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BASIS FOR SUBCRITICAL LIMITS IN PROPOSED CRITICALITY SAFETY STANDARD FOR MIXED OXIDES

CHEMICAL PROCESSING

KEYWORDS: *criticality, plutonium dioxide, uranium dioxide, safety, fuel cycle, homogeneous mixtures, water, data, subcriticality*

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Subcommittee 8 of the Standards Committee of the American Nuclear Society has proposed a standard providing subcritical limits for operations with mixed oxides of plutonium and uranium. The subcritical limit is the limiting value assigned to a controlled parameter that results in a system known to be subcritical, provided the limiting value of no other controlled parameter of the system is violated. The proposed standard includes subcritical limits for mixed oxides containing up to 30 wt% plutonium in Pu + U. A review was made of the available experimental data and validations undertaken that serve as the basis of the limits, and the assertion that they are, indeed, subcritical as given.

INTRODUCTION

The American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors (N16.1-1975) provides single-parameter limits for operations with ^{235}U, ^{233}U, and ^{239}Pu (Ref. 1). Subcommittee 8 of the Standards Committee of the American Nuclear Society is proposing a standard for operations with mixed oxides, extending N16.1 chiefly through the inclusion of additional subcritical limits. These limits may prove valuable for operations with mixed oxides of plutonium and uranium encountered in light water reactor, liquid-metal fast breeder reactor, and gas-cooled fast reactor fuel cycle operations. As defined in N16.1, a subcritical limit is the limiting value assigned to a controlled parameter that results in a system known to be subcritical provided the limiting value of no other controlled parameter of the system is violated. This limit contains margins designed to be sufficiently large to allow for uncertainties in calculations and experimental data used in its derivation, but, simultaneously, sufficiently small to discourage attempts to justify a larger limit. The margins contain no allowances for operating contingencies, e.g., double batching or a failure of analytical techniques to yield accurate values. Therefore, process specifications must incorporate margins to protect against the consequences of uncertainties in process variables and against a limit being accidentally exceeded.

The selection of limits with sufficiently, but not excessively, large margins is necessarily somewhat arbitrary, and requires the exercise of judgment, particularly, as in the present case, for mixed oxides, where pertinent experimental data are sparse. N16.1 offers the following guidance: "In the absence of directly applicable experimental measurements, the limits may be derived from calculations made by a method shown to be valid by comparison with experimental data, provided sufficient allowances are made for uncertainties in the data and in the calculations." The

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American National Standard, Validation of Calculational Methods for Nuclear Criticality Safety (N16.9-1975), offers further guidance. The material that follows gives the limits selected by the Work Group and the basis for asserting that they are indeed subcritical. Whenever numerical limits are given in a Standard, it is important that their basis be published in the open literature, where it may be examined by the user of the Standard.

SCOPE OF PROPOSED STANDARD

Although the limits may eventually be extended to lattices and other heterogeneous systems, they are presently restricted to homogeneous systems of plutonium and uranium dioxide. The mixtures may be solutions, suspended solids, precipitates, or a mechanical blend of powders, and are fully reflected by water. The following compositions were selected as having the greatest utility. The oxide mixtures contain 30, 15, 8, and 3% PuO_2 by weight. The uranium is natural. Isotopic compositions of plutonium are 100% ^{239}Pu; 79% ^{239}Pu, 15% ^{240}Pu, 6% ^{241}Pu; or 60% ^{239}Pu, 25% ^{240}Pu, and 15% ^{241}Pu. (Plutonium-238 and -242 may be considered present, but were conservatively ignored in calculating limits.)

Subcritical limits were derived for both dry and water-moderated systems. Since a completely dry oxide system may be difficult to maintain, subcritical limits were derived also for damp [$H/(Pu + U) \leq 0.45$] oxide.[2] Solutions and slurries were assumed to be uniform homogeneous mixtures of UO_2 [10.96 g/cm^3 (10.96×10^3 kg/m^3)] and PuO_2 (11.46 g/cm^3 for $^{239}PuO_2$) in water. Critical dimensions of such systems have minima as a function of concentration, provided the oxide mixture does not contain much more than 30% PuO_2 by weight.

SUBCRITICAL LIMITS

Uniform Aqueous Mixtures

Limits for uniform aqueous mixtures[3-5] of plutonium and uranium (^{235}U ≤ 0.71 wt%) fully reflected by water are given in Table I. A margin of ~2% (see next section) in k_{eff} was considered sufficient to account for uncertainties in calculations and experimental data used in deriving the limits on mass, volume, cylinder diameter, and slab thickness. Estimated critical values are shown in Figs. 1 through 4 along with the subcritical limits for a plutonium isotopic composition of 100% ^{239}Pu to indicate the corresponding margin in terms of mass or dimension. The margins for the subcritical limits with higher isotopes would be similar.

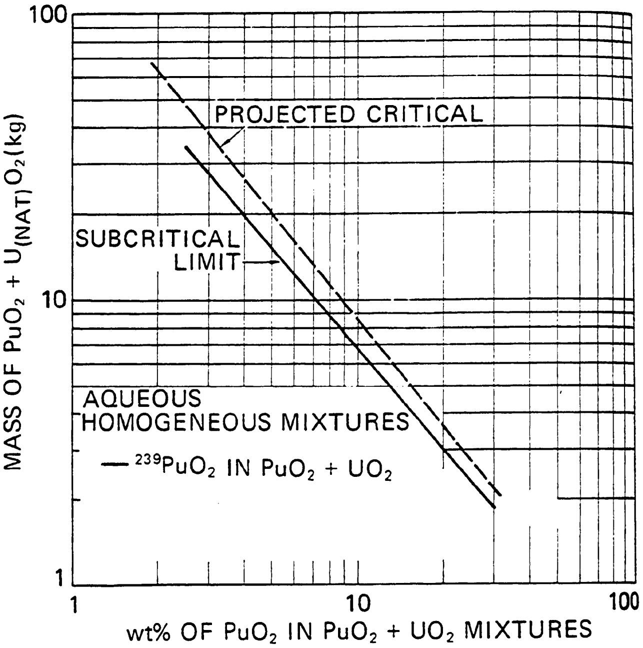

Fig. 1. Mass limit versus plutonium content.

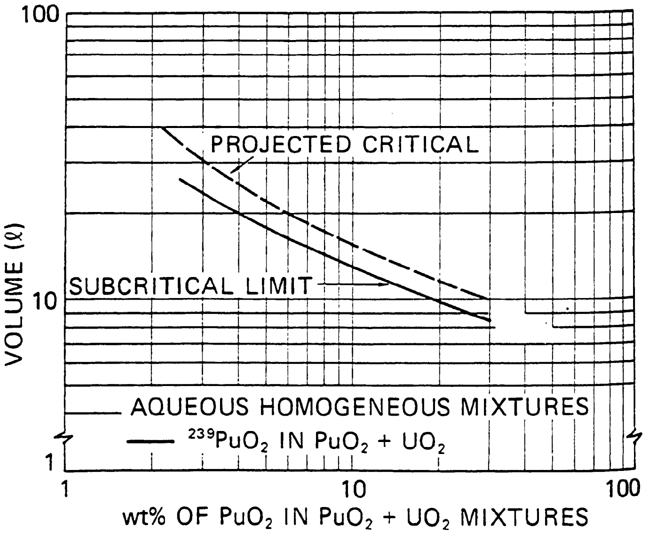

Fig. 2. Volume limit versus plutonium content.

