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Subcritical Limits for Uranium-235 Systems

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As a contribution to a required review of the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, limits for homogeneous ^{235}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 propose for inclusion in the Standard, namely limits for solutions of* $^{235}UO_2(NO_3)_2$ *and limits for solutions of both* UO_2F_2 *and* $UO_2(NO_3)_2$ *that allow credit for the presence of* ^{238}U*. Limits were also calculated for uranium oxides. The same three methods of calculation were used as in similar work done recently for plutonium systems. The validity of each was established by extensive correlations with pertinent critical experiments.*

INTRODUCTION

In a recent paper,[1] limits for plutonium systems were recalculated as a contribution to the mandatory quinquennial review of the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors.[2] The limits are intended to be "maximum" subcritical limits, i.e., actually subcritical under stated conditions, but by a minimal margin. Three calculational methods were used, and all were validated by extensive correlations with the results of pertinent critical experiments to establish bias. Where the present limits were thought to be too high to provide confidence in their subcriticality, lower limits were proposed. In a few cases where the margin of subcriticality seemed unnecessarily large, slightly less restrictive limits were proposed. A number of additional limits were calculated, namely limits for aqueous solutions of $Pu(NO_3)_4$ that allow credit for ^{240}Pu and limits for PuO_2.

This work has now been extended to ^{235}U systems. Limits in the Standard have been recalculated, and in some cases more restrictive values are being proposed. A large number of additional limits have been calculated and are being proposed for inclusion in the Standard. In particular, limits are being proposed for aqueous solutions of uranyl nitrate as well as for solutions of uranyl fluoride as a function of uranium isotopic composition. Limits are also being proposed for uranium oxides.

CALCULATIONAL METHODS

The same three one-dimensional calculational methods were used as for the plutonium limits: MGBS-TGAN, HRXN-ANISN, and GLASS-ANISN. The latter two were supplemented, as before, by SPBL in calculations for finite cylinders or cuboids in which separability of the spacial components of the flux was assumed. All codes are modules in the Savannah River Laboratory JOSHUA system and are executed by the driver subsystem KOKO, which links the codes and facilitates the preparation of input.[3] The codes MGBS, HRXN, and GLASS all

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[1] H. K. CLARK, *Nucl. Sci. Eng.*, **79**, 65 (1981).

[2] American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANS-8.1/ANSI N16.1-1975, American Nuclear Society.

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

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; GLASS collapses cross sections, taken from an 84-group library 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 more complete descriptions of the methods. All three methods were extensively correlated with the results of critical experiments to establish their biases for ^{235}U systems.

Some comments are in order relating specifically to the application of the methods to ^{235}U systems. In MGBS, Amster's[4] thermal neutron cross sections are extrapolated into the range $2 < \mathrm{H}/^{235}\mathrm{U} < 50$. In HRXN, resonance cross sections for all nuclides, including ^{238}U, are tabulated at potential scattering cross sections of 20, 40, 60, 100, 200, . . . , 10^6 b/atom of absorber with three-point Lagrange interpolation and extrapolation at the low end provided in terms of log σ_p. Hence, resonance cross sections of ^{238}U may differ slightly from values obtained directly from the Knight-modified Hansen-Roach sets furnished[5] with KENO-IV. Also, the cross-section sets for ^{234}U and ^{236}U, which, as furnished with KENO-IV, were merely modifications of ^{238}U absorption and production cross sections, σ_a and $\nu\sigma_f$, in the upper groups, were further modified by applying the ratio of 2200 m/s cross sections to groups 13 through 16 and the ratio of infinitely dilute resonance integrals to groups 8 through 12. For small concentrations of ^{234}U and ^{236}U, these approximations should not introduce much error and should be better than treating ^{234}U and ^{236}U as ^{238}U in groups 8 through 16. In GLASS, a ^{235}U fission source replaced the ^{239}Pu fission source used in the previous study.

EXPERIMENTAL DATA

The ^{235}U limits, like the plutonium limits, are one-, or in some cases, zero-dimensional as are the calculational methods. Therefore, in establishing the bias of the calculational methods, principal interest is in critical experiments with spheres and with finite cylinders and cuboids that can readily be extrapolated to infinite cylinders and slabs, or that fill in gaps in the data even if separability is assumed in analyzing them. Measurements of the infinite neutron multiplication factor of homogeneous mixtures in the Physical Constants Testing Reactor (PCTR) at the Hanford Laboratories are also of interest. The experimental data that were selected are described below.

Solution Densities

As was pointed out in the case of plutonium solutions, it is important to have a consistent recipe for computing the density of aqueous solutions that is applied both in the analysis of critical experiments to establish bias and in the subsequent calculation of limits. It is desirable that the recipe be accurate so that bias established for solutions may be applied to calculations for solutions of other solutes or for oxide-water slurries.

Johnson and Kraus[6] have measured the densities of aqueous solutions of UO_2F_2 for a wide range of concentrations at 25 and 30°C and have fitted the results to the empirical formula

$$\frac{1}{d} = \frac{1}{d_0} + aF_2 + bF_2^2 \ ,$$

where d is the density of the solution, d_0 is the density of pure solvent (H_2O), and F_2 is the weight fraction of UO_2F_2. The constants a and b were determined to be −0.9120 and 0.0567, respectively, at 25°C, and −0.9126 and 0.0569 at 30°C. The apparent molal volume can be shown to be[6]

$$V_m = M_2(1/d_0 + a + bF_2) \ ,$$

where M_2 is the molecular weight of UO_2F_2 (308.03 for natural uranium). This formula is, in effect, incorporated in HRXN and KOKO (the portion that prepares GLASS input). Linear interpolation or extrapolation is provided for determining a and b at temperatures other than 25 and 30°C. Concentrations are expressed in grams of uranium per litre rather than weight fraction and are adjusted to concentrations of natural uranium at the same molarity prior to computing the apparent molal volume, which is assumed to be independent of the isotopic composition of uranium. A full range of water densities as a function of temperature is incorporated and provision is made for three-point Lagrange interpolation. The calculation of solution density thus requires only the specification of uranium concentration (and isotopic composition) and temperature.

A similar treatment [also similar to that used

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[4] 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).

[5] L. M. PETRIE and N. P. CROSS, "KENO-IV, An Improved Monte Carlo Criticality Program," ORNL-4938, Oak Ridge National Laboratory (1975).

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

previously for $Pu(NO_3)_4$ solutions[1]] was provided for $UO_2(NO_3)_2$ solutions. These are considered to be solutions of UO_3 in nitric acid. Nitric acid density tables, derived from International Critical Tables, with density expressed as a function of acid molarity and temperature are incorporated in HRXN and KOKO. Analysis of density data for uranyl nitrate solutions ranging from 0.019 to 2.3 *M* at 25°C, reported by Kapustinsky and Lipilina,[7] leads to apparent molal volumes of UO_3 of 26.2, 28.5, 29.8, 30.6, and 31.0 cm^3, at concentrations of 0, 0.6, 1.2, 1.8, and 2.4 *M*, respectively. These apparent molal volumes, which are assumed to be temperature independent, are incorporated in HRXN and KOKO. Five-point Lagrange interpolation is used to obtain intermediate values. The apparent molal volume at 2.4 *M* is assumed to apply at higher concentrations, but 2.4 *M* is nearly saturated. The calculation of solution density then requires only the specification of uranium concentration (and isotopic composition), nitrate concentration, and temperature.

Densities calculated by this recipe were compared with recent measurements of densities of uranyl nitrate solutions made at Rocky Flats.[8] The recipe overestimates the Rocky Flats densities by 0.06, 0.16, and 0.31% at concentrations of ~0.25, 0.6, and 1.5 *M*, respectively. The Rocky Flats data, when interpreted as apparent molal volumes, show a maximum spread at all concentrations about equal to the minimum separation from a plot of the apparent molal volumes derived from the Russian data.[7] The deviation from the Russian data is about the same at all concentrations; the greater deviation in density at higher concentration is due to the increased importance of the exact value of the apparent molal volume The Rocky Flats data are reported with a probable error ranging from 0 to 0.25%. The Russian data have a probable error of ~0.01% and show very little scatter when expressed as apparent molal volumes. Conceivably, the discrepancy between the Rocky Flats and the Russian data may result from the free acid present in the former, which ranged from ~0.1 *M* at the lowest uranium concentration, to ~0.5 *M* at the highest. However, there is no obvious indication that this is the case. The assumptions are accordingly being made that the Russian data are correct and that there is no dependence of apparent molal volume of UO_3 on free acid concentration.

Spheres of Aqueous Solution

During the period from about 1954 to 1966, a number of critical experiments were done at the Oak Ridge National Laboratory (ORNL) with bare and water-reflected spheres of ^{235}U solutions. The water reflectors were effectively infinitely thick. A careful study was made of the reported data, and the critical conditions adopted are given in Table I. In some cases, the conditions were inferred, e.g., the isotopic composition at the higher concentrations in

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

[8] R. E. ROTHE and I. OH, *Nucl. Technol.*, **41**, 207 (1978).

[9] J. T. THOMAS, "Parameters for Two Group Analysis of Critical Experiments with Water Reflected Spheres of UO_2F_2 Aqueous Solutions," Applied Nuclear Physics Division Annual Report, ORNL-2081, p. 81, Oak Ridge National Laboratory (1956).

[10] J. K. FOX and L. W. GILLEY, "Critical Experiments with Aqueous Solutions of ^{235}U," Applied Nuclear Physics Division Annual Report, ORNL-2081, p. 61, Oak Ridge National Laboratory (1956).

[11] J. K. FOX, L. W. GILLEY, and D. CALLIHAN, "Critical Mass Studies, Part IX Aqueous ^{235}U Solutions," ORNL-2367, Oak Ridge National Laboratory (1958).

[12] J. K. FOX, L. W. GILLEY, R. GWIN, and J. T. THOMAS, "Critical Parameters of Uranium Solutions in Simple Geometry," Neutron Physics Annual Progress Report, ORNL-2609, p. 42, Oak Ridge National Laboratory (1958).

[13] J. T. THOMAS, J. K. FOX, and DIXON CALLIHAN, *Nucl. Sci. Eng.*, **1**, 20 (1955); see also "Comparison of Some Nuclear Properties of U-233 and U-235," ORNL-1992, Oak Ridge National Laboratory (1956).

[14] J. T. THOMAS and J. K. FOX, "Measurement of η for ^{233}U," Physics Division Semiannual Progress Report, ORNL-1715, p. 11, Oak Ridge National Laboratory (1954).

[15] J. K. FOX, J. T. THOMAS, and E. R. ROHRER, "UO_2F_2 Critical Experiments," Physics Division Semiannual Progress Report, ORNL-1820, p. 16, Oak Ridge National Laboratory (1955).

[16] J. T. THOMAS, "Critical Parameters of U^{235}-Enriched Solutions in Spherical Geometry," Physics Division Semiannual Progress Report, ORNL-1926, p. 6, Oak Ridge National Laboratory (1955).

[17] D. W. MAGNUSON, "DSN Transport Calculations of Critical Spheres of Uranium Solutions," Neutron Physics Division Annual Report, ORNL-3499, Vol. I, p. 68, Oak Ridge National Laboratory (1963).

[18] E. B. JOHNSON, "Criticality of a Sphere of $U(4.98)O_2F_2$ Solution," Neutron Physics Division Annual Progress Report, ORNL-3973, Vol. 1, p. 14, Oak Ridge National Laboratory (1966).

[19] J. W. WEBSTER and E. B. JOHNSON, *Trans. Am. Nucl. Soc.*, **9**, 514 (1966).

[20] J. K. FOX, L. W. GILLEY, R. GWIN, and D. W. MAGNUSON, "Critical Parameters of ^{235}U and ^{233}U Solutions in Simple Geometry," Neutron Physics Annual Progress Report, ORNL-2842, p. 76, Oak Ridge National Laboratory (1959).

[21] D. F. CRONIN and J. T. THOMAS, "Critical Parameters of Slightly Enriched Uranium Solutions in Spherical Geometry," Physics Division Semiannual Progress Report, ORNL-1926, p. 7, Oak Ridge National Laboratory (1955).

[22] D. F. CRONIN, "Critical Mass Studies Part X," ORNL-2968, Oak Ridge National Laboratory (1960).

[23] E. B. JOHNSON and D. F. CRONIN, *Trans. Am. Nucl. Soc.*, 7, 301 (1964).

