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SUBCRITICAL LIMITS FOR ^{233}U SYSTEMS

INTRODUCTION AND SUMMARY

To serve the dual purposes of validating computational methods in use at SRL and SRP and of examining and adding to subcritical limits currently in the American National Standard for "Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors", surveys have recently been completed for homogeneous ^{239}Pu and ^{235}U systems.¹⁻⁵ Experimental data suitable for one-dimensional computer codes have been selected and put in a form readily useable for code input so that correlations can easily be repeated when codes or cross sections are revised or when new codes are obtained. Bias has been established for the three computational methods commonly used at SRL and SRP, MGBS-TGAN⁶⁻⁷, HRXN-ANISN⁸⁻¹⁰, and GLASS-ANISN.¹¹⁻¹³ Subcritical limits have been calculated to be proposed for inclusion in the ANSI Standard, either as replacements where there are doubts as to the subcriticality of present limits, or as additional limits to extend the usefulness of the Standard. It is expected that these limits will also prove generally useful for SRL and SRP operations.

The present memorandum extends this work to ^{233}U systems. Doubts have been expressed as to the subcriticality of some of the limits in the Standard.¹⁴ These doubts have been found to be justified, and new limits are proposed. Although no operations are currently being

performed with ^{233}U at SRP, it is well to be prepared. Evaluations of previous operations¹⁵ were made prior to some of the experimental data now available and prior to the availability at SRL of transport theory methods. The present correlations should permit less restrictive limits if these operations are re-evaluated.

DISCUSSION

Experimental Data

Data for ^{233}U systems are much less extensive than for ^{235}U systems. No experiments have been done with a water-reflected sphere of ^{233}U metal. Experiments with solutions at the high concentrations at which minimum critical volumes and dimensions occur have not been done with spheres. For the one-dimensional computational methods⁶⁻¹³ being validated, the appropriate data are those obtained with spheres or with cylinders that can readily be extrapolated to critical diameters of cylinders with infinite height. Data obtained with vessels so large that assumptions of separability introduce little uncertainty are also suitable. Experiments with solutions have been done both with UO_2F_2 and with $\text{UO}_2(\text{NO}_3)_2$ containing some free acid. Solution densities were calculated from the recipes used for ^{235}U solutions^{2,5} and from reported concentrations. Agreement with reported densities is good. However calculated $\text{UO}_2(\text{NO}_3)_2$ solution densities are generally slightly greater than reported densities; UO_2F_2 densities slightly less.

Spheres of Aqueous Solution

A series of experiments were done in 1953 - 1954 with two spherical vessels containing aqueous solutions of $^{233}\text{UO}_2\text{F}_2$ and having volumes of 9.66 and 17.02 l at room temperature¹⁶. The smaller vessel was made critical, water-reflected, at several temperatures. The larger vessel was likewise made critical, water-reflected, at several temperatures and also was made critical bare at a single temperature. The same two spheres were also included in a series of experiments with UO_2F_2 and $\text{UO}_2(\text{NO}_3)_2$ solutions apparently done at about the same time, but not reported until 1959.¹⁷ (See Reference 18) In the later report the larger sphere is stated to have been coated internally with a polyvinyl chloride plastic, Unichrome, which is about 30 wt % chlorine. Removal of the Unichrome was found to decrease the critical concentration of $^{235}\text{UO}_2\text{F}_2$ by 2%. The Unichrome coating is apparently the systematic error referred to in the earlier report which resulted in masses and concentration "believed to be about 2% high".

Other experiments with spheres include bare and water reflected spheres of $\text{UO}_2(\text{NO}_3)_2$ solution ranging in volume from 5.8 to 26.0 l. The spheres were made critical within ± 0.0005 in k_{eff} . No free acid concentration is reported, but at 131 g U/l averaged 0.375 M HNO_3 (except for a bad value of 0.5M). The corresponding N/U ratio is 2.67, which presumably held at all uranium concentrations since the various concentrations were gotten by diluting the most concentrated solution.

Finally experiments with uranyl nitrate solutions were done in bare 174 and 949 liter spheres. In the smaller sphere boron concentration was a variable. These experiments were later analyzed to obtain slight corrections for lack of sphericity, etc. With or without the corrections the spheres were not exactly critical, i.e. k_{eff} deviated slightly from unity.

The critical experimental conditions are given in Table 1 for all the spheres. In the series with variable temperature, concentrations were calculated from reported masses and volumes since the concentrations are all reported at 25°C.

Cylinders of Aqueous Solution

The only experiments at concentrations approaching those at which minimum critical volumes and dimensions occur are some, already referred to, done with $UO_2(NO_3)_2$ and UO_2F_2 solutions in paraffin - reflected cylinders.¹⁷ Most of the cylinders were unreflected on top. An indirect method was used for measuring heights of the uranyl nitrate solutions, resulting in an estimated uncertainty of 3%. The estimated uncertainty for the uranyl fluoride solution heights was 1%. Three or four of the vessels containing UO_2F_2 solutions were coated with Urichrome. (The text says three; four are so indicated in the table of data.) In many cases there was insufficient material to make the system critical, and critical heights were extrapolated from source multiplication curves. The experimental data for the higher concentration $UO_2(NO_3)_2$ and UO_2F_2 solutions selected for the present work are given in Tables II and III, respectively. Temperature was assumed to be 25°C; cylinder walls, bottoms, and tops (when present), 1/16 inch aluminum.

The series of experiments with bare and water-reflected spheres of uranyl nitrate solution also included bare and water-reflected cylinders.¹⁹ The data reported for the reflected cylinders are for the case where each cylinder was supported by a 24.3 cm high cylinder of styrofoam of the same diameter.²⁰ Some of these data are in error: the mass for the 38.1 cm diameter cylinder at 132 g U/l should be 2.02 instead of 1.77 kg, and the height for the 20.3 cm diameter cylinder at 95.0 gU/l should be 27.02 rather than 20.02 cm. Of more interest are unreported data²⁰ for the case where the bottom and sides were reflected by water, i.e., the styrofoam was replaced by water. These data are given in Table IV. However, even in these experiments, concentrations were not great enough or cylinder diameters small enough to be of much interest in the present work. Critical experiments with bare and water-reflected cylinders have also been done in France²³. The greatest concentration was 206.5 g $^{233}U/l$, and the smallest cylinder diameter, 25 cm; hence again there is little interest in these data in the present work.

Cylinder data in which there is interest are measurements in large cylinders at concentrations close to the minimum critical value for an infinite system.^{2,1} The reported critical heights contain a correction for bottom structure and are truly bare critical heights. The radius was increased by an assumed wall thickness so that the dimensions in Table V are estimates of bare critical values. The dimensions are so large that small uncertainties in their exact values have little effect. Temperature was assumed to be 25°C.

Pertinent Metal Experiments

Since the critical mass of a water-reflected sphere of ^{233}U has not been measured, it is necessary to infer the appropriate bias for calculation for water-reflected metal and oxide from other experiments. Besides experiments with bare and water-reflected plutonium spheres for which correlations have been reported¹ and with a water-reflected ^{235}U sphere for which correlations also have been reported⁵, the experiments^{2,4} listed in Table VI were considered pertinent. Experiments in which ^{233}U , ^{235}U , and Pu cores were reflected by Be^{25} might also be pertinent, but were not considered.

Computational Techniques

The same three code combinations MGBS-TGAN, HRXN-ANISN, and GLASS-ANISN, with the latter two supplemented by SPBL, were used as in previous correlations and limit calculations.^{1,3,4,5} No changes were made in how they were used but a few remarks need to be made about MGBS. In Amster's compendium of thermal cross sections^{2,6}, which is partially incorporated in MGBS, the thermal spectrum is a function of $^{235}\text{U}/\text{H}$, $^{239}\text{Pu}/\text{H}$, $1/v$ barns/H and temperature. However, cross sections for 0, 2, and 4 barns per hydrogen atom, only, are incorporated in MGBS, although the compendium extends to 12 barns. In MGBS ^{233}U is treated as $1/v$ as regards its effect on the spectrum. Three point Lagrangian interpolation and extrapolation is provided in terms of barns/H. Although cross sections change nearly linearly with barns/H, quadratic extrapolation to ratios as high as 17.5 (as in the UO_2F_2 cylinder experiments) seems questionable. The ^{233}U absorption and fission cross sections, although not as far from varying as $1/v$ as ^{235}U cross section, deviate from strict $1/v$ behavior; hence relative thermal absorption may be in error at large extrapolations. On the other hand, as the spectrum hardens, the fraction of fission neutrons reaching the thermal group becomes small and the cross section errors may have little effect. However, at the high barns/H ratio of the volume and dimension limits (~ 33 for UO_2F_2), MGBS-TGAN should probably be considered the least reliable of the three methods.