For sufficiently dilute mixtures, criticality is impossible regardless of mass. A margin in k approaching 5% was used in deriving the infinite sea concentration limits in Table I, because uncertainties were considered greater than for mass or dimension limits. The corresponding margin in terms of concentration approaches 10%.

The product of critical slab thickness and concentration has a minimum, leading to a limiting areal density that is useful where precipitation or evaporation is a credible possibility. As for the infinite sea concentration, uncertainties

TABLE I

Subcritical Limits for Uniform Aqueous Mixtures of Plutonium and Uranium ($^{235}U \leq 0.71$ wt%)

	3 wt% PuO_2 in $PuO_2 + UO_2$			8 wt% PuO_2 in $PuO_2 + UO_2$			15 wt% PuO_2 in $PuO_2 + UO_2$			30 wt% PuO_2 in $PuO_2 + UO_2$		
	(A)[a]	(B)[a]	(C)[a]	(A)	(B)	(C)	(A)	(B)	(C)	(A)	(B)	(C)
Mass of plutonium contained in oxide (kg)	0.73	1.35	2.00	0.61	1.06	1.53	0.54	0.94	1.28	0.50	0.87	1.16
(Total oxide mass, $PuO_2 + UO_2$) (kg)	(27.5)	(51.3)	(75.9)	(8.6)	(15.1)	(21.7)	(4.1)	(7.1)	(9.7)	(1.9)	(3.3)	(4.4)
Cylinder diameter (cm) ($\times 10^{-2}$ = m)	24.3	30.8	34.8	19.8	24.9	27.5	17.8	22.5	24.8	16.2	21.0	23.4
Slab thickness (cm) ($\times 10^{-2}$ = m)	11.0	14.9	17.4	8.2	11.2	12.9	6.9	9.6	11.0	5.9	8.7	9.9
Volume (ℓ)	23.5	44.8	63.4	14.0	25.9	34.4	11.0	20.4	26.6	8.5	16.8	21.6
Infinite sea limiting[b] subcritical concentration of plutonium contained in oxide (g/ℓ) ($\times 10^{-3}$ = kg/ℓ)	6.8	8.06	9.27	6.9	8.19	9.43	7.0	8.16	9.39	7.0	8.12	9.32
(H/Pu atom ratio)[c]	(3780)	(3203)	(2780)	(3780)	(3210)	(2790)	(3780)	(3237)	(2818)	(3780)	(3253)	(2848)
(Total oxide limiting concentration, $PuO_2 + UO_2$) (g/ℓ) ($\times 10^{-3}$ = kg/ℓ)	(257)	(305)	(351)	(97.8)	(116)	(134)	(52.9)	(61.7)	(71.0)	(26.5)	(30.7)	(35.2)
Areal density of plutonium contained in oxide (g/cm²) (×10 = kg/m²)	0.27	0.38	0.47	0.25	0.34	0.42	0.25	0.33	0.41	0.24	0.32	0.37
(Total areal density of mixed oxides, $PuO_2 + UO_2$) (g/cm²) (×10 = kg m²)	(10.2)	(14.4)	(17.7)	(3.5)	(4.8)	(5.9)	(1.9)	(2.5)	(3.1)	(0.9)	(1.2)	(1.4)

[a] Conditions on plutonium isotopic ratios: (A) = $^{240}Pu > ^{241}Pu$; (B) = $^{240}Pu \geq 15$ wt%, $^{241}Pu \leq 6$ wt%; and (C) = $^{240}Pu \geq 25$ wt%, $^{241}Pu \leq 15$ wt%.

[b] For plutonium content in mixed oxides in the range below 3 wt% to 0.13 wt%, the subcritical limit of Table III is controlling. The plutonium concentration limit corresponding to 0.13 wt% is ≤4.9 g Pu/ℓ. For PuO_2 content ≤0.13 wt%, an aqueous, homogeneous mixture of mixed oxides will remain subcritical irrespective of any H/Pu ratio or concentration of $PuO_2 + UO_2$ in the mixture. (A reduction in the subcritical concentration below the value of 6.8 g/ℓ at 3 wt% PuO_2 is required to account for the presence of ^{235}U in the uranium, which becomes relatively more important at lower plutonium contents. The H/Pu ratio of 3780 will ensure subcriticality, if utilized for control, but at 0.13 wt% PuO_2, the corresponding plutonium concentration would be down to 4.2 g Pu/ℓ, which is a value somewhat less than required for the subcritical limit if the limit is expressed in terms of concentration or g Pu/ℓ.)

[c] Lower limit. All other limits are upper limits.

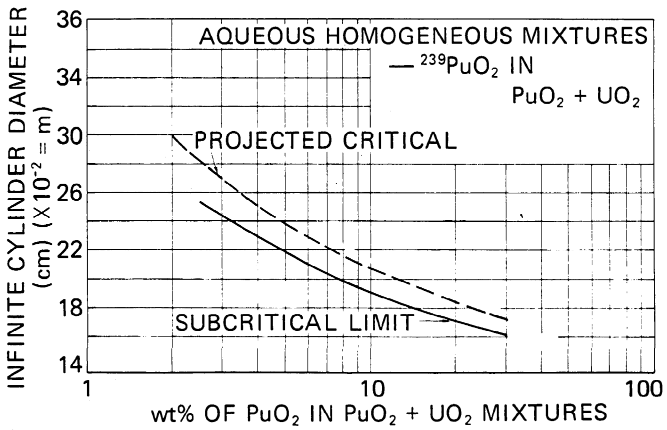

Fig. 3. Cylinder diameter limit versus plutonium content.

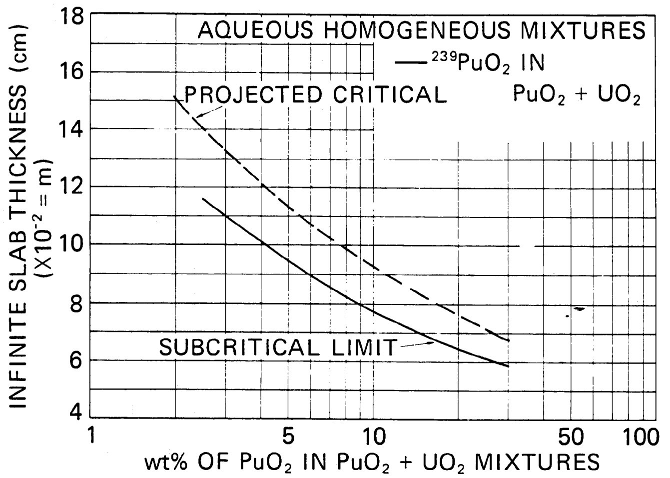

Fig. 4. Slab thickness limit versus plutonium content.

were considered greater than for mass or dimension limits, and a margin of ~4% in k_{eff} was used in deriving the areal density limits of Table I.

Dry and Damp Oxides

Subcritical mass limits for units of mixed oxides are given in Table II. Calculations were made for an isotopic composition of 100% ^{239}Pu, but the limits apply to other compositions. A considerable effort (see next section) was put forth in deriving these limits. In some cases, as many as six "independent" calculations were made. The smallest limits were selected, with a margin in k_{eff} estimated in these calculations to be ~3%. In terms of mass, the corresponding margin increases with mass. For dry 30% PuO_2, it is ~9%, and for damp, half-density 8% PuO_2, nearly 50%. The densities listed are the theoretical values for dry and damp mixed oxides. Subcritical limits are also included for damp mixed oxides at ½ theoretical density.

