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CRITICALITY AND NUCLEAR SAFETY OF SLIGHTLY ENRICHED URANIUM

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Fissile materials must be handled in such a manner as to prevent inadvertent criticality outside reactors. Criticality reviews are thus required in fuel element fabrication processes and reactor fuels reprocessing operations involving enriched uranium. Safe mass or volume limits can vary widely subject to the degree of moderation and homogeneity of the uranium-water systems under consideration. In connection with these criticality problems, mass and volume limits are discussed for slightly enriched uranium solution, solid uranium rods, and uranium tubes (hollow rods), including some comments on criticality problems in handling large billets. Factors affecting criticality are reviewed together with methods of criticality control for uranium with enrichment in the range 0.72 to 5%.

Many problems which are only of academic interest in the design of a reactor become of principal importance in nuclear safety applications—where the objective is to prevent inadvertent criticality in nonreactor environments. The criticality problems encountered in the handling and processing of fissile materials outside reactors are exceedingly complex; not only must criticality be considered for the standard operating conditions of the plant, but also for those off-standard conditions which are physically possible and perhaps more favorable for the chain reaction. In determining the nuclear safety of a uranium processing plant or metal fabrication operation, all aspects of reactor theory may be involved; the problem may be further enhanced because of the lack of certain experimental data, and the uncertainties which exist in theoretical values.

Consider the conditions under which a given quantity of fissile material will become chain reacting. The exact physical configuration, or in other words, the density in space of each kind of atom, must be known as a function of position. Criticality thus depends not only on the quantity of fissile material present, but on the size, shape, and material of the containment vessel; on the distribution of the material within the vessel; on the volume of solvent or solids in solution; and on the presence of materials which may act as neutron reflectors.

In the following presentation, the variables that affect the criticality and nuclear safety of slightly enriched uranium are reviewed. References to water mean light water.

CHARACTERISTICS OF SLIGHTLY ENRICHED URANIUM SYSTEMS

The critical mass for slightly enriched uranium depends in a very sensitive manner on the degree of enrichment and moderation, and on the form of the fissile material, due mainly to resonance absorption in U-238.

For *homogeneous* uranium-water solutions, there is one enrichment for which criticality is possible with only one hydrogen-U-235 ratio; this is the limiting enrichment for criticality. From k_∞ measurement data, this enrichment was found to be 1.034 ± 0.010% U-235 (*1*). At this particular enrichment, the largest value which can be obtained for the infinite reproduction factor (k_∞) under optimum conditions of moderation is unity. For nuclear safety applications, 1.02% U-235 (that is, adjusting the 1.03% value for the uncertainties in the measurement) is considered the enrichment below which uranium homogeneously dispersed in water cannot be made critical.

For *heterogeneous* systems of uranium in water, criticality considerations must be given to all enrichments *above* 0.72 wt. % U-235 (natural uranium). Although experimental data indicate that a heterogeneous system of natural uranium can be made critical in a water lattice (*2*, *3*), this fact is actually only of academic interest in nuclear safety applications, since the quantity of uranium would be large, and the possibility of obtaining the required amount inadvertently under optimum conditions would be extremely remote.

Note the sharp difference between the homogeneous and heterogeneous systems. A homogeneous mixture of UO_3 and water consisting of 1.00% U-235 enriched uranium cannot be made critical, whereas the minimum critical mass of a heterogeneous system consisting of fuel rods of this same enrichment in water is only about 2,000 kg. U (20 kg. U-235) (*1*, *4*).

In the case of *unmoderated* uranium metal, the limiting enrichment below which criticality is not possible, is between 5 and 6%. Exponential meas-

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urements made at Los Alamos indicate that the limiting critical enrichment is about 5.5% U-235 (*5*). These measurements were made on cylindrical columns of uranium metal ranging in U-235 content from 0.72 to 9.18%. The limiting critical enrichment for uranium metal has also been calculated by Richey and Carter with the Monte Carlo method (*6*, *7*) and by Chernick et al. (*8*). Richey and Carter obtained values of 5.8 and 5.3% U-235, and Chernick et al. obtained values of 5.5, 6.1, and 6.5%, using different cross-section sets.

Thus, if it were possible to exclude moderation in fuel processing and metal fabrication operations, no criticality limits or other nuclear safety restrictions would be required for uranium metal under 5% enrichment. On the other hand, if 5% enriched uranium fuel elements were dissolved in an aqueous solution ($UO_2F_2 + H_2O$ mixture, for example) as little as 1.8 kg. U-235 could be made chain reacting. The effect of moderation is to reduce the critical mass from an infinite value to about 1.8 kg. U-235, which is only about a factor or two larger than the minimum mass for a pure U-235 solution.

For uranium enrichments above 5%, the smallest critical masses will be obtained with homogeneous systems (*3*), rather than heterogeneous systems. Note the extreme reverse of this when the enrichment is 1% U-235. Criticality is not at all possible for a 1% homogeneous aqueous solution, whereas the minimum mass for a heterogeneous system of 1% enriched uranium rods in a water lattice is only about 20 kg. U-235.