CORRELATIONSAqueous Solution

Correlations were made of the three code combinations HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN with the sphere experiments of Table I, and the results are recorded in Table VII in the same order as the experiments are listed in Table I. As in correlations with ^{235}U solution experiments,⁵ UO_2F_2 was represented in MGBS by UO_4 . Densities of UO_3 and UO_2F_2 were calculated by HRXN and were adjusted to densities of natural UO_3 and UO_4 as required by MGBS. Since MGBS presumes a temperature of 20°C and since the experiments at lowest temperature in the two series in which temperature was a variable were essentially duplicated in the sphere experiments reported along with the paraffin - reflected cylinder experiments,^{1,7} no MGBS-TGAN correlations were made with these series. No attempt was made in MGBS to adjust to the temperature of any of the experiments by the introduction of voids. The correlations are expressed in Table VII in terms of the critical values of k_{eff} , i.e. as $1 + \text{Bias}$ where $\text{Bias} = k_{\text{eff}}(\text{calc}) - k_{\text{eff}}(\text{expt})$.

Prior to learning²⁰ that the N/U ratio was 2.67 in the series of experiments¹⁹ with $\text{UO}_2(\text{NO}_3)_2$, the effect of the ratio was studied. Increasing the ratio from 2.0 (no free acid) to 2.6 decreased k_{eff} for both the bare and reflected spheres at about 130 g U/l by about 0.004. At about 45 g/l, the reduction was about 0.002.

Four of the sphere experiments were calculated by McNeany and Jenkins¹⁴ Experiments 9, 10, 11, and 12 in their listing correspond, respectively, to experiments 19 ($\text{H}/^{233}\text{U}=192.3$), 11 ($\text{H}/^{233}\text{U}=381.5$), 23 ($\text{H}/^{233}\text{U}=1532$), and 28 ($\text{H}/^{233}\text{U}=1987$) as listed in Tables I and VII. Their results (by S_8 quadrature) with Hansen-Roach cross sections²⁷ were 0.994, 0.988, 1.004, and 1.005. The first two lie appreciably above the corresponding values of Table VII, and appear to indicate use of the dE/E weighted cross sections for H rather than the fission spectrum weighted values. Part of the reason 0.994 (they actually report 0.944 in their Table IX, but 0.994 is shown in their Figure 1.) lies so far above 0.972 as calculated here is their use of $\text{N}/\text{U}=2.0$. They also show F as being present, but this may be a typographical error. Their results (also by S_8) with ENDF/B-IV cross sections were, respectively, 1.028, 1.013, 0.996, and 0.991, and the first two lie appreciably below the corresponding values of Table VII, presumably reflecting differences in processing codes, resonance absorption calculation, and group structure. The same conclusion is reached, however, namely that Hansen-Roach cross sections under-estimate k_{eff} whereas ENDF/B-IV cross section over-estimate it.

Correlations with the paraffin-reflected cylinders of $\text{UO}_2(\text{NO}_3)_2$ solution are given in Table VIII and of UO_2F_2 solution in Table IX in the same order that the experiments are listed in Tables II and III. Since the density of paraffin is somewhat variable (The Chemical Rubber Handbook gives a range of 0.87 to 0.91 g/cm³), some consideration was given to the effect of variations in density. For a reflected sphere containing solution at about 50 g $^{233}\text{U}/\ell$, increasing the

the density from 0.87 to 0.91 g/cm³ increased k_{eff} (as calculated by HRXN-ANISN) by about 0.005. At this same concentration the experimenters found paraffin to be a slightly better reflector than water¹⁷; on the basis of their experiments, reflecting a sphere by paraffin rather than by water was calculated (again by HRXN-ANISN) to increase k_{eff} by about 0.003.

The approach incorporated in SPBL was used to correlate HRXN-ANISN and GLASS-ANISN with the cylinder experiments. An ANISN calculation was made for each dimension, and k_{eff} was determined with the transverse dimension assumed infinite (zero transverse buckling). Quadrature was S₁₆. Corresponding to each of the values of k_{eff} SPBL, by a B₁ calculation, computed the geometric buckling. The total geometric buckling was obtained by adding the axial and radial components, and the corresponding value of k_{eff} was calculated, again by B₁. Values of k_{eff} so determined are greater than would be obtained by a non-separable solution such as Monte Carlo or two-dimensional (R,Z) transport theory (See Appendix). However, by expressing k_{eff} as a function of axial buckling and extrapolating to zero axial buckling, the values appropriate for infinite cylinders can be obtained. For ²³⁵U solutions these values are in agreement with correlations made with spheres.⁵

The variation of k_{eff} with axial buckling exhibited in Tables VIII and IX is greater than found for ²³⁵U solutions, but does not appear inconsistent with that shown in the study reported in the Appendix. However, the variation with axial buckling is not nearly linear as zero is approached as the study indicates should be the case. Deviations from a straight line fit are outside the limits of error assigned to the data points. For the nitrate solutions the three highest concentration solutions (H/²³³U=57.9, 67.0, and 84.2) in the 7.55 cm radius cylinder have k_{eff} 's lower than would be expected from the other data. These three values are inconsistent with the assertion that 6.32 cm radius cylinders of these solutions would be subcritical at any height. (A similar disagreement with the assertion made by the experimenter that some cylinders would be subcritical at any height exists for ²³⁵U solution.⁵) Similar behavior is shown for the fluoride solutions. In particular, at H/²³³U=73.9 the values of k_{eff} determined for the 8.35 and 7.55 cm radius cylinder are inconsistent, as are those for the 6.85 and 6.34 cm radius cylinders. There is less reason to doubt that the smallest (5.60 cm radius) cylinder would be subcritical at any height at all concentrations, but at the four highest concentrations the margin appears small. In extrapolating to zero axial buckling consideration was given to the slope indicated by the study in the Appendix and to the maximum attainable heights in the smallest diameter cylinders. It is expected that the experimenters would have recognized it if these heights corresponded to k_{eff} close to critical; estimated critical heights were reported for cases where k_{eff} calculated for the available height was as much as 0.07 below the value calculated for the estimated height.

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

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

where B_c^2 is the calculated critical buckling, M^2 the associated migration area, and B_g^2 the geometric buckling. This approach according to the Appendix, should give less variation of k_{eff} with axial buckling. In these correlations paraffin was considered to be water, since the two appear nearly equivalent and paraffin is not easily introduced as a material in MGBS.

Although correlations were made with the experiments of Table IV, they contributed little to the determination of bias and are not reported here.

Correlations with the large bare cylinders described in Table V are given in Table 2. The assumptions of separability in SPBL and in the MGBS-TGAN approach introduce minimal error because of the large size. The correlations are given in the same order as the experiments are listed in Table V. The quadrature in the ANISN calculations was S_{16} .

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

Metal Systems

Correlations of HRXN-ANISN and GLASS-ANISN with the metal spheres of Table VI are given in Table XI. The calculations were made in exactly the same manner as for ^{235}U and ^{239}Pu , as reported previously.^{1,5} The effect of experimental uncertainties was evaluated with Hansen-Roach cross sections by S_4 quadrature. In the GLASS calculations for ^{233}U the resonance absorption rate exceeded the source rate in a number of groups, as was the case with ^{235}U and ^{239}Pu . The bare ^{233}U sphere was also calculated by McNeany and Jenkins¹⁴ with S_8 quadrature. Their results with Hansen-Roach and with ENDF/B-IV cross sections were, respectively, 1.008 and 0.967, in good agreement with Table XI. As they noted, ENDF/B-IV cross sections over-estimate k_{eff} for moderated ^{233}U systems and under-estimate it for metal.

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

Subcritical Limits

Aqueous Solution

All three computational methods (HRXN-ANISN, GLASS-ANISN, and MGBS-TGAN) were used to compute limits for solutions. A temperature of 200C was assumed, and all units were surrounded by an effectively infinite thickness of water. The ANISN quadrature was S_{16} . The margin from the curves of Figures 1-3 necessary to assure subcriticality is difficult to assess. For the concentration limit, the areal density limit, and the mass limit a margin in k_{eff} of 0.01 seems sufficient in view of experimental data at the corresponding concentrations. Scatter in the data as plotted in Figures 1-3 gives an indication of uncertainty. In similar experiments with spheres of ^{235}U solutions the uncertainty associated with quoted uncertainties in dimensions and solution concentration is well within $\pm 0.005^3$. A margin of 0.02 should be ample. The dimension limits occur at high concentration where the only data are those obtained with paraffin-reflected cylinders. However, the extrapolations to infinite cylinders are believed to have been done conservatively, and a margin of 0.02 seems sufficient.

Calculations were carried only as far as the saturated solutions, since limits apply only to homogeneous solutions. Johnson and Kraus,²⁸ whose density formula for UO_2F_2 solutions was used in the present work, indicate a 66% solution to be saturated. The equivalent molarity is 5.04, and for the present work a saturated solution was assumed to be 5.0 M. Kapustinsky and Lipilina²⁹, whose work serves as the basis for the formula adopted for computing the densities of uranyl nitrate solutions, in covering concentrations from almost saturation to extreme dilution report data up through 52.36% (2.3 M) and refer to work by others at as high a concentration as 54.77% (2.44 M). For the present work the saturated solution was assumed to be 2.5 M.