TABLE I

ORNL Critical Spheres of Aqueous Uranium Solution

Isotopic Composition[a]	Chemical Concentration, g/ℓ			Radius, cm	Aluminum Wall Thickness[d] cm	Reflector	Temperature, °C	Reference
	Uranium	Nitrate Ion[b]	Boron[c]					
1	696.93	0	0	11.52	0.16	H_2O	25	9
	543.43	0	0	11.47[e]	0.16	H_2O	25	10, 11
	518.77[f]	0	0	11.52	0.16	H_2O	25	9, 10, 11
2	348.60	0	0	11.52	0.16	H_2O	25	12
	213.28	0	0	11.84	0.16	H_2O	25	12
1	102.06	0	0	13.21	0.13	H_2O	27.5	9, 13, 14
	103.80	0	0	13.22	0.13	H_2O	39.5	13, 15
	109.42	0	0	13.24	0.13	H_2O	74.0	13, 14
	111.52	0	0	13.24	0.13	H_2O	85.5	13, 14
	52.91	0	0	15.96	0.13	H_2O	27	9, 13, 14, 15, 16
	53.88	0	0	15.96	0.13	H_2O	43	13, 14
	53.80	0	0	15.96	0.13	H_2O	43	13, 14
	54.29	0	0	15.96	0.13	H_2O	54	13, 14
	54.95	0	0	15.97	0.13	H_2O	64.5	13, 14
	55.48	0	0	15.97	0.13	H_2O	87.5	13, 14
	57.47	0	0	15.97	0.13	H_2O	87.5	13, 14
	134.31	0	0	15.96	0.13	None	27	13, 15
3	64.95	34.18	0	18.35	0.15	None	25	17
4	910.18	0	0	25.39	0.051[g]	None	19	18, 19
2	22.04	0	0	27.91	0.20	H_2O	25	12, 20
5	25.13	0	0	27.91	0.20	None	25	12
6	452.2	0	0	34.40[e]	0.32	H_2O	25	21, 22, 23
	491.7	0	0	34.40[e]	0.32	None	25	21, 22, 23
5	19.99	0	0	34.60	0.32	None	25	12
7	20.12	19.24	0	34.60	0.32	None	20	12, 24 to 28
	23.53	21.91	0.0935	34.60	0.32	None	20	25 to 28
	26.77	24.62	0.187	34.60	0.32	None	20	25 to 28
	28.45	26.22	0.230	34.60	0.32	None	20	25 to 28
8	15.14	11.54	0	61.0	0.77	None	20	25 to 28

[a] Isotopic compositions of the uranium of the various experiments in weight percent:

Number	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
1	0	1.10	93.13	0.50	5.27
2	0	0.98	93.18	0.50	5.34
3	0	0.99	92.43	0.56	6.02
4	0	0	4.98	0	95.02
5	0	1.14	93.20	0	5.66
6	0	0.02	4.89	0	95.09
7	0	1.04	93.18	0.27	5.51
8	0.01	1.05	93.21	0.54	5.19

[b] If the nitrate concentration is zero, the solute was UO_2F_2.

[c] Assumed present as B_2O_3 with an apparent density of 2.17 g/cm^3 derived from H_3BO_3.

[d] Unless noted to the contrary, walls were aluminum with a density of 2.7 g/cm^3. Impurities in actual 2S (Type 1100) or 3S aluminum alloy have an insignificant effect on reactivity and were ignored.

[e] The sphere was not quite full. The radius is that of a sphere having the solution volume.

[f] Critical concentration extrapolated from results for a nearly full sphere.

[g] Stainless steel is assumed to be 72% iron, 18% chrominum, 10% nickel with a density of 7.9 g/cm^3.

the 6.4-ℓ (11.52-cm-radius) sphere, for which ~90% ^{235}U(10) and 93.2% ^{235}U(10) were reported. Magnuson[17] gives a complete isotopic composition in calculations made for 9.7- and 17.0-ℓ (13.21- and 15.96-cm-radius) spheres, critical conditions for which were reported together with those for the 6.4-ℓ sphere; his values were adopted for all three. In most cases the uranium concentrations given in Table I were derived in this analysis from the ^{235}U concentrations and the isotopic compositions reported by the experimenters. (More significant figures are carried in Table I than are justified so as to avoid round-off errors.) Radii were calculated from reported volumes, including cases where the sphere was critical when not quite full.

To prevent corrosion by UO_2F_2 solutions, the spherical shells were coated internally with a phenol base plastic except in one case. The exception was the 17.0-ℓ sphere that was coated with Unichrome, a polyvinyl chloride plastic.[29] After discovery of the chlorine, the coating was removed[29] and the critical mass of ^{235}U, with a water reflector, was reduced by ~2%. This apparently is the systematic error referred to in Ref. 13 that resulted in masses believed to be ~2% high. The concentrations given in Table I for the reflected 17.0-ℓ sphere have been reduced to 98% of the reported values.[13] In what appears to be other references to the same experiments, the 2% correction is variously applied[9,16] and not applied.[14,15] It is not applied by Magnuson.[17]

In the experiments with the 9.7- and 17.0-ℓ spheres,[13] temperature was a variable. However, concentrations are reported at 25°C. The concentrations given in Table I for these experiments are the ratios of the reported masses and volumes. In the other experiments, temperature was not a variable and was not always reported; where it was not reported, 25°C was assumed. The system temperature for the two largest spheres was reported[27,28] to be 24 to 25°C, but values of k_{eff} are reported at 20°C. For these spheres containing uranyl nitrate solution, the reported values of k_{eff} were corrected for the filling port, for a newer value of the delayed neutron fraction, for deviations from sphericity, and for the vessel wall.[30] With the correction for the wall omitted, the respective values of k_{eff}, in the order in which the last five experiments of Table I are listed, are 1.00098, 1.00047, 1.00066, 0.99996, and 1.00100. These values were taken into account in establishing bias, but are close enough to unity to have been ignored in comparison with other experimental uncertainties. The only other experiment for which k_{eff} was reported is that with the bare, stainless steel wall 68.5-ℓ sphere containing UO_2F_2 solution with a uranium enrichment of 4.98%, for which correction for the container wall, for supporting members, and for the column of solution in the filling connection was estimated to reduce k_{eff} to 0.999. Otherwise, in correlations with the experiments, k_{eff} was assumed to have been unity.

The principal experimental error seems to have been the uranium concentration. For experiments 4, 5, 20, 21, 24, and 25 in the order listed in Table I, the volume is stated[12] to be accurate to ±0.5%, the concentration and mass to ±1%. Later publications[27,28] indicate a smaller error in concentration and mass for experiment 25 and for the subsequent experiments,[27-30] namely ±0.5%.

Besides the ORNL sphere experiments, the only others found were several done by the British.[31,32] Only two of these experiments, however, were done with highly enriched uranium; the uranium in the remainder was enriched to 30.45% ^{235}U. These experiments, particularly the two with 92.2% enriched uranium, are of interest in determining whether any bias is associated with the site where the experiments were done. There are slight discrepancies in the descriptions of the experiments given in the two references, which are probably within experimental error. The descriptions adopted here are given in Table II.

Slabs of Aqueous Solution

A series of experiments has been done with thin, water-reflected slabs of highly enriched UO_2F_2 solution at concentrations near that for minimum critical

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[24] D. W. MAGNUSON, "Determination of η of ^{233}U by Comparison of Critical Experiments in a 69.2-cm-dia Sphere," Neutron Physics Annual Progress Report, ORNL-2609, p. 29, Oak Ridge National Laboratory (1958).

[25] D. W. MAGNUSON and R. GWIN, "The Determination of η of U^{233} by Direct Comparison of Critical Experiments in Large Spheres," Neutron Physics Annual Progress Report, ORNL-2842, p. 96, Oak Ridge National Laboratory (1959).

[26] D. W. MAGNUSON and R. GWIN, *Trans. Am. Nucl. Soc.*, **2**, 146 (1959).

[27] R. GWIN and D. W. MAGNUSON, "Critical Experiments for Reactor Physics Studies," ORNL-CF-60-4-12, Oak Ridge National Laboratory (1960).

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

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

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

[31] F. ABBEY, "Handbook of Criticality Data" AHSB(S) Handbook 5, U.K. Atomic Energy Authority, Risley (1967-1968).

[32] 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 II

British Critical Spheres of Aqueous Uranium Solution*

Isotopic Composition, wt%			Reflector	Uranium Concentration, g/ℓ	Radius, cm
^{234}U	^{235}U	^{238}U			
1.0[a]	92.2	6.8[a]	None	76.25	17.45
				44.14	20.28
0.35[a]	30.45	69.2[a]	None	944.17	17.45
				371.10	17.45
				156.85	20.28
				81.44	27.94
			H_2O	235.14	15.25
				146.14	17.45
				107.55	20.28
				70.94	27.94

*The spheres were fabricated from 0.32-cm-thick aluminum and contained aqueous solutions of UO_2F_2. The temperature was assumed to be 20°C.

[a] Assumed.

volume in cylinders.[33] The solution thicknesses were small enough to permit reasonably good extrapolation to the infinite slab. The experiments are described in Table III.

TABLE III

Critical Water-Reflected Slabs of Uranium Solution*

Uranium, g/L	Dimensions[a]	
	Thickness, cm	Height, cm
571.24	5.385 ± 0.025	71.58
	5.232 ± 0.025	88.67
		88.95
	5.08 ± 0.025	106.91
		109.42
	5.067 ± 0.013	115.27
		119.89
503.60	5.232 ± 0.025	93.29
		95.55
	5.067 ± 0.013	130.25
		132.05

*The slab vessel was constructed of Lucite acrylic resin ($C_5H_8O_2$ with the density assumed to be 1.185 g/cm^3). The bottom, end, and one side were 1.904 cm thick. The remaining side was 7.62 − t cm thick, where t is the reported solution thickness. The slab was water reflected except on top. Differing solution heights at the same thickness resulted from differing reflector heights, which, in all cases, exceeded the solution heights. Greater solution heights corresponded to greater reflector heights indicating slightly thinner slabs resulting from greater hydrostatic pressure. The uranium was reported to be 93.2% enriched in the ^{235}U isotope; a composition of 1.1% ^{234}U, 93.13% ^{235}U, 0.5% ^{236}U, 5.27% ^{238}U by weight was assumed, characteristic of uranium used in experiments with spheres at that time.[11] The solution temperature was ~23°C.

[a] The length in all cases was 147.32 cm.

Cylinders of Aqueous Solutions

Many critical experiments have been done with cylinders of aqueous solutions. Of chief interest here are (a) experiments in which the solution concentration was held constant while the cylinder diameter and height were varied and (b) experiments with both uranyl nitrate and uranyl fluoride in vessels of similar size. The former experiments, if done with vessels of sufficiently small diameter, permit extrapolation to infinite cylinders and provide a firmer basis for cylinder diameter limits. The latter experiments indicate whether bias established largely with UO_2F_2 solution is applicable to $UO_2(NO_3)_2$ solutions. No sphere experiments with uranyl nitrate solutions have been done at concentrations as high as that at which minimum critical volume occurs. Since the Standard limits are for systems reflected by water, data for bare cylinders were not generally considered of interest. Table IV lists the conditions for the critical experiments with water-reflected cylinders that were selected. Subcritical results are also frequently helpful in establishing the subcritical limit on cylinder diameter. Table V lists maximum attainable heights (generally limited by quantity of solution

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[33] J. K. FOX, L. W. GILLEY, and J. H. MARABLE, *Nucl. Sci. Eng.*, **3**, 694 (1958).

TABLE IV

Critical Water-Reflected* Cylinders of Uranium Solution+

Chemical Concentration, g/ℓ					
Uranium[a]	Nitrate Ion[b]	Wall[c]	Radius, cm	Height, cm	Reference
890	0	Aluminum	15.35	173.2	34
		Aluminum	15.55	139.6	
		Aluminum	16.50	72.4	
		Aluminum	19.05	41.7	
888.32	0	Aluminum	7.62	89.3	11
		Aluminum	12.70	12.4	
		Aluminum	19.05	7.7	
870.3	0	Aluminum	19.05	44.78	34
		Stainless steel	19.05	50.37	
		Stainless steel	25.40	29.3	
774.95	0	Stainless steel	8.26	49.0	35
		Stainless steel	8.89	34.0	
		Stainless steel	10.16	22.6	
		Stainless steel	11.43	17.8	
728.5	0	Stainless steel	19.05	75.5	34
		Stainless steel	25.40	34.7	
650.3	0	Stainless steel	19.05	153.01	34
		Stainless steel	25.40	40.09	
577.26	0	Stainless steel	7.62	118.4	11
		Aluminum	8.26	38.7	
576.61	0	Aluminum	7.62	70.1	36
		Aluminum	10.16	18.6	
		Aluminum	12.70	12.5	
575.91	0	Stainless steel	8.26	47.1	35
		Stainless steel	8.89	32.7	
		Stainless steel	10.16	21.9	
		Stainless steel	11.43	17.8	
523.72	0	Aluminum	10.16	18.8	37
		Aluminum	12.70	12.6	
495.5	0	Stainless steel	25.40	85.72	34
491.26	0	Aluminum	7.62	70.9	35
		Aluminum	8.26	39.2	
		Aluminum	10.16	19.5	
		Aluminum	12.70	13.4	
423.98	0	Stainless steel	8.26	47.9	35
		Stainless steel	10.16	22.9	
		Stainless steel	11.43	18.0	
		Stainless steel	12.70	15.2	
385.18	271.12	Aluminum	10.16	25.5	38
		Aluminum	11.43	19.7	
		Aluminum	12.70	16.6	
		Aluminum	19.05	9.4	
338.06	0	Aluminum	8.26	42.6	37
		Aluminum	10.16	19.4	

See footnotes at the end of the table.

