REFERENCE 6

H. K. CLARK, "SUBCRITICAL LIMITS FOR URANIUM-233 SYSTEMS," NUCL. SCI ENG. 81, 379-395 (1982).

NUCLEAR SCIENCE AND ENGINEERING®

(ISSN: 0029-5639)

THE RESEARCH JOURNAL OF THE AMERICAN NUCLEAR SOCIETY

Indexed in "Engineering Index" and
Abstracted in "INIS Atom Index"

VOLUME 81, NUMBER 3, JULY 1982
NSENAO 81 (3) 305-490 (1982) ISSM: 0029-5639

NUCLEAR SCIENCE AND ENGINEERING (ISSN: 0029-5639) is published monthly by the American Nuclear Society, Inc., with business offices at 555 N. Kensington Ave., La Grange Park, Illinois 60525; telephone 312/352-6611. The subscription rate is $90 per volume or $237 for three volumes per calendar year; overseas subscribers add $61 per year for postage and handling. Single copy price is $32 for current and back issues. Microfiche subscriptions are also available at the same rates. Inquiries about the distribution and delivery of NUCLEAR SCIENCE AND ENGINEERING and requests for changes of address should be directed to the American Nuclear Society. Allow 6 weeks for a change to become effective. NUCLEAR SCIENCE AND ENGINEERING is printed in Danville, Illinois 61832, by Interstate Printers and Publishers, Inc. Second-class postage is paid at La Grange, Illinois, and at additional mailing offices.

NUCLEAR SCIENCE AND ENGINEERING: 81, 379-395 (1982)

Subcritical Limits for Uranium-233 Systems

Hugh K. Clark

E. I. du Pont de Nemours and Company, Savannah River Laboratory
Aiken, South Carolina 29808

Received November 3, 1981
Accepted February 3, 1982

As a contribution to the required quinquennial review of the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors (ANSI N16.1-1975/ANS-8.1), limits for homogeneous ^{233}U systems have been recalculated to confirm their subcriticality or, where there were doubts, to propose more restrictive values. In addition, other limits were calculated to be proposed for inclusion, namely, limits for aqueous solutions of $UO_2(NO_3)_2$ and limits for uranium oxides. The same three methods of calculation were used as in similar work done recently for plutonium and ^{235}U systems. The validity of each was established by correlation with the results of pertinent critical experiments.

I. INTRODUCTION

In recent papers,[1,2] limits for plutonium systems and for ^{235}U systems were calculated for comparison with limits presently in the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors.[3] Where there was doubt as to subcriticality of limits in the Standard, more restrictive values were proposed. Occasionally, where the margin of subcriticality seemed unnecessarily large, slightly less restrictive values were proposed. Additional limits were proposed for inclusion in the Standard, such as those for oxides of uranium and for aqueous solutions of uranyl nitrate.

Part of the stimulus for this work was doubt expressed by McNeany and Jenkins[4] as to the subcriticality of the dimensional limits for aqueous solutions of ^{233}U. Attention has now been turned to ^{233}U systems. The same three methods of calculation have been used as in the previous studies. All have been validated by comparison with pertinent critical (or nearly critical) experiments. Dimensional limits in the Standard have indeed been found to be too large, and more restrictive values are being proposed.

II. CALCULATIONAL METHODS

The same three computer code combinations (MGBS-TGAN, HRXN-ANISN, and GLASS-ANISN) were used in this work as in the previous correlations and limit calculations. All codes are modules in the Savannah River Laboratory (SRL) JOSHUA system and are executed by the driver subsystem KOKO, which links the codes and facilitates the preparation of input.[5] The codes MGBS, HRXN, and GLASS all serve the same function, the generation of problem-dependent macroscopic cross sections from composition data and microscopic cross-section libraries. The MGBS code collapses cross sections from a built-in 12-group library to two groups in a B_0 spectrum for use in the two-group diffusion theory code TGAN. The HRXN code incorporates the 16-group Hansen-Roach library. The GLASS code collapses cross sections, taken from an 84-group library

[1] H. K. CLARK, *Nucl. Sci. Eng.*, **79**, 65 (1981).
[2] H. K. CLARK, *Nucl. Sci. Eng.*, **81**, 351 (1982).
[3] "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI N16.1-1975/ANS-8.1," American Nuclear Society.
[4] S. R. McNEANY and J. D. JENKINS, *Nucl. Sci. Eng.*, **65**, 441 (1978); see also *Nucl. Sci. Eng.*, **81**, 303 (1982).
[5] H. C. HONECK, "The JOSHUA System," DP-1380, Savannah River Laboratory (1975).

processed from ENDF/B-IV data, to 16 groups in a B_1 spectrum. The ANISN code performs S_n transport theory calculations with either set of 16-group cross sections. Reference 1 contains fuller descriptions of the methods.

No changes were made in how the codes were used, but a few remarks need to be made about MGBS. In Amster's compendium of thermal neutron cross sections,[6] which is partially incorporated in MGBS, the thermal spectrum is a function of the atomic ratios $^{235}U/H$ and $^{239}Pu/H$, of the cross section of $1/v$ absorbers present expressed in barns per hydrogen atom (b/H) and of temperature. However, only cross sections for 0, 2, and 4 b/H are incorporated in MGBS, although the compendium extends to 12. In MGBS, ^{233}U is treated as a $1/v$ absorber with regard to its effect on the spectrum. Three-point Lagrangian interpolation and extrapolation is provided in terms of b/H. Although cross sections change nearly linearly with b/H, quadratic extrapolation to ratios as high as 17.5 (as in critical experiments with cylinders of aqueous solutions of UO_2F_2) seems questionable. The ^{233}U absorption and fission cross sections deviate from strict $1/v$ behavior; hence, relative thermal absorption may be in error at large extrapolations. On the other hand, as the spectrum hardens, the fraction of fission neutrons reaching the thermal group becomes smaller, and the cross-section errors may have little effect. However, at the high b/H ratio of the volume and dimension limits (~33 for UO_2F_2 solutions), MGBS-TGAN should probably be considered the least reliable of the three methods.

It should be noted that all three methods are one-dimensional. An assumption of separability of the neutron flux into spatial components is required to extend them to finite cylinders or to cuboids. The schemes for doing this, implemented by SPBL for the two S_n methods and by TGAN, are investigated in the Appendix.

III. EXPERIMENTAL DATA

Data for ^{233}U systems are much less extensive than for ^{235}U. No experiments have been done with a water-reflected sphere of ^{233}U metal. Experiments with solutions at the high concentrations at which minimum critical volumes and dimensions occur have not been done with spheres. For the one-dimensional computational methods being validated, the appropriate data are those obtained with spheres or with cylinders that can readily be extrapolated to critical diameters of cylinders with infinite height. Data obtained with vessels so large that assumptions of separability introduce little uncertainty are also suitable. Experiments with solutions have been done with UO_2F_2 and with $UO_2(NO_3)_2$ containing some free acid. Solution densities were calculated from the recipes used for ^{235}U solutions[2] and from reported concentrations. Agreement with reported densities is good. However, calculated $UO_2(NO_3)_2$ solution densities are generally slightly greater than reported densities; UO_2F_2 densities, slightly less.

III.A. Spheres of Aqueous Solution

A series of experiments was done in 1953-1954 with two spherical vessels containing aqueous solutions of $^{233}UO_2F_2$ and having volumes of 9.66 and 17.02 ℓ at room temperature.[7] Both vessels were made critical at several temperatures when water reflected. The larger vessel was also made critical bare at a single temperature. The same two spheres were included in a series of experiments with UO_2F_2 and $UO_2(NO_3)_2$ solutions apparently done at about the same time, but not reported until 1959 (Refs. 8 and 9). In the later report, the larger sphere is stated to have been coated internally with a polyvinyl chloride plastic, Unichrome, which is ~30 wt% chlorine. Removal of the Unichrome was found to decrease the critical concentration of $^{235}UO_2F_2$ by 2%. The Unichrome coating is apparently the systematic error, referred to in the earlier report,[7] which resulted in masses and concentrations "believed to be about 2% high."

Other experiments include bare and water-reflected spheres of $UO_2(NO_3)_2$ solution ranging in volume from 5.8 to 26.0 ℓ (Ref. 10). The spheres were made critical within ±0.0005 in k_{eff}. No free acid concentration is reported, but at a uranium concentration of 131 g/ℓ, the solution averaged[11] 0.375 M HNO_3. The corresponding N/U atomic ratio was 2.67, which presumably described all uranium concentrations, since the various concentrations were obtained by diluting the most concentrated solution.

Finally, experiments with uranyl nitrate solutions

[6]H. J. AMSTER, "A Compendium of Thermal Neutron Cross Sections Averaged Over the Spectra of Wigner and Wilkins," WAPD-185, Westinghouse Atomic Power Division (1958).

[7]J. T. THOMAS, J. K. FOX, and DIXON CALLIHAN, *Nucl. Sci. Eng.*, **1**, 20 (1956).

[8]J. K. FOX, L. W. GILLEY, and E. R. ROHRER, "Critical Mass Studies, Part VIII Aqueous Solutions of ^{233}U," ORNL-2143, Oak Ridge National Laboratory (1959).