It is therefore evident that criticality (and nuclear safety) of slightly enriched uranium depends strongly on the degree of moderation and homogenization of the fuel-moderator system. If the possibility of moderation can be completely excluded, there is no criticality problem at all for enrichments less than about 5%; on the other hand, if optimum conditions of moderation and lattice spacing must be assumed, then criticality controls in handling and processing operations may be extremely restrictive. To be practical, nuclear safety controls should be based on a realistic evaluation of actual operating conditions in the plant, taking into account the possibility of credible accidents, such as water flooding, over-accumulation of material, departure from safe geometry, error in fuel enrichment, etc.

CRITICALITY OF SLIGHTLY ENRICHED HOMOGENEOUS SYSTEMS

CRITICAL MASS AND VOLUME MEASUREMENTS

Critical mass and volume measurements for homogeneous, hydrogen moderated uranium systems have been performed at enrichments of 4.9, 2.0, and 1.42 wt.% U-235 (Figures 1 and 2). Extensive measurements have been made at the Oak Ridge National Laboratory (*3*, *9*) with 4.9% U-235 enriched UO_2F_2 solution and 4.9% U-235 enriched U_3O_2-sterotex blocks. Sterotex (glyceryl tristearate, $C_{57}H_{110}O_6$) has a hydrogen density and nuclear properties very similar to those of water and was used to extend the measurements into the lower range of moderator ratios not obtainable with UO_2F_2 solution. From these measurements, the minimum critical mass (spherical geometry-water reflected) for 4.9% U-235 enriched uranium solutions was found to be 1.79 kg. U-235 (36.5 kg. U).

Measurements with 2.0% U-235 enriched uranium were also made at Oak Ridge (*3*, *10*). In these experiments, UF_4-paraffin blocks were used to obtain the desired hydrogen–U-235 ratios. The minimum critical mass for 2.0% U-235 enriched uranium solutions was found to be 5.0 kg. U-235 (250 kg. U).

Arrays of water reflected UF_4-paraffin compacts containing 1.42% U-235 enriched uranium were studied at Dounreay (*5*). The results for two hydrogen–U-235 ratios, 418 and 562, are shown in Figures 1 and 2. From these data, minimum critical mass for uranium atoms in water is estimated to be about 20 kg. U-235 (1,408 kg. U).

Based on these measurements, a curve of minimum critical mass as a function of U-235 enrichment for slightly enriched homogeneous systems is shown in Figure 3.

Critical mass calculations for slightly enriched homogeneous systems have been made by Mills and Bell (*11*). In their study, critical masses were calculated for solutions and compacts similar to those used in the experimental measurements. The calcu-

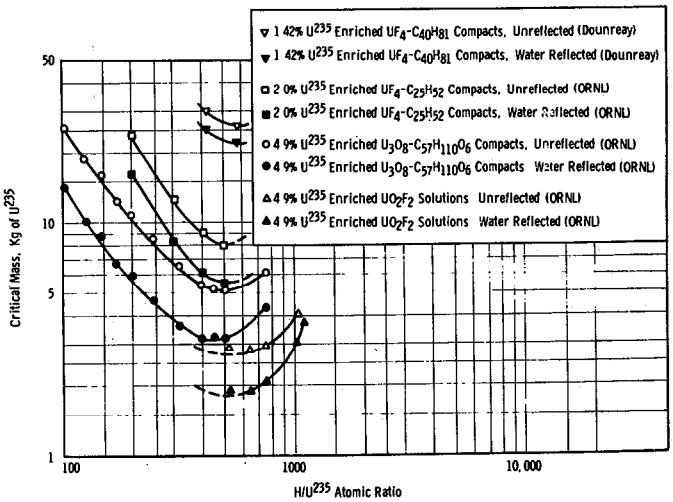

Fig. 1. Critical mass of U-235 enriched uranium in spherical geometry as a function of hydrogen–U-235 atomic ratio as determined from experimental measurements.

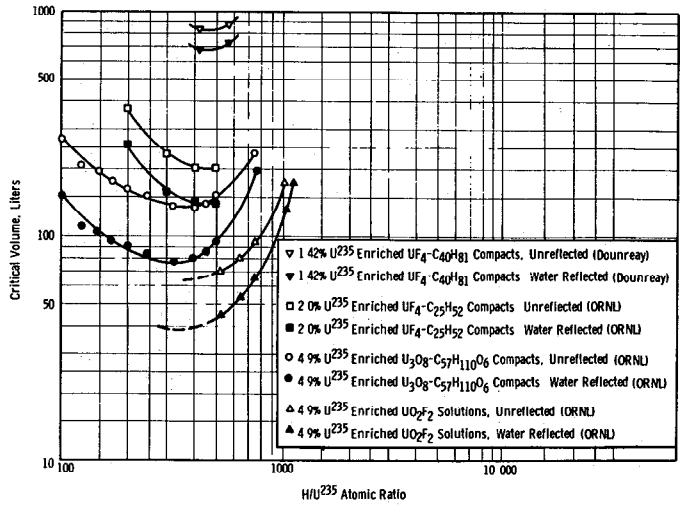

Fig. 2. Critical volume of U-235 enriched uranium in spherical geometry as a function of hydrogen-U-235 ratio as determined from experimental measurements

lations gave k_{eff} = 0.995 for the 1.4% U-235 enriched uranium critical experiments of Dounreay and k_{eff} = 1.03 ± 0.02 for the bare 2.0% and 4.9% critical assemblies of Oak Ridge, which is good agreement. To be in exact agreement with the experiments, k_{eff} would be unity in each case.