(Continued)

TABLE IV (Continued)

Chemical Concentration, g/ℓ					
Uranium[a]	Nitrate Ion[b]	Wall[c]	Radius, cm	Height, cm	Reference
111.71	78.63	Aluminum	10.16	36.7	38
		Aluminum	11.43	25.3	
		Aluminum	12.70	20.1	
		Aluminum	15.24	15.1	
		Aluminum	19.05	12.2	
77.48	54.54	Aluminum	11.43	35.0	38
		Aluminum	12.70	26.2	
		Aluminum	15.24	19.3	
		Aluminum	19.05	14.4	
56.59	39.83	Aluminum	12.70	37.0	38
		Aluminum	15.24	24.6	
		Aluminum	19.05	17.9	
55.91	0	Aluminum	12.70	35.2	35
		Aluminum	19.05	16.9	
37.84	26.64	Aluminum	15.24	43.2	38
		Aluminum	19.05	26.7	

[*] A smooth fitting tank inserted in each cylinder provided water reflection on the top.

[+] The temperature was 25°C in the experiments with ~4.9% enrichment. Otherwise, the temperature was assumed to be 23°C. Reference 11 reports that variations in temperature did not add greatly to experimental errors. Reference 35 reports seasonal variations of as much as 15°C.

[a] The uranium isotopic composition was usually given only as percent ^{235}U by weight. The complete composition was generally inferred, as for the sphere experiments. The various assumed compositions in weight percent were:

Reference	^{234}U	^{235}U	^{236}U	^{238}U
11	1.1	93.13	0.5	5.27
34	0.02	4.89	0	95.09
35	1.1	93.4	0	5.5
36	1.1	93.13	0.5	5.27
37	0.98	93.18	0.5	5.34
38	1.1	93.3	0	5.6

Private communication from E. B. Johnson, however, stated that for the highest uranium concentration in Ref. 34 (890 g/ℓ), the enrichment was 4.98%.

[b] If a nonzero nitrate concentration is specified, the uranium was present as $UO_2(NO_3)_2$; otherwise, as UO_2F_2.

[c] The wall thickness was 0.16 cm.

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[34] E. B. JOHNSON and D. F. CRONIN, "Critical Dimensions of Aqueous UO_2F_2 Solutions Containing 4.9% ^{235}U-Enriched Uranium," Neutron Physics Division Annual Progress Report, ORNL-3714, Vol. 1, p. 31, Oak Ridge National Laboratory (1964).

[35] C. K. BECK, A. D. CALLIHAN, J. W. MORFITT, and R. L. MURRAY, "Critical Mass Studies, Part III," K-343, Oak Ridge Gaseous Diffusion Plant (1949).

[36] J. K. FOX and L. W. GILLEY, "Critical Parameters of Aqueous Solutions of U^{235}," Applied Nuclear Physics Division Annual Progress Report, ORNL-2389, p. 71, Oak Ridge National Laboratory (1957).

[37] J. K. FOX, L. W. GILLEY, R. GWIN, and D. W. MAGNUSON, "Critical Parameters of U^{235} and U^{233} Solutions in Simple Geometry," Neutron Physics Annual Progress Report, ORNL-2842, p. 76, Oak Ridge National Laboratory (1959).

[38] DIXON CALLIHAN, D. F. CRONIN, J. K. FOX, and J. W. MORFITT, "Critical Mass Studies, Part V," K-643, Oak Ridge Gaseous Diffusion Plant (1950).

TABLE V

Subcritical Water-Reflected Cylinders of Uranium Solution

Uranium Concentration, g/ℓ	Wall	Radius, cm	Height, cm	
			Maximum Attainable	Estimated Critical
930.19	Stainless steel	7.62	56.9	[a]
885.91	Aluminum	7.62	51.6	>75
870.3[b]	Stainless steel	15.24	152.5	[a]
774.95	Stainless steel	7.62	66.6	[a]
571.24[c]	Aluminum	6.99	203	[a]
454.29	Aluminum	6.99	74.3	[a]
421.80	Stainless steel	7.62	87.6	[a]
227.31	Aluminum	7.62	72.6	[a]
219.49	Stainless steel	8.26	77.2	80
157.99	Stainless steel	8.26	72.3	[a]
150.85	Stainless steel	8.89	54.4	63
123.76	Aluminum	8.26	61.6	[a]
122.51	Stainless steel	8.89	69.9	>100
55.91	Aluminum	10.16	74.5	[a]
55.91	Stainless steel	11.43	40.5	>75
36.75	Aluminum	12.70	56.4	[a]

[a] Subcritical at any height.

[b] From Ref. 34. Except as noted the rest are from Ref. 35.

[c] From Ref. 36. Except as noted the rest are from Ref. 35.

available) and heights estimated to be critical by the experimenters.

Experiments with bare cylinders that are of interest are some that have been done with uranyl nitrate solutions at concentrations close to the minimum critical value.[27,28] The critical heights reported for the smaller cylinder contain a correction for bottom structure and are intended to be truly bare critical heights. The same correction was added to the heights reported for the larger cylinder, and cylinder radii were increased by an assumed wall thickness so that the dimensions as given in Table VI are estimates of the bare critical values. The dimensions are so large that small uncertainties in their exact values and uncertainties introduced by assumptions of separability required with one-dimensional computer codes have little effect.

Homogeneous Hydrogenous Mixtures

A series of experiments, useful for establishing the bias appropriate for calculations of enrichment limits for homogeneous aqueous mixtures, was done[39] in the PCTR with UO_3-H_2O mixtures at three enrichments close to 1.0%. The reported H/U atomic ratios are inconsistent with the reported fractional water contents as given in Appendix II of the reference. The latter were assumed to be correct and the H/U atomic ratios were recalculated. The present recalculation, with atomic weights on a $^{12}C = 12.0000$ basis, gave slightly different results from those obtained in a previous examination of these data.[40,41] The results are given[39] as k on the four-factor formula basis. Discrepancies between k calculated by this formula and from the ratio of production rate to source rate, as in the computer codes, are conceivable. The four-factor formula and a boron cross section of 755 b together with reported cross sections[39] were therefore used[40,41] to convert the deviations of k from unity into amounts of boron (positive or negative) required to make k unity. (The conversion was not reexamined.) The adjusted results in terms of k and of the B/U atomic ratio are given in Table VII. The experimental uncertainty in individual data points in terms of k is about ±0.005.

Another series of PCTR experiments that is somewhat useful was done with UO_3 and $UO_2(NO_3)_2$

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[39] V. I. NEELEY and H. E. HANDLER, "Measurement of Multiplication Constant for Slightly Enriched Homogeneous UO_3-Water Mixtures and Minimum Enrichment for Criticality," HW-70310, Hanford Atomic Products Operation (1961).

[40] Savannah River Laboratory Quarterly Report, Light Water Reactor Fuel Recycle, July-September 1976, DPST-LWR-76-1-3 (Dec. 1976).

[41] E. D. CLAYTON et al., *Nucl. Technol.*, **35**, 97 (1977).

TABLE VI

Critical Unreflected Cylinders* of Uranium Solution

Radius, cm	Uranium, g/ℓ	Isotopic Composition,[a] wt%				Nitrate Ion, g/ℓ	Height, cm
		^{234}U	^{235}U	^{236}U	^{238}U		
77.75	17.31	1.05	93.22	0.55	5.18	13.09	46.51
	17.02	1.03	93.03	0.51	5.43	14.55	48.77
	15.27	1.04	93.12	0.54	5.30	12.25	74.24
	14.60	1.06	93.11	0.52	5.31	12.45	106.83
	14.06	1.06	93.01	0.52	5.41	12.85	204.93
137.11	14.27	1.08	92.79	0.66	5.47	14.48	92.28
	13.96	1.06	92.78	0.65	5.51	14.10	120.73
	13.61	1.05	92.82	0.63	5.50	13.78	242.39

*The temperature is assumed to be 25°C.

[a] Fractional differences are probably within analytical precision.

TABLE VII

Neutron Multiplication Factor and Natural Boron Concentrations for UO_3-H_2O Mixtures*

^{235}U content of Uranium, wt%	H/U Atomic Ratio	k	B/U to Make $k = 1$
1.0059	3.833	0.9920	-0.000116
	5.062	0.9925	-0.000113
	6.229	0.9875	-0.000194
	6.947	0.9821	-0.000283
	7.516	0.9702	-0.000480
1.0704	3.781	1.0063	0.000095
	5.842	1.0064	0.000102
	7.141	0.9957	-0.000071
1.1586	3.789	1.0298	0.000472
	5.990	1.0330	0.000555
	6.904	1.0313	0.000539
	7.516	1.0209	0.000365

*The experiment was performed in the PCTR.

mixed principally with polyethylene in the form of $\frac{1}{8}$-in.-diam spheres for $H/U < 30$, and with polyethylene powder for higher atomic ratios.[42] The ^{235}U enrichment of the uranium was 3.04%. A number of experiments were performed that showed that k was the same for a particular H/U atomic ratio regardless of whether water or polyethylene was used to achieve the ratio. However, in some of the earlier experiments, the error in the analysis for water was so great that the agreement was not conclusive. Typically, the UO_3 mixtures contained ~0.8% water; the $UO_2(NO_3)_2$ mixtures contained ~11.5%. Both mixtures contained 0.34% of unidentified impurities. Analysis of the uranyl nitrate gave a N/U atomic ratio of 1.94. The data are given in Table VIII in terms of the B/U atomic ratio required to make k unity.

A final series of PCTR experiments, intended to to establish the minimum critical enrichment of $UO_2(NO_3)_2$, was done in which $UO_2(NO_3)_2$ of two enrichments was mixed homogeneously with $\frac{1}{8}$-in.-diam spheres of polyethylene.[43] The uranyl nitrate was present as approximately a dihydrate. The

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[42] V. I. NEELEY, J. A. BERBERET, and R. H. MASTERSON, "K_∞ of Three Weight Per Cent U^{235} Enriched UO_3 and $UO_2(NO_3)_2$ Hydrogenous Systems," HW-66882, Hanford Atomic Product Operation (1961).

[43] S. R. BIERMAN and G. M. HESS, "Minimum Critical ^{235}U Enrichment of Homogeneous Hydrogenous Uranyl Nitrate," ORNL-CDC-5, Oak Ridge National Laboratory (1968).

TABLE VIII

Natural Boron Concentration Required to Establish a Neutron Multiplication Factor of Unity in UO$_3$-CH$_2$ and UO(NO$_3$)$_2$-CH$_2$ Mixtures*

UO$_3$		UO$_2$(NO$_3$)$_2$	
Atomic Ratio		Atomic Ratio	
H/U	B/U	H/U	B/U
3.58	0.00960	6.10	0.00438
	0.00969	8.81	0.00532
5.86	0.01116	13.02	0.00542
	0.01100	31.00	0.00000
	0.01118		
	0.01119		
6.38	0.01102		
8.01	0.01126		
8.60	0.01114		
9.94	0.01107		
10.08	0.01091		
10.12	0.01117		
12.36	0.01073		
12.37	0.01072		
30.20	0.00550		
35.26	0.00376		
39.77	0.00138		
40.05	0.00171		
43.85	-0.00022		
47.98	-0.00103[a]		

*The ^{235}U content of the uranium was 3.04 wt%. The experiments were performed in the PCTR.

[a] The reported value of the B/U atomic ratio is -0.00235. It is inconsistent, however, with the cross section -0.774 b/uranium-atom also reported for the 1/v absorber (boron). The value of the B/U atomic ratio listed was determined from this cross section.

uranium enrichments were 2.14 and 2.26 wt% ^{235}U. The data are given as atom densities and as B/^{235}U atomic ratios required to make k unity. The compositions are given as atom densities in Table IX. Copper, zirconium, and iron are impurities. Note that the N/U atomic ratio is not exactly 2. For the two experiments at 2.26% ^{235}U enrichment (see the last two lines of Table IX), the ratio is 2.19. For the other experiments, it varies from 2.00 to 2.06. Although there is reference to analyses for free acid, none is reported and there is no indication that a ratio other than 2 should be expected.[43] Consultation with the experimenters revealed that the 2.19 ratio was due to an inconsistently low uranium which persisted upon repetition. They did not pursue the matter further.

Uranium Metal Sphere

The critical mass of a water-reflected sphere of uranium enriched to 97.67% in ^{235}U has recently been measured.[44] The sphere was formed by two hemispheres and with corrections to ideal conditions would have attained a value of k_{eff} = 1.0003 ± 0.0005 at 16.3°C. These data are quite precise and provide a very useful benchmark. The composition is reported in terms of atom densities and the critical conditions are given in Table X.