[9]J. K. FOX, J. T. THOMAS, and E. R. ROHRER, "Critical Mass Studies of Aqueous Solutions of ^{233}U," ORNL-1715, p. 11, Oak Ridge National Laboratory (1954).

[10]J. T. THOMAS, "Critical Experiments with Aqueous Solutions of $^{233}UO_2(NO_3)_2$," Neutron Physics Division Annual Progress Report, ORNL-4280, p. 53, Oak Ridge National Laboratory (1968).

[11]J. T. THOMAS, Oak Ridge National Laboratory, Private Communication (1980).

were performed in bare 174- and 949-ℓ spheres.[12] In the smaller sphere, the concentration of boron added to the solution was a variable. These experiments were later analyzed to obtain slight corrections for lack of sphericity, etc.[13] With or without the corrections, the spheres were not exactly critical, i.e., k_{eff} deviated slightly from unity.

The critical experimental conditions are given in Table I for all the spheres. In the series with variable temperature, concentrations were calculated from reported masses and volumes, since the concentrations are all reported at 25°C.

III.B. Cylinders of Aqueous Solution

The only experiments at concentrations approaching those at which minimum critical volumes and dimensions occur were performed with $UO_2(NO_3)_2$ and UO_2F_2 solutions in paraffin-reflected cylinders.[8] Most of the cylinders were unreflected on top. An indirect method was used for measuring heights of the uranyl nitrate solutions, resulting in an estimated uncertainty of 3%. The estimated uncertainty for the uranyl fluoride solution heights was 1%. Three or four of the vessels containing UO_2F_2 solutions were coated with Unichrome. (The text says three; four are so indicated in the table of data.) In many cases, there was insufficient material to make the system critical, and critical heights were extrapolated from source multiplication curves. The experimental data for the higher concentration $UO_2(NO_3)_2$ and UO_2F_2 solutions selected for the present work are given in Tables II and III, respectively.

The series of experiments with bare and water-reflected spheres of uranyl nitrate solution also included bare and water-reflected cylinders.[10] According to Thomas,[11] the data reported[10] for the reflected cylinders are for the case where each cylinder was supported by a 24.3-cm-high cylinder of Styrofoam of the same diameter, i.e., both the top and bottom of the cylinder were essentially unreflected, and some of these data are in error. The mass for the 38.1-cm-diam cylinder at 132 g/ℓ should be 2.02 instead of 1.77 kg, and the height for the 20.3-cm-diam cylinder at 95.0 g/ℓ should be 27.02 rather than 20.02 cm. Of more interest are unreported data[11] for the case where the bottom and sides were reflected by water, i.e., the Styrofoam was replaced by water. These data are given in Table IV. However, even in these experiments, concentrations were not great enough or cylinder diameters small enough to be of much interest in the present work. Critical experiments with both bare and water-reflected cylinders have also been done in France.[14] The greatest concentration was 206.5 g ^{233}U/ℓ, and the smallest cylinder diameter was 25 cm. Hence, there is little interest in these data in the present work.

Cylinder data in which there is interest, reported by Gwin and Magnuson,[12] are measurements in large cylinders of aqueous uranyl nitrate solutions at concentrations close to the minimum critical value for an infinite system. The critical heights contain a correction for cylinder bottom structure. The radius was increased in the analysis by an assumed wall thickness so that the dimensions in Table V are estimates of bare critical values. The cylinders are so large that small uncertainties in their exact dimensions have little effect on the results. Temperature was assumed to be 25°C.

III.C. Pertinent Metal Experiments

Since the critical mass of a water-reflected sphere of ^{233}U has not been measured, it is necessary to infer the appropriate bias in calculations for water-reflected metal and oxide from other experiments. Besides experiments with bare and water-reflected plutonium and ^{235}U spheres, for which correlations have been reported,[1,2] the experiments[15] listed in Table VI were considered pertinent. Experiments[16] in which ^{233}U, ^{235}U, and plutonium cores were reflected by beryllium might also be pertinent, but were not considered.

IV. CORRELATIONS

IV.A. Aqueous Solutions

Correlations were made of the three code combinations (HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN) with the sphere experiments of Table I. The results are recorded in Table VII in the same order as the experiments are listed in Table I. Densities of UO_2F_2 and $UO_2(NO_3)_2$ solutions were calculated as for the ^{235}U solutions.[2] The UO_2F_2 solution was represented in MGBS by UO_4. In all codes, aqueous solutions of $UO_2(NO_3)_2$ were treated as solutions of UO_3 in nitric acid solutions. For MGBS, densities of UO_3 and UO_2F_2 were calculated by HRXN and

[12]R. GWIN and D. W. MAGNUSON, *Nucl. Sci. Eng.*, **12**, 364 (1962).

[13]ALAN STAUB, D. R. HARRIS, and MARK GOLDSMITH, *Nucl. Sci. Eng.*, **34**, 263 (1968).

[14]JEAN-GEORGES BRUNA et al., "Alecto-Resultats des Experiences Critiques Homogenes Realisees sur le ^{239}Pu, ^{235}U et ^{233}U," CEA-R 2814, Centre d'Etudes Nucleaires de Saclay (1965).

[15]G. E. HANSEN and H. C. PAXTON, "Reevaluated Critical Specifications of Some Los Alamos Fast-Neutron Systems," LA-4208, Los Alamos National Laboratory (1969).

[16]H. C. PAXTON, "Los Alamos Critical-Mass Data," LA-3067-MS, Rev., Los Alamos National Laboratory (1975).

TABLE I

Critical Spheres of Aqueous ^{233}U Solution

Isotopic Composition[a]	Chemical Concentration, g/ℓ				Radius, cm	Aluminum Wall Thickness,[e] cm	Reflector[f]	Temperature, °C	Reference
	Uranium	Nitrate Ion (NO_3^-)[b]	Thorium[c]	Boron[d]					
1	61.95 62.44 63.79 64.92 66.39	0	0	0	13.21 13.22 13.23 13.24 13.25	0.13	H_2O	32.0 39.5 65.5 83.2 96.5	7
1	39.23 40.01 41.72	0	0	0	15.96 15.96 15.97	0.13[g]	H_2O	26.3 56.0 99.5	7
1	68.22	0	0	0	15.96	0.13[g]	None	27.0	7
2	62.8	43.9	0	0	13.21	0.13	H_2O	25[h]	8
1	67.9	0	0	0	15.95[i]	0.13[g]	None	25[h]	8
1	66.9 61.8 60.8	0	0	0	13.04[j] 13.20[i] 13.28[k]	0.13	H_2O	25[h]	8
1	39.5	0	0	0	15.96	0.13[g]	H_2O	25[h]	8
3	132 95 47.9	93.7 67.5 34.0	0 0 0	0 0 0	11.170 11.847 14.579	0.122	H_2O	25[h]	10,11
3	131 102 74.6 44.6	93.0 72.4 53.0 31.7	0 0 0 0	0 0 0 0	14.579 15.078 15.821 18.378	0.122	None	25[h]	10,11
4	17.14 17.86 18.52 19.18 19.82	12.17 12.61 13.15 13.56 13.99	0.076 0.079 0.082 0.085 0.087	0 0.0239 0.0465 0.0688 0.0912	34.6[l]	0.32	None	20.0	12
5	13.25	7.72	0.057	0	61.0[m]	0.77	None	20.0	12

[a]Isotopic composition in weight percent:

	^{233}U	^{234}U	^{235}U	^{238}U
1	98.7	0.54	0.04	0.72
2	98.7	0.5	0.01	0.79
3	97.53	1.05	0.03	1.39
4	97.70	1.62	0.04	0.64
5	97.67	1.54	0.03	0.76

[b]If the NO_3^- concentration is zero, the solute was UO_2F_2.

[c]Assumed present as ThO_2 at a density of 9.86 g/cm^3.

[d]Assumed present as B_2O_3 at a density of 2.17 g/cm^3.

[e]All vessel walls were aluminum.

[f]The water reflector was effectively infinitely thick (⩾20 cm).

[g]The vessel was coated internally with Unichrome, mocked up by a 0.016-cm-thick layer of CH_2CH Cl with a density 1.4 g/cm^3 or equivalently in GLASS by 0.0092% ^{10}B by weight in the vessel wall and in MGBS by a 0.034-cm-thick layer of iron, the amount required to increase critical ^{235}U concentration by 2%.

[h]Assumed temperature.

[i]Sphere volume was reduced 40 cm^3 to compensate for void above solution.

[j]Sphere volume was reduced 380 cm^3 to compensate for void above solution.

[k]Sphere volume was extrapolated from source multiplication curves.

[l]Corrected values of k_{eff} in order of increasing boron concentration: 1.0002, 1.0008, 1.0009, 1.0000, 1.0001.

[m]Corrected value of k_{eff} 1.0001.