Table XII contains "limits", i.e. minimum values calculated to correspond to k_{eff} 0.02 below the curves of Figures 1-3, as calculated by the three computational methods. The quadrature in ANISN was S_{16} . Limits now in the Standard, based on Webster's calculations,³⁰ are shown for comparison, and limits are proposed for the revised Standard.

Agreement is very good in the case of the concentration limit and would be even better if the curve in Figure 3 were given a sharp upturn at $H/^{233}\text{U}=1800$, similar to that for ^{235}U so as to fit the data more closely, rather than drifting downward. The critical concentrations, calculated by the three methods are 11.22, 11.20, and 11.12 g $^{233}\text{U}/\ell$.

MGBS-TGAN does not agree well with the other two methods in the case of areal density. The minimum occurs at a concentration of about 0.11 M ($H/^{233}\text{U} \approx 1000$). Drawing the curve of Figure 3 through the neighboring data points at $H/^{233}\text{U} < 1000$ rather than below them and providing a sharp dip at $H/^{233}\text{U}=1800$, as indicated above, would increase the critical value of k_{eff} by about 0.01 and would increase 0.334 to about 0.344 g/cm². The critical value would increase from 0.355 to 0.365. The least change in slope as a function of $H/^{233}\text{U}$ near 1000 is shown by the HRXN-ANISN correlations. Interpolation, by way of the curve, to yield critical values of k_{eff} near $H/^{233}\text{U}$ should be least open to question in this case. There appears to be no reason to suspect that a margin of 0.02 is insufficient to provide subcriticality or that the Standard limit of 0.35 g/cm² might be critical.

The spread in mass values is surprising. The minimum mass occurs at $H/^{233}\text{U} \approx 450$. Redrawing the curve in Figure 3 as indicated above would increase the critical k_{eff} by about 0.006 and increase the critical mass calculated by MGBS-TGAN by about 16 g from 550 to 566 g. (As has been noted previously^{1,5} a margin in k_{eff} of 0.02 corresponds to a larger increment in mass or other parameter as calculated by MGBS-TGAN than by HRXN-ANISN or GLASS-ANISN. Here the difference in mass is 53 g by MGBS-TGAN, 43 by HRXN-ANISN.) The fictitious transverse buckling applied in MGBS-TGAN calculations for spheres makes aluminum walls appear to be worth more than they actually are when the critical k_{eff} deviates appreciably from unity. Since aluminum walls were present in the experiment, their removal, as in the limit calculations, results in too low a critical mass, in the present case about 8 g too low. The resulting critical mass, 574 g, is in good agreement with that, 573, calculated by HRXN-ANISN with the critical value of k_{eff} read from Figure 1. Webster³⁰ calculated a critical mass of 570 g. His few correlations with experiment indicate this mass might be subcritical by a margin of about 0.005 in k_{eff} . Previous calculations by Clark^{15,31} led to a critical mass of about 600 g, in agreement with that reported by Paxton et al.³² The critical mass was not calculated by GLASS-ANISN, but would probably be about 564 g. The curve in Figure 2, however, tends to fall a little below the correlations near $H/^{233}\text{U}$ in 450. Although it appears doubtful that 550 g could be critical, more confidence is provided by reducing the limit and 540 g is being proposed. It also is proposed that the limit for possibly non-uniform slurries³¹ be reduced from 520 to 500 g.

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

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

Metal Oxides

Limits (i.e. values corresponding to k_{eff} 0.02 below the critical value selected by analogy with ^{235}U and plutonium experiments) for dry metal and oxide calculated by HRXN-ANISN and GLASS-ANISN are given in Table XIV. The metal or oxide cores were surrounded by 20 cm H_2O at 20°C. The quadrature was S_{16} , the small difference between S_{16} and S_{∞} being ignored. Since the larger change in the critical k_{eff} between bare and water-reflected systems was selected, a margin of 0.02 was considered sufficient to assure subcriticality for metal. It was also considered sufficient for oxide since experiments with plutonium oxide indicate no lower critical k_{eff} for oxide than for metal.¹ The limits in the Standard are based on calculations by Roach and Smith³³ and are values they calculate from Hansen-Roach cross sections by S_8 at k_{eff} (uncorrected for bias) = 0.97. Not surprisingly, they are consistent with the HRXN-ANISN results by S_{16} at k_{eff} = 0.977. The agreement between HRXN-ANISN and GLASS-ANISN is poorer for ^{233}U than for ^{235}U or ^{239}Pu and may indicate selection of too low a critical value of k_{eff} for water-reflected systems. However, in the absence of a definitive experiment or of a compelling reason for increasing the critical value, the prudent course to follow is to base the limits on the GLASS-ANISN calculations.

Limits, calculated similarly, for moist oxides at full and half density are given in Table XV. The moisture is limited to 1.5% as for ^{235}U and ^{239}Pu . Volumes of moisture and oxide are assumed additive. Comparison of Table XIV and XV shows that moisture reduces the limiting mass of uranium for all oxides as calculated by either

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method, but only in the case of the cylinder diameter for UO_3 by GLASS-ANISN is a dimension reduced. The moisture content is an upper limit; it would not be practical to require a moisture content of 1.5% H_2O . The proposed limits in Table XV are then the lower of the dry and moist values. (Although not tabulated here, calculations were also made for dry half-density oxides).

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Table 1
Critical Spheres of ^{233}U Solution

Isotopic Comp. ^a	$\text{g U}/\ell$	$\text{g NO}_3/\ell$ ^b	$\text{g Th}^{\text{C}}/\ell$	$\text{g B}^{\text{d}}/\ell$	Radius, cm	Wall Thickness, cm ^e	Refl ^f	Temp, $^{\circ}\text{C}$	Ref
1	61.95	0	0	0	13.21	0.13	H ₂ O	32.0	16
	62.44				13.22			39.5	
	63.79				13.23			65.5	
	64.92				13.24			83.2	
	66.39				13.25			96.5	
1	39.23	0	0	0	15.96	0.13 ^g	H ₂ O	26.3	16
	40.01				15.96			56.0	
	41.72				15.97			99.5	
1	68.22	0	0	0	15.96	0.13 ^g	None	27.0	16
2	62.8	43.9	0	0	13.21	0.13	H ₂ O	25 ^h	17
1	67.9	0	0	0	15.95 ⁱ	0.13 ^g	None	25 ^h	17
1	66.9	0	0	0	13.04 ^j	0.13	H ₂ O	25 ^h	17
	61.8				13.20 ⁱ				
	60.8				13.28 ^k				
1	39.5	0	0	0	15.96	0.13 ^g	H ₂ O	25 ^h	17
3	132	93.7	0	0	11.170	0.122	H ₂ O	25 ^h	18
	95	67.5	0	0	11.847				
	47.9	34.0	0	0	14.579				
3	131	93.0	0	0	14.579	0.122	None	25 ^h	18

Table 1 (cont.)

Isotopic Comp. ^a	g U/l	g NO ₃ ⁻ /l ^b	g TH ^c /l	g B ^d /l	Radius, cm	Wall Thickness, cm ^e	Refl ^f	Temp, °C	Ref
	102	72.4	0	0	15.078				
	74.6	53.0	0	0	15.821				
	44.6	31.7	0	0	18.378				
4	17.14	12.17	0.076	0	34.6 ^l	0.32	None	20.0	20
	17.86	12.61	0.079						
	18.52	13.15	0.082	0.0465					
	19.18	13.56	0.085	0.0688					
	19.82	13.99	0.087	0.0912					
5	13.25	7.72	0.057	0	61.0 ^m	0.77	None	20.0	20

a) Isotopic Composition in weight %

	<u>233_U</u>	<u>234_U</u>	<u>235_U</u>	<u>238_U</u>
1	98.7	0.54	0.04	0.72
2	98.7	0.5	0.01	0.79
3	97.53	1.05	0.03	1.39
4	97.70	1.62	0.04	0.64
5	97.67	1.54	0.03	0.76

b) If NO₃⁻ concentration is zero, solute was UO₂F₂

Table 1 (cont.)

