used in GLASS (except for chromium, nickel, and zirconium, which were processed from older files) were derived from ENDF/B-IV files. The collapsed cross sections were used in the transport theory code ANISN (Ref. 12) (S_{16} quadrature) and in the two-group diffusion theory code TGAN (Ref. 11). Both codes are one-dimensional.

Correlations were made of these two methods with the same 42 experiments analyzed by Libby and with two other sets.[13,14] For the three-dimensional experiments, separability of the flux into spatial components was assumed, and results of critical transverse buckling searches were combined with the calculated critical buckling to yield the geometric buckling from which k_{eff} was calculated. As for Libby's method, bias was generally positive, with the critical value of k_{eff} ranging from ~1.00 to 1.03, and was not taken into account in the calculations for the Standard.

Because of the large number of calculations required to find minima in three dimensions and in mass as functions of rod diameter and pitch and because of the reasonably good agreement between TGAN and ANISN, TGAN rather than ANISN was selected for most of the calculations. In calculations for dry oxide at theoretical density (TD), Clark used ANISN (S_{16}) with Hansen-Roach cross sections. For plutonium metal spheres this method has little bias[15] and again no allowance for bias was made in calculations for dry MOX. Clark also did some calculations for homogeneous systems and used the same methods, MGBS-TGAN and ANISN (S_4) with Hansen-Roach cross sections, as he used previously[16] for the original Standard[1] except that the averaging of the transport cross section for the fast group was altered in MGBS, and a k_{eff} search similar to that in other codes, such as ANISN, was incorporated in TGAN. These changes altered the correlations with plutonium solution experiments, with the critical value of k_{eff} now falling from ~1.00 at low hydrogen/plutonium (H/Pu) to ~0.98 near H/Pu = 1000. This bias was incorporated in calculations for homogeneous mixtures.

Calculations were also done by Walker, of the U.K. Atomic Energy Authority, using the WIMS code option TWOTRAN for critical cylinder diameters and the MONK 6.3 Monte Carlo code for spheres and slabs, which uses "point" nuclear data derived from the U.K. Nuclear Data File and the GENEX files that form a supplement to the U.K. Nuclear Data Library. Correlations of the latter method with mixed uranium/plutonium critical experiments have shown it to overestimate k_{eff} by ~0.02. As in the calculations by Libby and Clark, no allowance was made for this bias.

Results of the various calculations are given in Table I, together with limits being suggested for the Standard. These limits were read from smooth curves initially drawn through Libby's values, calculated for a nominal k_{eff} of 0.95, i.e., with no allowance for the apparently positive bias in his method, but later adjusted downward where the discrepancy between Libby's and Clark's results was large. Clark's calculations were for 3, 9.5, and 30% PuO_2 in natural UO_2 plus PuO_2. The extremes are the lower and upper ends of the range covered by the Standard. His results were plotted and graphically interpolated and extrapolated to yield the values in Table I at the concentrations chosen by Libby. Clark's results were obtained from searches for minima as a function of rod diameter and triangular pitch. His rod diameters and pitches agreed well with those reported originally by Libby and Clayton.[2] Walker did not search for minima but calculated critical dimensions at the rod diameters and pitches obtained by Libby. Agreement among the calculations is quite good, in some cases excellent. Clark compared GLASS-ANISN and GLASS-TGAN results at 3% PuO_2 and 100% ^{239}Pu and found a slight tendency of the diffusion theory method to undercalculate the slab thickness; therefore his slab thicknesses in Table I may be slightly low.

At 30% plutonium in $PuO_2 + UO_2$ with 25% ^{240}Pu, 15% ^{241}Pu, Libby found that critical dimensions of dry TD MOX surrounded by a water reflector were lower than the minima he found for rod lattices as indicated in Table II. At 30% PuO_2 in $PuO_2 + UO_2$ and 25% ^{240}Pu, Clark found approximate equivalence except for the slab, but his lattice minima are not true minima. They are values indicated from searches at rod diameters of 0.25, 0.38, and 0.51 cm, but attempts to determine the minima more precisely by proceeding to larger diameters resulted in further decreases in critical sizes. These decreases were accompanied by a decrease in the water-to-fuel ratio at which the minima as a function of this variable occurred into a range far below the area covered by critical experiments, and doubts arose as to the ability of the calculational method to yield good results. At 30% PuO_2 and 15% ^{240}Pu, 6% ^{241}Pu, Clark found fairly flat minima, but did not investigate further to see whether once dimensions started to rise with increasing rod size they again began to decrease. For these reasons, limits are not being proposed in the Standard for 30% PuO_2, 25% ^{240}Pu.