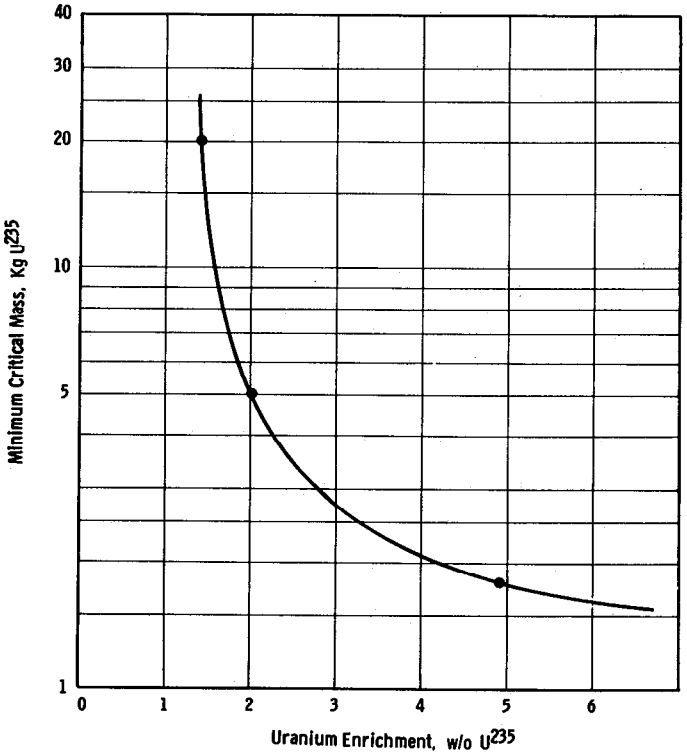

Fig. 3. Miniumum critical mass for uranium solution as a function of U-235 enrichment (spherical geometry-water reflected).

MAXIMUM k_∞ VS. ENRICHMENT

Measurements of k_∞ for slightly enriched UO_3 solution systems have been made with 1.006, 1.070, 1.159, and 3.04% U-235 in the Hanford PCTR (*1*, *12*). Also, k_∞ for 2.0% U-235 enriched UF_4-paraffin blocks has been measured at a hydrogen-uranium ratio of 3.9 at both Hanford (*13*) and Oak Ridge (*14*). The results of these measurements are shown in Figure 4.

Of particular interest in these measurements are the maximum values of k_∞ and the H-U ratios at which k_∞ is unity. A curve of maximum k_∞ as a function of enrichment is shown in Figure 5. The letter U signifies total uranium U-235 and U-238.

MODERATOR RATIOS AT WHICH k_∞ = 1

For homogeneous water systems, it is possible to obtain criticality with slightly enriched uranium only over a limited range of moderation or hydrogen-U-235 ratios; these ratios, for which k_∞ = unity, are of particular interest in nuclear safety applications. At high values of hydrogen-U-235, excess neutron absorption in hydrogen reduces k_∞ to values less than unity, whereas for low values of hydrogen-U-235, the high resonance capture in U-238 reduces k_∞ to less than unity. Consequently, if a uranium solution is either sufficiently diluted or highly enough concentrated, criticality will not occur.

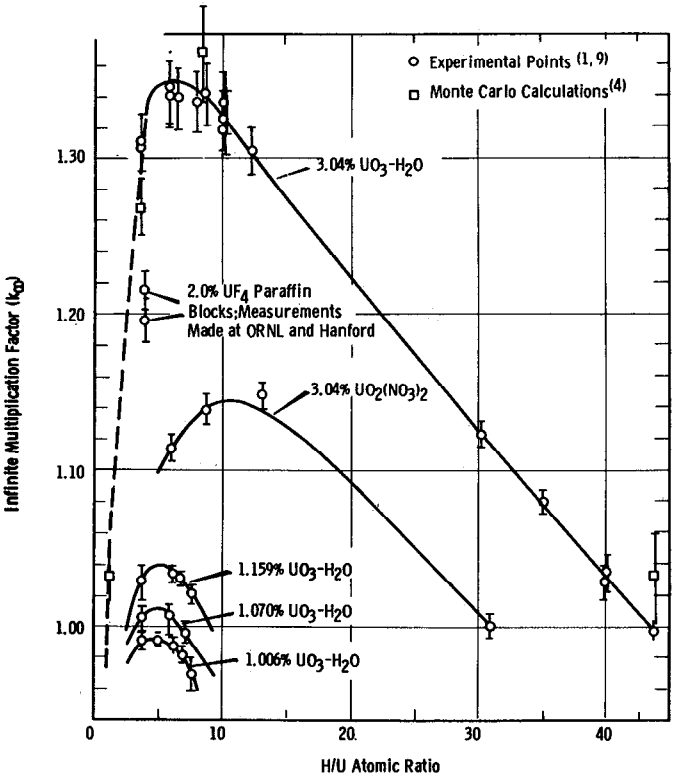

Fig. 4. k_∞ for UO_3-water and uranyl nitrate systems as a function of hydrogen-uranium atomic ratio.