- c) Assumed present as ThO_2 at 9.86 g/cm^3
- d) Assumed present as B_2O_3 at 2.17 g/cm^3
- e) All vessel walls were aluminum
- f) Water reflector effectively infinitely thick ($\geq 20 \text{ cm}$)
- g) Vessel was coated internally with Unichrome, mocked up by 0.016 cm of $\text{CH}_2\text{CH Cl}$ with density 1.4 g/cm^3 or equivalently in GLASS by 0.0092% ^{10}B by weight in the vessel wall and in MGBS by 0.034 cm Fe . (Amount required to reduce critical ^{235}U concentration by 2%).
- h) Assumed temperature
- i) Sphere volume reduced 40 cm^3 to compensate for void above solution
- j) Sphere volume reduced 380 cm^3 to compensate for void above solution
- k) Sphere volume extrapolated from source multiplication curves
- l) Corrected values of k_{eff} in order of increasing B concentration: 1.0002, 1.0008, 1.0009, 1.0000, 1.0001
- m) Corrected value of k_{eff} 1.0001

Table II
Critical Paraffin - Reflected Cylinders^a of Uranyl Nitrate Solution

<u>gU/l^b</u>	<u>gNO₃⁻/l</u>	<u>Radius, cm</u>	<u>Critical Height, cm</u>	<u>Maximum Expt. Ht., cm</u>
496.5	346.8	10.25	16.1±0.2	14.0
386.0	269.7	6.32	∞ ^c	51
		7.55	27.9	
		9.53	16.3	
		10.25	14.4	
340.4	237.8	6.32	∞ ^c	59
		7.55	29.0	
		9.53	16.2	
278.6	194.6	6.32	∞ ^c	61
		7.55	30.7	
		10.25	14.7	
200.6	140.1	7.55	38.5±0.5	36.8
		10.25	16.4	
169.2	118.2	6.32	∞ ^c	55
		9.53	18.6	
		10.25	16.7	
162.1	113.2	7.55	46.8±0.5	45.4
		10.25	16.7	
128.7	89.9	7.55	73±2	55.4
		10.25	18.8	

a) Paraffin was assumed to be CH₂ with density 0.89 g/cm³. Where cylinder radii differ from reported values, they were derived from reported volumes and heights. Only the 9.53 and 10.25 cm. radius cylinders had top reflectors. Walls, bottom, and top (where present) were assumed to be 0.16 cm. aluminum; temperature, 25°C.

b) Uranium contained 98.7% ²³³U, 0.5% ²³⁴U, 0.01% ²³⁵U, 0.79% ²³⁸U by weight.

c) To be interpreted as apparently subcritical at any height.

Table III

Critical Paraffin - Reflected Cylinders^a of Uranyl Fluoride Solution

<u>g U^b/ℓ</u>	<u>Radius, cm</u>	<u>Critical Height, cm</u>	<u>Maximum Expt. Ht., cm</u>
693.0	5.60	∞ ^c	29.9
	6.34	38±2	23.8
	8.35	20±1	13.5
608.9	5.60	∞ ^c	34.9
	6.34	41±2	27.6
	8.35	16.7±0.2	16.3
526.8	5.60	∞ ^c	42.6
	6.34	41±1	32.4
	8.35	16.9	
456.9	5.60	∞ ^c	49.0
	8.35	18.0±0.3	16.9
336.4	5.60	∞ ^c	68.5
	6.34	56.5±0.5	53.3
	6.85	48.7±0.5	46.3
	7.55	24.0	
	8.35	19.1±0.4	16.9

a) Paraffin was assumed to be CH₂ with density 0.89 g/cm³. Only the 8.35 cm radius cylinder had a top reflector. Walls, bottom, and top (where present) were assumed to be 0.16 cm aluminum; temperature, 25°C. All except the 6.34 cm radius (and perhaps the 7.55 cm radius) cylinder were coated with Unichrome, mocked up by 0.016 cm of CH₂CH Cl with density 1.4 g/cm³ or equivalently in GLASS by 0.0074% ¹⁰B in the vessel wall and in MGBS by 0.034 cm Fe.

b) Uranium contained 98.7% ²³³U, 0.54% ²³⁴U, 0.04% ²³⁵U, 0.72% ²³⁸U

c) To be interpreted as apparently subcritical at any height.

Table IV
 Water-reflected Cylinders^a of $^{233}\text{UO}_2(\text{NO}_3)$ Solution

Conc. gU/ℓ ^b	Critical height (cm) for Diameter (cm) of:		
	<u>38.1</u>	<u>25.3</u>	<u>20.3</u>
132	11.80	15.49	21.16
95.0	c	17.92	25.40
47.9	18.06	25.90	c

- a) Aluminum cylinders with 0.15 cm wall, 1.27 cm thick bottom, no top reflector.
- b) Uranium contained 97.53% ^{233}U , 1.05% ^{234}U , 0.03% ^{235}U , 1.39% ^{238}U by weight.
- c) Insufficient material for criticality.

Table V

Bare Critical Cylinders of Low Concentration Solution

<u>Radius, cm</u>	<u>gU/l</u>	<u>wt %</u>				<u>gTh/l^a</u>	<u>gNO₃/l</u>	<u>Ht, cm</u>
		<u>²³³U</u>	<u>²³⁴U</u>	<u>²³⁵U</u>	<u>²³⁸U</u>			
155.5	14.50	97.37	1.50	0.04	1.09	0.014	8.47	50.85
	13.89	97.35	1.52	0.05	1.08	0.012	8.77	60.58
	13.22	97.30	1.49	0.05	1.16	0.014	8.24	79.04
	12.53	97.24	1.55	0.05	1.16	0.100	8.23	140.16

a) Assumed present as ThO₂ at 9.86 g/cm³

Table VI
Critical Metal Spheres

<u>Region</u>	<u>Isotopic Composition</u>	<u>Density g/cm³</u>	<u>Core radius or Reflector Thickness, cm</u>
Core	98.13% ²³³ U, 1.24% ²³⁴ U, 0.03% ²³⁵ U, 0.6% ²³⁸ U (by wt)	18.424	5.983±0.008
Core	1.02% ²³⁴ U, 93.8% ²³⁵ U, 5.18% ²³⁸ U (by wt)	18.75	8.732±0.009
Core ^a	94.79% ²³⁹ Pu, 4.9% ²⁴⁰ Pu, 0.31% ²⁴¹ Pu (by atom)	15.778	5.042
Ref1.	1.02% ²³⁴ U, 93.2% ²³⁵ U, 5.78% ²³⁸ U (by wt)	18.80	1.664±0.016
Core	98.2% ²³³ U, 1.1% ²³⁴ U, 0.7% ²³⁸ U (by wt)	18.621	5.044
Ref1.	1.02% ²³⁴ U, 93.2% ²³⁵ U, 5.78% ²³⁸ U (by wt)	18.8	1.222±0.012
Core	98.2% ²³³ U, 1.1% ²³⁴ U, 0.7% ²³⁸ U (by wt)	18.644	4.600
Ref1.	1.02% ²³⁴ U, 93.2% ²³⁵ U, 5.78% ²³⁸ U (by wt)	18.8	1.989±0.020

a) Contained 1.0% Gd by wt.

Table VII

Values of k_{eff} (1+Bias) Calculated for Critical Spheres of Table I

$H/^{233}\text{U}^a$	HRXN-ANISN				GLASS-ANISN				MGBS-TGAN
	S_4	S_8	S_{16}	S_{∞}	S_4	S_8	S_{16}	S_{∞}	
417.5	0.9955	0.9909	0.9895	0.9890	-	-	1.0381	1.0376	
413.2	0.9966	0.9919	0.9904	0.9898	-	-	1.0394	1.0388	
399.1	0.9953	0.9905	0.9891	0.9886	-	-	1.0391	1.0386	
387.5	0.9937	0.9889	0.9874	0.9868	-	-	1.0383	1.0377	
375.1	-	-	0.9875	0.9870	-	-	-	-	
662.5	1.0018	0.9985	0.9975	0.9971	-	-	1.0314	1.0310	
641.7	1.0036	1.0002	0.9992	0.9988	-	-	1.0324	1.0320	
598.0	-	-	1.0011	1.0007	-	-	-	-	
379.4	0.9821	0.9792	0.9781	0.9777	-	-	1.0363	1.0359	
406.9	0.9905	0.9858	0.9844	0.9839	1.0389	1.0343	1.0329	1.0324	1.0542
381.5	0.9813	0.9784	0.9774	0.9770	1.0390	1.0360	1.0350	1.0346	1.0736
387.2	1.0043	0.9995	0.9981	0.9976	1.0547	1.0499	1.0485	1.0480	1.0679
419.4	0.9951	0.9905	0.9891	0.9886	1.0436	1.0389	1.0375	1.0370	1.0577
426.4	0.9959	0.9913	0.9899	0.9894	1.0438	1.0393	1.0379	1.0374	1.0576
658.2	-	1.0005	0.9994	0.9990	1.0380	1.0348	1.0337	1.0333	1.0458
190.7	0.9828	0.9767	0.9749	0.9742	1.0481	1.0419	1.0401	1.0395	1.0789
268.8	0.9829	0.9773	0.9757	0.9751	1.0412	1.0356	1.0339	1.0333	1.0650
542.6	0.9913	0.9875	0.9863	0.9859	1.0315	1.0277	1.0265	1.0261	1.0430
192.3	0.9756	0.9721	0.9709	0.9704	1.0502	1.0466	1.0454	1.0449	1.1100
249.7	0.9793	0.9760	0.9749	0.9745	1.0476	1.0443	1.0431	1.0426	1.0960
345.0	0.9757	0.9728	0.9717	0.9713	1.0358	1.0328	1.0317	1.0313	1.0726
583.5	0.9925	0.9903	0.9895	0.9892	1.0357	1.0334	1.0326	1.0323	1.0526

Table VII (cont.)