TABLE II

Critical Paraffin-Reflected Cylinders* of Aqueous $^{233}UO_2(NO_3)_2$ Solution

Chemical Concentration, g/ℓ		Radius, cm	Critical Height, cm	Maximum Attainable Experimental Height, cm
Uranium[a]	Nitrate Ion (NO_3^-)			
496.5	346.8	10.25	16.1 ± 0.2	14.0
386.0	269.7	6.32	[b]	51
		7.55	27.9	
		9.53	16.3	
		10.25	14.4	
340.4	237.8	6.32	[b]	59
		7.55	29.0	
		9.53	16.2	
278.6	194.6	6.32	[b]	61
		7.55	30.7	
		10.25	14.7	
200.6	140.1	7.55	38.5 ± 0.5	36.8
		10.25	16.4	
169.2	118.2	6.32	[b]	55
		9.53	18.6	
		10.25	16.7	
162.1	113.2	7.55	46.8 ± 0.5	45.4
		10.25	16.7	
128.7	89.9	7.55	73 ± 2	55.4
		10.25	18.8	

*In this analysis, paraffin was assumed to be CH_2 with a density of 0.89 g/cm^3. Where cylinder radii differ from reported values, they were derived from reported volumes and heights. Only the cylinders of 9.53- and 10.25-cm radius had top reflectors. Wall, bottom, and top (where present) were assumed to be 0.16-cm-thick aluminum; temperature, 25°C.

[a]The uranium contained 98.7% ^{233}U, 0.5% ^{234}U, 0.01% ^{235}U, 0.79% ^{238}U by weight.

[b]Apparently subcritical at any height.

were converted to densities of natural UO_3 and UO_4. No MGBS-TGAN correlations were made with the two series of sphere experiments in which temperature was a variable, since MGBS presumes a temperature of 20°C. The measurements at the lowest temperature were essentially duplicated in the sphere experiments reported along with the paraffin-reflected cylinders.[8] No attempt was made in MGBS to adjust to the temperature of the experiments by the introduction of voids. The correlations are expressed in Table VII in terms of the critical values of k_{eff}, i.e., as 1 + Bias where Bias = k_{eff}(calc) − k_{eff}(expt).

Prior to learning[11] that the N/U ratio was 2.67

TABLE III

Critical Paraffin-Reflected Cylinders* of $^{233}UO_2F_2$ Solution

Uranium,[a] g/ℓ	Radius, cm	Critical Height, cm	Maximum Attainable Experimental Height, cm
693.0	5.60	[b]	29.9
	6.34	38 ± 2	23.8
	8.35	20 ± 1	13.5
608.9	5.60	[b]	34.9
	6.34	41 ± 2	27.6
	8.35	16.7 ± 0.2	16.3
526.8	5.60	[b]	42.6
	6.34	41 ± 1	32.4
	8.35	16.9	
456.9	5.60	[b]	49.0
	8.35	18.0 ± 0.3	16.9
336.4	5.60	[b]	68.5
	6.34	56.5 ± 0.5	53.3
	6.85	48.7 ± 0.5	46.3
	7.55	24.0	
	8.35	19.1 ± 0.4	16.9

*In this analysis, paraffin was assumed to be CH_2 with a density of 0.89 g/cm^3. Only the 8.35-cm-radius cylinder had a top reflector. Wall, bottom, and top (where present) were assumed to be 0.16-cm-thick aluminum; temperature, 25°C. All cylinders except that of 6.34-cm radius (and perhaps of 7.55-cm radius) were coated with Unichrome, mocked up by a 0.016-cm-thick layer of CH_2CH Cl with a density of 1.4 g/cm^3 or equivalently in GLASS by 0.0074% ^{10}B in the vessel wall and in MGBS by a 0.034-cm-thick layer of iron.

[a]The uranium contained 98.7% ^{233}U, 0.54% ^{234}U, 0.04% ^{235}U, 0.72% ^{238}U by weight.

[b]Apparently subcritical at any height.

TABLE IV

Critcal Water-Reflected Cylinders* of $^{233}UO_2(NO_3)_2$ Solution

Uranium[a] Concentration, g/ℓ	Critical Height, cm		
	Cylinder Diameter, cm		
	38.1	25.3	20.3
132	11.80	15.49	21.16
95.0	[b]	17.92	25.40
47.9	18.06	25.90	[b]

*The containers were aluminum with a 0.15-cm-thick wall and a 1.27-cm-thick bottom; the top of the cylinder was not reflected.

[a]The uranium contained 97.53% ^{233}U, 1.05% ^{234}U, 0.03% ^{235}U, and 1.39% ^{238}U by weight.

[b]Insufficient material was available for criticality.

TABLE V

Bare Critical Cylinders of Dilute Aqueous $^{233}UO_2(NO_3)_2$ Solutions

Radius of Cylinders: 155.5 cm

Chemical Concentration of Solution, g/ℓ			Isotopic Content of Uranium,[b] wt%				Critical Height, cm
Uranium	Thorium[a]	Nitrate Ion (NO_3^-)	^{233}U	^{234}U	^{235}U	^{238}U	
14.50	0.014	8.47	97.37	1.50	0.04	1.09	50.85
13.89	0.012	8.77	97.35	1.52	0.05	1.08	60.58
13.22	0.014	8.24	97.30	1.49	0.05	1.16	79.04
12.53	0.100	8.23	97.24	1.55	0.05	1.16	140.16

[a]Assumed present as ThO_2 at a density of 9.86 g/cm^3.
[b]The variations in these values probably represent experimental scatter in the analyses.

TABLE VI

Critical Metal Spheres of Uranium or Plutonium, Some Reflected by ^{235}U-Enriched Uranium

Dominant Isotope in the Assembly	Isotopic Composition, wt%				Density, g/cm^3	Core Radius or Reflector Thickness, cm
	^{233}U	^{234}U	^{235}U	^{238}U		
^{233}U core	98.13	1.24	0.03	0.60	18.424	5.983 ± 0.008
No reflector	---	---	---	---	---	---
^{235}U core	---	1.02	93.80	5.18	18.75	8.732 ± 0.009
No reflector	---	---	---	---	---	---
^{239}Pu core[a]	---	---	---	---	15.778	5.042
^{235}U reflector	---	1.02	93.20	5.78	18.80	1.664 ± 0.016
^{233}U core	98.20	1.10	---	0.70	18.621	5.044
^{235}U reflector	---	1.02	93.20	5.78	18.8	1.222 ± 0.012
^{233}U core	98.20	1.10	---	0.70	18.644	4.600
^{235}U reflector	---	1.02	93.20	5.78	18.8	1.989 ± 0.020

[a]The isotopic composition of the plutonium, in atomic percent, was: ^{239}Pu, 94.79; ^{240}Pu, 4.9; and ^{241}Pu, 0.31. The core contained 1.0 wt% gallium.

in the series of experiments[10] with $UO_2(NO_3)_2$ of Table I, the effect of the N/U ratio on the effective neutron multiplication factor was studied. Increasing the ratio from 2.0 (no free acid) to 2.6 decreased k_{eff} for both the bare and reflected spheres at ~130 and 45 g/ℓ by ~0.004 and ~0.002, respectively.

Four of the sphere experiments were calculated by McNeany and Jenkins.[4] Experiments 9, 10, 11, and 12 in their listing correspond, respectively, to entries 19 ($H/^{233}U$ = 192.3, 131 g/ℓ), 11 ($H/^{233}U$ = 381.5, 67.9 g/ℓ), 23 ($H/^{233}U$ = 1532, 17.14 g/ℓ), and 28 ($H/^{233}U$ = 1987, 13.25 g/ℓ) as listed in Tables I and VII. Their results (by S_8 quadrature) with Hansen-Roach cross sections[17] were 0.994, 0.988, 1.004, and 1.005, respectively. The first two lie appreciably above the corresponding values of Table VII, and appear to indicate use of the dE/E weighted cross sections for hydrogen, rather than the fission spectrum weighted values used here. Part of the reason 0.994 lies so far above 0.972, as calculated here by S_8, is their use of N/U = 2.0. Their results (also by S_8) with ENDF/B-IV cross sections were

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

TABLE VII

Values of k_{eff} Calculated for the Critical ^{233}U Solution Spheres of Table I by the HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN Code Combinations

Uranium Concentration, g/ℓ	H/^{233}U[a]	k_{eff}		
		HRXN-ANISN (S_{∞})	GLASS-ANISN (S_{∞})	MGBS-TGAN
61.95	417.5	0.9890	1.0376	---
62.44	413.2	0.9898	1.0388	---
63.79	399.1	0.9886	1.0386	---
64.92	387.5	0.9868	1.0377	---
66.39	375.1	0.9870	---	---
39.23	662.5	0.9971	1.0310	---
40.01	641.7	0.9988	1.0320	---
41.72	598.0	1.0007	---	---
68.22	379.4	0.9821	1.0359	---
62.8	406.9	0.9839	1.0324	1.0542
67.9	381.5	0.9770	1.0346	1.0736
66.9	387.2	1.0043	1.0480	1.0679
61.8	419.4	0.9886	1.0370	1.0577
60.8	426.4	0.9894	1.0374	1.0576
39.5	658.2	0.9990	1.0333	1.0458
132	190.7	0.9742	1.0395	1.0789
95	268.8	0.9751	1.0333	1.0650
47.9	542.6	0.9859	1.0261	1.0430
131	192.3	0.9704	1.0449	1.1100
102	249.7	0.9745	1.0426	1.0960
74.6	345.0	0.9713	1.0313	1.0726
44.6	583.5	0.9872	1.0323	1.0526
17.14	1532	1.0007	1.0049	1.0076
17.86	1470	1.0001	1.0043	1.0075
18.52	1418	0.9995	1.0036	1.0073
19.18	1368	1.0002	1.0044	1.0084
19.82	1324	0.9994	1.0036	1.0080
13.25	1987	1.0039	0.9964	1.0078

[a]Actually H/fissile. Includes trace of ^{235}U where present. The ratio was calculated from concentrations and density formulas.