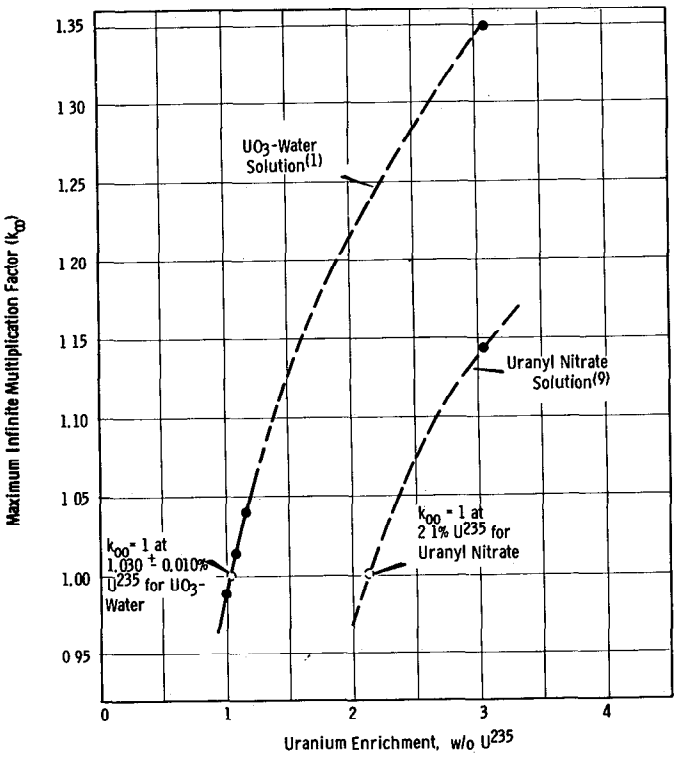

Fig. 5. Maximum k_∞ for UO_3-water and uranyl nitrate systems as a function U-235 enrichment.

Several H-U ratios at which k_∞ = unity are shown in Figure 4. These points are plotted in Figure 6 to show limiting H-U (total U) ratios as a function of U-235 enrichment. The minimum point of this curve is the limiting enrichment for criticality of 1.03% U-235 (which was mentioned above), for which criticality is possible only with one H-U ratio. From Figure 6, one may obtain, for a given enrichment, the lowest and highest values of H-U for which criticality is possible with uranium solutions.

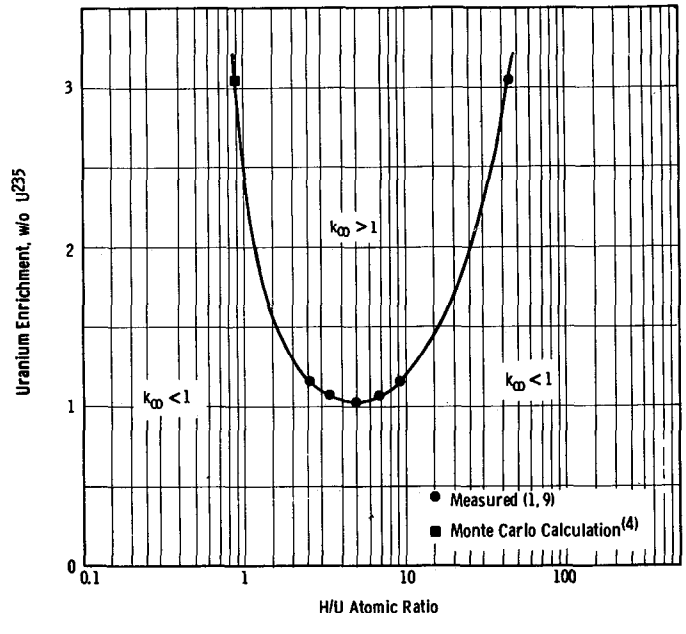

Fig. 6. Hydrogen-uranium atomic ratios at which ∞ = 1 for UO_3-water systems as a function of U-235 enrichment.

Mills and Bell have computed the hydrogen-U-235 ratios for which k_∞ = 1 as a function of the enrichment by means of a multigroup transport code (*11*). In these computations, a limiting critical enrichment of about 1.0% U-235 was obtained, which is in agreement with the value of 1.03% U-235 measured by Neeley and Handler (*1*).