TABLE II

Subcritical Limits for Units of Mixed Oxides*

	3 wt% PuO_2	8 wt% PuO_2	15 wt% PuO_2	30 wt% PuO_2
Dry mixed oxides at theoretical density	Subcritical in any amount	122 kg Pu (1729 kg $PuO_2 + UO_2$ at density ≤ 11.00 g $PuO_2 + UO_2/cm^3$)	47.0 kg Pu (355 kg $PuO_2 + UO_2$ at density ≤ 11.04 g $PuO_2 + UO_2/cm^3$)	26.1 kg Pu (98.6 kg $PuO_2 + UO_2$ at density ≤ 11.11 g $PuO_2 + UO_2/cm^3$)
Damp mixed oxides at theoretical density, H/(Pu + U) ≤ 0.45	236 kg Pu (8919 kg $PuO_2 + UO_2$ at density ≤ 9.42 g $PuO_2 + UO_2/cm^3$)	49.4 kg Pu (700 kg $PuO_2 + UO_2$ at density ≤ 9.44 g $PuO_2 + UO_2/cm^3$)	32.9 kg Pu (249 kg $PuO_2 + UO_2$ at density ≤ 9.47 g $PuO_2 + UO_2/cm^3$)	23.3 kg Pu (88.1 kg $PuO_2 + UO_2$ at density ≤ 9.52 g $PuO_2 + UO_2/cm^3$)
Damp mixed oxides at one-half theoretical density[a], H/(Pu + U) ≤ 0.45	885 kg Pu (33 447 kg $PuO_2 + UO_2$ at density ≤ 4.71 g $PuO_2 + UO_2/cm^3$)	161 kg Pu (2282 kg $PuO_2 + UO_2$ at density ≤ 4.72 g $PuO_2 + UO_2/cm^3$)	102 kg Pu (771 kg $PuO_2 + UO_2$ at density ≤ 4.73 g $PuO_2 + UO_2/cm^3$)	67.9 kg Pu (256.6 kg $PuO_2 + UO_2$ at density ≤ 4.76 g $PuO_2 + UO_2/cm^3$)

*Masses given are for the plutonium contained in the mixed oxide; the total mass of $PuO_2 + UO_2$, together with the oxide density, is given in parentheses. The limits apply to mixed oxides of ^{239}Pu and natural uranium (^{235}U ≤ 0.71 wt%).

[a] **Note that if any of these numbers is to be applied, the total oxide density must not exceed the densities given for 3, 8, 15, and 30 wt% PuO_2.**

Plutonium Enrichment

Just as uranium oxide cannot be made critical in a homogeneous aqueous mixture without being enriched in ^{235}U or as dry oxide without further enrichment, so there are concentrations of ^{239}Pu in natural uranium that are necessary for criticality, regardless of mass. Subcritical concentration limits of ^{239}Pu in natural uranium are given in Table III for dry and damp mixed oxides, for mixed nitrates in water, and for mixed oxides in water. For the dry and damp oxides, the margin in k was ~5%, and for the aqueous mixtures, ~2%.

TABLE III

Subcritical Limits for ^{239}Pu Content in Uranium ($^{235}U \leq 0.71$ wt%) Applicable to Unrestricted Quantities

	Plutonium Content
Dry Mixed Oxides[a]	
Limiting subcritical wt% ^{239}Pu in Pu + U	4.4 wt%
Damp Mixed Oxides[a]	
$H/(Pu + U) \leq 0.45$; limiting subcritical wt% ^{239}Pu in Pu + U	1.8 wt%
Wet Mixed Oxides[a]	
Limiting subcritical wt% of ^{239}Pu in Pu + U	0.13 wt%
Aqueous Nitrate Solutions	
Limiting subcritical wt% of ^{239}Pu in Pu + U in the presence of 4 nitrate ions per Pu atom [$Pu(NO_3)_4$], and two nitrate ions per uranium atom [$UO_2(NO_3)_2$]	0.65 wt%

[a] These limits are not applicable to atom mixtures of plutonium and uranium, but are restricted to the oxides of these nuclides ($PuO_2 + UO_2$).

Validation of Methods and Establishment of Bias (Ref. 6)

For the purpose of deriving the subcritical limits, several calculations were independently performed, utilizing various codes and cross-section sets. To assess the confidence with which the results of the calculations can be applied, the calculations were validated against pertinent experimental data. Unfortunately, the "areas of applicability" (quoting N16.9) defined by available data do not include many of the conditions of interest for mixed oxides, and "extensions" were required. Agreement between results of diverse methods with different cross-section sets (although no guarantee) was taken to be a good indication of accuracy. Limits were not taken to be greater than the smallest critical values obtained with a reasonably good method, but in some cases were not much less where other methods gave generally larger values. Particular care was exercised to calculate limits by the various codes and cross-section sets in a manner consistent with that adopted for performing correlations.

The MONK Monte Carlo code, a neutronics code derived from GEM, has been used extensively in the U.K. for criticality calculations.[7,8] A sample selection of correlations with this code by two of us (Chalmers and Walker) is given in Table IV. The choice of experiments includes mixed oxides with PuO_2 contents of 30, 14.62 and 7.89 wt% and $H/(Pu + U)$ ratios of 47.4, 30.6, and 51.8, respectively[9]; and plutonium oxides with moderation ratios of 0.04, 15, and 50 (Refs. 10, 11, and 12). The results of the calculations on the experimental systems are included in Table IV. Note that the bias is mainly to overestimate reactivity.

The computational methods selected by another of us (Clark, Savannah River Laboratory) for dry and damp mixed oxides (Table II limits) was S_n as implemented by the ANISN code.[13] The cross sections were Hansen-Roach[14] with ^{238}U resonance cross sections modified by J. R. Knight (Oak Ridge National Laboratory). Cross sections for ^{241}Pu at infinite dilution were furnished by Smith (Los Alamos Scientific Laboratory). Cross sections for ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu at resonance energies were selected by linear interpolation of tabulated values as a function of the logarithm of the total potential scattering cross section per absorber atom. The cross-section set for hydrogen was that obtained by fission spectrum weighting. The fission spectrum for ^{235}U or for ^{239}Pu was used, depending on which nuclide was predominant. For infinite systems (Table III limits; infinite sea concentration, Table I) the B_1 method was selected, as implemented by HRXN with Hansen-Roach cross sections and by GLASS (Ref. 15), with essentially HAMMER (Ref. 16) cross sections.