Uranium Metal with Reduced Enrichment

Several critical experiments have been done with bare cylinders containing uranium of ^{235}U content ranging from 10.9 to 53.3% by weight.[45] These reduced enrichments were achieved by interleaving plates of natural uranium and of uranium enriched to ~93% in ^{235}U. The data have been tabulated in a form suitable for computer code input.[40,41] Besides these critical experiments, exponential experiments have been done with bare 21-in.-diam cylinders, having average enrichments of 9.12, 6.53, and 4.29% ^{235}U, also formed by stacking plates of natural and highly enriched uranium.[46-48] Measured axial bucklings are given in Table XI.

The minimum critical enrichment of pure uranium, consisting only of the ^{235}U and ^{238}U isotopes, has been proposed as a benchmark for fast reactor calculations.[49] As the result of studies in France, the Federal Republic of Germany, and the United Kingdom of five experimental lattices with minimum

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[44] C. C. BYERS, J. J. KOELLING, G. E. HANSEN, and D. R. SMITH, *Trans. Am. Nucl. Soc.*, **27**, 412 (1977).

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

[46] C. G. CHEZEM and R. G. STEINKE, *Nucl. Sci. Eng.*, **31**, 549 (1968).

[47] R. G. STEINKE, "Spectral Indices of a 4.29% ^{235}U Enriched Uranium Metal Exponential Column," LA-3406-MS, Los Alamos Scientific Laboratory (1965).

[48] R. G. STEINKE, "Neutron Flux Measurements in Uranium Metal Exponential Columns of 6.53 and 9.12 Percent U-235," LA-3934, Los Alamos Scientific Laboratory (1968).

[49] M. DARROUZET, J. P. CHAUDAT, E. A. FISCHER, G. INGRAM, J. E. SANDERS, and W. SCHOLTYSSEK, "Studies of Unit K_∞ Lattices in Metallic Uranium Assemblies Zebra 8H, Sneak 8, Ermine and Harmonie UK," *Proc. Int. Conf. Physics of Fast Reactors*, October 16-23, 1973, Power Reactor and Nuclear Fuel Development Corporation (1973).

TABLE IX

Composition in atom/b·cm of Uranyl Nitrate–Polyethylene Mixtures Required to Make *k* Unity*

$H \times 10^2$	$^{235}U \times 10^5$	$^{238}U \times 10^3$	$N \times 10^3$	$O \times 10^2$	$C \times 10^3$	$B^a \times 10^7$	$Cu \times 10^5$	$Zr \times 10^5$	$Fe \times 10^6$
2.1005	7.1306	3.2298	6.6004	3.2665	4.2161	1.78 ± 0.86	3.9243	9.6649	8.7782
2.1005	7.1306	3.2298	6.6004	3.2665	4.2161	1.71 ± 1.28	3.9243	9.6649	8.7782
2.8087	5.8888	2.6522	5.5901	2.7337	8.9650	−3.53 ± 6.48	5.8740	14.508	8.2363
2.8210	5.8888	2.6522	5.5901	2.7337	8.9650	−3.47 ± 6.48	5.8740	14.508	8.2363
2.8210	5.8888	2.6522	5.5901	2.7337	8.9650	−3.24 ± 6.30	5.8740	14.508	8.2363
2.5603	6.5348	2.9610	6.0854	3.0592	6.5057	9.02 ± 0.59	3.5477	8.9581	10.091
2.5603	6.5348	2.9610	6.0854	3.0592	6.5057	8.69 ± 1.63	3.5477	8.9581	10.091
2.2177	6.6982	3.0261	6.3749	3.1259	5.1203	4.02 ± 0.80	6.7562	16.472	10.249
2.4969	6.9419	2.9571	6.6326	3.2520	5.8897	33.3 ± 0.2	4.9986	8.5462	7.7551
3.0341	6.2063	2.6470	5.9390	2.9112	9.2700	25.9 ± 0.9	4.2507	7.6982	7.2543

*The experiments were performed in the PCTR.

[a]Derived from the reported B/^{235}U atomic ratio required to make *k* unity.

diluents and with enrichments such that k_∞ was close to unity, an enrichment of 5.66 ± 0.02 at.% (5.59 wt%) has been derived as a benchmark.[50]

CORRELATIONS

Correlations of the three methods of calculation were made with the various experiments to establish bias, although not all methods were applied to all experiments. The effects of container walls were included in the calculations. The S_n methods were generally extrapolated to S_∞. The correlations are expressed in terms of the values of k_{eff} calculated for the experiments. Where the experiments were not exactly critical, these values of k_{eff} were adjusted so that the bias is always (k_{eff} − 1).

Aqueous Solutions

Correlations with the ORNL sphere experiments are given in Table XII in the same order as the experiments are listed in Table I (except that the two samples at 43°C were averaged). The H/^{235}U atomic ratios were computed from concentrations and density formulas, and generally differ slightly from reported values. In MGBS, UO_2F_2 was mocked up by UO_4 since fluorine is not in its cross-section library. The principal experimental uncertainty appears to have been in concentration, and the effect, as investigated with HRXN-ANISN, is greatest at low concentration. At H/^{235}U = 76, a 1% increase in uranium concentration increases k_{eff} by 0.0003, whereas at H/^{235}U = 1836 a 0.5% increase increases k_{eff} by 0.0024. The 3.6% difference in concentration

TABLE X

Critical Water-Reflected Sphere of Uranium Metal

Temperature, 16.3°C

Dimensions			
Core Radius, cm		Reflector Thickness, cm	
6.5537		∞	
Composition, atom/b·cm			
^{234}U	0.00053	H	0.06679
^{235}U	0.04703	O	0.03340
^{236}U	0.00010		
^{238}U	0.00049		

TABLE XI

Exponential Experiments with Bare, 26.67-cm-Radius Uranium Cylinders*

^{235}U Content,[a] wt%	Axial Buckling, B_H^2, cm^{-2}
9.12	−0.00171 ± 0.00009
6.53	−0.00489 ± 0.00030
4.29	−0.00878 ± 0.00045

*The reported density was 18.72 g/cm^3. This density was assumed to apply to natural uranium and was adjusted for the isotopic composition.

[a]The remainder was assumed to be ^{238}U.

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[50]The author is indebted to Gordon Hansen for pointing out this reference.

TABLE XII

Values of k_{eff} Calculated for the Critical Spherical Aqueous Uranium Solutions of Table I

Atomic Ratio, H/^{235}U	Calculated k_{eff}		
	HRXN-ANISN (S_∞)	GLASS-ANISN (S_∞)	MGBS-TGAN
35.7	0.9750	1.0318	1.0369
47.2	0.9759	1.0336	1.0397
49.6	0.9793	---	1.0426
76.1	0.9756	1.0333	1.0407
127.1	0.9739	1.0296	1.0364
270.0	0.9802	1.0276	1.0300
264.2	0.9806	---	---
245.7	0.9778	---	---
239.0	0.9754	---	---
524.3	0.9908	1.0235	1.0191
512.2	0.9924	---	---
505.3	0.9922	---	---
496.2	0.9929	---	---
484.2	0.9886	---	---
467.2	0.9962	---	---
204.2	0.9701	1.0283	1.0417
425.2	0.9807	1.0233	1.0240
491.4	0.9856	1.0139	0.9993
1263	1.0061	1.0085	0.9960
1107	1.0039	1.0139	0.9984
1099	1.0067	1.0010	0.9945
1004	0.9996	0.9992	0.9893
1393	1.0061	1.0055	0.9905
1378	1.0027	1.0028	0.9873
1176	1.0022	1.0026	0.9875
1032	0.9983	0.9989	0.9842
970.7	1.0003	1.0008	0.9864
1836	1.0105	0.9968	0.9898

of the two samples at 87.5°C in the 17.0-ℓ sphere results in a difference in k_{eff} of 0.0076. Correlations with the British sphere experiments of Table II are given in Table XIII.

Correlations with the slab experiments of Table III were made with HRXN-ANISN and GLASS-ANISN, supplemented by SPBL. For each dimension, k_{eff} was calculated with the other dimensions assumed to be infinite. The geometric bucklings corresponding to each of these values of k_{eff} were then calculated and, finally, k_{eff} for the experiment was determined from this combined geometric buckling. The results of the correlations are given in Table XIV, along with calculated values of the transverse buckling. The variation in height has little effect on the neutron multiplication factor. (The top was assumed to be bare in the calculations.) The reported 0.025-cm uncertainty in solution thickness was calculated to correspond to an uncertainty in k_{eff} of ~0.0014. The values of k_{eff} extrapolated to zero transverse buckling were obtained from linear least-squares fits to the values of k_{eff} as a function of transverse buckling.

Correlations with the cylinder experiments of Table IV were made with HRXN-ANISN and GLASS-ANISN, supplemented by SPBL. The quadrature was S_{16}. Differences between S_{16} and S_∞ were considered insignificant in comparison with experimental uncertainties and those associated with extrapolation to infinite cylinders. Correlations with the cylinder experiments were also made with MGBS-TGAN. The procedure followed in obtaining k_{eff} for infinite cylinders was essentially the same as for the slabs, except that k_{eff} was plotted as a function of axial buckling and was extrapolated graphically to zero axial bucklings. The extrapolated values and

TABLE XIII

Values of k_{eff} Calculated for the Critical Spherical Aqueous Uranium Solutions of Table II

Atomic Ratio, $H/^{235}U$	Calculated k_{eff}		
	HRXN-ANISN (S_∞)	GLASS-ANISN (S_∞)	MGBS-TGAN
367.0	0.9780	1.0262	1.0272
636.6	0.9822	1.0151	1.0064
76.84	0.9753	1.0321	1.0310
218.3	0.9847	1.0315	1.0365
534.3	0.9889	1.0198	1.0149
1040	1.0051	1.0141	0.9999
352.2	0.9975	1.0314	1.0275
574.3	0.9934	1.0181	1.0122
784.6	0.9998	1.0160	1.0069
1195	1.0072	1.0090	0.9976

TABLE XIV

Values of the Transverse Buckling and of k_{eff} Calculated for the Critical Aqueous Uranium Solution Slabs of Table III

Atomic Ratio, $H/^{235}U$	B_r^2, cm^{-2}		k_{eff} (S_∞)	
	HRXN-ANISN	GLASS-ANISN	HRXN-ANISN	GLASS-ANISN
44.7	0.00187	0.00189	0.9933	1.0352
	0.00142	0.00144	0.9918	1.0332
	0.00141	0.00143	0.9919	1.0333
	0.00114	0.00116	0.9874	1.0282
	0.00112	0.00113	0.9879	1.0288
	0.00105	0.00107	0.9880	1.0289
	0.00101	0.00103	0.9889	1.0294
	0	0	0.9808	1.0200
51.3	0.00134	0.00135	0.9904	1.0319
	0.00130	0.00132	0.9910	1.0324
	0.00094	0.00095	0.9869	1.0278
	0.00093	0.00094	0.9870	1.0280
	0	0	0.9780	1.0176

the values calculated for the finite cylinders are given in Table XV in the same order as the data are listed in Table IV. The $H/^{235}U$ values given are those calculated from solution concentrations at 23°C and in most cases differ slightly from reported values.

It is apparent from a study of Table XV that extrapolations to zero axial buckling are often large, in some cases too large to be attempted, and that the extrapolated values are somewhat uncertain and subject to the judgment of the extrapolator. However, there appears to be no reason to expect the bias (k_{eff} − 1) derived from correlations with sphere experiments not to be applicable to infinite cylinders or to expect bias derived from correlation with solutions of UO_2F_2 not to be applicable to solutions of $UO_2(NO_3)_2$.