At 30% PuO_2 minimum masses occur at very small rod diameters, and doubts arise as to the ability of the resonance absorption treatment for lattices to move smoothly into that for homogeneous media. In this connection, it is of some interest to compare limits[15,17] calculated for homogeneous PuO_2-H_2O mixtures with no uranium present, namely, 450, 740, and 990 g plutonium for the three isotopic compositions of the Standard, with the values in Table I.

The behavior at 30% PuO_2, which is undoubtedly associated with the increased fissionability of ^{240}Pu as the spectrum hardens, prompted Clark to investigate homogeneous mixtures. In the present Standard, mass limits for dry and damp oxides are given only for 100% ^{239}Pu; no credit is allowed for the presence of ^{240}Pu. The limits are given in terms of mass so it is not immediately apparent that the corresponding volume at 30% PuO_2, 8.87 ℓ, falls well below the volume limits for aqueous mixtures at 15 and 25% ^{240}Pu. To investigate this further, Clark calculated critical volumes for homogeneous mixtures of oxides and water with the results given in Table III. The maximum difference

TABLE I

Minimum Critical Masses and Dimensions of Lattices of MOX Rods in Water and Proposed Subcritical Limits

PuO_2 (wt%)[a]	^{240}Pu[b] (%)	Code[c]	Plutonium Mass (g)	Sphere Diameter (cm)	Cylinder Diameter (cm)	Slab Thickness (cm)
3.40	0	L	710 (540)[d]	32.5 (29.8)	21.9 (20.1)	10.1 (8.9)
		C	725 (635)	32.6 (31.0)	21.9 (20.8)	9.5 (8.9)
		W	---	34.6 (33.6)	23.6	11.0 (10.4)
		S	540	29.8	20.1	8.9
	15	L	1100 (820)	37.3 (34.0)	24.6 (23.1)	12.4 (10.8)
		C	1215 (1050)	37.9 (36.2)	26.1 (24.7)	12.3 (11.4)
		W	---	39.4 (37.2)	27.5	12.9 (12.4)
		S	820	34.0	23.1	10.8
	25	L	1450 (1070)	39.6 (35.9)	27.4 (24.7)	13.6 (11.8)
		C	1580 (1345)	40.6 (38.7)	28.2 (26.7)	13.7 (12.6)
		W	---	42.6 (41.4)	29.5	14.6 (13.9)
		S	1070	35.9	24.7	11.8
9.07	0	L	600 (470)	28.1 (26.1)	18.7 (17.1)	7.9 (6.9)
		C	595 (520)	27.5 (26.3)	17.9 (17.1)	7.1 (6.5)
		W	---	29.0 (27.8)	19.1	8.0 (7.4)
		S	470	25.8	16.9	6.7
	15	L	930 (700)	32.0 (29.4)	21.6 (19.7)	9.8 (8.5)
		C	980 (860)	31.9 (30.7)	21.5 (20.3)	9.3 (8.6)
		W	---	34.0 (32.0)	22.9	10.2 (9.8)
		S	680	29.2	19.7	8.4
	25	L	1180 (880)	33.6 (30.9)	22.8 (20.7)	10.5 (9.1)
		C	1280 (1110)	34.1 (32.6)	22.9 (21.8)	10.3 (9.5)
		W	---	36.0	24.4	11.4 (10.5)
		S	880	30.7	20.7	9.1
17.0	0	L	560 (440)	26.4 (24.6)	17.4 (16.0)	7.1 (6.2)
		C	535 (475)	25.4 (24.5)	16.2 (15.5)	6.1 (5.5)
		W	---	27.2 (26.2)	17.6	7.0
		S	440	24.1	15.3	5.7
	15	L	850 (660)	30.0 (27.7)	20.1 (18.3)	8.8 (7.7)
		C	895 (785)	29.8 (28.6)	19.6 (18.8)	8.2 (7.6)
		W	---	32.1 (31.0)	21.1	9.3 (8.8)
		S	660	27.5	18.2	7.4
	25	L	1090 (820)	31.4 (28.8)	21.2 (19.2)	9.5 (8.2)
		C	1180 (1020)	31.4 (30.2)	20.9 (19.8)	9.0 (8.2)
		W	---	33.6 (32.4)	22.5	9.9 (9.5)
		S	820	28.8	19.1	8.2
34.0	0	L	520 (410)	25.0 (23.3)	16.3 (15.0)	6.4 (5.5)
		C	485 (440)	23.6 (22.7)	15.0 (14.3)	5.2 (4.7)
		W	---	25.4 (24.4)	16.1	6.1 (5.9)
		S	410	22.3	13.8	4.8
	15	L	800 (620)	28.4 (26.3)	18.9 (17.3)	8.0 (7.0)
		C	830 (730)	27.8 (26.8)	18.2 (17.4)	7.2 (6.7)
		W	---	30.0 (29.0)	19.6	8.5 (8.0)
		S	620	26.1	16.8	6.3
	25	L	1010 (770)	<28.6	19.8 (18.0)	<7.9
		C	1095 (955)	29.0 (27.9)	19.2 (18.2)	7.8 (7.1)
		W	---	29.0 (27.6)	---	6.9 (6.4)
		S	770	---	---	---