1.028, 1.013, 0.996, and 0.991, respectively. The first two lie appreciably below the corresponding values of Table VII, presumably reflecting differences in processing codes, resonance absorption calculation, and group structure. However, the same conclusion as in Ref. 4 is reached, namely, that Hansen-Roach cross sections underestimate k_{eff}, whereas ENDF/b-IV cross sections overestimate it.

Correlations with the paraffin-reflected cylinders of the $UO_2(NO_3)_2$ solution are given in Table VIII and of the UO_2F_2 solution in Table IX in the same order that the experiments are listed in Tables II and III. Since the density of paraffin is somewhat variable (*The Handbook of Physics and Chemistry* gives a range of 0.87 to 0.91 g/cm^3), some consideration was given to the effect of variations in reflector density. For a reflected sphere of solution at ~50 g ^{233}U/ℓ, increasing the paraffin density from 0.87 to 0.91 g/cm^3 increased k_{eff} (as calculated by HRXN-ANISN) by ~0.005. At this concentration, the experimenters found paraffin to be a slightly better reflector than water.[8] On the basis of their experiments, reflecting a sphere by paraffin rather than by water was calculated (again by HRXN-ANISN) to increase k_{eff} by ~0.003.

The approach incorporated in SPBL (Ref. 1) was used to correlate the HRXN-ANISN and GLASS-ANISN codes with the cylinder experiments. An ANISN calculation was made for each dimension while holding the other dimension infinite, and k_{eff} was determined with zero transverse buckling. The quadrature was S_{16}. Corresponding to each of the values of k_{eff}, SPBL computed the geometric buckling by a B_1 calculation. The total geometric buckling was obtained by adding the axial and radial components, and the corresponding value of k_{eff} was

TABLE VIII

Values of Axial Buckling and k_{eff} Calculated for the Critical $^{233}UO_2(NO_3)_2$ Cylinders of Table II by the Three Code Combinations

Atomic Ratio, H/^{233}U[a]	B_H^2, cm^{-2}			k_{eff}		
	HRXN	GLASS	MGBS	HRXN[b]	GLASS	MGBS
42.6	0	0	0	0.98[c,d]	1.05[c,d]	---
	0.01067	0.01114	0.01324	1.0616 ± 0.0091	1.1518	1.1679
57.9	0	0	0	0.96[d]	1.03[d]	1.11[d]
	0	0	0	0.9801[e]	1.0563[e]	1.1259[e]
	<0.00262[f]	---	<0.00281[f]	>0.9408[f]	>1.0170[f]	>1.0881[f]
	0.00685	0.00695	0.00748	0.9793 ± 0.0045	1.0617	1.1055
	0.01075	0.01114	0.01324	1.0358 ± 0.0063	1.1197	1.1254
	0.01235	0.01284	0.01522	1.0422 ± 0.0063	1.1262	1.1268
67.0	0	0	0	0.96[d]	1.03[d]	1.11[d]
	0	0	0	0.9778[e]	1.0517[e]	1.1166[e]
	<0.00205[f]	---	<0.00219[f]	>0.9496[f]	>1.0208[f]	>1.0871[f]
	0.00649	0.00657	0.00709	0.9845 ± 0.0047	1.0645	1.1018
	0.01093	0.01130	0.01341	1.0340 ± 0.0064	1.1154	1.1142
84.2	0	0	0	0.965[d]	1.03[d]	1.10[d]
	0	0	0	0.9705[e]	1.0408[e]	1.1002[e]
	<0.00195[f]	---	<0.00207[f]	>0.9415[f]	>1.0118[f]	>1.0724[f]
	0.00598	0.00605	0.00652	0.9877 ± 0.0042	1.0639	1.0929
	0.01239	0.01279	0.01506	1.0418 ± 0.0063	1.1194	1.1038
121	0	0	0	0.97[d]	1.04[d]	1.095[d]
	0.00422	0.00424	0.00457	0.9973 ± 0.0047	1.0668	1.0884
	0.01117	0.01146	0.01342	1.0489 ± 0.0068	1.1207	1.0952
145	0	0	0	0.98[d]	1.045[d]	1.085[d]
	0	0	0	0.9323[e]	0.9942[e]	1.0512[e]
	<0.00234[f]	---	<0.00250[f]	>0.8988[f]	>0.9607[f]	>1.0185[f]
	0.00970	0.00910	0.01158	1.0250 ± 0.0065	1.0934	1.0726
	0.01105	0.01132	0.01318	1.0385 ± 0.0067	1.1071	1.0796
152	0	0	0	0.98[d]	1.045[d]	1.085[d]
	0.00308	0.00309	0.00330	0.9969 ± 0.0033	1.0619	1.0829
	---	---	0.01319	---	---	1.0744
194	0	0	0	0.99[d]	1.05[d]	1.08[d]
	0.00145	0.00145	0.00151	0.9968 ± 0.0022	1.0563	1.0801
	0.00973	0.00992	0.01148	1.0340 ± 0.0067	1.0972	1.0696

[a]This ratio was calculated from concentrations reported by the experimenters and from density formula. They may differ slightly from the reported ratio. These entries are listed in the same order as are the concentrations of Table II.

[b]The uncertainty in k_{eff} corresponds to the reported uncertainty of 3% in the measured height and the uncertainty associated with the extrapolation to criticality from the source multiplication curves, and was calculated by HRXN-ANISN-SPBL codes only.

[c]These values are uncertain.

[d]The extrapolated calculated value of k_{eff} of a critical infinite cylinder of this solution.

[e]The calculated value of k_{eff} of a cylinder described in the experiments as "apparently cannot be critical at any height," were it to have actually been critical at infinite height.

[f]The calculated values of the axial buckling and k_{eff} of a cylinder of height equal to the maximum height achievable with the inventory of solutions available to the experiment. The GLASS-ANISN-SPBL values were inferred from those obtained with the HRXN-ANISN-SPBL codes.

TABLE IX

Values of Axial Buckling and k_{eff} Calculated for the Critical Uranyl-Fluoride-Solution Cylinders of Table III by the Three Code Combinations

	B_H^2, cm^{-2}			k_{eff}		
H/^{233}U[a]	HRXN	GLASS	MGBS	HRXN[b]	GLASS	MGBS
33.6	0	0	0	0.96	1.05	1.15
	0	0	0	0.9590[c]	1.0448[c]	1.1409[c]
	<0.00614[d]	---	<0.00667[d]	>0.8832[d]	>0.9690[d]	>1.0652[d]
	0.00421	0.00428	0.00453	0.9804 ± 0.0054	1.0719	1.1463
	0.00839	0.00875	0.01045	1.0639 ± 0.0092	1.1624	1.1913
38.8	0	0	0	0.96	1.05	1.15
	0	0	0	0.9559[c]	1.0389[c]	1.1326[c]
	<0.00484[d]	---	<0.00525[d]	>0.8953[d]	>0.9783[d]	>1.0722[d]
	0.00373	0.00379	0.00401	0.9850 ± 0.0045	1.0735	1.1444
	0.01047	0.01094	0.01313	1.0317 ± 0.0042	1.1250	1.1484
45.6	0	0	0	0.96	1.045	1.14
	0	0	0	0.9511[c]	1.0313[c]	1.1215[c]
	<0.00353[d]	---	<0.00380[d]	>0.9063[d]	>0.9865[d]	>1.0772[d]
	0.00375	0.00379	0.00402	0.9813 ± 0.0028	1.0667	1.1331
	0.01041	0.01083	0.01300	1.0308 ± 0.0021	1.1208	1.1385
53.3	0	---	0	0.96	---	1.14
	0	---	0	0.9451[c]	---	1.1093[c]
	<0.00281[d]	---	<0.00301[d]	>0.9092[d]	---	>1.0739[d]
	0.00974	---	0.01208	1.0378	---	1.1372
73.9	0	0	0	0.965	1.05	1.12
	0	0	0	0.9274[c]	0.9995[c]	1.0803[c]
	0.00160[d]	---	0.00168[d]	>0.9068[d]	>0.9789[d]	>1.0603[d]
	0.00222	0.00223	0.00234	0.9830 ± 0.0010	1.0594	1.1127
	0.00285	0.00287	0.00306	1.0166 ± 0.0015	1.0947	1.1352
	0.00874[e]	0.00886[e]	0.00950[e]	0.9874 ± 0.0016[e]	1.0688[e]	1.0958[e]
	0.00921[e]	0.00947[e]	0.01128[e]	1.0342 ± 0.0054[e]	1.1155[e]	1.1164[e]

[a]This ratio was calculated from concentrations reported by the experimenters and from density formula. They may differ slightly from the reported ratio. These entries are listed in the same order as are the concentrations of Table III.