The former conclusion is supported by examining the subcritical cases in Table V and, on the basis of sphere correlations, making judgments concerning the critical heights estimated by the experimenters. With HRXN cross sections, there was disagreement between this analysis and the experiment report whether the cylinder would be subcritical at any height in three cases. These were at uranium concentrations of 774.95, 421.80, and 227.31 g/ℓ, corresponding to $H/^{235}U$ = 31.5, 62.0, and 119, respectively. There was uncertainty in the agreement

TABLE XV

Values of the Axial Buckling and of k_{eff} Calculated for the Critical Cylindrical Aqueous Uranium Solutions of Table IV

Atomic Ratio H/^{235}U	B_h^2, cm^{-2}			k_{eff}		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
503.5	0	0	0	0.987	1.009	1.000
	0.00032	0.00034	0.00029	0.9868	1.0084	0.9995
	0.00045	0.00046	0.00043	0.9894	1.0110	1.0009
	0.00134	0.00135	0.00138	0.9923	1.0143	1.0002
	0.00323	0.00328	0.00338	0.9986	1.0215	1.0004
27.0	0	0	0	~0.978	~1.030	1.042
	0.00091	0.00092	0.00095	0.9838	1.0347	1.0379
	0.01383	0.01437	0.01496	0.9984	1.0504	1.0048
	0.02140	0.02251	0.02179	0.9814	1.0293	1.0007
526.5	0	0	0	0.987	~1.009	1.001
	0.00290	0.00294	0.00303	0.9983	1.0190	1.0000
	0.00251	0.00254	0.00258	0.9976	1.0187	1.0022
	0.00577	0.00587	0.00589	1.0020	1.0228	1.0031
31.5	0	0	0	0.973	1.027	1.041
	0.00244	0.00247	0.00259	0.9925	1.0468	1.0411
	0.00420	0.00427	0.00452	1.0034	1.0581	1.0420
	0.00731	0.00746	0.00789	1.0175	1.0725	1.0424
	0.00981	0.01007	0.01055	1.0266	1.0814	1.0438
647.5	0	0	0	0.996	~1.009	1.001
	0.00130	0.00131	0.00131	0.9988	1.0131	1.0015
	0.00456	0.00462	0.00463	1.0048	1.0189	1.0025
736.6	0	0	0	0.997	~1.006	1.000
	0.00040	0.00041	0.00037	0.9980	1.0076	1.0000
	0.00367	0.00371	0.00373	1.0047	1.0144	1.0002
44.2	0	0	0	0.973	1.029	1.041
	0.00059	0.00059	0.00058	0.9781	1.0326	1.0411
	0.00343	0.00349	0.00382	0.9963	1.0503	1.0368
44.2	0	0	0	~0.973	~1.031	1.044
	0.00137	0.00138	0.00146	0.9846	1.0378	1.0404
	0.00911	0.00935	0.01027	1.0052	1.0603	1.0198
	0.01419	0.01468	0.01560	1.0091	1.0632	1.0143
44.2	0	0	0	0.981	1.036	1.044
	0.00261	0.00264	0.00280	0.9940	1.0493	1.0438
	0.00450	0.00456	0.00488	1.0039	1.0599	1.0435
	0.00774	0.00789	0.00843	1.0176	1.0739	1.0431
	0.01001	0.01025	0.01085	1.0321	1.0879	1.0427
49.1	0	0	0	---	---	~1.044
	0.00905	0.00927	0.01022	1.0082	1.0634	1.0229
	0.01418	0.01465	0.01563	1.0119	1.0662	1.0169
995.1	0	0	0	1.002	1.000	0.995
	0.00106	0.00106	0.00105	1.0047	1.0029	0.9955
52.6	0	0	0	0.976	1.030	1.045
	0.00135	0.00136	0.00144	0.9858	1.0392	1.0426
	0.00339	0.00344	0.00378	0.9986	1.0529	1.0405
	0.00869	0.00889	0.00984	1.0152	1.0704	1.0295
	0.01334	0.01377	0.01481	1.0275	1.0817	1.0301

(Continued)

TABLE XV (Continued)

Atomic Ratio $H/^{235}U$	B_h^2, cm^{-2}			k_{eff}		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
61.7	0	0	0	0.970	1.027	1.041
	0.00257	0.00260	0.00276	0.9925	1.0483	1.0444
	0.00742	0.00756	0.00813	1.0238	1.0803	1.0499
	0.01002	0.01024	0.01097	1.0341	1.0902	1.0511
	0.01230	0.01262	0.01331	1.0380	1.0937	1.0519
62.0	0	0	0	~0.975	~1.030	~1.03
	0.00616	0.00627	0.00697	1.0102	1.0632	1.0327
	0.00854	0.00873	0.00965	1.0210	1.0739	1.0329
	0.01046	0.01072	0.01173	1.0290	1.0814	1.0352
	0.01880	0.01953	0.01921	0.9985	1.0479	1.0145
78.7	0	0	0	---	---	~1.05
	0.00303	0.00306	0.00308	0.9979	1.0522	1.0423
	0.00891	0.00910	0.00984	1.0080	1.0634	1.0231
240	0	0	0	0.976	1.025	~1.032
	0.00390	0.00394	0.00434	1.0008	1.0466	1.0271
	0.00662	0.00671	0.00743	1.0115	1.0577	1.0237
	0.00895	0.00912	0.01001	1.0162	1.0621	1.0204
	0.01269	0.01301	0.01390	1.0162	1.0615	1.0165
	0.01614	0.01662	0.01720	1.0178	1.0614	1.0219
350	0	0	0	0.980	1.025	~1.024
	0.00424	0.00429	0.00468	1.0056	1.0453	1.0224
	0.00641	0.00651	0.00710	1.0149	1.0546	1.0215
	0.00962	0.00982	0.01054	1.0274	1.0664	1.0251
	0.01376	0.01411	0.01464	1.0169	1.0551	1.0173
483	0	0	0	~0.975	~1.011	~1.015
	0.00395	0.00400	0.00429	1.0046	1.0374	1.0155
	0.00711	0.00722	0.00771	1.0202	1.0527	1.0190
	0.01079	0.01103	0.01147	1.0200	1.0518	1.0164
495	0	0	0	---	---	~1.019
	0.00426	0.00432	0.00451	1.0042	1.0375	1.0145
	0.01164	0.01190	0.01216	1.0093	1.0417	1.0074
728	0	0	0	~0.990	~1.015	~1.005
	0.00316	0.00319	0.00335	1.0046	1.0263	1.0069
	0.00647	0.00657	0.00681	1.0145	1.0359	1.0083

in four additional cases at uranium concentrations of 930.19, 157.99, 123.76, and 36.75 g/ℓ, corresponding to $H/^{235}U$ = 25.4, 173, 222, and 755, respectively. With GLASS cross sections, the cylinder at $H/^{235}U$ = 119 moved from "disagreement" to "uncertainty" and the cylinder at $H/^{235}U$ = 222 moved from uncertainty to agreement. In only one case was there doubt as to the subcriticality of a cylinder at the maximum attainable height, and there the estimated height only slightly exceeded the attainable height. The projections based on correlations with sphere experiments thus tend to be more conservative than those of the experimenters.

Correlations with the experiments of Table VI were made with all three methods. For these large, bare cylinders, errors in the effective geometric bucklings for the finite cylinders derived by SPBL or by TGAN should have little effect. The ANISN calculations were made with S_4 quadrature, which is adequate at the low concentrations involved. The correlations are given in Table XVI.

The correlations of Tables XII through XVI are plotted against the $H/^{235}U$ atomic ratio in Figs. 1, 2, and 3 for the HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN codes, respectively. The $H/^{235}U$ atomic ratio has a large bearing on the neutron spectrum and hence is an important parameter. However, it is not necessarily the only parameter

TABLE XVI

Values of k_{eff} Calculated for the Critical Cylindrical Aqueous Uranium Solutions of Table VI

Atomic Ratio H/^{235}U	Calculational Method		
	HRXN-ANISN	GLASS-ANISN	MGBS-TGAN
1602	1.0090	1.0011	0.9892
1632	1.0089	1.0002	0.9888
1819	1.0132	0.9990	0.9928
1903	1.0150	0.9986	0.9950
1978	1.0122	0.9942	0.9937
1952	1.0110	0.9936	0.9924
1996	1.0127	0.9943	0.9950
2047	1.0138	0.9940	0.9978

of which bias is a function. The various correlations were accordingly inspected for other trends that appeared significant. As has already been stated, the bias for spheres seems appropriate for infinite cylinders, and there seems to be no dependence on whether the solution is UO_2F_2 or $UO_2(NO_3)_2$. Likewise, there seems to be no dependence on whether a sphere is bare or water reflected. When calculated by HRXN-ANISN, the slab and sphere experiments are in good agreement but they are not when calculated by GLASS-ANISN. With the HRXN-ANISN codes, there seems to be no dependence of k_{eff} on the ^{235}U enrichment of the uranium, but with GLASS-ANISN and MGBS-TGAN, the values of k_{eff} for ^{235}U enrichments of ~5% lie consistently below those for uranium of high ^{235}U content. The curves in the

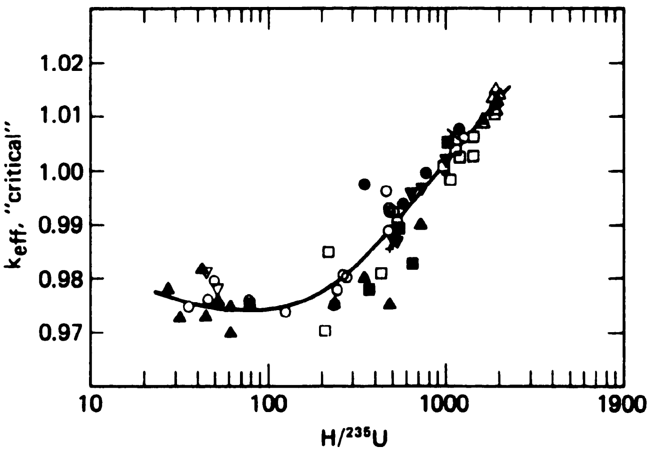

Fig. 1. The k_{eff} of volumes of aqueous solutions of ^{235}U-enriched uranium salts as a function of the H/^{235}U atomic ratio calculated by the HRXN-ANISN (S_{∞}) codes. The line is an "eyeball" fit.

- □ Bare sphere, >90% ^{235}U content, ORNL, Tables I and XII
- ○ Water-reflected sphere, >90% ^{235}U content, ORNL, Tables I and XII
- + Bare sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- X Water-reflected sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- △ Bare cylinder, >90% ^{235}U content, ORNL, Tables VI and XVI
- ■ Bare sphere, British, Tables II and XIII
- ● Water-reflected sphere, British, Tables II and XIII
- ▽ Water-reflected slab, >90% ^{235}U content, Tables III and XIV
- ▲ Water-reflected cylinder, >90% ^{235}U,content, ORNL, Tables IV and XV
- ▼ Water-reflected cylinder, ~5% ^{235}U content, ORNL, Tables IV and XV

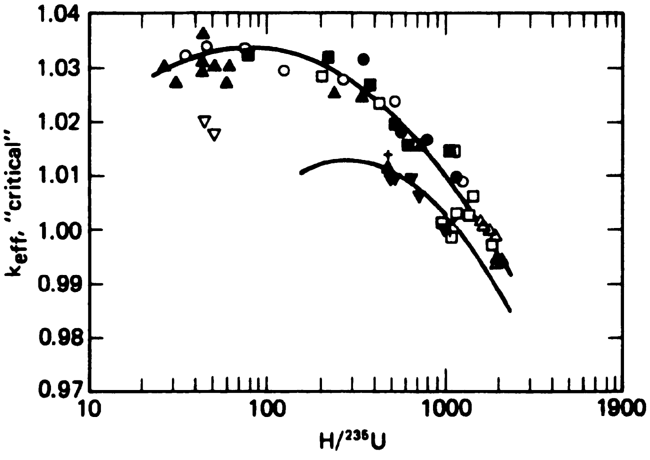

Fig. 2. The k_{eff} of volumes of aqueous solutions of ^{235}U-enriched uranium salts as a function of the H/^{235}U atomic ratio calculated by the GLASS-ANISN (S_{∞}) codes. The lines are "eyeball" fits; the lower curve represents uranium of ^{235}U content ~5%.

- □ Bare sphere, >90% ^{235}U content, ORNL, Tables I and XII
- ○ Water-reflected sphere, >90% ^{235}U content, ORNL, Tables I and XII
- + Bare sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- X Water-reflected sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- △ Bare cylinder, >90% ^{235}U content, ORNL, Tables VI and XVI
- ■ Bare sphere, British, Tables II and XIII
- ● Water-reflected sphere, British, Tables II and XIII
- ▽ Water-reflected slab, >90% ^{235}U content, Tables III and XIV
- ▲ Water-reflected cylinder, >90% ^{235}U,content, ORNL, Tables IV and XV
- ▼ Water-reflected cylinder, ~5% ^{235}U content, ORNL, Tables IV and XV

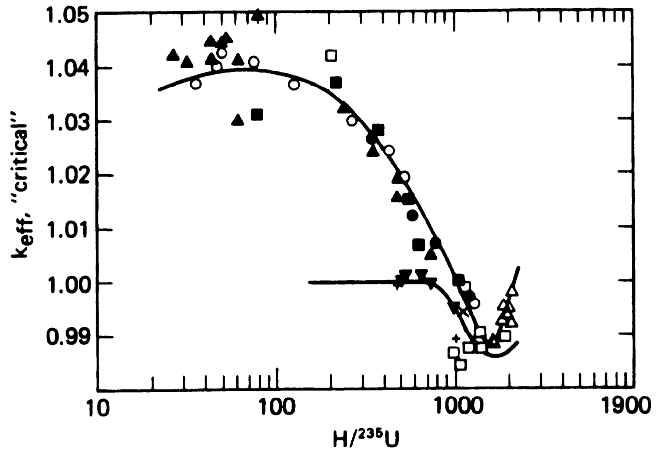

Fig. 3. The k_{eff} of volumes of aqueous solutions of ^{235}U-enriched uranium salts as a function of the $H/^{235}U$ atomic ratio calculated by the MGBS-TGAN codes. The lines are "eyeball" fits; the lower curve represents uranium of ^{235}U content ~5%.