[a]Weight percent PuO_2 in PuO_2 plus natural UO_2.
[b]The three isotopic compositions represented by 0, 15, and 25% ^{240}Pu are, by weight: 100% ^{239}Pu; 79% ^{239}Pu, 15% ^{240}Pu, 6% ^{241}Pu; and 60% ^{239}Pu, 25% ^{240}Pu, 15% ^{241}Pu.
[c]In this table, L denotes Libby, C denotes Clark, W denotes Walker, and S denotes subcritical limit proposed for Standard.
[d]The value in parentheses for L is for $k_{eff} = 0.95$, for C is for $k_{eff} = 0.98$, and for W corresponds to 3σ in Monte Carlo calculation.

TABLE II

Critical Dimensions of Water-Reflected Dry MOXs at Theoretical Density

PuO_2 (wt%)	^{240}Pu[b] (%)	Code	Sphere Diameter (cm)	Cylinder Diameter (cm)	Slab Thickness (cm)
30.00	0	Clark	26.9 (23.9)[c]	17.1 (15.2)	5.8 (5.3)
	15	Clark	28.8 (28.2)	18.6 (18.4)	6.9 (7.4)
	25	Clark	29.6 (29.4)	19.2 (19.4)	7.2 (8.0)
34.01	25	Libby	28.5 (29.7)	18.8 (19.8)	7.75 (8.8)
		Walker	29.0 (27.6)	---	6.9 (6.4)

[a]Weight percent PuO_2 in PuO_2 plus natural UO_2.
[b]The three isotopic compositions represented by 0, 15, and 25% ^{240}Pu are, by weight: 100% ^{239}Pu; 79% ^{239}Pu, 15% ^{240}Pu, 6% ^{241}Pu; and 60% ^{239}Pu, 25% ^{240}Pu, 15% ^{241}Pu.
[c]The value in parentheses for Clark and Libby is the minimum critical value for heterogeneous oxide-water mixture but for Walker corresponds to 3σ in Monte Carlo calculation.

between the two methods is ~0.04 in k_{eff}. Substitution of ^{239}Pu for ^{241}Pu increased the critical volumes substantially with both methods and indicated that the differences in Table III are largely due to the treatment given ^{240}Pu. It is apparent that some minimum limit needs to be placed on oxide concentration for the dimensional limits of the Standard to be valid. As a practical matter, however, such a limit would have little impact since TD oxide will not be attained as dryness is approached in a homogeneous aqueous mixture.