[b]The uncertainty in k_{eff} corresponds to the reported uncertainty of 1% in measured height and uncertainty associated with extrapolation to criticality from the source multiplication curves, and was calculated by the HRXN-ANISN-SPBL codes only.

[c]The calculated value of k_{eff} of a cylinder described in the experiments as "apparently cannot be critical at any height," were it to have actually been critical at infinite height.

[d]The calculated values of the axial buckling and k_{eff} of a cylinder of height equal to the maximum height achievable with the inventory of solutions available to the experiment. The GLASS-ANISN-SPBL values were inferred from those obtained with the HRXN-ANISN-SPBL codes.

[e]Unichrome assumed present, but may have been absent.

calculated, also by B_1. Values of k_{eff} thus determined are greater than would be obtained by a nonseparable solution such as Monte Carlo or two-dimensional (r,z) transport theory (see the Appendix). However, by expressing k_{eff} as a function of axial buckling and extrapolating to zero axial buckling, the values appropriate for infinite cylinders can be obtained. Such values are in agreement with correlations made with spheres of ^{235}U solution.[2]

The variation of k_{eff} with axial buckling exhibited in Tables VIII and IX is greater than that found for ^{235}U solutions, but does not appear inconsistent with that shown in the study reported in the Appendix. However, the variation as the axial buckling approaches zero is not nearly linear, as the Appendix indicates should be the case. Deviations from a straight-line fit are outside the limits of error assigned to the data points. For the nitrate solutions, the three solutions of highest concentration (H/^{233}U = 57.9, 67.0, and 84.2) in the 7.55-cm-radius cylinder have calculated k_{eff} values that are lower than would be expected from the other data. These three values are inconsistent with the assertion[8] that a cylinder of these solutions

having a 6.32-cm-radius cylinder would be subcritical at any height. For ^{235}U solution, a similar disagreement exists with the assertion made by the experimenters that some cylinders would be subcritical at any height.[2] A similar behavior is shown for the fluoride solutions. In particular, at $H/^{233}U = 73.9$, the values of k_{eff} determined for the 8.35- and 7.55-cm-radius cylinders are internally inconsistent, as are those for the 6.85- and 6.34-cm-radius cylinders. There is less reason to doubt that the smallest (5.60-cm-radius) cylinder would be subcritical at any height at all concentrations, but at the four highest concentrations of Table IX, the margin appears small. In extrapolating to zero axial buckling, consideration was given to the slope indicated by the study presented in the Appendix and to the maximum attainable heights in the smallest diameter cylinders. It is expected that the experimenters would have recognized whether these heights corresponded to k_{eff} close to unity. Estimated experimental critical heights were reported for cylinders having k_{eff}, calculated for the maximum experimental height, as much as 0.07 below the value calculated for a cylinder of the estimated critical height.

Comparison of the results of the MGBS-TGAN calculations with the cylinder experiments was performed differently. For each critical cylinder dimension, a search was made for the critical transverse buckling. Subtraction of each of these bucklings from the calculated critical buckling yielded the geometric buckling associated with that dimension. The geometric bucklings were added, and k_{eff} was calculated as

$$k_{eff} = \frac{1 + M^2 B_c^2}{1 + M^2 B_g^2} ,$$

where

B_c^2 = calculated critical buckling

M^2 = associated migration area

B_g^2 = geometric buckling.

This approach, according to the Appendix, should give less variation of k_{eff} with axial buckling. In these comparisons, paraffin was considered to be water, since the two appear nearly equivalent and paraffin is not easily introduced as a material in MGBS.

Although calculations were made of the cylindrical-solution experiments of Table IV, their comparisons with experimental results contribute little to the determination of bias, and they are not reported here.

The values of k_{eff} calculated by three code combinations for the large bare cylinders of uranyl nitrate solutions described in Table V are given in Table X in the order of their earlier listing. The assumptions of separability in SPBL and in the MGBS-TGAN analysis introduce minimal error because of the size of the units. The quadrature in the ANISN calculation was S_{16}.

TABLE X

Values of k_{eff} Calculated for the Critical Bare Aqueous Uranyl Nitrate Cylinders of Table V by the Three Code Combinations

$H/^{233}U$[a]	k_{eff}		
	HRXN-ANISN	GLASS-ANISN	MGBS-TGAN
1818	1.0014	0.9977	1.0049
1898	1.0039	0.9981	1.0078
1996	1.0040	0.9961	1.0085
2108	1.0021	0.9918	1.0081

[a]All fissile atoms present, including trace quantities of ^{235}U, are considered as ^{233}U.

Correlations of HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN with the experiments with spheres and cylinders of aqueous solution are plotted in Figs. 1, 2, and 3. The curves are "eyeball" fits to the data with a tendency to be on the conservative side, especially for GLASS-ANISN and MGBS-TGAN. The steep slope and the coarser (by a factor of 2) vertical scale in Fig. 3 should be noted.

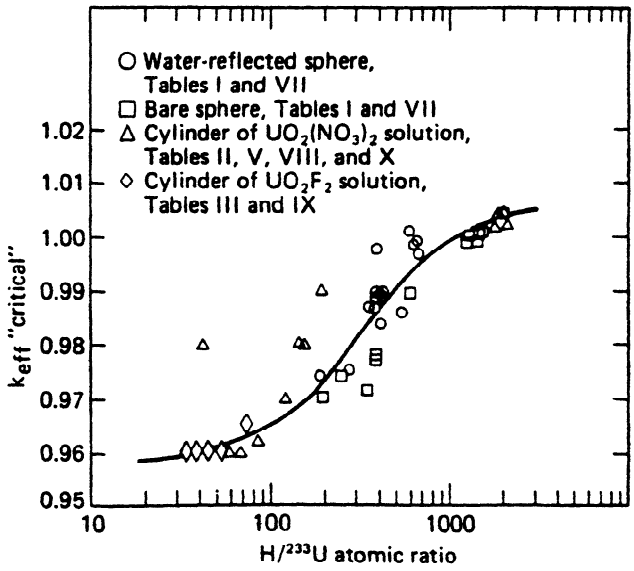

Fig. 1. The k_{eff} of volumes of aqueous solutions of ^{233}U salts as a function of the $H/^{233}U$ atomic ratio calculated by the HRXN-ANISN (S_∞) codes. The line is an "eyeball" fit.

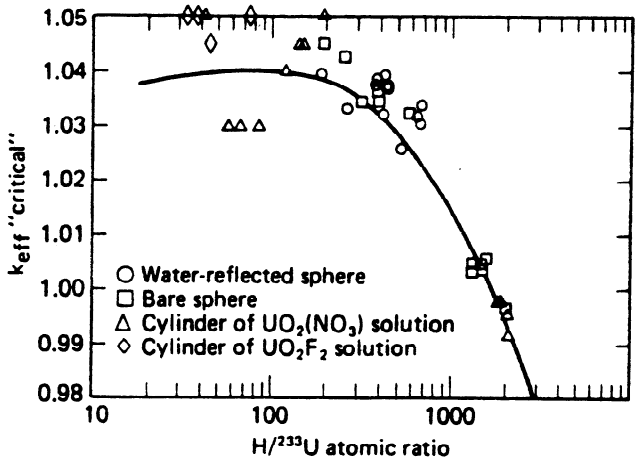

Fig. 2. The k_{eff} of volumes of aqueous solutions of ^{233}U salts as a function of the H/^{233}U atomic ratio calculated by the GLASS-ANISN (S_∞) codes. The line is an "eyeball" fit.

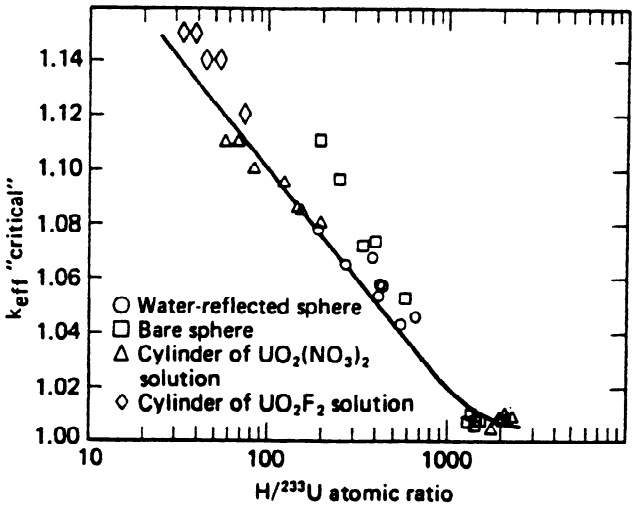

Fig. 3. The k_{eff} of volumes of aqueous solutions of ^{233}U salts as a function of the H/^{233}U atomic ratio calculated by the MGBS-TGAN codes. The line is an "eyeball" fit.