Monte Carlo calculations have been made by Richey for 3.04% U-235 enriched UO_3 systems having hydrogen-to-uranium atom ratios in the range of 3.57 to 43.87 (*6*). One purpose of these calculations was to estimate the H-U ratio for which k_∞ = 1 in the very low hydrogen-uranium ratio range not covered by the experiments. These calculated results, also shown in Figures 4 and 6, indicate k_∞ = 1 at hydrogen-uranium ratios of 0.9 and 46, which is in agreement with experimental values and the values calculated by Mills and Bell (*11*).

NEUTRON ABSORBER—BORON REQUIRED TO REDUCE k_∞ TO UNITY

During the course of the Hanford experiments, the quantity of boron required to reduce k_∞ (max) of the homogeneous solutions to unity was determined (*12*). For the 3.04% U-235 enriched UO_3, the amount required was 0.0113 ± 0.0003 atom of natural boron per atom of uranium (0.36 atom of boron per atom of U-235). For 3.04% U-235 enriched $UO_2(NO_3)_2$, 0.0055 ± 0.0003 atom of boron per atom of uranium (0.18 atom of boron per atom of U-235) was required. Since it is also known that k_∞ = 1 at 1.03% U-235 for UO_3 in water and about 2.1% U-235 for uranyl nitrate, curves of boron poisoning as a function of enrichment for the two systems are estimated in Figure 7.

EFFECT OF NITRATE ON CRITICALITY

In chemical processing operations, the uranium fuel is usually dissolved in nitric acid solution. There are some data which may be used to evaluate the effect of nitrate on the criticality of the uranium-nitrate solution systems (fuel elements after dissolution).

From Hanford PCTR experiments, the maximum value of k_∞ for 3.04 wt. % U-235 enriched $UO_2(NO_3)_2$ was found to be 1.145 ± 0.01 and the H-U ratio for which k_∞ = unity was 31.2 ± 1.0. The maximum value of k_∞ measured for the corresponding 3% UO_3 hydrogenous mixture was 1.350 ± 0.013 and the hydrogen-uranium ratio for k_∞ equal unity was 43.9 ± 0.5. These results are shown in Figure 4.

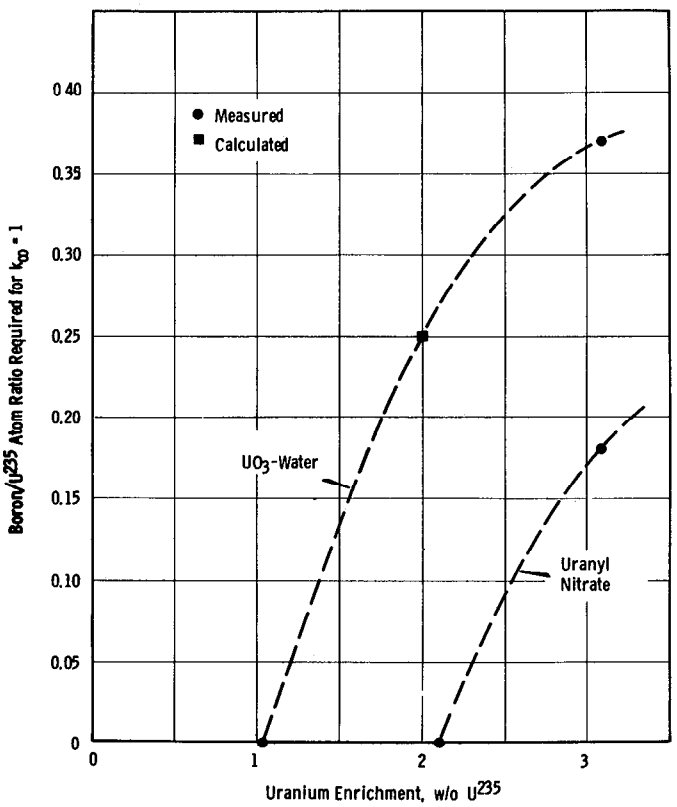

Fig. 7. Estimated amount of boron required in UO_3-water and uranyl nitrate systems to reduce ∞ to unity as a function of U-235 enrichment.

Measurements of k_∞ were also made in the Hanford PCTR for uranyl nitrate solutions ($UO_2(NO_3)_2$-hydrogenous mixtures) in the enrichment range of 2.1 to 2.3% U-235. Unfortunately, the quantity of material used in these measurements was insufficient to permit a reliable estimate of the experimental error. The data, however, imply that the limiting critical enrichment for a $UO_2(NO_3)_2$ solution is about 2.1% U-235. It should be emphasized that this is an estimated value based on the results of these preliminary experiments. The experiments, however, do represent the only data available for uranyl nitrate systems of this enrichment. The results indicate that nitrate has an appreciable effect on the limiting critical enrichment, since the limiting enrichment for UO_3 in water is 1.03% U-235.

EFFECT OF OXYGEN ON CRITICALITY

The effect of oxygen on the criticality of unmoderated uranium (UO_3) is of particular interest in connection with the minimum hydrogen-uranium atomic ratio for criticality of an infinite system ($k_\infty = 1$). Some interesting results were obtained from Monte Carlo calculations of k_∞ for unmoderated 3.04% enriched uranium metal and for UO_3. The results are given below (*6*).