ACKNOWLEDGMENTS

This technical note is based on work performed under the U.S. Department of Energy contracts DE-AC06-76RLO 1830 with Pacific Northwest Laboratory, and DE-AC09-76R00001 with E. I. du Pont de Nemours and Company.

REFERENCES

1. "American National Standard for Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors," ANSI/ANS-8.12, American Nuclear Society, La Grange Park, Illinois (1978).

2. R. A. LIBBY and E. D. CLAYTON, "Nuclear Criticality of Heterogeneous Mixed-Oxide Systems," *Trans. Am. Nucl. Soc.*, **41**, 354 (1982).

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

4. C. L. BENNETT and W. L. PURCELL, "BRT-1: Battelle-Revised Thermos," BNWL-1434, Pacific Northwest Laboratory (1970).

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. K. 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,"

TABLE III

Critical Volumes (in litres) of Homogeneous Mixtures of Oxides and Water with 30% PuO_2

	^{240}Pu (%)					
	0		15		25	
g(U + Pu)/ℓ	MGBS-TGAN	H-A[a]	MGBS-TGAN	H-A	MGBS-TGAN	H-A
9800[b]	---	9.9	---	12.3	---	13.2
	---	9.7[c]	---	---	---	---
8000	9.8	10.4	13.8	13.6	12.8	14.2
6000	9.1	10.7	18.2	17.1	17.6	17.6
4000	10.0	10.9	30.2	24.1	33.2	26.6
3000	10.7	11.0	37.4	28.2	46.5	34.0
2000	11.0	10.9	38.9	29.9	53.1	39.6
1000	10.6	10.1	28.5	24.0	38.8	32.9
500	10.1[d]	---	20.5	---	27.0	---
	9.5[c]	9.9[c]	---	---	---	---
300	10.6	---	19.0[d]	---	24.6[d]	---
	---	---	18.7[c]	19.6[c]	24.0[c]	25.2[c]
200	12.0	---	20.4	---	26.4	---
Hetero[e]	7.1		11.7		13.4	

[a]ANISN (S_4) with Hansen-Roach cross sections.
[b]Dry oxide at TD.
[c]From Ref. 16.
[d]Minimum.
[e]GLASS-TGAN for lattices of MOX rods.

WCAP-3845-1 (ENDF-114), Westinghouse Electric Corporation (1969).

7. J. R. LILLEY, "Computer Code HFN-Multigroup, Multiregion Neutron Diffusion Theory in One Space Dimension," HW-71545, General Electric Corporation (1971).

8. H. C. HONECK, "The JOSHUA System" DP-1380, Savannah River Laboratory (1975).

9. G. E. HANSEN and W. M. ROACH, "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos National Laboratory (1961).

10. H. K. CLARK, "Correlation of Nuclear Criticality Safety Computer Codes with Plutonium Benchmark Experiments and Derivation of Subcritical Limits," DP-1565, Savannah River Laboratory (1981).

11. H. K. CLARK, "Computer Codes for Nuclear Criticality Safety Calculations," DP-1121, Savannah River Laboratory (1967).

12. W. W. ENGLE, Jr., "A User's Manual for ANISN," A One-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering," K-1693, Oak Ridge Gaseous Diffusion Plant (1967).

13. H. TSURUTA et al., "Critical Sizes of Light-Water Moderated UO_2 and PuO_2 Lattices," JAERI 1259, Japan Atomic Energy Research Institute (1978).

14. S. R. BIERMAN et al., "Critical Experiments with Fast Test Reactor Fuel Pins in Water," *Nucl. Technol.*, **44**, 141 (1979).

15. H. K. CLARK, "Subcritical Limits for Plutonium Systems," *Nucl. Sci. Eng.*, **79**, 65 (1981).

16. E. D. CLAYTON et al., "Basis for Subcritical Limits in Proposed Criticality Safety Standard for Mixed Oxides," *Nucl. Technol.*, **35**, 97 (1977).

17. "American National Standard for Nuclear Criticality Safety in Operation with Fissionable Materials Outside Reactors," ANSI/ANS-8.1, American Nuclear Society, La Grange Park, Illinois (1983).