- □ Bare sphere, >90% ^{235}U content, ORNL, Tables I and XII
- ○ Water-reflected sphere, >90% ^{235}U content, ORNL, Tables I and XII
- + Bare sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- X Water-reflected sphere, ~5% ^{235}U content, ORNL, Tables I and XII
- △ Bare cylinder, >90% ^{235}U content, ORNL, Tables VI and XVI
- ■ Bare sphere, British, Tables II and XIII
- ● Water-reflected sphere, British, Tables II and XIII
- ▲ Water-reflected cylinder, >90% ^{235}U content, ORNL, Tables IV and XV
- ▼ Water-reflected cylinder, ~5% ^{235}U content, ORNL, Tables IV and XV

figures are "eyeball" fits to the data and are weighted most heavily by the reflected spheres. For the results from GLASS-ANISN and MGBS-TGAN, separate curves are drawn for high and low enrichment.

Homogeneous Hydrogenous Mixtures

Correlations of results from the HRXN, GLASS, and MGBS codes with the experiments at the PCTR on UO_3-H_2O mixtures and reported in Table VII are given in Table XVII in terms of the values of k (at zero buckling and hence equal to k_{eff}) calculated for the compositions experimentally determined to make k unity. The $H/^{235}U$ atomic ratios are given so that the results can be placed in Figs. 1, 2, and 3. The HRXN values lie substantially above the curve, particularly at the lower $H/^{235}U$ atomic ratios. The GLASS and MGBS results lie below the curve and appear consistent with the trend with enrichment in Figs. 2 and 3.

The HRXN and GLASS correlations differ from previously reported results.[40,41] In the case of HRXN, this difference is greater at low atomic ratios and is probably due to the ^{238}U cross sections now accompanying KENO-IV differing from previous values and to the treatment given to them. In the case of GLASS, the previous results were obtained with cross sections older than those of ENDF/B-IV used in the present work, and the differences in the neutron multiplication factor are substantial (~0.02).

Examination of the correlations shows a dependence of k_{eff} on both the ^{235}U enrichment and the hydrogen content (H/U atomic ratio) of these test mixtures of uranium with low ^{235}U content. The enrichment dependence is in the same direction with all three codes, with k_{eff} increasing with enrichment. This is the direction of the trend shown by comparison of the values of the k_{eff} calculated for highly enriched uranium by the GLASS and MGBS codes, but opposite to that by the HRXN code. The enrichment range of the experiments is small, and the trend among the experiments appears almost to be within experimental error. It was concluded that the trend is probably not real and that extrapolation of the trend outside the range of the experimental data is not required. Limits, however, should allow for this possible dependence and should not be values that extrapolation would predict to be critical.

Linear least-squares fits to the values of k_{eff}, obtained by the three codes, as a function solely of H/U; give the following expressions:

$$\text{HRXN: } k_{eff} = 1.0463 - 0.004514(H/U)$$

$$\text{GLASS: } k_{eff} = 0.9905 + 0.0001626(H/U)$$

$$\text{MGBS: } k_{eff} = 0.9738 + 0.0018121(H/U).$$

(The biases are these values of k_{eff} minus unity.)

Correlations of the analyses by the HRXN, GLASS, and MGBS codes with the experimental values at 3.04% ^{235}U enrichment, as listed in Table VIII, are given in Table XVIII. Again, the $H/^{235}U$ atomic ratios are reported so that comparison can be made with Figs. 1, 2, and 3. The HRXN calculations support the experimental observation that k is independent of whether the H/U atomic ratio is achieved with water or with polyethylene, but this is not the case with GLASS as demonstrated by the results obtained with $UO_2(NO_3)_2$ mixtures. The HRXN results lie substantially above the curve of Fig. 1, generally further above than do the results near 1% ^{235}U enrichment (Table XVII). The GLASS and MGBS results are more nearly consistent with

TABLE XVII

Values of k Calculated by the Three Codes for the UO_3-H_2O Mixtures of Table VII

Atomic Ratio H/^{235}U	HRXN		GLASS		MGBS	
	k	Δk^a	k	Δk^a	k	Δk^a
376.4	1.0236	-0.0054	0.9868	-0.0043	0.9750	-0.0057
497.4	1.0205	-0.0030	0.9900	-0.0013	0.9814	-0.0016
612.1	1.0127	-0.0055	0.9876	-0.0039	0.9808	-0.0043
682.6	1.0079	-0.0070	0.9857	-0.0059	0.9795	-0.0069
738.6	1.0109	-0.0015	0.9908	-0.0009	0.9848	-0.0026
349.2	1.0306	0.0014	0.9921	0.0010	0.9812	0.0005
538.9	1.0227	0.0028	0.9948	0.0034	0.9884	0.0040
659.2	1.0163	0.0022	0.9938	0.0022	0.9886	0.0019
323.3	1.0342	0.0050	0.9939	0.0028	0.9846	0.0039
511.1	1.0245	0.0052	0.9960	0.0046	0.9910	0.0063
588.9	1.0156	0.0005	0.9907	-0.0009	0.9869	0.0006
641.0	1.0172	0.0048	0.9948	0.0031	0.9911	0.0037

[a] Deviation of k from a least-squares fit to a linear dependence of k on H/U, i.e., $\Delta k = k_{calc} - k_{least\text{-}squares}$.

the curves in Figs. 2 and 3, respectively, but tend to rise above the curves as H/^{235}U increases.

Correlations of the results from the HRXN and GLASS codes with the PCTR experiments with $UO_2(NO_3)_2$-polyethylene mixtures near 2% ^{235}U enrichment as listed in Table IX are given in Table XIX. As for the nitrate experiments with uranium at 3.04% enrichment, no correlation of MGBS was made with the experiments since nitrate molarities in an equivalent H_2O-$UO_2(NO_3)_2$ mixture exceed the ~8 *M* limit provided in that code. The uncertainties assigned to the HRXN values of k_{eff} in Table XIX represent the reported uncertainties in the B/^{235}U atomic ratio. The reported uncertainties in the H/U atomic ratio translate into uncertainties in k_{eff} of 0.0005. Except for the two points at 2.26% ^{235}U enrichment, k_{eff} calculated by the HRXN code lies among the values for UO_3-H_2O mixtures at ~1% enrichment and are well above the curve of Fig. 1. With the GLASS code these two points lie among the 1% values and the others lie above the lower curve of Fig. 2. Linear least-squares fits to the correlations yield these relations for k_{eff} from the two codes:

$$\text{HRXN: } k_{eff} = 1.0322 - 0.002015(\text{H/U})$$

$$\text{GLASS: } k_{eff} = 1.0068 - 0.00005257(\text{H/U}).$$

All the data are included despite some doubts concerning the N/U atomic ratio in the mixture containing uranium enriched to 2.26 wt% in the ^{235}U isotope.

Uranium Metal Sphere

Correlations of HRXN-ANISN and GLASS-ANISN were made with the essentially critical water-reflected sphere of uranium metal described in Table X. Values of k_{eff} calculated by the former method with S_4, S_8, and S_{16} quadratures were 1.0089, 0.9989, and 0.9961, respectively. Extrapolation to S_∞ gave 0.9952. With the latter method, the four corresponding values were 1.0229, 1.0132, 1.0103, and 1.0093.

Uranium Metal with Reduced Enrichment

Correlations with the critical experiments on cylinders of uranium metal formed by interleaving natural and highly ^{235}U-enriched uranium plates (Table XI of Ref. 41) and with the exponential experiments with similarly formed cylinders (Table XI) were made with HRXN-ANISN (S_{16})-SPBL and with MGBS. (Attempts were made to correlate with GLASS in place of HRXN, but results were inconsistent and seemed to be related to the handling of unresolved resonances in ^{238}U; the conditions in these experiments, however, are far removed from those GLASS was designed to treat.) In the HRXN correlations with the exponential experiments, ANISN and SPBL were used to obtain the radial buckling; the axial buckling was that measured in the experiments. In the MGBS correlations, the geometric bucklings were calculated from the dimensions and from a bare extrapolation distance, λ, calculated by MGBS as

$$\lambda = \frac{1}{B} \tan^{-1} \frac{B}{\overline{\Sigma}_{tr}} ,$$

where B^2 is the geometric buckling and $\overline{\Sigma}_{tr}$ is the macroscopic transport cross section of uranium obtained by flux weighting $1/\Sigma_{tr}$ as a function of

TABLE XVIII

Values of k Calculated by the Three Codes for the Uranium Mixtures of Table VIII

Atomic Ratio $H/^{235}U$	UO_3				
	HRXN		GLASS[a]		MGBS
	B_4C-CH_2	B_2O_3-H_2O	$^{10}B_4C$-CH_2	$^{10}B_2O_3$-H_2O	B_2O_3-H_2O
116.3	1.0354	1.0348	1.0064	---	0.9798
	1.0335	1.0328	1.0044	---	0.9780
190.4	1.0296	1.0290	1.0108	---	0.9927
	1.0330	1.0324	1.0142	---	0.9960
	1.0292	1.0286	1.0104	---	0.9923
	1.0290	1.0284	1.0102	---	0.9920
207.3	1.0349	1.0342	1.0153	---	1.0009
260.3	1.0314	1.0310	1.0161	---	1.0062
279.4	1.0326	1.0321	1.0194	---	1.0107
323.0	1.0309	1.0302	1.0207	---	1.0143
327.5	1.0339	1.0332	1.0239	---	1.0177
328.8	1.0283	1.0276	1.0183	---	1.0123
401.6	1.0309	1.0300	1.0225	---	1.0193
401.9	1.0311	1.0301	1.0227	---	1.0194
981.3	1.0416	1.0405	1.0250	---	1.0294
1146	1.0415	1.0406	1.0232	---	1.0281
1292	1.0571	1.0563	1.0375	---	1.0426
1301	1.0481	1.0474	1.0284	---	1.0336
1425	1.0590	1.0583	---	---	1.0435
1559	1.0435	1.0430	---	---	1.0273
	$UO_2(NO_3)_2$				
198.2	1.0344	1.0340	1.0250	1.0158	---
286.3	1.0247	1.0239	1.0220	1.0135	---
423.0	1.0213	1.0202	1.0183	1.0106	---
1007	1.0431	1.0421	1.0258	1.0231	---

[a] Since the GLASS code contains the cross sections of ^{10}B and not of natural boron, these calculations assumed the boron of Table VIII to contain 20 at.% ^{10}B.

energy. The thermal and resonance groups were omitted in the MGBS calculations, leaving a ten-group structure, a valid procedure for a bare metallic system since few neutrons are scattered into these groups.

Results of the correlations are given in Table XX. On the basis of SPBL results for other systems, the HRXN results probably overestimate k_{eff} of the critical experiments, especially the first two with the smaller dimensions, but may underestimate k_{eff} of the exponential experiments. The uncertainty in k_{eff} for the critical experiments associated with the reported uncertainty in critical height is ~0.002 (Refs. 40 and 41). The uncertainty associated with homogenization of the stack of plates can be as much as 0.01, but for the cylinder with average enrichment of 10.9%, it appears to be perhaps as small as 0.003 (Ref. 40). In the exponential experiments, this uncertainty seems likely to be insignificant compared with the uncertainty in axial buckling,

TABLE XIX

Values of k Calculated by Two Codes for the Uranyl Nitrate-Polyethylene Mixtures of Table IX

Nominal Enrichment ^{235}U, wt%	Atomic Ratio		HRXN		GLASS	
	H/U	H/^{235}U	k	Δk^a	k	Δk^a
2.14	6.36	294.6	1.0235 ± 0.0009	0.0041	1.0090	0.0026
			1.0236 ± 0.0012	0.0042	1.0090	0.0026
	10.36	447.0	1.0192 ± 0.0072	0.0079	1.0142	0.0080
	10.41	479.0	1.0190 ± 0.0072	0.0078	1.0140	0.0078
			1.0187 ± 0.0070	0.0075	1.0137	0.0075
	8.46	391.8	1.0151 ± 0.0006	-0.0001	1.0078	0.0015
			1.0155 ± 0.0016	0.0003	1.0078	0.0015
	7.17	331.1	1.0153 ± 0.0008	-0.0025	1.0048	-0.0016
2.26	8.25	359.7	0.9989 ± 0.0002	-0.0161	0.9910	-0.0153
	11.20	488.9	0.9973 ± 0.0009	-0.0123	0.9918	-0.0144

[a] Deviation of k from a least-squares fit to a linear dependence of k on H/U, i.e., $\Delta k = k_{\text{calc}} - k_{\text{least-squares}}$.

to which the uncertainties in Table XX correspond. When consideration is given to these uncertainties, agreement between the critical and exponential experiments is fairly good. The result at 6.53% ^{235}U enrichment may be slightly low, however. Agreement with the enrichment benchmark[49] is also fairly good, especially with HRXN. At 5.59 wt% ^{235}U, HRXN gave $k = 0.9921$ at zero buckling and 0.9901 at the critical (negative) buckling. The MGBS code gave, respectively, 1.0030 and 1.0039.