These methods were validated by correlation with a number of critical experiments pertinent to dry and damp mixed oxides. Except for experiments with PuO_2 (Ref. 10), which were not considered by Clark, no experiments have been performed with dry or even damp [$H/(U + Pu) = 0.45$] oxides, but experiments have been performed at fairly low (~3) ratios of hydrogen to fissionable atoms with various ratios of fissionable to fissile atoms. Experiments have also been performed

TABLE IV

Correlations with Experiment Using MONK Monte Carlo Code* (Refs. 7 and 8)

Experiment Number	Composition	Reflector	k_{eff}	Standard Deviation	Reference
1	PuO_2(30)-UO_2-Polystyrene H/(Pu + U) = 47.4	Plexiglas	1.0635	0.0124	9
2	PuO_2(14.62-UO_2-Polystyrene H/(Pu + U) = 30.6	Plexiglas	1.0611	0.0126	9
3	PuO_2(7.89)-UO_2-Polystyrene H/(Pu + U) = 51.8	Plexiglas	1.0357	0.0116	9
4[a]	PuO_2(7.89-UO_2-Polystyrene H/(Pu + U) = 51.8	Plexiglas	1.0624	0.0157	
5	PuO_2 H/Pu = 0.04	Plexiglas	0.9926	0.0177	10
6	PuO_2-Polystyrene H/Pu = 15	Plexiglas	1.0298	0.0134	11
7	PuO_2-Polystyrene H/Pu = 50	Plexiglas	1.0400	0.0124	12

*Calculations by J. H. Chalmers and G. Walker.

[a] This calculation was made on the actual experimental array of compacts from data provided by S. R. Bierman. All other calculations were made on the homogeneous cuboid, as reported.

with dry metal. For nonspherical experiments, correlations were made with the KENO code,[17] again with Hansen-Roach cross sections, since it is equivalent to S_∞. Some of the experiments contained ^{234}U, ^{238}Pu, ^{242}Pu, and ^{241}Am, which are not in the Hansen-Roach tabulation. Cross sections for ^{238}Pu were furnished by Smith. Fairly rough approximations were made for the remaining three nuclides, but they were considered adequate for the minor concentrations of these nuclides in the experiments.

There is a series of critical experiments with Plexiglas cuboids built from blocks of PuO_2 and UO_2 compacted with polystyrene in which the H/(Pu + U) ratio is 2.8, the concentration of plutonium in the U + Pu is 29.3%, and the plutonium contains 11.5% ^{240}Pu (Ref. 18). Correlations with these experiments are given in Table V. The ANISN calculations, involving transverse bucklings appropriate for a bare system, are not very meaningful except as a guide for the fairly large and somewhat uncertain extrapolation to the infinte slab.

There are PCTR experiments with UO_3 enriched to ~1% (Ref. 19) and with UO_3 enriched to 3.04% (Ref. 20) ^{235}U at H/U ratios extending upward from ~3.5. No plutonium was present, but correlation with these experiments ought to give a good indication of the bias at high ratios of fissionable to fissile atoms. The results for the experiments near 1% enrichment are reported as values of k_∞ as a function of H/U ratio with an uncertainty in k_∞ of about ±0.005. Since there may be some discrepancy in k_∞ calculated by the four-factor formula of the report and k_∞ calculated as the ratio of neutrons produced per neutron absorbed by B_1 method, reported cross sections and k_∞'s were used to determine the amount of boron required to make k_∞ unity. The results, expressed as B/U ratios, are given in Table VI. The H/U ratios listed in Table VI differ somewhat from the reported values, which are inconsistent with reported fractional water contents. Also given in Table VI are values of k_∞ calculated for these compositions. These values were fitted by least-squares to linear expressions in H/U. For the Hansen-Roach correlation, k = 1.0375 - 0.003427 H/U, and for GLASS, k = 1.0240 - 0.002145 H/U. The data show too much scatter to display any departure from linearity over the limited range of H/U. The deviation of k from unity represents the bias of the calculational method. For the experiments at 3.04%, the values in Table VII are the values computed for the reported barns of $1/v$ absorber per uranium atom required to make k_∞ unity, with the absorber taken to be boron. Correlations beyond an H/U ratio of 8.01 are not given, since these are not pertinent here. A similar analysis was carried out for nitrate experiments.[21] The correlation is

TABLE V

Correlations with PuO_2-UO_2 Compacts Reflected by 15 cm of Plexiglas

(Density 1.185 - 8% hydrogen, 60% carbon, 32% oxygen)*

Experimental Dimensions[a] (cm)	k_{eff}			
	S_4	S_8	S_{16}	KENO
35.57 × 35.63 × 36.50			0.9085	1.0163 ± 0.0062
28.86 × 40.66 × 40.72			0.9396	1.0073 ± 0.0053
22.10 × 50.82 × 50.90			0.9737	1.0023 ± 0.0053
20.49 × 50.82 × 61.08			0.9832	1.0090 ± 0.0050
19.01 × 60.98 × 61.08			0.9923	0.9954 ± 0.0061
18.49 × 61.08 × 66.07			0.9949	1.0072 ± 0.0058
18.04 × 66.07 × 71.26			1.0050	1.0129 ± 0.0061
13.01 × ∞ × ∞	1.0286	1.0299	1.0301	1.0220 ± 0.0053
12.60 × ∞ × ∞	1.0203	1.0217	1.0219	1.0218 ± 0.0063

*ANISN or KENO with Hansen-Roach cross sections. The errors associated with the KENO calculations are one standard deviation and do not include reported experimental uncertainties in dimensions. Adjoint biasing was used in the Plexiglas reflector. Calculations were made with the reported atom densities.[18] The experimental data for the 18.04-cm slab do not appear in Ref. 18; these were obtained in a later experiment by Bierman. He also revised his estimate of the infinite slab thickness to 13.01 cm (0.1301 m). The 12.60-cm (0.1260-m) thickness was obtained independently by Clark from an analysis of Bierman's data, and appears more consistent with the trend shown by the S_n calculations. The KENO results may indicate an even smaller thickness for the infinite slab.

[a] From Ref. 18.

TABLE VI

Correlations with UO_3-H_2O Experiments (Ref. 19)

% ^{235}U	H/U	B/U	$k_{\infty calc}/k_{exptl}$	
			HRXN	GLASS
1.0059	3.834	-0.000116	1.0194	1.0120
	5.067	-0.000113	1.0168	1.0114
	6.235	-0.000194	1.0104	1.0069
	6.953	-0.000283	1.0072	1.0039
	7.524	-0.000480	1.0104	1.0087
1.0704	3.785	0.000095	1.0262	1.0174
	5.841	0.000102	1.0195	1.0140
	7.145	-0.000071	1.0157	1.0113
1.1586	3.793	0.000472	1.0297	1.0189
	5.996	0.000555	1.0215	1.0140
	6.909	0.000539	1.0146	1.0071
	7.520	0.000365	1.0166	1.0106

TABLE VII

Correlations with PCTR Experiments (Ref. 20)

% ^{235}U	H/U	$k_{\infty calc}/k_{exptl}$ HRXN
3.04	3.58	1.0320
		1.0302
	5.86	1.0296
		1.0308
		1.0270
		1.0268
	6.38	1.0336
	8.01	1.0324

TABLE VIII

Correlation with Nitrate Experiments (Ref. 21)

% ^{235}U	H/U	$k_{\infty calc}/k_{exptl}$	
		HRXN	GLASS
2.14	6.36	1.0237	1.0211
	7.17	1.0179	1.0159
	8.46	1.0180	1.0182
	10.36	1.0216	1.0234
	10.40	1.0212	1.0231
2.26	8.25	1.0025	1.0022
	11.2	1.0001	1.0010

given in Table VIII and is fitted by k = 1.0307 - 0.001604 H/U for Hansen-Roach, and by k = 1.0229 - 0.0007028 H/U for GLASS.