IV.B. Metal Systems

Correlations of HRXN-ANISN and GLASS-ANISN with the metal spheres of Table VI are given in Table XI. The calculations were made in exactly the same manner as for ^{235}U and ^{239}Pu, reported previously.[1,2] The effect of experimental uncertainties was evaluated with Hansen-Roach cross sections by S_4 quadrature. In the GLASS calculations for ^{233}U, the resonance absorption rate exceeded the source rate from slowing down in a number of groups, as was the case with ^{235}U and ^{239}Pu. (In those groups, the absorption rate was arbitrarily set equal to 99% of the source rate, as before.) The bare ^{233}U sphere was also calculated by McNeany and Jenkins[4] with S_8 quadrature. Their results with Hansen-Roach and with ENDF/B-IV cross sections were, respectively, 1.008 and 0.967, in good agreement with Table XI. As they noted, ENDF/B-IV cross sections overestimate k_{eff} for moderated ^{233}U systems and underestimate it for metal.

The bias appropriate for water-reflected metal and oxide cores was selected by combining the results of Table XI with previous results for ^{235}U and ^{239}Pu spheres.[1,2] With Hansen-Roach cross sections, the value of k_{eff} (S_∞) for a critical bare plutonium sphere was 1.0018, and for a critical water-reflected sphere, 0.9951. The corresponding values for a ^{235}U sphere are 1.0004 and 0.9952. The maximum decrease in k_{eff} (occurring for plutonium) was applied to the bare sphere result for ^{233}U to obtain a critical value of 0.9970. With GLASS cross sections, k_{eff} increased by 0.0098 for water reflection of plutonium, and decreased by 0.0024 for water reflection of ^{235}U. For conservatism, the decrease was applied to the bare sphere of ^{233}U to obtain a critical k_{eff} of 0.9635.

V. SUBCRITICAL LIMITS

V.A. Aqueous Solution

All three computational methods (HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN) were used to compute limits for solutions. A temperature of 20°C was assumed, and all units were surrounded by an effectively infinite thickness of water. The ANISN quadrature was S_{16}. The margin necessary to assure subcriticality is difficult to assess from the curves of Figs. 1, 2, and 3. For the concentration limit, the areal density limit, and the mass limit, a margin in k_{eff} of 0.01 seems sufficient in view of experimental data at the corresponding concentrations. Scatter in the data plotted in Figs. 1, 2, and 3 gives an indication of the uncertainty. Since in similar experiments[2] with spheres of ^{235}U solutions the uncertainty in k_{eff} associated with quoted uncertainties in dimensions and solution concentrations is well within ±0.005, a margin of 0.02 should be ample for safety. The dimension limits occur at high concentration where the only data are those obtained with paraffin-reflected cylinders. However, the extrapolations to infinite cylinders are believed to have been done conservatively. Hence, a margin of 0.02 in k_{eff} seems sufficient here also.

Calculations were carried only as far as the saturated fluoride and nitrate solutions, since limits apply only to homogeneous solutions. Johnson and Kraus[18] indicate a 66% aqueous solution of uranyl

[18] J. S. JOHNSON and K. A. KRAUS, *J. Am. Chem. Soc.*, 75, 4594 (1953).

TABLE XI

Values of k_{eff} Calculated for the Critical Metal Spheres of Uranium and Plutonium Given in Table VI

Case	HRXN-ANISN				GLASS-ANISN			
	S_4	S_8	S_{16}	S_∞	S_4	S_8	S_{16}	S_∞
1	1.0164 ± 0.0010	1.0074	1.0047	1.0037	0.9785	0.9696	0.9669	0.9659
2	1.0102 ± 0.0009	1.0033	1.0012	1.0004	1.0217	1.0149	1.0129	1.0117
3	1.0171 ± 0.0015	1.0063	1.0032	1.0021	1.0167	1.0060	1.0030	1.0019
4	1.0175 ± 0.0010	1.0074	1.0045	1.0035	0.9908	0.9810	0.9782	0.9772
5	1.0195 ± 0.0016	1.0091	1.0061	1.0050	0.9992	0.9891	0.9862	0.9852

TABLE XII

Limits for Uniform Homogeneous Aqueous Solutions of UO_2F_2 Containing 100% ^{233}U

Dimension	Present Standard	Calculational Method			Proposed Standard
		HRXN-ANISN	GLASS-ANISN	MGBS-TGAN	
Mass ^{233}U, g	550	530	521	497	540
Cylinder diameter, cm	11.5	10.81	10.50	10.19	10.5
Slab thickness, cm	3.0	2.47	2.67	2.82	2.5
Volume, ℓ	3.5	3.09	2.77	2.52	2.8
^{233}U chemical concentration, g/ℓ	10.8	10.83	10.79	10.73	10.8
$H/^{233}U$ atomic ratio	---	2383	2392	2404	2390
^{233}U areal density, g/cm^2	0.35	0.353	0.351	0.334	0.35

fluoride to be saturated. The equivalent molarity is 5.04, and for the present work a saturated solution was assumed to be 5.0 *M*. Kapustinsky and Lipilina[19] indicate a 52.36% (2.3 *M*) solution of uranyl nitrate to be almost saturated and refer to work by others at as high a concentration as 54.77% (2.44 *M*). For the present work, the saturated solution was assumed to be 2.5 *M*.

V.A.1. Uranyl Fluoride

Table XII contains "limits" for aqueous solutions of UO_2F_2, i.e., minimum values calculated to correspond to a k_{eff} value 0.02 below the curves of Figs. 1, 2, and 3, calculated by the three computational methods. The quadrature in ANISN was S_{16}. Limits now in the Standard, based on Webster's calculations,[20] are shown for comparison, and limits are proposed for the revised Standard.

Agreement is very good in the case of the concentration limit and would be even better if the curve in Fig. 3 were given a sharp upturn at $H/^{233}U$ = 1800, so as to fit the data more closely. The critical concentration calculated by HRXN is 11.22; by GLASS, 11.20; and by MGBS, 11.12 g $^{233}U/ℓ$.

The MGBS-TGAN result for areal density does not agree well with the other two values. The minimum occurs at a concentration of ~0.11 *M* ($H/^{233}U$ = 1000). The 0.334 g/cm^2 limit would increase to ~0.344 g/cm^2 if the curve of Fig. 3 were drawn through the neighboring data points at $H/^{233}U <$ 1000 rather than below them and if a sharp dip were provided at $H/^{233}U$ = 1800, as indicated above. This would increase the critical value of k_{eff} by ~0.01, and the critical density would increase from 0.355 to 0.365 g/cm^2. Slope of the dependence of k_{eff} on $H/^{233}U$ near 1000 is minimal in the HRXN-ANISN calculations, as shown in Fig. 1. Interpolation of the curve to yield critical values of k_{eff} near $H/^{235}U$ = 1000 should be least uncertain in this case. There appears to be no reason to suspect that a margin of 0.02 is insufficient to provide subcriticality or that the Standard limit of 0.35 g/cm^2 might be critical.

The spread in ^{233}U mass values in Table XII is

[19] A. F. KAPUSTINSKY and I. I. LIPILINA, *Bull. Acad. Sci. USSR, Div. Chem. Sci.*, **661** (1956).

[20] J. W. WEBSTER, "Calculated Neutron Multiplication Factors of Uniform Aqueous Solutions of ^{233}U and ^{235}U," ORNL-CDC-2, Oak Ridge National Laboratory (1967).

surprising. The minimum mass noted in the first line occurs at $H/^{233}U \cong 450$. Redrawing the curve in Fig. 3 as indicated above would increase the critical k_{eff} by ~0.006 and increase the critical mass calculated by MGBS-TGAN by ~16 g from 550 to 566 g. (As has been noted previously,[1,2] a margin in k_{eff} of 0.02 corresponds to a larger increment in mass or other parameter as calculated by MGBS-TGAN than by HRXN-ANISN and GLASS-ANISN. For ^{233}U, the difference in mass corresponding to $\Delta k_{eff} = 0.02$ is 53 g by MGBS-TGAN, 43 g by HRXN-ANISN.) The fictitious transverse buckling applied in MGBS-TGAN calculations for spheres makes aluminum walls appear to be worth more than they actually are when the critical k_{eff} deviates appreciably from unity. Since aluminum walls were present in the experiment, their removal, as in the limit calculations, results in too low a critical mass, in the present case ~8 g too low. These two adjustments lead to a critical mass, 574 g, in good agreement with that, 573 g, calculated by HRXN-ANISN with the critical value of k_{eff} read from Fig. 1. Webster[20] calculated a critical mass of 570 g. His few correlations with experiment indicate this mass might be subcritical by a margin of ~0.005 in k_{eff}. Previous calculations by Clark[21] led to a critical mass of ~600 g, in agreement with that reported by Paxton et al.[22]

The critical mass was not calculated by GLASS-ANISN, but would probably be ~564 g. The curve in Fig. 2, however, tends to fall a little below the correlations near $H/^{233}U = 450$. Although it appears doubtful that 550 g could be critical, more confidence is provided by reducing the limit, and, accordingly, 540 g is being proposed in the revised Standard. It also is proposed that the limit for possibly nonuniform slurries[21] be reduced from 520 to 500 g.