$H/^{233}U$	HRXN-ANISN				GLASS-ANISM				MGBS-TGAN
	S_4	S_8	S_{16}	S_{∞}	S_4	S_8	S_{16}	S_{∞}	
1532	1.0014	1.0010	1.0008	1.0007	-	-	1.0050	1.0049	1.0076
1470	1.0008	1.0004	1.0002	1.0001	-	-	1.0044	1.0043	1.0075
1418	1.0002	0.9998	0.9996	0.9995	-	-	1.0037	1.0036	1.0073
1368	1.0009	1.0005	1.0003	1.0002	-	-	1.0045	1.0044	1.0084
1324	1.0001	0.9997	0.9995	0.9994	-	-	1.0037	1.0036	1.0080
1987	1.0041	1.0040	1.0039	1.0039	-	-	0.9964	0.9964	1.0078

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

Table VIII

Values of k_{eff} Calculated for Critical Cylinders of Table II

$H/^{233}\text{U}^a$	B_H^2, cm^{-2}			k_{eff}^b		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
42.6	0	0	0	0.98(?)	1.05(?)	-
	0.01067	0.01114	0.01324	1.0616±0.0091	1.1518	1.1679
57.9	0	0	0	0.96	1.03	1.11
c	0	0	0	0.9801	1.0563	1.1259
	< 0.00262	-	< 0.00281	> 0.9408	> 1.0170	> 1.0881
	0.00685	0.00695	0.00748	0.9793±0.0045	1.0617	1.1055
	0.01075	0.01114	0.01324	1.0358±0.0063	1.1197	1.1254
	0.01235	0.01284	0.01522	1.0422±0.0063	1.1262	1.1268
67.0	0	0	0	0.96	1.03	1.11
c	0	0	0	0.9778	1.0517	1.1166
	< 0.00205	-	< 0.00219	> 0.9496	> 1.0208	> 1.0871
	0.00649	0.00657	0.00709	0.9845±0.0047	1.0645	1.1018
	0.01093	0.01130	0.01341	1.0340±0.0064	1.1154	1.1142
84.2	0	0	0	0.965	1.03	1.10
c	0	0	0	0.9705	1.0408	1.1002
	< 0.00195	-	< 0.00207	> 0.9415	> 1.0118	> 1.0724
	0.00598	0.00605	0.00652	0.9877±0.0042	1.0639	1.0929
	0.01239	0.01279	0.01506	1.0418±0.0063	1.1194	1.1038
121	0	0	0	0.97	1.04	1.095
	0.00422	0.00424	0.00457	0.9973±0.0047	1.0668	1.0884
	0.01117	0.01146	0.01342	1.0489±0.0068	1.1207	1.0952

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Table VII(cont.)

Values of k_{eff} Calculated for Critical Cylinders of Table II

$H/^{233}U^a$	B_H^2, cm^{-2}			k_{eff}^b		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
145	0	0	0	0.98	1.045	1.085
c {	0	0	0	0.9323	0.9942	1.0512
	<0.00234	-	< 0.00250	> 0.8988	> 0.9607	> 1.0185
	0.00970	0.00910	0.01158	1.0250 0.0065	1.0934	1.0726
	0.01105	0.01132	0.01318	1.0385 0.0067	1.1071	1.0796
	152	0	0	0	0.98	1.045
	0.00308	0.00309	0.00330	0.9969 0.0033	1.0619	1.0829
	-	-	0.01319	-	-	1.0744
194	0	0	0	0.99	1.05	1.08
	0.00145	0.00145	0.00151	0.9968 0.0022	1.0563	1.0801
	0.00973	0.00992	0.01148	1.0340 0.0067	1.0972	1.0696

- a.) Ratio calculated from reported concentrations and from density formula. May differ slightly from reported ratio.
- b.) First line for each mixture is extrapolated critical value of k_{eff} for an infinite cylinder of the mixture. Uncertainty in k_{eff} corresponds to reported uncertainty of 3% in measured height and uncertainty associated with extrapolation to criticality from source multiplication curves, and was calculated by HRXN-ANISN-SPBL only. Order of listing is the same as in Table II.
- c.) First line enclosed by brace gives k_{eff} if cylinder which "apparently cannot be made critical at any height" were exactly critical at infinite height. Second line gives axial buckling and k_{eff} corresponding to maximum height achieved with available solution. GLASS-ANISN-SPBL values were inferred from HRXN-ANISN-SPBL.

Table IX

Values of k_{eff} Calculated for Critical Cylinders of Table III

$\text{H}/^{233}\text{U}^{\text{a}}$	$B_{\text{H}}^2 \text{cm}^{-2}$			$k_{\text{eff}}^{\text{b}}$		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
33.6	0	0	0	0.96	1.05	1.15
c {	0	0	0	0.9590	1.0448	1.1409
	<0.00614	-	<0.00667	>0.8832	>0.9690	>1.0652
	0.00421	0.00428	0.00453	0.9804±0.0054	1.0719	1.1463
	0.00839	0.00875	0.01045	1.0639±0.0092	1.1624	1.1913
	38.8	0	0	0	0.96	1.05
c {	0	0	0	0.9559	1.0389	1.1326
	<0.00484	-	<0.00525	>0.8953	>0.9783	>1.0722
	0.00373	0.00379	0.00401	0.9850±0.0045	1.0735	1.1444
	0.01047	0.01094	0.01313	1.0317±0.0042	1.1250	1.1484
	45.6	0	0	0	0.96	1.045
c {	0	0	0	0.9511	1.0313	1.1215
	<0.00353	-	<0.00380	>0.9063	>0.9865	>1.0772
	0.00375	0.00379	0.00402	0.9813±0.0028	1.0667	1.1331
	0.01041	0.01083	0.01300	1.0308±0.0021	1.1208	1.1385
	53.3	0	-	0	0.96	-
c {	0	-	0	0.9451	-	1.1093
	<0.00281	-	<0.00301	>0.9092	-	>1.0739
	0.00974	-	0.01208	1.0378	-	1.1372

Table IX (cont.)

Values of k_{eff} Calculated for Critical Cylinders of Table III

$H/^{233}\text{U}^{\text{a}}$	$B_{\text{H}}^2, \text{cm}^{-2}$			$k_{\text{eff}}^{\text{b}}$		
	HRXN	GLASS	MGBS	HRXN	GLASS	MGBS
73.9	0	0	0	0.965	1.05	1.12
c	0	0	0	0.9274	0.9995	1.0803
	{ 0.00160	-	0.00168	> 0.9068	> 0.9789	> 1.0603
	0.00222	0.00223	0.00234	0.0930±0.0010	1.0594	1.1127
	0.00285	0.00287	0.00306	1.0166±0.0015	1.0947	1.1352
d	0.00874	0.00886	0.00950	0.9874±0.0016	1.0688	1.0958
	0.00921	0.00947	0.01128	1.0342±0.0054	1.1155	1.1164

- a.) Ratio calculated from reported concentration and from density formula. May differ slightly from reported ratio.
- b.) First line for each mixture is extrapolated critical value of k_{eff} for an infinite cylinder of the mixture. Uncertainty in k_{eff} corresponds to reported uncertainty of 1% in measured height and uncertainty associated with extrapolation to criticality from source multiplication curves, and was calculated by HRXN-ANISN-SPBL only. Order of listing is the same as in Table III.
- c.) First line enclosed by brace gives k_{eff} if cylinder which "apparently cannot be made critical at any height with the absence of a top reflector and the presence of Unichrome" were exactly critical at infinite height. Second line gives axial buckling and k_{eff} corresponding to maximum height achieved with available solution. GLASS-ANISN-SPBL values were inferred from HRXN-ANISN-SPBL.
- d.) Unichrome assumed present, but may have been absent.

Table X

Values of k_{eff} Calculated for Critical Cylinders of Table V

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

a) Actually H/Fissile U. Includes trace of ^{235}U .