The correlations with the PCTR experiments in Tables VI, VII, and VIII indicate that S_∞, Monte Carlo, or B_1 calculations with Hansen-Roach or HAMMER cross sections are conservative by ~2 to 3% in k_{eff} at low H/U ratios. Similarly, the correlations with the compacts in Table V indicate

TABLE IX

Critical Experiments with Spheres*

Experiment Number	Region	Composition	Density	Dimension	Reference
1	Core	Pu (5.18, 0.30, 0.02; a)	19.74	4.122 ± 0.006	22
	Reflector	H_2O	0.9982	>30	
2	Core	U(93.8)	18.75	8.732 ± 0.009	23
3	Core	Pu (4.5, 0.3; a), 1.02% Ga	15.61	6.385 ± 0.013	23
4	Core	Pu (20.1, 3,1, 0.4; a), 1.01% Ga	15.73	6.660 ± 0.017	23
5	Core	Pu (4.80, 0.30), 1.10% Ga	15.53	4.533 ± 0.008	23
	Reflector	U(Nat)	19.00	19.609	
6	Core	U(93.24)	18.62	6.116 ± 0.004	23
	Reflector	U(Nat)	19.00	18.009	
7	Core	U(93.90)	18.69	6.326 ± 0.011	23
	Reflector	U(Nat)	19.00	9.982	
8	Core	U(93.99)	18.67	6.977 ± 0.011	23
	Reflector	U(Nat)	18.67	4.425	
9	Core	U(93.91)	18.70	7.755 ± 0.013	23
	Reflector	U(Nat)	19.00	1.735	

*Plutonium composition is % ^{240}Pu, ^{241}Pu, ^{242}Pu; the remainder is ^{239}Pu. All percentages are by weight except where "a" denotes at.%. Uranium composition is % ^{235}U; except for 1.02% ^{234}U, remainder is ^{238}U. Natural uranium contains 0.71% ^{235}U; the remainder is ^{238}U. The dimensions are in centimetres and are the core radius and the reflector thickness. In many cases, they were derived from reported masses and densities. Densities are in g/cm^3.

TABLE X

Correlations with Critical Spheres*

Experiment Number	k_{eff} Calculated for Experiment				
	S_4	S_8	S_{16}	S_∞	KENO
1	1.0159 ± 0.0012	1.0008	0.9968	0.9954	0.9845 ± 0.0056
2	1.0103 ± 0.0008	1.0034	1.0013	1.0009	1.0094 ± 0.0053
3	1.0159 ± 0.0017	1.0060	1.0030	1.0019	0.9973 ± 0.0053
4	1.0222 ± 0.0022	1.0124	1.0095	1.0084	0.9989 ± 0.0055
5	1.0153 ± 0.0015	1.0018	0.9983	0.9971	1.0018 ± 0.0052
6	1.0108 ± 0.0005	1.0011	0.9986	0.9978	0.9933 ± 0.0043
7	1.0139 ± 0.0013	1.0040	1.0014	1.0005	1.0017 ± 0.0036
8	1.0122 ± 0.0012	1.0029	1.0003	0.9994	0.9990 ± 0.0049
9	1.0124 ± 0.0013	1.0042	1.0018	1.0014	1.0017 ± 0.0044

*The errors associated with the S_4 calculations correspond to the probable errors in the experimental radii and would presumably be about the same for S_∞.

conservatism in calculations at low H/(U + Pu). The maximum ratio to be considered for damp oxide, however, is only $\sim$0.5, which is much less than the minimum ratio in these experiments. Critical experiment data available for dry, metal spheres are listed in Table IX. For these systems, the correlations in Table X indicate good agreement between calculation and experiment and no apparent dependence on whether the fissile material is ^{235}U or ^{239}Pu.

Experimental data for bare uranium cylinders are listed in Table XI. The correlations (Table XII) appear to indicate an increasing degree of nonconservatism as the concentration of fissile material in uranium decreases.

On the basis of these various correlations,

TABLE XI

Critical Experiments with Bare Cylinders*

Experiment Number	Composition	Density	Radius	Height	Reference
10	Ave: U(0.58, 53.33) 11 pairs: 0.6076 U(Nat), 0.7978 U(93.41)	18.83	13.335	15.459 ± 0.077	23 23
11	Ave: U(0.40, 37.46) 13 triplets: 0.5900 U(Nat), 0.7751 U(93.43), 0.5900 U(Nat)	18.88	13.335	25.416 ± 0.127	23
12	Ave: U(0.17, 16.01) 19 pairs: 1.5262 U(Nat), 0.3021 U(93.3)	18.68	26.67	34.4 ± 0.31	23,20
13	Ave: U(0.15, 14.11) 21 pairs: 1.8098 U(Nat), 0.3062 U(93.3)	18.41	26.67	44.44 ± 0.31	23, 24
14	Ave: U(0.13, 12.32) 25 triplets: 0.6076 U(Nat), 0.3052 U(93.3) 1.5216 U(Nat)	18.64	26.67	60.86 ± 0.30	23, 24
15	Ave: U(0.11, 10.9) 22 quadruplets: 1.8041 U(Nat), 0.2975 U(93.3) 3.0077 U(Nat), 0.2975 U(93.3)	18.63	26.67	118.95 ± 3.0	23, 24

*First figure in average uranium composition is % ^{234}U, next is ^{235}U, remainder is ^{238}U. Dimensions are in centimetres ($\times 10^{-2}$ = m). Heights were derived from reported masses, diameters, and densities, which the experimenters adjusted to compensate for warpage of plates. The dimensions (cm) ($\times 10^{-2}$ = m) of the stacked units were adjusted slightly from the experimental values to give an integral number of groups at critical. The average density was assumed for each component of a group.

TABLE XII

Correlations with Bare Cylinders*

Experiment Number	k_{eff} Calculated for Experiment			
			KENO	
	S_4	S_8	Homogenized	Heterogeneous
10	0.9854 ± 0.0019	0.9840	0.9897 ± 0.0040	1.0057 ± 0.0044
11	0.9850 ± 0.0013	0.9831	0.9864 ± 0.0045	1.0015 ± 0.0039
12	0.9805 ± 0.0026	0.9800	0.9798 ± 0.0039	0.9840 ± 0.0038
13	0.9777 ± 0.0015	0.9771	0.9765 ± 0.0030	0.9823 ± 0.0036
14	0.9781 ± 0.0007	0.9774	0.9845 ± 0.0031	0.9794 ± 0.0046
15	0.9769 ± 0.0010	0.9762	0.9745 ± 0.0028	0.9753 ± 0.0037

*The S_n calculations were made for the homogenized cylinders. The errors associated with the S_4 calculations correspond to the probable errors in the experimental heights. The experimental heights were used directly in the S_n calculations. The KENO calculations for the cylinders having the average compositions were for single cylindrical units. Enclosing the first three in void cuboids gave, respectively, 0.9894, 0.9854, and 0.9833. The KENO calculations for the stacks of alternating Oralloy and natural uranium plates were made for multiple units. Treating the first three as single units with all interfaces specified gave, respectively, 0.9949, 0.9890, and 0.9899.