As indicated in the discussion of calculational methods, MGBS-TGAN should not be considered highly reliable for calculating dimensional limits. Diffusion theory is presumably less accurate than S_{16} transport theory for converting from one shape to another (e.g., from cylinders, for which bias was established, to slabs). The effect of the aluminum walls is overestimated in diffusion theory calculations. Limits calculated by the three methods are appreciably below the values in the Standard. The minima as calculated by MGBS-TGAN occur at a concentration of ~3.5 *M*. With HRXN-ANISN, the volume minimum occurs at 3.5 *M*, the cylinder diameter minimum is at 4.5 *M*, and the slab thickness is still decreasing at 5.0 *M* (saturation). With GLASS-ANISN, all three dimensions are still decreasing at 5.0 *M*.

V.A.2. Uranyl Nitrate

Limits calculated in the same manner for uranyl nitrate solutions are given in Table XIII, along with values proposed for the revised Standard. The slight differences in concentration and areal density are not worth taking advantage of, and identical limits are proposed for UO_2F_2 and $UO_2(NO_3)_2$. The proposed mass limit for $UO_2(NO_3)_2$ is simply the value proposed for UO_2F_2 in Table XII increased by the nitrate increment calculated by HRXN-ANISN and GLASS-ANISN. The dimensional limits as calculated by MGBS-TGAN and by GLASS-ANISN and the slab thickness calculated by HRXN-ANISN are still decreasing at 2.5 *M* (saturation). However, by HRXN-ANISN, the minimum cylinder diameter occurs at 2.25 *M*, and the minimum volume at 2.0 *M*.

[21] H. K. CLARK, *Nucl. Sci. Eng.*, **24**, 133 (1966).

[22] H. C. PAXTON, J. T. THOMAS, DIXON CALLIHAN, and E. B. JOHNSON, "Critical Dimensions of Systems Containing U^{235}, Pu^{239}, and U^{233}," TID-7028, U.S. Atomic Energy Commission (1964).

TABLE XIII

Limits for Uniform Homogeneous Aqueous Solutions of $UO_2(NO_3)_2$ Containing 100% ^{233}U

Dimension	Calculational Method			Proposed Standard
	HRXN-ANISN	GLASS-ANISN	MGBS-TGAN	
Mass ^{233}U, g	543	536	523	550
Cylinder diameter, cm	11.73	11.69	11.41	11.7
Slab thickness, cm	3.13	3.41	3.48	3.1
Volume, ℓ	3.74	3.61	3.36	3.6
^{233}U concentration, g/ℓ	10.86	10.82	10.76	10.8
$H/^{233}U$ atomic ratio	2371	2379	2393	2390
^{233}U areal density, g/cm^2	0.357	0.355	0.339	0.35

TABLE XIV

Limits Calculated for Metal and Dry Oxide* Containing 100% ^{233}U

Material	Dimension[a]	Present Standard	Calculational Method		Proposed Standard
			HRXN-ANISN	GLASS-ANISN	
Metal	M	6.7	6.95	6.05	6.0
	D	4.6	4.90	4.53	4.5
	T	0.54	0.61	0.38	0.38
UO_2	M	---	13.05	10.90	10.9
	MO	---	14.84	12.39	12.4
	D	---	7.89	7.20	7.2
	T	---	1.28	0.80	0.80
U_3O_8	M	---	18.57	15.10	15.1
	MO	---	21.97	17.86	17.8
	D	---	9.94	8.98	9.0
	T	---	1.79	1.12	1.1
UO_3	M	---	21.89	17.56	17.5
	MO	---	26.40	21.17	21.1
	D	---	11.07	9.95	9.9
	T	---	2.09	1.31	1.3

*Densities of U, UO_2, U_3O_8, and UO_3 may not exceed 18.65, 10.76, 8.15, and 7.16 g/cm^3, respectively.

[a]M = mass of ^{233}U in kilograms, MO = mass of uranium oxide in kilograms, D = cylinder diameter in centimetres, and T = slab thickness in centimetres.

TABLE XV

Limits Calculated for Moist* Oxides Containing 100% ^{233}U

Oxide	Dimensions[a]	Full Density[b]			Half-Density[c]		
		HRXN-ANISN	GLASS-ANISN	Proposed Standard	HRXN-ANISN	GLASS-ANISN	Proposed Standard
UO_2	M	13.00	10.15	10.1	32.69	23.40	23.4
	MO	15.01	11.72	11.7	37.75	27.02	27.0
	D	8.35	7.44	7.2	14.26	12.31	11.9
	T	1.42	0.87	0.80	2.84	1.74	1.6
U_3O_8	M	17.62	13.38	13.4	44.06	30.50	30.5
	MO	21.17	16.07	16.0	52.92	36.64	36.6
	D	10.22	9.01	9.0	17.48	14.91	14.8
	T	1.90	1.17	1.1	3.80	2.34	2.2
UO_3	M	20.39	15.26	15.2	50.93	34.68	34.7
	MO	24.96	18.69	18.7	62.35	42.46	42.4
	D	11.26	9.88	9.9	19.28	16.36	16.3
	T	2.19	1.34	1.3	4.37	2.68	2.6

*The oxide contains 1.5 wt% water.

[a]M = mass of ^{233}U in kilograms, MO = mass of moist oxide in kilograms, D = cylinder diameter in centimetres, and T = slab thickness in centimetres.

[b]Full density of moist oxide is based on the assumption that its volume is the sum of the volume of dry oxide (at physical densities of 10.76, 8.15, and 7.16 g/cm^3, respectively, for UO_2, U_3O_8, and UO_3) and the volume of water at 20°C with a density of 0.99823 g/cm^3.

[c]The above densities of oxide and water are halved, i.e., the moist oxide contains 50% voids.

V.B. Metal and Oxides

Limits for metal and dry oxide, calculated by HRXN-ANISN and GLASS-ANISN, are given in Table XIV. These limits correspond to a k_{eff} value 0.02 below the critical value selected by analogy with ^{235}U and plutonium experiments. The metal or oxide cores were surrounded by 20-cm-thick water at 20°C. The quadrature was S_{16}, the small difference between S_{16} and S_∞ being ignored. Since the larger change in the critical k_{eff} between bare and water-reflected systems was selected in Sec. IV.B, a margin of 0.02 was considered sufficient to assure subcriticality for metal. It was also considered sufficient for oxides since experiments with plutonium oxide indicate no lower critical k_{eff} for oxide than for metal.[1] The limits in the present Standard are based on calculations by Roach and Smith[23] and are values they calculate from Hansen-Roach cross sections by S_8 at k_{eff} (uncorrected for bias) = 0.97. Not surprisingly, they are consistent with the HRXN-ANISN results by S_{16} at $k_{eff} = 0.977$. The agreement between the HRXN-ANISN and the GLASS-ANISN calculations is poorer for ^{233}U than it was for ^{235}U or ^{239}Pu and may indicate the selection of too low a critical value of k_{eff} for water-reflected ^{233}U systems. However, in the absence of a definitive experiment or of a compelling reason for increasing the critical value, the prudent course to follow is to base the proposed limits on the GLASS-ANISN calculations.

Limits, calculated similarly, for moist oxides at full and half-density are given in Table XV. The moisture is limited to 1.5 wt% as for ^{235}U and ^{239}Pu. Volumes of moisture and oxide are assumed to be additive. Comparison of Tables XIV and XV shows that moisture reduces the limiting mass of uranium for all oxides as calculated by either method, but the only dimension reduced is the cylinder diameter of UO_3 as calculated by GLASS-ANISN. The proposed limits in Table XV are the lower of the dry and moist values. (Although not tabulated here, calculations were also made for dry half-density oxides and were consistent with other results.)

[23]W. H. ROACH and D. R. SMITH, "Estimate of Maximum Subcritical Dimensions of Single Fissile Metal Units," ORNL-CDC-3, Oak Ridge National Laboratory (1967).

APPENDIX

To gain a better understanding of the application of one-dimensional methods to two-dimensional problems, i.e., finite cylinders, some critical, mathematical benchmark cases were calculated by the TWOTRAN code[24] and were analyzed in various ways by one-dimensional methods. The cases selected were cylinders of $^{233}UO_2F_2$ solution containing 400 g $^{233}U/\ell$, with various height-to-diameter ratios, and reflected by 15-cm-thick water. In some cases, an aluminum wall was interposed. To limit computer time, the calculations were made with two energy groups, isotropic scattering, and no upscatter. The macroscopic cross sections were generated by GLASS from ENDF/B-IV cross sections and are given in Table A.I. Calculations were made with a uniform mesh in each material, using 0.2 times the number of radial intervals and 0.8 times the number of axial intervals prescribed in empirical formulas.[25] Typically, the number of mesh volumes was ~600. Quadrature was S_{16} to give an accurate solution. In addition, ANISN was used to calculate the infinite slab. Calculations have shown a disagreement between TWOTRAN and ANISN for the infinite cylinder corresponding to ~1% in k_{eff} with only

[24]K. D. LATHROP and F. W. BRINKLEY, "TWOTRAN-II: An Interfaced, Exportable Version of the TWOTRAN Code for Two-Dimensional Transport," LA-4848-MS, Los Alamos National Laboratory (1973).