Table XI
 Values of k_{eff} Calculated for Metal Spheres of Table VI

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

Table XII

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

<u>Parameter</u>	<u>Standard</u>	<u>HRXN-ANISN</u>	<u>GLASS-ANISN</u>	<u>MGBS-TGAN</u>	<u>Proposed</u>
Mass U, g	550	530	521	497	540
Cylinder Dia, cm	11.5	10.81	10.50	10.19	10.5
Slab Thickness, cm	3.0	2.47	2.67	2.82	2.5
Volume, ℓ	3.5	3.09	2.77	2.52	2.8
Conc, g U/ ℓ	10.8	10.83	10.79	10.73	10.8
H/U	-	2383	2392	2404	2390
Areal Density, g U/cm ²	0.35	0.353	0.351	0.334	0.35

Table XIII

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

<u>Parameter</u>	<u>HRXN-ANISN</u>	<u>GLASS-ANISN</u>	<u>MGBS-TGAN</u>	<u>Proposed</u>
Mass U,g	543	536	523	550
Cylinder Diameter, cm	11.73	11.69	11.41	11.7
Slab Thickness, cm	3.13	3.41	3.48	3.1
Volume, ℓ	3.74	3.61	3.36	3.6
Conc,g U/ℓ	10.86	10.82	10.76	10.8
H/U	2371	2379	2393	2390
Areal Density, g U/cm ²	0.357	0.355	0.339	0.350

Table XIV
Limits Calculated for Dry Metal and Oxide^a

100% ²³³U

<u>Material</u>	<u>Parameter</u> ^b	<u>HRXN - ANISN</u>	<u>GLASS-ANISN</u>	<u>Standard</u>	<u>Proposed</u>
Metal	M	6.95	6.05	6.7	6.0
	D	4.90	4.53	4.6	4.5
	T	0.61	0.38	0.54	0.38
UO ₂	M	13.05	10.90		10.9
	MO	14.84	12.39		12.4
	D	7.89	7.20		7.2
	T	1.28	0.80		0.80
U ₃ O ₈	M	18.57	15.10		15.1
	MO	21.97	17.86		17.8
	D	9.94	8.98		9.0
	T	1.79	1.12		1.1
UO ₃	M	21.89	17.56		17.5
	MO	26.40	21.17		21.1
	D	11.07	9.95		9.9
	T	2.09	1.31		1.3

a) Densities of U, UO₂, U₃O₈, and UO₃ may not exceed 18.65, 10.76, 8.15, and 7.16 g/cm³.

b) M = Mass of U in kg. MO = Mass of uranium oxide in kg.

D = Cylinder diameter in cm. T = Slab thickness in cm.

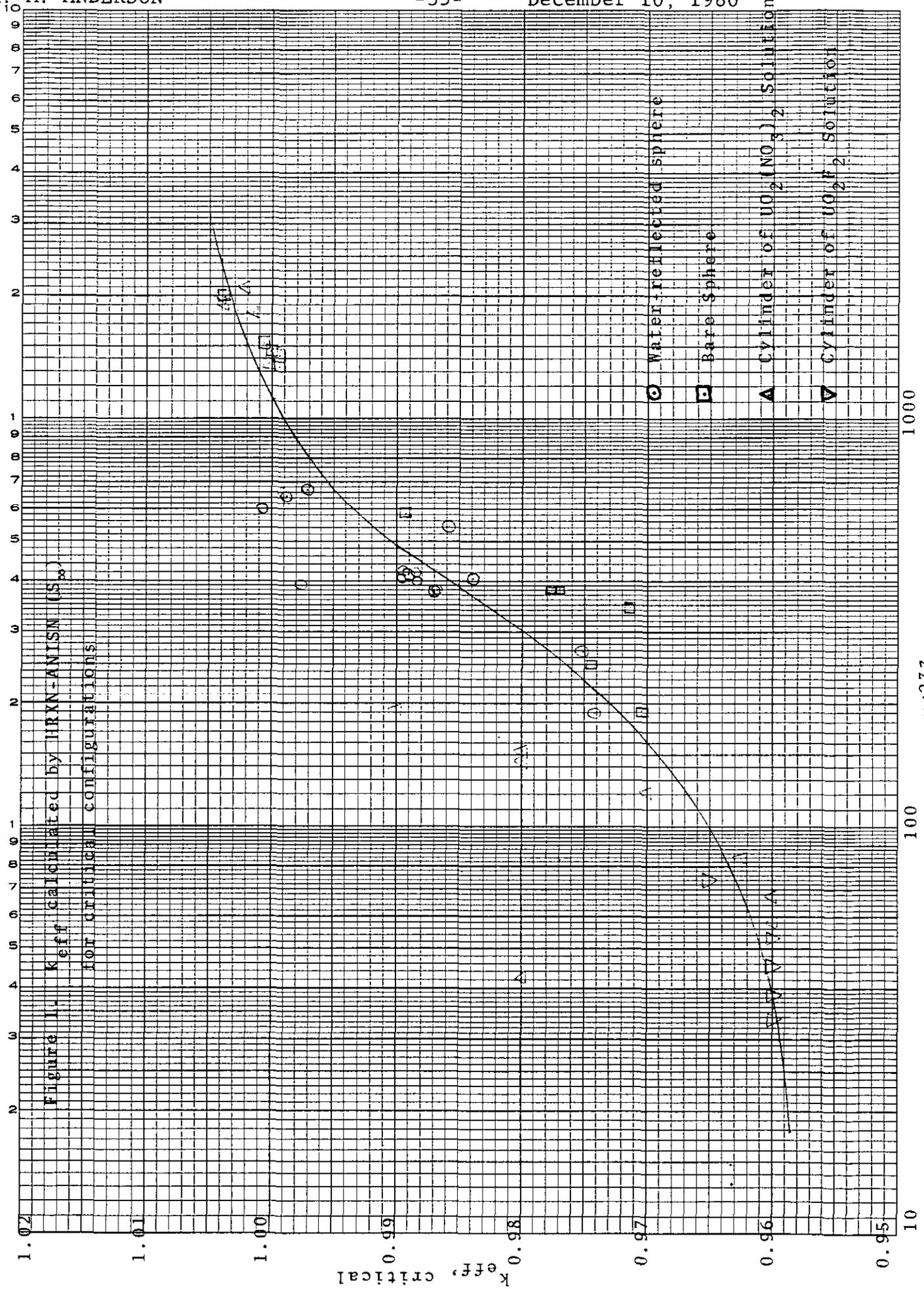
Table XV

Limits Calculated for Moist^a Oxide

<u>Density</u> ^b	<u>Oxide</u>	<u>Parameter</u> ^c	<u>HRXN - ANISN</u>	<u>GLASS - ANISN</u>	<u>Proposed</u>
Full	UO ₂	M	13.00	10.15	10.1
		MO	15.01	11.72	11.7
		D	8.35	7.44	7.2
		T	1.42	0.87	0.80
	U ₃ O ₈	M	17.62	13.38	13.4
		MO	21.17	16.07	16.0
		D	10.22	9.01	9.0
		T	1.90	1.17	1.1
	UO ₃	M	20.39	15.26	15.2
		MO	24.96	18.69	18.7
		D	11.26	9.88	9.9
		T	2.19	1.34	1.3
Half ^d	UO ₂	M	32.69	23.40	23.4
		MO	37.75	27.02	27.0
		D	14.26	12.31	11.9
		T	2.84	1.74	1.6
	U ₃ O ₈	M	44.06	30.50	30.5
		MO	52.92	36.64	36.6
		D	17.48	14.91	14.8
		T	3.80	2.34	2.2
	UO ₃	M	50.93	34.68	34.7
		MO	62.35	42.46	42.4
		D	19.28	16.36	16.3
		T	4.37	2.68	2.6

Table XV (cont.)

- a) Oxide contains 1.5% H₂O.
- b) Full density of moist oxide is based on the assumption that the volume of moist oxide is the sum of the volume of dry oxide at theoretical density (10.76, 8.15, and 7.16 g/cm³, respectively, for UO₂, U₃O₈, and UO₃) and the volume of water at 20° with density 0.99823 g/cm³.
- c) M = Mass of U in kg, MO = Mass of moist oxide in kg, D = cylinder diameter in cm, T = slab thickness in cm.
- d) Densities of oxide and water are halved, i.e. moist oxide contains 50% voids.