MONTE CARLO CALCULATIONS OF k_∞ FOR DRY 3.04 WT.% U-235 ENRICHED URANIUM

	k_∞
Uranium metal	0.720 ± 0.012
UO_3	0.584 ± 0.019

The dry UO_3 salt is seen to have a value of k_∞ which is actually less than the value for uranium metal. The smaller k_∞ value for the UO_3 system is primarily due to scattering and moderation by the oxygen. The oxygen degrades the fast neutron spectrum slightly, which reduces fast fission in U-238 and enhances resonance absorption in U-238. It is estimated that for UO_3 the median capture energy shifts from 0.1 to 0.2 MeV down to 0.025 to 0.050 MeV, and the median fission energy shifts from 0.4 to 0.5 MeV down to 0.075 to 0.1 MeV. In the case of uranium metal, the only significant moderating effect the neutrons experience is due to inelastic scattering.

The net effect of the oxygen in dry 3.04 wt.% U-235 enriched UO_3 appears to be a reduction in k_∞ of approximately 136 mk.

CRITICALITY OF SLIGHTLY ENRICHED HETEROGENEOUS SYSTEMS

HETEROGENEOUS SYSTEMS OF SOLID FUEL RODS

Several hundred exponential and neutron multiplication measurements have been performed at Hanford with uranium rods enriched up to 3% U-235 (*15* to *17*). The data from these measurements, together with bibliography of detailed reports, have been compiled by Lloyd (*18*). The most extensive series of experiments was conducted with 3.06 wt.% U-235 enriched uranium rods, in which exponential and neutron multiplication measurements were made with four different rod diameters ranging from 0.175 to 0.925 in.

Maximum buckling and minimum critical mass values derived from multiplication and exponential experiments, which serve as a basis in nuclear safety evaluations, are presented in Figure 8. Where there were insufficient experimental data, the results have been extended by calculations (*4*).

Curves of minimum critical mass and minimum critical volume (spherical geometry) as a function of enrichment have been constructed from the experimental data. Similarly, constant buckling conversions have been used to compute the minimum critical dimensions of infinite length cylinders and infinite slabs. These results are presented in Figures 9, 10, 11, and 12. Also given are curves for homogeneous systems (*3*) based on UO_2 and UO_2F_2 in water (not uranium atoms in water).

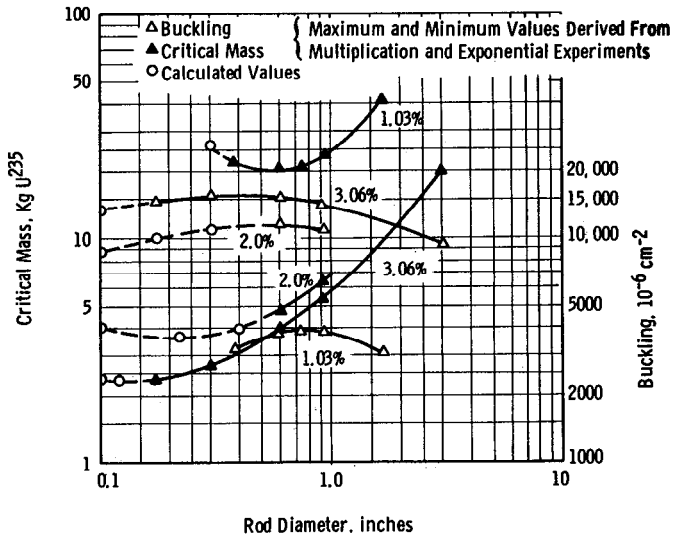

Fig. 8. Minimum water reflected critical mass and maximum buckling for lattices of 1.03, 2.0, and 3.06 wt. % U-235 enriched uranium rods in water as a function of rod diameter.

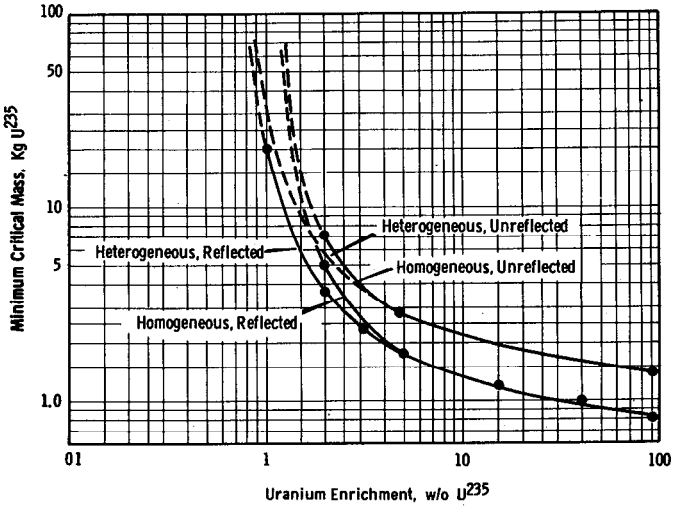

Fig. 9. Minimum critical mass for heterogeneous and homogeneous uranium-water systems as a function of U-235 enrichment.