TABLE XX

Values of k_{eff} Calculated for Bare Uranium Cylinders of Various ^{235}U Enrichments

Average ^{235}U Enrichment, wt%	k_{eff}	
	HRXN-ANISN	MGBS
53.33	1.0091	1.0412
37.46	0.9966	1.0233
16.01	0.9815	0.9973
14.11	0.9797	0.9954
12.32	0.9786	0.9943
10.9	0.9761	0.9912
9.12	$0.9809^{+0.0035}_{-0.0035}$	$0.9935^{+0.0041}_{-0.0041}$
6.53	$0.9643^{+0.0142}_{-0.0138}$	$0.9643^{+0.0169}_{-0.0164}$
4.29	$0.9830^{+0.0298}_{-0.0278}$	$0.9615^{+0.0355}_{-0.0331}$

SUBCRITICAL LIMITS

With the three calculational methods, as validated above, limits in the Standard[2] were examined, and, where subcriticality under stated conditions appears doubtful, revised limits are being proposed. Also some extensions of the limits are being proposed for the Standard.

Uranium Solutions

All three methods 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. In the two S_n methods, the quadrature was S_{16} since results are not significantly different from those with S_∞. For full enrichment, i.e., pure ^{235}U, a margin of 0.01 in k_{eff} below the appropriate curve in Figs. 1, 2, and 3 appears sufficient to ensure subcriticality, but, as for plutonium,[1] an additional margin of 0.01 was arbitrarily included making the total margin in "calculated limits" 0.02. However, this margin applied to the most conservative method seems excessively conservative, and, in selecting "proposed limits," a margin of at least 0.01 for the most conservative method was usually selected. Where agreement between methods was good and little extension beyond experimental conditions was involved, as in the case of the mass and the diameter of an infinitely long cylinder of UO_2F_2 solutions, a margin as small as 0.01 was occasionally considered acceptable, even though with no method was the margin as great as 0.02.

Critical parameters, corresponding to values of k_{eff} read from Figs. 1, 2, and 3, were first calculated for UO_2F_2 solutions. Results obtained by the three methods are given in Table XXI where they are compared with other calculations. The minimum values listed were obtained graphically from plots of the parameter of interest as a function of the uranium concentration. The slab thickness barely shows a minimum. With GLASS-ANISN, it was still decreasing slightly at 5 *M* (66% UO_2F_2). However, since this solution is saturated,[6] extension to higher concentrations is pointless. Calculations by Webster[51] served as the basis for the American National Standards Institute (ANSI) Standard.[2] His sphere and slab calculations are S_6, his cylinder calculations S_8. He used Hansen-Roach cross sections,[52] presumably with the *DE/E* set for hydrogen, judging from his correlation with experiment. His calculations are not normalized to experiment, but he cites six correlations and concludes that his computed values of k_{eff} are accurate to about ±1%. However, except for the slab experiment,[33] his correlations show a trend toward higher values of k_{eff} with increasing $H/^{235}U$, and he appears not to have allowed for the Unichrome coating on the 17-ℓ sphere.[13,29] He fails to state how his solution densities were computed. The values of Paxton et al.[32] are read from curves and hence are somewhat approximate. Moreover, the curves are stated to be for uranium enriched to 93.2% rather than 100% ^{235}U. Reference 53 is a study of the effect of concentration distribution from which some of the limits in the Standard were drawn.

Agreement among the methods is fairly good, but MGBS-TGAN gives somewhat smaller critical dimensions. The GLASS-ANISN method gives a larger critical slab thickness than HRXN-ANISN, but this is not unexpected since the GLASS-ANISN correlations with the slab experiments[33] fall below the curve of Fig. 2. Why there should apparently be this shape dependence in the bias is not known.

The conditions of Table XXI depart slightly from those of the experiments that established the biases of the methods of calculations. The ^{235}U enrichment has been increased from ~93 to 100%, and there is no aluminum wall interposed between the solutions and the water reflector. Some calculations were therefore made to see whether these departures from experimental conditions might account for some of the descrepancies in the results, particularly the smaller critical dimensions calculated by MGBS-TGAN. The extension of the isotopic composition from approximately that of most of the experiments to 100% ^{235}U had differing effects on dimensions and volume for all three methods, but MGBS-TGAN computed the greatest decrease only for cylinder diameter and there only slightly (0.02 cm) greater than GLASS-ANISN. The magnitudes of the decreases in volume ranged from 0.34 to 0.43 ℓ, in cylinder diameter from 0.34 to 0.45 cm, and in slab thickness from 0.23 to 0.33 cm. Decreases in minimum critical mass ranged from 13 to 15 g. In the HRXN-ANISN and GLASS-ANISN calculations, interposition of a 0.16-cm-thick aluminum wall increased the critical volume by 0.02 to 0.04 ℓ, increased the cylinder diameter by 0 to 0.02 cm, and decreased the slab thickness by 0.02 to 0.03 cm; but with MGBS-TGAN the volume was increased by 0.25 ℓ, the cylinder diameter by 0.12 cm, and the slab thickness by 0.06 cm. The large effect of the wall in the MGBS calculations is likely due to a fictitious neutron streaming in the aluminum associated with the fictitious transverse leakage introduced to achieve criticality. Since walls were present in the experiments with which comparisons were made, the MGBS-TGAN results without the container wall are conservatively too small.

TABLE XXI

Minimum Critical Parameters of Aqueous Solutions of UO_2F_2 Containing 100% ^{235}U

Dimension	HRXN-ANISN	GLASS-ANISN	MGBS-TGAN	Webster (Ref. 51)	Paxton et al.[a] (Ref. 32)	Clark (Ref. 53)
Mass ^{235}U, g	806	800	805	820	810	815
Cylinder diameter, cm	14.06	14.13	13.79	14.3	14.2	
Slab thickness, cm	4.64	4.83	4.47	4.9	4.6	
Volume, ℓ	5.83	5.80	5.65	6.1	6.4	
Uranium concentration, g/ℓ	12.08	12.07	12.17	11.8		
H/U, atomic ratio	2153	2154	2137			
Areal density, g/cm²	0.427	0.430	0.420	0.42		0.421

[a]The results of Paxton et al. (Ref. 32) are for uranium enriched to 93.2 wt% in the ^{235}U isotope.

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

[52] 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).

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

A similar effect was found for the minimum critical mass where the presence of the wall increased the mass by 6 g ^{235}U with HRXN-ANISN, 4 g with GLASS-ANISN, but by 13 g with MGBS-TGAN.

Calculated limits, i.e., minimum values of parameters corresponding to a margin of 0.02 in k_{eff} below the curves of Figs. 1, 2, and 3, are given in Table XXII together with limits now in the Standard and the proposed limits for the revised Standard. As pointed out in connection with plutonium limits,[1] due to the difference in the manner in which k_{eff} is computed, a margin of 0.02 produces a greater change in a parameter based on the MGBS-TGAN codes than on the other methods.

Similar calculations were made for uranyl nitrate solutions. Calculations were extended to sufficiently high concentrations to determine the minimum infinite slab thickness, which occurs with an approximately saturated solution[7] (~2.5 *M*). Calculated limits are given in Table XXIII. The difference in the calculated concentration limits between Tables XXII and XXIII is trivial. Accordingly, the limits proposed for both the fluoride and nitrate solutions are the same. The limits on the H/U atomic ratio and on the areal density of $UO_2(NO_3)_2$ are not very practical. The former is useful as a lower limit of the ratio for homogeneous hydrogenous nonaqueous mixtures where the uranium compound is likely not uranyl nitrate. The latter is useful as a secondary limit where precipitation may occur and where the compound is likely not uranyl nitrate.

Limit calculations for aqueous solutions of UO_2F_2 and $UO_2(NO_3)_2$ were extended to enrichments below 10% ^{235}U. Since the Standard requires maintenance of uniform aqueous solutions, the calculations were not carried to concentrations greater than those of saturated solutions. [Saturated solutions are assumed to be 5 *M* UO_2F_2 and 2.5 *M* $UO_2(NO_3)_2$.] Most of the limits occur at these concentrations. The areal density is less meaningful for solutions at these lower enrichments since its minimum occurs at high concentrations where the precipitate becomes important in determining mixture densities. The calculations were made for ^{235}U enrichments ranging from 1.5 to 10%. The uranium was assumed to consist solely of ^{235}U and ^{238}U. Replacing ^{238}U by ^{234}U or ^{236}U would increase the margin of subcriticality because

TABLE XXII

Limiting Specifications for Uniform Homogeneous Aqueous Solutions of UO_2F_2 Containing 100% ^{235}U

Dimension	Present Standard (Ref. 2)	Calculational Method			Proposed Standard
		HRXN-ANISN	GLASS-ANISN	MGBS-TGAN	
Mass, g	760	738	730	720	760
Cylinder diameter, cm	13.9	13.6	13.66	13.16	13.7
Slab thickness, cm	4.6	4.33	4.51	4.04	4.4
Volume, ℓ	5.8	5.36	5.32	5.05	5.5
Uranium concentration, g/ℓ	11.5	11.63	11.58	11.74	11.6
H/U, atomic ratio		2237	2246	2216	2250
Areal density, g/cm^2	0.4	0.407	0.408	0.392	0.4

TABLE XXIII

Limiting Specifications for Uniform Homogeneous Aqueous Solutions of $UO_2(NO_3)_2$ Containing 100% ^{235}U

Dimension	Calculational Method			Proposed Standard
	HRXN-ANISN	GLASS-ANISN	MGBS-TGAN	
Mass, g	757	751	742	780
Cylinder diameter, cm	14.41	14.59	14.12	14.4
Slab thickness, cm	4.9	5.19	4.67	4.9
Volume, ℓ	6.19	6.25	5.99	6.2
Uranium concentration, g/ℓ	11.67	11.62	11.77	11.6
H/U, atomic ratio	2225	2234	2206	2250
Areal density, g/cm^2	0.411	0.412	0.395	0.4

of their larger cross sections. The single curve of Fig. 1 and the lower curves of Figs. 2 and 3, representing uranium of ~5% ^{235}U content, were assumed for the calculations. The results of the PCTR experiments appear to give no reason for suspecting that the use of these curves might be nonconservative. Because of the greater uncertainty in these calculations, the margin selected in determining limits was not much less than 0.02 for the results by the most conservative method. Limits for UO_2F_2 solutions are given in Table XXIV, and for $UO_2(NO_3)_2$ solutions are given in Table XXV.

Enrichment Limits

The ANSI Standard[2] contains a limit on the ^{235}U content of uranium homogeneously mixed with water with no limit on the water-to-uranium ratio and no specification of the uranium compound. An examination of that limit revealed a dependence on the compound and indicated that the limit in the Standard is too large.[40] There is a similar limit in the Standard for $UO_2(NO_3)_2$ with, again, no limit on the H/U atomic ratio. It, too, was examined and judged to be too large.[40] These examinations have been repeated, and critical enrichments and limits have been calculated, based on the correlations of Tables XVII and XIX as fitted by least-squares linear functions of the H/U atomic ratio. The procedure followed was to calculate k (at zero buckling) as a function of both the H/U atomic ratio and ^{235}U enrichment and to determine the deviation from the critical value as obtained from the linear functions. Plotting the deviations at each enrichment against the H/U atomic ratio permitted the maximum deviation to be determined. Plotting these maxima

TABLE XXIV

Limits for Uniform Homogeneous Aqueous Solutions of UO_2F_2 at ^{235}U Enrichment of 10% and Less

Dimension	^{235}U Enrichment, wt%	Calculational Method			ΔP[a]	Proposed Standard
		HRXN-ANISN	GLASS-ANISN	MGBS-TGAN		
^{235}U mass, g	10	1138	1121	1065	70	1070
	5	1672	1638	1643	125	1640
	4	2024	1975	2024	160	1980
	3	2784	2731	2886	270	2750
	2	7960	8050	8620	1680	8000
Cylinder diameter, cm	10	20.8	20.5	20.1	0.47	20.1
	5	26.9	26.7	26.6	0.65	26.6
	4	30.4	30.2	30.3	0.85	30.2
	3	37.5	37.4	37.9	1.20	37.4
	2	62.9	63.2	64.5	3.95	63.0
Slab thickness, cm	10	8.9	9.0	8.3	0.30	8.3
	5	13.0	13.1	12.6	0.45	12.6
	4	15.3	15.4	15.1	0.55	15.1
	3	19.9	20.0	20.1	0.80	20.0
	2	36.4	36.8	37.5	2.55	36.5
Volume, ℓ	10	16.1	15.3	14.8	0.92	14.8
	5	31.5	30.6	30.8	2.20	30.6
	4	43.7	42.7	43.8	3.45	42.7
	3	78.0	76.5	80.8	7.70	77.0
	2	335	339	362	70.0	340.0
Uranium concentration, g/ℓ	10	125.4	125.1	123	3.0	123
	5	266.0	266.7	260.7	6.0	261
	4	341.4	343.3	334.7	7.8	335
	3	475.9	480.9	365.2	12.0	470
	2	787.8	793.7	767.6	22.6	770
Limiting ^{235}U enrichment[b], wt%		1.48	---	1.45	---	1.45

[a] The approximate change in the parameter resulting from $\Delta k_{eff} = 0.01$, at $k_{eff} = 1.00$, calculated by the MGBS-TGAN code.