[25]R. G. SOLTESZ et al., "Nuclear Rocket Shielding Methods Modification, Updating, and Input Data Preparation, Vol. IV, One Dimensional Discrete Ordinates Transport Technique. Final Progress Report," WANL-PR(LL)-034, Westinghouse Astronuclear Laboratory (1970).

TABLE A.I

Two-Group Cross Sections

Material	Group	Σ_a, cm^{-1}	$\nu\Sigma_f$, cm^{-1}	Σ, cm^{-1}	$\Sigma_{(g\to g)}$, cm^{-1}	$\Sigma_{(g\to g+1)}$, cm^{-1}
Uranium solution	1	0.020794	0.044029	0.267053	0.222033	0.024226
	2	0.362821	0.800351	1.39704	1.03421	0
Water	1	0.000458	0	0.255747	0.204324	0.050965
	2	0.018972	0	2.23097	2.21200	0
Aluminum	1	0.000419	0	0.135803	0.135163	0.000221
	2	0.011993	0	0.089297	0.077304	0

S_4 quadrature; agreement is much better with S_{16}. The central processing unit time required for the calculation of a cylinder was ~20 min on an IBM Model 195. Results are given in Table A.II. The code indicated that the problems were converged in all cases, despite the specified inner iteration limit of 10 always being reached in the thermal group.

The first method applied to these benchmarks was ANISN-SPBL with the P_0 cross sections of Table A.I and with S_{16} quadrature. In this approach, k_{eff} is calculated for each dimension of a finite cylinder with the other dimension assumed to be infinite. Geometric bucklings are calculated (by B_1) corresponding to each value of k_{eff} and are added to obtain the total geometric buckling. The value of k_{eff} corresponding to this buckling is then calculated (again by B_1). Table A.III gives results obtained by this method for the benchmark cases of Table A.II. The method overestimates k_{eff} for finite cylinders, but the overestimation decreases as the infinite cylinder is approached (i.e., as the axial buckling approaches zero) and k_{eff} then becomes very nearly a linear function of the axial buckling. (The failure of k_{eff} to be exactly unity for the infinite cylinder in Table A.III represents the slight discrepancy between ANISN and TWOTRAN with S_{16} quadrature.) Thus, linear extrapolation of k_{eff} as a function of axial buckling should be a valid procedure for obtaining the critical value of k_{eff} for an infinite cylinder and hence of the bias of the calculational method. An additional test of this thesis was made by repeating the ANISN-SPBL analysis of the benchmarks of Table A.II, but with Hansen-Roach cross sections (16 groups, P_1 scattering.) The cylinders contained in aluminum and the cylinders with height-to-diameter ratios of zero and 0.25 were omitted. Results are given in Table A.IV, and again k_{eff} is nearly linear with B_h^2 at small B_h^2, albeit with slightly larger slope. The low values of k_{eff} are consistent with the finding that, at high concentrations of ^{233}U, ENDF/B-IV cross

TABLE A.II

Dimensions of Critical Benchmark Cylinders Calculated by Two-Dimensional Codes

Height-to-Diameter Ratio	Wall[a] Thickness, cm	Diameter, cm	Height, cm
0[b]	None	∞	2.73126
	0.16	∞	2.75136
0.25	None	25.9620	6.4905
0.50	None	19.1352	9.5676
	0.16	19.2670	9.6335
1.0	None	14.9304	14.9304
2.0	None	12.4237	24.8474
	0.16	12.5212	25.0424
4.0	None	11.0869	44.3476
∞	None	10.2008	∞
	0.16	10.2496	∞

[a]The wall material was aluminum.

[b]The infinite slab was calculated by the ANISN code; other cylinders were calculated by TWOTRAN.

TABLE A.III

Calculation of the Cylinders of Table A.II by the ANISN-SPBL Codes

Height-to-Diameter Ratio	Wall[a] Thickness, cm	Axial Buckling B_h^2, cm^{-2}	Radial[b] k_D	Axial[b] k_H	Total k_{eff}
0	None	0.04698	2.1644	1.0000	1.0000
	0.16	0.04698	2.1644	1.0000	1.0000
0.25	None	0.02604	1.5545	1.2903	1.0705
0.5	None	0.01859	1.3722	1.4494	1.0719
	0.16	0.01839	1.3736	1.4544	1.0767
1.0	None	0.01182	1.2212	1.6402	1.0570
2.0	None	0.00641	1.1118	1.8403	1.0340
	0.16	0.00631	1.1146	1.8445	1.0374
4.0	None	0.00284	1.0462	2.0066	1.0148
∞	None	0.	0.9995	2.1644	0.9995
	0.16	0	0.9994	2.1644	0.9994

[a]The wall material was aluminum.

[b]These are values of k for cylinders with one dimension assumed to be infinity.

TABLE A.IV

Calculation of the Cylinders of Table A.II by the ANISN-SPBL Codes with Hansen-Roach Cross Sections

Height-to-Diameter Ratio	B_h^2, cm^{-2}	Radial[a] k_D	Axial[a] k_H	k_{eff}
0.5	0.01924	1.2955	1.3768	0.9604
1.0	0.01213	1.1282	1.5762	0.9429
2.0	0.00651	1.0043	1.7757	0.9159
4.0	0.00286	0.9292	1.9337	0.8935
∞	0	0.8756	---	0.8756

[a]These are values of k for cylinders with one dimension assumed to be infinity.

TABLE A.V

Calculation of the Cylinders of Table A.II by Critical Transverse Buckling Implemented by ANISN

Height-to-Diameter Ratio	Wall[a] Thickness, cm	B_h^2,[b] cm^{-2}	k_{eff}
0	None	0.04698	1.0000
	0.16	0.04698	1.0000
0.25	None	0.03065	0.9982
0.50	None	0.02261	0.9980
	0.16	0.02363	0.9813
1.00	None	0.01440	0.9988
2.00	None	0.00756	1.0001
	0.16	0.00785	0.9937
4.00	None	0.00318	1.0003
∞	None	0	0.9995
	0.16	0	0.9994

[a]The wall material was aluminum.

[b]Here, B_h^2 is the calculated critical buckling minus the calculated critical radial buckling.

TABLE A.VI

Calculation of the Cylinders of Table A.II by the TGAN Code

Height-to-Diameter Ratio	Wall[a] Thickness, cm	k_{eff}	
		GLASS Code	MGBS Code
0	None	1.0098	1.0657
	0.16	1.0093	1.0593
0.50	None	0.9964	1.0726
	0.16	0.9784	1.0498
1.00	None	0.9968	1.0729
2.00	None	0.9990	1.0722
	0.16	0.9922	1.0605
4.00	None	1.0008	1.0712
∞	None	1.0014	1.0699
	0.16	1.0017	1.0648

[a]The wall material is aluminum.

sections underestimate the critical mass, whereas Hansen-Roach cross sections overestimate it.

Another method of analyzing two-dimensional critical bodies by one-dimensional codes, the one incorporated in TGAN, is to search for the critical transverse buckling corresponding to each critical dimension. The geometric buckling of a finite cylinder is then $B_g^2 = 2B_c^2 - B_r^2 - B_h^2$. The term B_c^2 is the critical buckling calculated from composition and cross sections, and B_r^2 and B_h^2 are, respectively, the transverse (radial) buckling calculated to make a slab with thickness equal to the cylinder height critical and the transverse (axial) buckling calculated to make the cylinder critical. The value of k_{eff} calculated for the critical finite cylinder corresponds to this geometric buckling. This method, implemented by ANISN with the cross sections of Table A.I, was applied to the benchmarks[26] of Table A.II. The transverse leakage is calculated as DB_{tr}^2 and is treated as an equivalent absorption. With the diffusion coefficient, D, equal to $1/3\Sigma_{tr}$, poor results were obtained ($k_{eff} \cong 0.95$). Much better results were obtained with

$$D = \left[\frac{\Sigma}{B^2}\frac{B/\Sigma}{\tan^{-1}(B/\Sigma)}\right] - 1 ,$$

the correct transport theory expression for isotropic scattering. Results of the calculations are given in Table A.V. With only water reflection, the method gives very good results for the finite cylinders, but with the aluminum wall interposed, k_{eff} is too low due to streaming in the aluminum resulting from the assumption of separability of the neutron flux.

The same method as implemented by TGAN with diffusion theory constants was applied to the benchmarks. In one case, the constants were derived by GLASS; in the other, by MGBS. Results are given in Table A.VI. The results from the diffusion theory calculations agree fairly well with those from transport theory. The large values of k_{eff} calculated with MGBS cross sections are consistent with the biases found in the correlations with experiment. Again, the calculated effect of the aluminum wall is too large.

[26]In similar calculations (Ref. 27), it was found that k_{eff} calculated by ANISN at the critical transverse buckling determined by ANISN deviated somewhat from unity, but no such discrepancies were found in the present case.

[27]H. K. CLARK, "Snake Bites from Code Misuse and Overuse," *Proc. Topl. Mtg. Nuclear Criticality Safety*, El Paso, Texas, April 8-10, 1980, SAND-80-1675, Sandia National Laboratories (1980).

ACKNOWLEDGMENT

The information contained in this article was developed during the course of work sponsored by the U.S. Department of Energy under contract with the E. I. du Pont de Nemours and Company.