The criticality of a heterogeneous system of enriched uranium rods in enriched uranyl nitrate solution is also of interest. This would be the actual case for fuel elements during the dissolution process. Such an exponential experiment was conducted

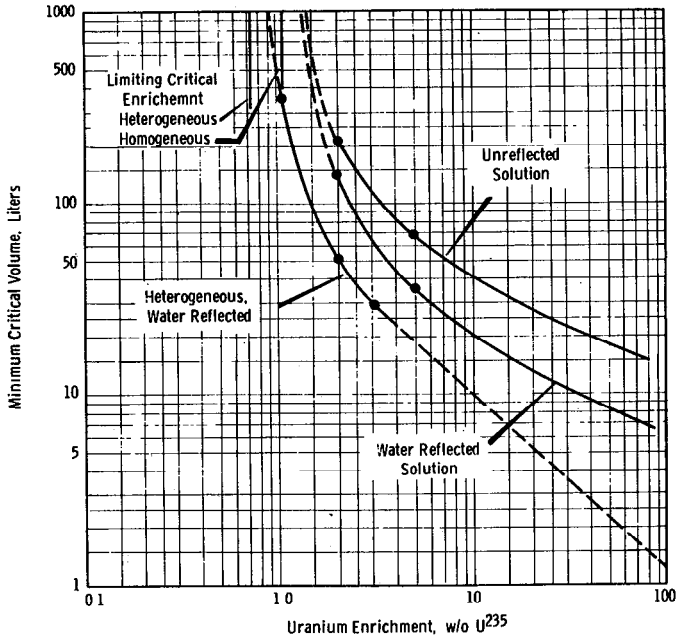

Fig. 10. Minimum critical volume for heterogeneous and homogeneous uranium-water systems as a function of U-235 enrichment.

at Hanford with 0.925 in. diameter rods of 1.007% U-235 enrichment immersed in a uranyl nitrate solution of the same enrichment (*20*). A similar experiment was made with the same fuel elements in a water lattice. The data from these measurements are compared in Table 1.

At 1.007 wt.% U-235, therefore, the buckling is reduced appreciably and the critical mass correspondingly increased because of the uranyl nitrate. This reduction is not as great as implied, however, since the maximum buckling for the uranyl nitrate lattice will be obtained with a different (larger) lattice spacing than for the water lattice, and it is the maximum bucklings which should be compared. Calculations for a uranyl nitrate lattice of the above enrichment show the water lattice to have a higher maximum buckling than the uranyl nitrate lattice.

In another experiment, the 1.007 wt.% U-235 fuel rods were immersed in solutions of boric acid (H_3BO_3) (*20*). Both the lattice spacing and the boron concentration in the moderator were varied. For water-uranium volume ratios in the range 1.37

TABLE 1. EXPONENTIAL MEASUREMENTS OF 1.007% ENRICHED URANIUM-WATER AND UNH LATTICES 0.925 IN. DIAMETER FUEL RODS

Lattice spacing	Solution to uranium rod volume ratio	Uranium in the moderator solution, U/liter of water	Nitrate content of uranyl nitrate g./liter	Buckling, 10^{-5}cm.$^{-2}$	Critical mass (water reflected sphere)
1.5 in.	1.74	431	243	1,304	30,500 lb. total U (rods only)
1.5 in.	1.74	Zero	None	3,470	5,889 lb. total U

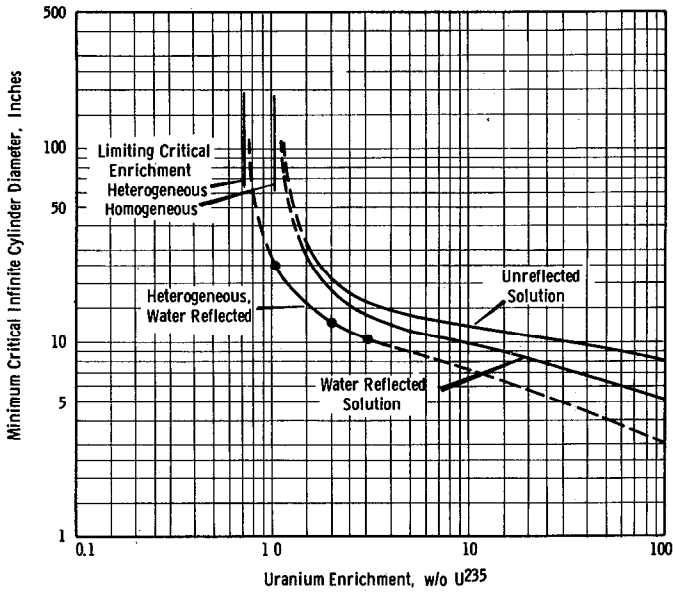

Fig. 11. Minimum critical cylinder diameter for heterogeneous and homogeneous uranium-water systems as a function of U-235 enrichment.