Clark concluded that concentrations of ^{239}Pu in U(Nat) + ^{239}Pu in dry and damp homogeneous mixtures of oxides for which B_1 calculations with Hansen-Roach cross sections give 0.95 for k_∞ will indeed by subcritical regardless of the mass of material; hence these concentrations may be chosen as subcritical limits (first two limits of Table III). In view of the paucity of data and of the downward trend in k_{eff} as a function of decreasing uranium enrichment in bare cylinders, a larger

TABLE XIII

Summary of Calculations on Mixed Oxides—Dry and Damp* Powders

	Dry—Theoretical Density	Damp—Theoretical Density	Damp—$\frac{1}{2}$ Theoretical Density
		3 wt% PuO_2	
D. W. Magnuson[a]	$k_\infty = 0.992$	688 kg Pu ($k_{eff} = 0.95$) (26 061 kg MO)	2389 kg Pu ($k_{eff} = 0.95$) (90 492 kg MO)
H. K. Clark[b]	---	236 kg Pu ($k_{eff} = 0.95$) (8939 kg MO)	885 kg Pu ($k_{eff} = 0.95$) (33 523 kg MO)
		8 wt% PuO_2	
D. W. Magnuson[a]	167 kg Pu ($k_{eff} = 0.95$) 312.5 kg Pu ($k_{eff} = 1.00$)	56 kg Pu ($k_{eff} = 0.95$) 83.3 kg Pu ($k_{eff} = 1.00$)	--- 290.9 kg Pu ($k_{eff} = 1.00$)
H. K. Clark[b]	122 kg Pu ($k_{eff} = 0.95$) 184 kg Pu ($k_{eff} = 1.00$)	49.4 kg Pu ($k_{eff} = 0.95$) 69.4 kg Pu ($k_{eff} = 1.00$)	161 kg Pu ($k_{eff} = 0.95$) 239.8 kg Pu ($k_{eff} = 1.00$)
L. C. Davenport[c]	160.6 kg Pu ($k_{eff} = 0.95$) 265.6 kg Pu ($k_{eff} = 1.00$)	57.8 kg Pu ($k_{eff} = 0.95$) 88.9 kg Pu ($k_{eff} = 1.00$)	203.8 kg Pu ($k_{eff} = 0.95$) 323.9 kg Pu ($k_{eff} = 1.00$)
J. H. Chalmers[d]	337.5 kg Pu ($k_{eff} = 1.00$)	---	160.9 kg Pu ($k_{eff} = 0.95$) 268.0 kg Pu ($k_{eff} = 1.00$)
		15 wt% PuO_2	
D. W. Magnuson[a]	46 kg Pu ($k_{eff} = 0.95$) 62 kg Pu ($k_{eff} = 1.00$)	32 kg Pu ($k_{eff} = 0.95$) 43.7 kg Pu ($k_{eff} = 1.00$)	103 kg Pu ($k_{eff} = 0.95$) 142.5 kg Pu ($k_{eff} = 1.00$)
H. K. Clark[b]	47 kg Pu ($k_{eff} = 0.96$) 56 kg Pu ($k_{eff} = 1.00$)	32.9 kg Pu ($k_{eff} = 0.96$) 39.4 kg Pu ($k_{eff} = 1.00$)	102 kg Pu ($k_{eff} = 0.96$) 126.9 kg Pu ($k_{eff} = 1.00$)
L. C. Davenport[c]	50.4 kg Pu ($k_{eff} = 0.95$) 66.7 kg Pu ($k_{eff} = 1.00$)	34.4 kg Pu ($k_{eff} = 0.95$) 46.0 kg Pu ($k_{eff} = 1.00$)	113.4 kg Pu ($k_{eff} = 0.95$) 157.0 kg Pu ($k_{eff} = 1.00$)
J. H. Chalmers[d]	---	---	143.0 kg Pu ($k_{eff} = 1.00$)
S. R. Bierman[e]	62.2 kg Pu ($k_{eff} = 1.00$)	48.4 kg Pu[f] ($k_{eff} = 1.00$)	---
G. Walker[d]	64.2 kg Pu ($k_{eff} = 1.00$)	---	141.0 kg Pu ($k_{eff} = 1.00$)
		30 wt% PuO_2	
D. W. Magnuson[a]	25 kg Pu ($k_{eff} = 0.95$) 30.2 kg Pu ($k_{eff} = 1.00$)	23 kg Pu ($k_{eff} = 0.95$) 27.5 kg Pu ($k_{eff} = 1.00$)	63 kg Pu ($k_{eff} = 0.95$) 83.7 kg Pu ($k_{eff} = 1.00$)
H. K. Clark[b]	26.1 kg Pu ($k_{eff} = 0.97$) 28.4 kg Pu ($k_{eff} = 1.00$)	23.3 kg Pu ($k_{eff} = 0.97$) 25.5 kg Pu ($k_{eff} = 1.00$)	67.9 kg Pu ($k_{eff} = 0.97$) 76.3 kg Pu ($k_{eff} = 1.00$)
L. C. Davenport[c]	25.4 kg Pu ($k_{eff} = 0.95$) 31.6 kg Pu ($k_{eff} = 1.00$)	23.0 kg Pu ($k_{eff} = 0.95$) 28.9 kg Pu ($k_{eff} = 1.00$)	70.4 kg Pu ($k_{eff} = 0.95$) 91.7 kg Pu ($k_{eff} = 1.00$)
J. H. Chalmers[d]	23.5 kg Pu ($k_{eff} = 0.95$) 28.3 kg Pu ($k_{eff} = 1.00$)	21.9 kg Pu ($k_{eff} = 0.95$) 27.8 kg Pu ($k_{eff} = 1.00$)	64.2 kg Pu ($k_{eff} = 0.95$) 86.9 kg Pu ($k_{eff} = 1.00$)
S. R. Bierman[e]	32.2 kg Pu ($k_{eff} = 1.00$)	28.9 kg Pu[f] ($k_{eff} = 1.00$)	---

*Damp powders contain ~1.5 wt% water, $H/(Pu + U) \simeq 0.45$.

[a] Computed with ENDF/B-III cross-section data and 123-group XSDRN transport code; the radii for $k_{eff} = 0.95$ were estimated as a function of k_{eff}, which was iterated during the calculational search to $k_{eff} = 1.0$.

[b] Computed with ANISN, a one-dimensional discrete ordinates transport code with anisotropic scattering, utilizing Hansen-Roach cross sections with ^{238}U resonance cross sections modified by J. R. Knight.

[c] Computed with DTF-IV transport code and ENDF/B-III cross sections; ENDF/B cross sections processed by ETOG and FLANGE codes for input to GAMTEC-II code, 18 groups used in DTF-IV.

[d] Computed with MONK-4 Monte Carlo code and British cross sections.

[e] Computed with DTF-IV transport code and ENDF/B-III cross sections; ENDF/B cross sections processed by ETOG and FLANGE codes and averaged over 17 epithermal groups and 1 thermal group (0 to 0.683 eV) by the EGGNIT code.

[f] From interpolation.

TABLE XIV

Calculated Limiting Critical Enrichment for Uranium and Limits for ^{239}Pu Content in Uranium (^{235}U ≤ 0.71 wt%)

	Limiting Critical Enrichment (wt% ^{235}U)		Critical Plutonium Content [wt% ^{239}Pu in (Pu + U)]	
	Experimental	Calculated	Calculated	Bias Corrected
UO_3-H_2O	1.034[a]	1.010		
$UO_2(NO_3)_2$-H_2O	2.104[b]	1.970		
$U(0.71)O_2$-$^{239}PuO_2$-H_2O			0.159	0.183
$U(0.71)O_2(NO_3)_2$-$^{239}Pu(NO_3)_4$-H_2O			0.678	0.812

[a] Reference 19.
[b] Reference 21.