10

100

1000

H/233U

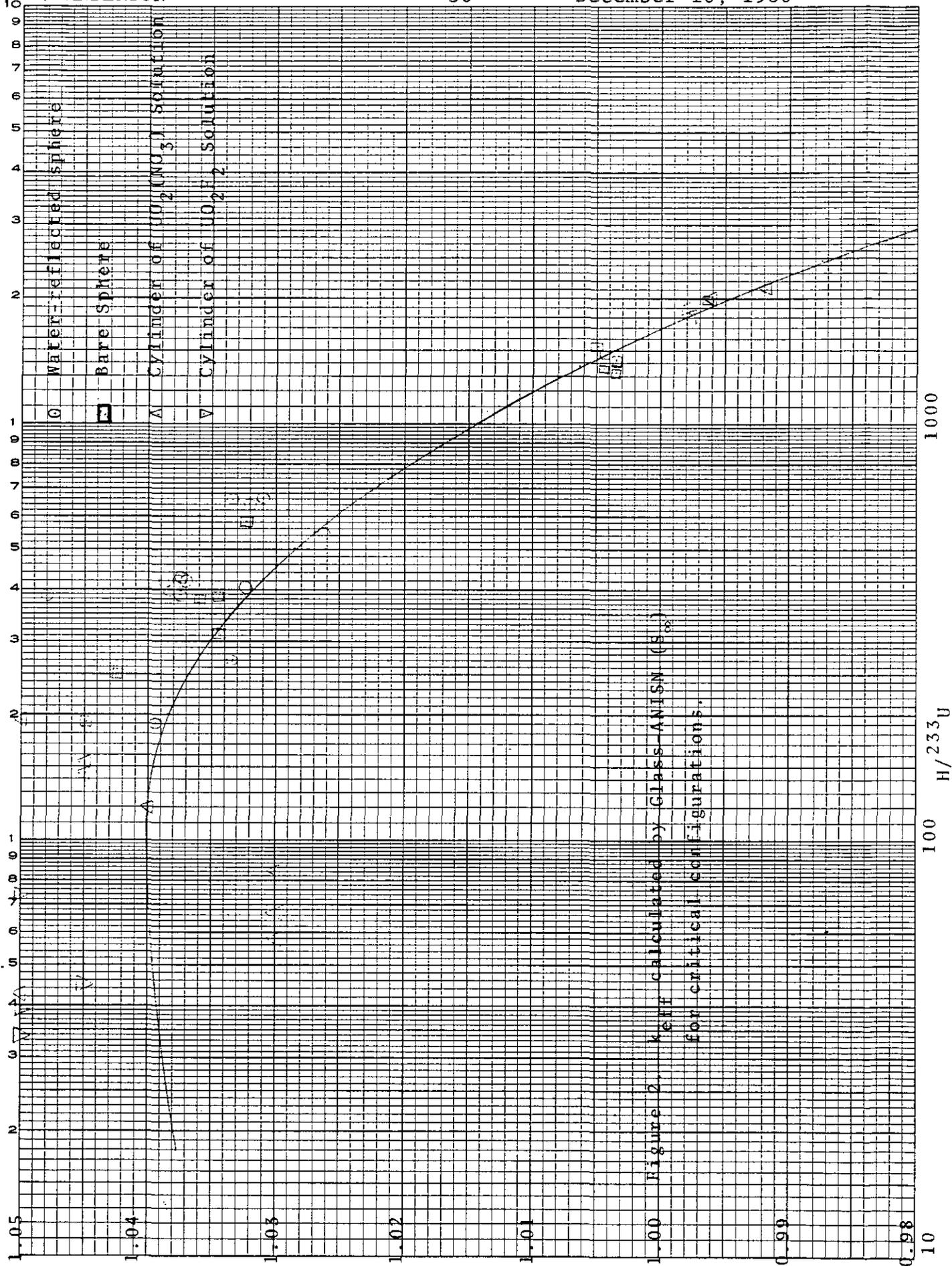


Figure 2. k_{cfl} calculated by Glass-ANISN (S_{∞}) for critical configurations.

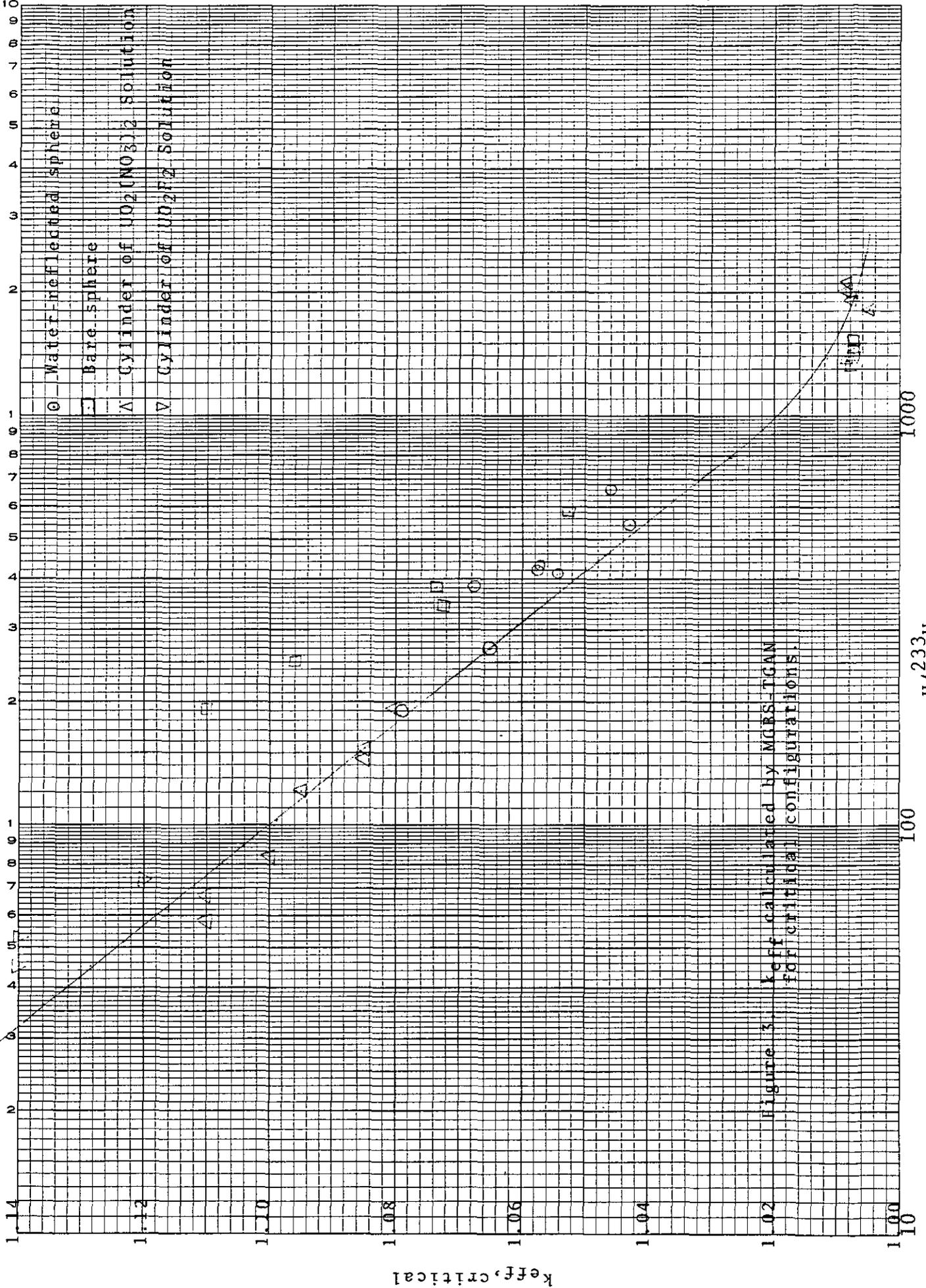


Figure 3. k_{eff} calculated by MGBS-TGAN for critical configurations.

H/233U

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Appendix

To gain a better understanding of the application of one-dimensional methods to two-dimensional problems, i.e. finite cylinders, some benchmark cases were calculated by TWOTRAN¹⁻³ and were analyzed in various ways by one-dimensional methods. The cases selected were cylinders of $^{233}\text{UO}_2\text{F}_2$ solution containing 400 g $^{233}\text{U}/\ell$ with various height (H) to diameter (D) ratios reflected by 15 cm of water. In some cases an aluminum wall was interposed. To limit computer time, the calculations were made with two energy groups, isotropic scattering, and no upscatter. The macroscopic cross sections were generated by GLASS from ENDF/B-IV cross sections and are given in Table A.1. Empirical mesh formulas⁴ for R and Z have been incorporated in the KOKO subroutines³ that prepare TWOTRAN input. The radial mesh is finer by a factor of perhaps 4. As with ANISN the mesh is often impractically fine, and a scheme is provided for making the mesh coarser where the flux varies least rapidly. Initial calculations were made using this scheme. Subsequent calculations were made with a uniform mesh in each material, using 0.2 times the number of radial intervals and 0.8 times the number of axial intervals the formulas prescribed. Results differed insignificantly, and it was concluded that the mesh was sufficiently fine. Typically, the number of mesh volumes was of the order of 600. Quadrature was S_{16} to give an accurate solution. Moreover, ANISN was used to calculate the infinite slab and the sphere, and previous calculations^{3,5} have shown a disagreement between TWOTRAN and ANISN for the infinite cylinder corresponding to about 1% in k_{eff} with only S_4 quadrature; agreement is much better with S_{16} . The CPU time was about 20 minutes for each cylinder. Results are given in Table A.2. The code indicated that the problems were converged in all cases, despite the specified inner iteration limit of 10 always being reached in the thermal group.