[b] The specified limit on the ^{235}U content of uranium in a saturated (5 M) aqueous solution of UO_2F_2 to preserve subcriticality.

TABLE XXV

Limits for Uniform Homogeneous Aqueous Solutions of $UO_2(NO_3)_2$ at ^{235}U Enrichment of 10% and Less

Dimension	^{235}U Enrichment, wt%	Calculational Method			ΔP[a]	Proposed Standard
		HRXN-ANISN	GLASS-ANISN	MGBS-TGAN		
^{235}U mass, g	10	1495	1528	1470	100	1470
	5	3300	3590	3560	390	3300
	4	6500	7640	7230	1250	6500
Cylinder diameter, cm	10	25.7	25.9	25.2	0.6	25.2
	5	42.7	44.0	43.7	1.6	42.7
	4	58.6	62.0	60.7	3.3	58.6
Slab thickness, cm	10	12.3	12.6	11.9	0.4	11.9
	5	23.4	24.4	24.0	1.0	23.4
	4	33.7	36.1	35.1	2.1	33.7
Volume, ℓ	10	27.7	28.1	26.7	1.8	26.7
	5	111	121	120	13.1	111
	4	273.1	321	304	52.0	273
Uranium concentration, g/ℓ	10	130	130	128	3.0	128
	5	288	294	283	7.5	283
	4	380	391	375	10.4	375
Limiting ^{235}U enrichment,[b] wt%		2.9	2.98	2.88	---	2.88

[a] The approximate change in the parameter resulting from $\Delta k_{eff} = 0.01$ at $k_{eff} = 1.00$ calculated by the MGBS-TGAN code.

[b] The specified limit on the ^{235}U content of uranium in a saturated (2.5 *M*) aqueous solution of $UO_2(NO_3)_2$ to preserve subcriticality.

against enrichment determined the critical enrichment (maximum deviation of 0) and the limiting enrichment (maximum deviation of −0.02). Based on the experimental uncertainties, on the deviations from the least-squares fits as recorded in Tables XVII and XIX, and on the likelihood of the two results at 2.26% ^{235}U in the $UO_2(NO_3)_2$ experiments being in error, a margin of 0.02 in the effective neutron multiplication factor was judged sufficient to ensure subcriticality. The results are given in Table XXVI for homogeneous mixtures of uranium and compounds with water along with limits proposed for the Standard. If the apparent trend with enrichment in the UO_3-H_2O experiments is extrapolated linearly, the proposed margin in the multiplication factor for uranium is reduced to slightly more than 0.01.

Uranium-235 enrichment limits accompanied by limitations on the amount of water present, the H/U atomic ratio, are useful, particularly the limit for uranium metal with no interspersed water. Although similar values for oxides may not find wide application since it may be difficult to exclude moisture, limits for oxides with controlled quantities of moisture may find application. The nominal quantity selected here and for mixtures of uranium and plutonium oxides[41] was 1.5%. For UO_2, U_3O_8, and UO_3, this amount corresponds to H/U atomic ratios of 0.456, 0.474, and 0.483, respectively. Although the data on which to base such limits are meager, there are the critical and exponential experiments with metal cylinders, the fast reactor enrichment benchmark,[49] and the PCTR experiments[42] with UO_3 enriched to 3.04%. The inconsistent behavior of the GLASS code for ^{238}U in dry or nearly dry materials tends to render it useless for deriving these limits. Consequently, HRXN and MGBS were the codes employed in these calculations.

The ANSI Standard contains a ^{235}U enrichment limit for dry uranium metal of 5 wt% ^{235}U. Because of experimental uncertainties, it would be difficult to justify a limit this large solely on the basis of the exponential experiments. However, the establishment of the enrichment benchmark[49] provides ample justification. According to results from the HRXN code, relative to the benchmark, the limit is subcritical by a margin in *k* of 0.05. If the same normalization is used and the same margin, the enrichment limit computed by HRXN (at zero buckling) for dry uranium oxides, essentially independent of the oxygen content, is 5.8%. Limits similarly computed by MGBS were 5.93% for UO_2, 6.04% for U_3O_8, and 6.10% for UO_3. The critical

TABLE XXVI

Critical and Limiting ^{235}U Enrichment of Uranium and of Various Compounds in Mixtures with Water*

Method	Condition	Chemical Composition				
		Metal	UO_2	U_3O_8	UO_3	$UO_2(NO_3)_2$
HRXN	Critical	0.996	1.02	1.028	1.033	2.063
	Limit	0.94	0.966	0.973	0.977	1.956
GLASS	Critical	0.992	1.019	1.025	1.028	2.075
	Limit	0.936	0.962	0.969	0.972	1.972
MGBS	Critical	0.99	1.017	1.026	1.03	---
	Limit	0.932	0.96	0.968	0.972	---
	Proposed limit for the Standard	0.93	0.96	0.97	0.97	1.96

*The ^{235}U content is given in weight percent.

value of k computed for the enrichment benchmark by MGBS is somewhat further above the values computed for the exponential and critical experiments than is that calculated by HRXN; the latter agrees fairly well. In the absence of further information, the prudent course is to propose 5.8% as the enrichment limit for dry oxide.

Moderation by water rapidly increases k for uranium oxide of this ^{235}U content, and the enrichment limit decreases as the associated hydrogen increases. Most directly applicable are the PCTR experiments with 3.04% enriched UO_3, but the lowest H/U atomic ratio in those experiments is ~3.6, far above the limiting ratio of ~0.5 adopted here. Calculation of these experiments with the HRXN code gives k equal to ~1.03 with no apparent trend with the H/U atomic ratio. However, correlation with the exponential and critical metal-cylinder experiments and with the benchmark experiment indicate[6] a value of k equal to ~0.99 with a trend toward larger values as the ^{235}U content decreases; hence, a decrease in k as H/U approaches 0.5 from 3.6 appears likely. It was assumed that a value of k equal to 0.95 provides adequate ensurance of subcriticality. The corresponding subcritical enrichment limits for moist oxides are 3.21% ^{235}U for UO_2, 3.27% for U_3O_8, and 3.29% for UO_3. The calculations of k by MGBS for the PCTR experiments show a downward trend in k with decreasing H/U. Extrapolation of a semilog plot of k as a function of the atomic ratio gives $k \cong 0.91$ at $H/U = 0.45$; on a linear plot the value is ~0.96. If a value of k of 0.86 is assumed, the limiting enrichments for moist oxide are 3.38% for UO_2 and 3.57% for UO_3. Despite the inconsistencies associated with the GLASS code calculations for these conditions, there is no indication that either the HRXN or MGBS results are nonconservative. As for dry oxide, the prudent course is to propose the HRXN results for limits, namely an enrichment limit of 3.2% ^{235}U in moist UO_2 and 3.3% in moist U_3O_8 and UO_3, provided the oxide contains no more than 1.5% water by weight.

Metal and Oxide Limits

Limits for ^{235}U as metal and as dry and moist oxides, the oxides at both full and half density, were calculated by HRXN-ANISN and by GLASS-ANISN, with the same combined spectrum treatment[1] for the latter method as used in the comparisons with experiment. The quadrature in the ANISN calculations was S_{16} in all cases. Some GLASS-ANISN calculations for moist oxides were made with cross sections collapsed by the single spectrum developed for the investigation of the moist oxides, but the results differed only slightly from those obtained with the combined spectrum. A margin of 0.02 below the value of k_{eff} determined in the calculations of the water-reflected sphere was judged to be sufficient to ensure subcriticality. For metal, this margin is certainly ample in view of the precision of the experiment and the small extrapolation to 100% ^{235}U. The only pertinent experiments with oxide appear to be some with PuO_2, correlations with which give no indication of a lower value of k_{eff} for oxide than for metal[1]; hence, this margin should be adequate for uranium oxides as well. Densities of metal, UO_2, U_3O_8, and UO_3 were normalized to the corresponding densities of natural uranium and its oxides, taken to be 19.05, 10.96, 8.30, and 7.29 g/cm^3, respectively. Moist oxides contained 1.5% water by weight and their densities were determined for mixtures of water and oxide with volumes additive. For half density limits, the mixture densities were simply halved, i.e., the moist oxides were

TABLE XXVII

Calculated Limits for Dry Uranium Metal and Oxides Containing 100% ^{235}U

Material	Dimension[a]	Calculational Method		Present Standard[b]	Proposed Standard
		HRXN-ANISN	GLASS-ANISN		
Metal	M	19.75	19.52	20.1	20.1
	D	7.29	7.30	7.3	7.3
	T	1.40	1.41	1.3	1.3
UO_2	M	35.16	35.73	---	35.2
	MO	39.95	40.60	---	40.0
	D	11.59	11.71	---	11.6
	T	2.90	2.97	---	2.9
U_3O_8	M	50.47	51.94	---	50.5
	MO	59.63	61.37	---	59.7
	D	14.66	14.87	---	14.7
	T	4.01	4.12	---	4.0
UO_3	M	59.88	62.00	---	59.9
	MO	72.11	74.66	---	72.1
	D	16.38	16.64	---	16.4
	T	4.65	4.80	---	4.6

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

[b] From Ref. 54.

assumed to contain 50% voids. All units were reflected by an effectively infinite thickness of water at 20°C.

Limits for metal and dry oxides are given in Table XXVII. The limits in the present Standard are derived from Ref. 54. Agreement between the respective values of the various dimensions calculated by the two codes is quite good. Additionally, the critical masses of a water-reflected ^{235}U metal sphere calculated by HRXN-ANISN and by GLASS-ANISN were 21.50 and 21.27 kg, respectively; hence, the margin of the present limit is in excess of 0.01. Because of the precision of the recent experiment,[44] a margin of 0.005 for metal and a mass limit of ~21.0 kg would not be difficult to justify, except for the possibility that a reflected cube may have a lower critical mass than a reflected sphere.[1] The present limit appears to make sufficient allowance for this possibility. There is no particular incentive to increase the slab thickness limit; hence, it is proposed to leave it unchanged despite the permissible increase. The critical thicknesses calculated by the two methods are 1.52 and 1.55 cm.

Calculations of critical spherical masses were made for uranium containing 90% ^{235}U with the remaining 10% ^{234}U, ^{236}U, or ^{238}U. From both HRXN-ANISN and GLASS-ANISN calculations, the mass of ^{235}U was less with 10% ^{234}U present than when the sphere was 100% ^{235}U. The total mass was, of course, greater. With ^{236}U or ^{238}U, the ^{235}U mass was greater. Thus, mass limits calculated for 100% ^{235}U may conservatively be assumed to apply to the sum of the masses of ^{234}U and ^{235}U in any isotopic mixtures, with ^{236}U and ^{238}U ignored.

Limits for moist oxides at full and half density are given in Table XXVIII. Comparison of Tables XXVII and XXVIII for full density oxides shows that the introduction of moisture reduces the mass limit for all three oxides. It increases the cylinder diameter limit for UO_2 and the slab thickness limits for UO_2 and U_3O_8. However, the limit on moisture content is an upper limit. It would not be practical to require a moisture content of 1.5%. The cylinder diameter and slab thickness limits proposed for the revised Standard are, therefore, those calculated for dry oxides. (Although not tabulated here, calculations were also made for dry half-density oxides.)

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

TABLE XXVIII

Limits Calculated for Moist Oxides* of Uranium Containing 100% ^{235}U

Density	Oxide	Dimension[a]	Calculational Method		Proposed Standard
			HRXN-ANISN	GLASS-ANISN	
Full[b]	UO$_2$	M	32.26	32.28	32.3
		MO	37.20	37.22	37.2
		D	11.91	11.97	11.6
		T	3.07	3.13	2.9
	U$_3$O$_8$	M	44.02	44.36	44.0
		MO	52.81	53.22	52.8
		D	14.62	14.73	14.6
		T	4.07	4.16	4.0
	UO$_3$	M	51.21	51.78	51.2
		MO	62.61	63.31	62.6
		D	16.15	16.28	16.2
		T	4.65	4.76	4.6
Half[c]	UO$_2$	M	88.6	89.55	88.6
		MO	102.2	103.29	102.2
		D	21.03	21.20	20.4
		T	6.14	6.26	5.8
	U$_3$O$_8$	M	122.05	124.36	122.0
		MO	146.40	149.17	146.4
		D	25.97	26.24	26.0
		T	8.14	8.32	8.0
	UO$_3$	M	142.86	146.11	142.9
		MO	174.65	178.63	174.7
		D	28.77	29.10	28.8
		T	9.30	9.52	9.3

*The oxide contains no more than 1.5 wt% water.

[a] M = mass of ^{235}U in kilograms, MO = mass of uranium oxide, including the moisture, 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 10.84, 8.21, and 7.22 for UO$_2$, U$_3$O$_8$, and UO$_3$, respectively) and the volume of water at 20°C at 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.