TABLE XV

Calculated Values of k_∞ for Low Enriched UO_3 and Nitrate Aqueous Mixtures

Mixture	Enrichment (wt% ^{235}U)	k_∞ from Experimental Data	Computed k_∞
UO_3-H_2O	1.006 (H/U = 5)	0.99 ± 0.01	0.97
	(H/U = 4)	0.99 ± 0.01	0.97
	1.034 (H/U = 5)	1.00	0.98
$UO_2(NO_3)_2$-H_2O	2.104	1.00	0.989

value was not considered justified. Clark similarly concluded that mass limits for oxides mixtures in which the concentration of plutonium in (U + Pu) is 3 and 8% (Table II) should also be masses for which k_{eff} is calculated by S_∞ to be 0.95. For concentrations of 15 and 30%, he judged k_{eff}'s of 0.96 and 0.97 sufficiently far below the correlating values to provide adequate assurance of subcriticality. The limits in Table II are those calculated by Clark, since his values are the smallest. As many as six "independent" calculations were made by members of the Work Group or their fellow workers in deriving these limits. The results of the various calculations are summarized in Table XIII.

An important nuclear criticality safety parameter for the processing of mixed-oxide fuel is the amount of plutonium which can be added to an aqueous mixture, with and without nitrate, before criticality is achieved (last two limits of Table III). Since there have been no experiments performed to determine these concentrations, limits were deduced from calculations validated against the experimental limiting ^{235}U enrichments for criticality as determined from measurements in the Physical Constants Test Reactor at Hanford. Several analyses were made utilizing different computer codes and cross-section sets. Clark's correlations have already been described. Another of us (Magnuson, ORNL) did calculations using Hansen-Roach cross sections in the ANISN transport code,[13] but also included some calculations with the XSDRN transport code,[25] and 123-group cross sections.[26] His results are given in Table XIV.

The resultant biases in the calculations are 0.024 and 0.134, respectively, for the oxide and nitrate mixtures.

Similar type calculations were independently made by Durst (Battelle-Northwest Laboratories) utilizing ENDF/B-III cross sections in the GAMTEC-II code.[27] The 17 epithermal energy groups were obtained via application of the ETOG code[28] and the thermal group data via application of the FLANGE code.[29] The results are given in Table XV.

The bias in the computed k_∞ is ~2% in the case of UO_3 and 1% for the nitrate. Correcting for the bias, the critical plutonium content in PuO_2 + UO_2 corresponding to a k_∞ of unity would be 0.17 wt% plutonium in the Pu + U. The plutonium content for a k_∞ = 0.98 would be 0.136. For nitrate [$^{239}Pu(NO_3)_4$-$UO_2(NO_3)_2$] water mixtures, the corresponding plutonium content would be 0.77 for a k_∞ of unity and 0.70 for a k_∞ = 0.98.

Clark concluded from his correlations with PCTR experiments that a 2% margin in k was sufficient to compensate for uncertainties and to ensure subcriticality. His limiting values for ^{239}Pu in U + Pu as oxides were 0.134 and 0.142 as calculated from Hansen-Roach cross sections and by GLASS, respectively. The corresponding limits for nitrates were 0.654 and 0.708.

The computed concentrations of ^{239}Pu in (Pu +

TABLE XVI

<table>
<tr><th colspan="3">Limiting Subcritical wt% of ^{239}Pu in Pu + U as Oxides in Water</th></tr>
<tr><td colspan="3">For k_∞ of 0.98</td></tr>
<tr><td colspan="2">Clark</td><td>Durst</td></tr>
<tr><td colspan="2">0.134 (H-R)[a] 0.142 (GLASS)</td><td>0.136</td></tr>
<tr><td colspan="3">For k_∞ of 1.00</td></tr>
<tr><td>Clark</td><td>Durst</td><td>Magnuson</td></tr>
<tr><td>0.166 (H-R) 0.176 (GLASS)</td><td>0.169</td><td>0.183</td></tr>
</table>

[a] Hansen-Roach.

<table>
<tr><th colspan="3">Limiting Subcritical wt% of ^{239}Pu in Pu + U in Presence of Four Nitrate Ions per Plutonium Atom [$Pu(NO_3)_4$], and Two Nitrate Ions per Uranium Atom [$UO_2(NO_3)_2$]</th></tr>
<tr><td colspan="3">For k_∞ of 0.98</td></tr>
<tr><td colspan="2">Clark</td><td>Durst</td></tr>
<tr><td colspan="2">0.654 (H-R) 0.708 (GLASS)</td><td>0.70</td></tr>
<tr><td colspan="3">For k_∞ of 1.00</td></tr>
<tr><td>Clark</td><td>Durst</td><td>Magnuson</td></tr>
<tr><td>0.772 (H-R) 0.771 (GLASS)</td><td>0.77</td><td>0.812</td></tr>
</table>

TABLE XVII

Calculated Minimum Critical Conditions for Water-Reflected PuO_2-UO_2-H_2O Mixtures*

wt% Pu	wt% ^{240}Pu	wt% ^{241}Pu	Minimum Critical						
			Mass g Pu ($\times 10^{-3}$ = kg)	Cylinder Diam, cm ($\times 10^{-2}$ = m)	Slab Thickness, cm ($\times 10^{-2}$ = m)	Volume, ℓ ($\times 10^{-6}$ = m^3)	Areal Density, g Pu/cm^2 ($\times 10$ = kg/m^2)	Concentration, g Pu/ℓ ($\times 10^{-3}$ = kg/ℓ)	Methods
3	0	0	969	26.4	12.4	30.4	0.308	7.23	a
			965	26.5	12.7	30.8		7.29	b
	15	6	1875	33.9	17.2	60.3	0.433	8.91	a
			1738	33.0	16.9	56.5		9.03	b
	25	15	2778	38.5	20.1	85.7	0.533	10.12	a
			2563	37.4	19.6	79.3		10.42	b
8	0	0	701	20.8	8.9	16.1	0.285	7.35	a
			709	21.3	9.4	17.3		7.36	b
	15	6	1275	26.5	12.4	30.9	0.393	9.07	a
			1197	26.4	12.6	30.8		9.10	b
	25	15	1800	29.5	14.3	41.6	0.476	10.30	a
			1657	29.1	14.4	40.4		10.45	b
15	0	0	616	18.6	7.5	12.1	0.276	7.35	a
			629	19.3	8.1	13.3		7.34	b
	15	6	1099	23.8	10.7	23.3	0.378	9.06	a
			1048	24.0	11.1	23.9		9.06	b
	25	15	1505	26.4	12.3	30.8	0.453	10.29	a
			1434	26.4	12.6	30.7		10.39	b
30	0	0	561	16.9	6.4	9.5	0.268	7.32	a
			552	17.2	6.8	9.9		7.30	b
			609[c]	17.2[c]	6.6[c]				
	15	6	975	21.9	9.5	18.7	0.364	9.0	a
			961	22.3	10.0	19.6		9.0	b
			926[c]	21.0[c]					
	25	15	1320	24.1	10.8	24.0	0.436	10.22	a
			1285	24.5	11.4	25.2		10.30	b
			1233[c]	23.0[c]					

*Calculations in this table by Clark, except as noted.

[a] MGBS-TGAN normalized to plutonium solution experiments.