The first method applied to these benchmarks was ANISN-SPBL⁶ with the P_0 cross sections of Table A.1 and with S_{16} quadrature. In this approach k_{eff} is calculated for each dimension of a finite cylinder with the other dimension assumed infinite. Geometric bucklings are calculated (by B_1) corresponding to each value of k_{eff} and are added to obtain the total geometric buckling. The value of k_{eff} corresponding to this buckling is then calculated (again by B_1). Table A.3 gives results obtained by this method for the benchmark cases of Table A.2. The method overestimates k_{eff} for finite cylinders, but the overestimate decreases as the infinite cylinder is approached (i.e. as axial buckling approaches zero) and k_{eff} becomes very nearly a linear function of axial buckling. (The failure of k_{eff} to be exactly unity for the infinite cylinder in Table A.3 represents the slight discrepancy between ANISN and TWOTRAN with S_{16} quadrature). Thus, linear extrapolation of k_{eff} as a function of axial buckling should be a valid procedure for obtaining the critical value of k_{eff} for an infinite cylinder and hence the bias of the calculational method. An additional test of this thesis was made by repeating the ANISN-SPBL analysis of the benchmarks, but with Hansen-Roach cross sections (16 groups, P_1 scattering.) The aluminum-walled cylinders and the cylinders with $H/D=0$ and 0.25 were omitted. Results are given in Table A.4, and again k_{eff} is nearly linear with B_H^2 at small B_H^2 , albeit with

slightly larger slope. The low values of k_{eff} are consistent with the finding that at high concentrations of ^{235}U ENDF/B-IV cross sections underestimate the critical mass whereas Hansen-Roach cross sections overestimate it.

Another method of analyzing two dimensional critical bodies by one-dimensional codes, the one incorporated in TGAN, is to search for the critical transverse buckling corresponding to each critical dimension. The geometric buckling of a finite cylinder is then $B_g^2 = 2B_c^2 - B_r^2 - B_h^2$ where B_c^2 is the critical buckling calculated from composition and cross sections and B_r^2 and B_h^2 are, respectively, the transverse (radial) buckling calculated to make a slab with thickness equal to the cylinder height critical and the transverse (axial) buckling calculated to make the cylinder critical. The value of k_{eff} calculated for the critical finite cylinder is then the value calculated to correspond to this geometric buckling. This method, implemented by ANISN with the cross sections of Table A.1, was applied to the benchmarks. In similar calculations,⁵ discrepancies have been found in that k_{eff} calculated by ANISN at the critical transverse buckling determined by ANISN deviated somewhat from unity, but such discrepancies were not found in the present case. It is not known whether the previous discrepancies were due to the magnitude of the transverse bucklings (some were negative) or the number of energy groups (16 versus 2). The transverse leakage is calculated as $D B_r^2$ and is treated as an equivalent absorption. As the codes are formulated, $D = 1/3 \Sigma_{tr}$ when scattering is isotropic, but poor results were obtained ($k_{eff} \approx 0.95$). Much better results were obtained with

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

the correct transport theory expression for isotropic scattering. As the codes are formulated this value of D is placed in the P_1 table when scattering is linearly anisotropic. (The P_0 and P_1 tables have the same structure, but positions occupied by Σ_a , $\nu \Sigma_f$, and Σ in the P_0 table are available for other parameters in the P_1 table. D is placed in the Σ location.) To perform the calculation P_1 cross sections were generated by GLASS so as to obtain D , P_1 transfer cross sections were set to zero, and P_0 cross sections were modified to conform to Table A.1. (The cross section changes were made in the records in the jobdata set). Results of the calculations are given in Table A.5. With only water reflection, the method gives very good results for the finite cylinders, but with the aluminum wall interposed, k_{eff} is too low due to streaming in the aluminum resulting from the assumption of separability.

The same method as implemented by TGAN with diffusion theory constants was also applied to the benchmarks. In one case the constants were derived by GLASS; in the other, by MGBS. Results are given in Table A.6. Diffusion theory agrees fairly well with transport theory. The large values of k_{eff} calculated with MGBS cross sections are consistent with

the biases found in correlations with experiment. Again, the effect calculated for the aluminum wall is too large.

The benchmarks also permit a comparison to be made between the minimum critical volume of a cylinder and of a sphere. It has been suggested^{7,8} that for highly undermoderated systems surrounded by a moderating reflector the critical volume of a cube may be slightly less than that of a sphere. The minimum volume of a cylinder should then also be expected to be less than that of a sphere although by a smaller amount. The reciprocal of the height in Table A.2 is nearly a linear function of the reciprocal of the diameter. Five point Lagrange interpolation was used to obtain intermediate values of H and D, and a minimum volume of 2602.0 cm³ was determined at H/D=0.87. The volume of the sphere, 2579.0 cm³, is 0.89% less. In this case, then, the sphere has the smallest volume. An XY calculation was also made with TWOTRAN for comparison with a cylinder. The value of keff was set at 1.1996, corresponding to two-thirds the critical buckling to approximate a cube. The cylinder radius, calculated by ANISN, was 7.2018 cm for a area of 162.94 cm². The dimensions of the square cuboid, calculated by TWOTRAN, were 6.3785 x 6.3785 cm for an area of 162.74 cm². The square cross section has 0.12% less area than the circular cross section, but perhaps this difference results from the slight difference between ANISN and TWOTRAN with S₁₆ quadrature.

Table A.1

Two Group Cross Sections

<u>Material</u>	<u>Group</u>	<u>Σ_a</u>	<u>$\nu\Sigma_f$</u>	<u>Σ</u>	<u>$\Sigma_{g \rightarrow g}$</u>	<u>$\Sigma_{g \rightarrow g+1}$</u>
U Solution	1	0.020794	0.044029	0.267053	0.222033	0.024226
	2	0.362821	0.800351	1.39704	1.03421	0
H ₂ O	1	0.000458	0	0.255747	0.204324	0.050965
	2	0.018972	0	2.23097	2.21200	0
Al	1	0.000419	0	0.135803	0.135163	0.000221
	2	0.011993	0	0.089297	0.077304	0

Table A.2

Benchmark Cylinders

<u>H/D</u>	<u>Wall</u>	<u>D(cm)</u>	<u>H(cm)</u>
0 ^a	None	∞	2.73126
	0.16 cm Al	∞	2.75136
0.25	None	25.9620	6.4905
0.50	None	19.1352	9.5676
	0.16 cm Al	19.2670	9.6335
1.0	None	14.9304	14.9304
2.0	None	12.4237	24.8474
	0.16 cm Al	12.5212	25.0424
4.0	None	11.0869	44.3476
∞	None	10.2008	∞
	0.16 cm Al	10.2496	∞
Sphere ^a	None	17.0144	-
	0.16 cm Al	17.1014	-

a) Calculated by ANISN

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Table A.3

Application of ANISN-SPBL to Benchmarks

<u>H/D</u>	<u>Wall</u>	<u>B_H^2, cm^{-2}</u>	<u>k_D</u>	<u>k_H</u>	<u>k_{eff}</u>
0	None	0.04698	2.1644	1.0000	1.0000
	A λ	0.04698	2.1644	1.0000	1.0000
0.25	None	0.02604	1.5545	1.2903	1.0705
0.5	None	0.01859	1.3722	1.4494	1.0719
	A λ	0.01839	1.3736	1.4544	1.0767
1.0	None	0.01182	1.2212	1.6402	1.0570
2.0	None	0.00641	1.1118	1.8403	1.0340
	A λ	0.00631	1.1146	1.8445	1.0374
4.0	None	0.00284	1.0462	2.0066	1.0148
∞	None	0	0.9995	2.1644	0.9995
	A λ	0	0.9994	2.1644	0.9994

Table A.4

ANISN-SPBL with Hansen-Roach Cross Sections Applied to Benchmarks

<u>H/D</u>	<u>B_H², cm⁻²</u>	<u>k_D</u>	<u>k_H</u>	<u>k_{eff}</u>
0.5	0.01924	1.2955	1.3768	0.9604
1.0	0.01213	1.1282	1.5762	0.9429
2.0	0.00651	1.0043	1.7757	0.9159
4.0	0.00286	0.9292	1.9337	0.8935
∞	0	0.8756	-	0.8756

Table A.5

Analysis of Benchmarks by Critical Transverse Buckling
Implemented by ANISN

<u>H/D</u>	<u>Wall</u>	<u>B_H^{2 a}</u>	<u>k_{eff}</u>
0	None	0.046980	1.0000
	A _l	0.046980	1.0000
0.25	None	0.03065	0.9982
0.50	None	0.02261	0.9980
	A _l	0.02363	0.9813
1.00	None	0.01440	0.9988
2.00	None	0.00756	1.0001
	A _l	0.00785	0.9937
4.00	None	0.00318	1.0003
∞	None	0	0.9995
	A _l	0	0.9994

a) B_H = Calculated critical buckling - calculated critical radial buckling.

Table A.6

Analysis of Benchmarks by TGAN

<u>H/D</u>	<u>Wall</u>	k_{eff}	
		<u>GLASS</u>	<u>MGBS</u>
0	None	1.0098	1.0657
	A ℓ	1.0093	1.0593
0.50	None	0.9964	1.0726
	A ℓ	0.9784	1.0498
1.00	None	0.9968	1.0729
2.00	None	0.9990	1.0722
	A ℓ	0.9922	1.0605
4.00	None	1.0008	1.0712
	None	1.0014	1.0699
	A ℓ	1.0017	1.0648
Sphere	None	1.0001	1.0728
	A ℓ	1.0003	1.0682