[b] Hansen-Roach cross sections, S_4.

[c] Calculations by Chalmers.

TABLE XVIII

Calculated k_{eff} for Selected Critical Experiments Using the XSDRN-S_8 Transport Code*

Material	Composition ($\times 10^{-3}$ = kg/ℓ)	Geometry	Reflection	Calculated k_{eff}
$U(5)O_2F_2$-H_2O	915 g U/ℓ	25.39-cm sphere	None	1.016
$Pu(95.4)(NO_3)_4$-H_2O	58 g Pu/ℓ	8.455-cm half-slab	None	1.025
$Pu(71.9)(NO_3)_4$-H_2O	202 g Pu/ℓ	10.595-cm half-slab	None	1.013
$Pu(95.4)(NO_3)_4$-H_2O	268.7 g Pu/ℓ	14.57-cm sphere	0.124-cm stainless steel Inf. H_2O	1.009
$Pu(95)(NO_3)_4$-H_2O	35.1 g Pu/ℓ 110.8 g U/ℓ	19.30-cm sphere	0.122-cm stainless steel Inf. H_2O	1.014
$Pu(91.4)O_2$-UO_2-$(C_8H_8)_m$	85.0 g Pu/ℓ 495 g U/ℓ	5.78-cm half-slab	15-cm Plexiglas	1.029
			Average	1.018

*Calculations by Magnuson.

U) that provide k_∞ = unity and 0.98, adjusting for calculational bias, are summarized in Table XVI.

The computational methods selected by Clark for homogeneous aqueous mixtures (Table I limits) were S_4 with Hansen-Roach cross sections (as already described) and MGBS-TGAN (Ref. 30). The latter combination of codes was applied to the three sets of mixed oxide experiments[9] considered by Chalmers and Walker. Extrapolations of the data to infinite slabs were made, and values of k_{eff} were calculated for the slabs. These values were ~0.01 larger than values similarly obtained[6,30] for aqueous solutions of plutonium nitrate at the same $H/^{239}Pu$ ratio. Clark concluded that the bias established for nitrate solutions is appropriate for aqueous mixed-oxide mixtures. Little bias exists in the less extensive correlations of S_4 and Hansen-Roach cross sections with the solution experiments,[6] but there appears to be a trend toward larger values of the correlating k_{eff} with larger $H/^{239}Pu$ ratios. Both methods were used to compute minimum critical masses, dimensions, and concentrations for PuO_2-UO_2-H_2O mixtures. The bias characteristics of plutonium nitrate solutions were used in the MGBS-TGAN calculations. No bias was assumed in the S_4 calculations. However, for the infinite sea concentrations calculated by MGBS and by B_1 from Hansen-Roach cross sections, bias was assumed for both methods and was obtained by extrapolation of trends outside the range of experimental data. Results are given in Table XVII along with a few results obtained by Chalmers. Most of the differences between MGBS-TGAN and ANISN results correspond to a difference of <1% in k_{eff}. The maximum difference is ~2% and occurs mainly for the slabs.

The limiting concentrations for homogeneous mixtures of plutonium and natural uranium, i.e., the "infinite sea" concentrations, were also calculated by Magnuson utilizing ENDF/B-III cross-section data and the XSDRN transport theory code.[31] Values of k_{eff} were calculated for a number of critical experiments to provide an estimate of the bias in the calculational method. The results of these calculations and the experimental conditions are given in Table XVIII.

It is seen that there are biases that are conservative for nuclear criticality safety, i.e., critical systems are calculated to be supercritical, and calculated critical systems would then be subcritical. The biases are in general the same order of magnitude as those previously found with either ANISN and Hansen-Roach 16-group cross sections or with XSDRN and an earlier ENDF/B cross-section set.

The compositions and the k_∞ values calculated with the transport code XSDRN are given in Table XIX. The values of the concentrations for various k_∞ values in Table XX were obtained from Table XIX by linear interpolation and extrapolation. It is noted for the mixtures having Pu/(Pu + U) ratios of 1.0, 0.30, 0.15, and 0.08 that the addition of natural uranium decreases k_∞ or increases the plutonium concentration for criticality (k_∞ = 1). For Pu/(Pu + U) = 0.03, a reversal in the trend is noted, and the k_∞ values are higher than those for the 0.08 Pu/(Pu + U) weight ratio for the same plutonium concentrations.

TABLE XIX

XSDRN Calculations of k_∞ for PuO_2-UO_2-H_2O Mixtures

Weight Fraction, Pu/(Pu + U)	1.0	0.30	0.15	0.08	0.03
Pu (g/ℓ) ($\times10^{-3}$ kg/ℓ)	Calculated k_∞ Values (XSDRN-S_2)				
100 wt% ^{239}Pu					
6.5	0.9474	0.9441	0.9421	0.9414	0.9466
7.0	0.9855	0.9811	0.9783	0.9766	0.9809
79 wt% ^{239}Pu—15 wt% ^{240}Pu—6 wt% ^{241}Pu					
8.0	0.9480	0.9433	0.9411	0.9402	0.9472
8.5	0.9768	0.9710	0.9682	0.9664	0.9714
60 wt% ^{239}Pu—25 wt% ^{240}Pu—15 wt% ^{241}Pu					
9.0	0.9407	0.9354	0.9339	0.9321	0.9397
10.0	0.9875	0.9802	0.9766	0.9742	0.9777

TABLE XX

Plutonium Concentrations for Several Calculated k_∞ Values

k_∞	1.0	0.97	0.95
Weight Fraction, Pu/(Pu + U)	Plutonium Concentrations (g/ℓ) ($\times10^{-3}$ kg/ℓ), with H/Pu Atom Ratios in Parentheses		
100 wt% ^{239}Pu			
1.0	7.19 (3676)	6.80 (3892)	6.47 (4095)
0.30	7.26 (3625)	6.85 (3863)	6.58 (4020)
0.15	7.30 (3594)	6.89 (3831)	6.61 (3996)
0.08	7.33 (3561)	6.91 (3805)	6.62 (3974)
0.03	7.28 (3533)	6.84 (3789)	6.55 (3958)
79 wt% ^{239}Pu—15 wt% ^{240}Pu—6 wt% ^{241}Pu			
1.0	8.90 (2965)	3.38 (3169)	8.03 (3306)
0.30	9.02 (2909)	8.43 (3140)	8.12 (3262)
0.15	9.09 (2873)	8.43 (3131)	8.12 (3253)
0.08	9.14 (2837)	8.57 (3061)	8.19 (3210)
0.03	9.09 (2799)	8.47 (3042)	8.06 (3203)
60 wt% ^{239}Pu—25 wt% ^{240}Pu—15 wt% ^{241}Pu			
1.0	10.27 (2576)	9.63 (2765)	9.20 (2892)
0.30	10.44 (2517)	9.77 (2715)	9.32 (2848)
0.15	10.54 (2479)	9.85 (2682)	9.39 (2818)
0.08	10.61 (2442)	9.90 (2651)	9.42 (2793)
0.03	10.59 (2391)	9.80 (2614)	9.27 (2780)

CONCLUSION

The work leading to the development of a proposed standard involves the efforts, contributions, and views of many persons. In addition to the derivation of subcritical limits, the validations undertaken in this regard have provided heretofore unpublished data on correlations against experimental data that may be used to assess the validity of the codes used, and cross sections, in certain applications.

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