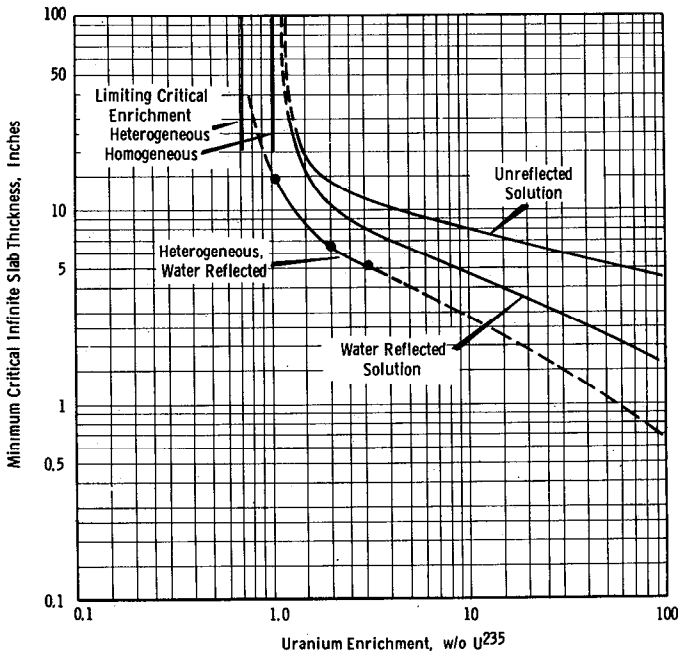

Fig. 12. Minimum critical slab thickness for heterogeneous and homogeneous uranium-water systems as a function of U-235 enrichment.

to 2.15, 4 g./liter of H_3BO_3 were sufficient to reduce the buckling to negative values.

CRITICALITY OF URANIUM TUBES IN WATER

The critical mass and critical volume data presented above are for solid uranium rods in water. In the case of slightly enriched tubular fuel elements, there are much less data available.

Exponential experiments with tubular fuel elements in water lattices have been conducted at Hanford in the enrichment range of 1.0 to 1.6% U-235 (*18*) and at Oak Ridge with 0.95% U-235 (*21*). In these experiments, the uranium tubes ranged in size from 1.37 in. OD; 0.49 in. I.D. to 1.66 in. O.D.; 0.94 in I.D. The maximum bucklings obtained in these measurements are compared with calculated maximum bucklings (*22*) in Figure 13.

Bucklings for fuel tubes are more difficult to calculate than those of solid rods, because of resonance escape probability, fast effect, and thermal utilization are affected by the water within the fuel cores. A hollow rod or fuel tube with the same volume as a solid rod will have a smaller value for both the resonance escape probability and the fast effect; however, as a result of the lower thermal disadvantage factor, the thermal utilization will be increased. Relative to the resonance escape probability, a fuel tube immersed in water will have two effective surfaces for resonance absorption.

There are insufficient experimental data to determine whether tubular elements can be made to have larger bucklings or lower critical masses than the maximum buckling or minimum critical mass for solid rods of optimum diameter; however, calculational estimates have been made. For comparative purposes, the nearest equivalent solid rod would be one having the same effective surface-to-volume ratio as the fuel tube. The effective surface of a tube. The effective surface of a tube (S_{eff}) is obtained from the expression

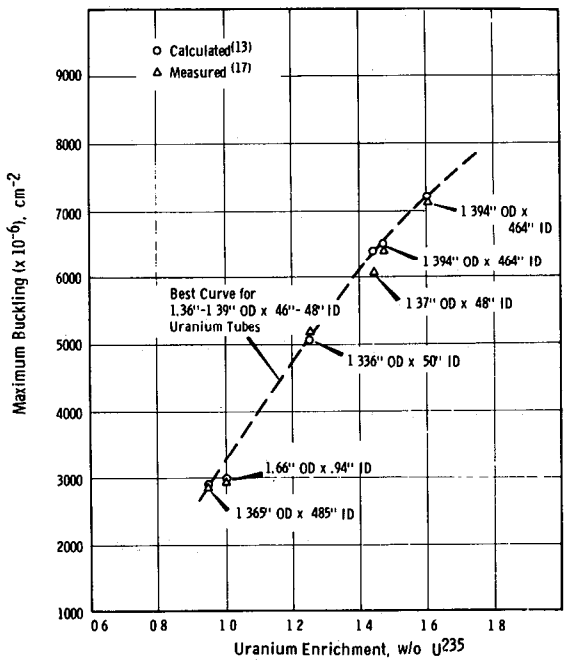

Fig. 13. Measured and calculated maximum material bucklings for uranium tubes in water as a function of U-235 enrichment.

$$S_{eff} = S_{out} + \Sigma_s V_1 [1 - P_c (\Sigma_s, r_1)]$$

where S_{out} = outer surface of tube, Σ_s = macroscopic scattering cross section of water, V_1 = volume of central water region, and P_c = collision probability in the central region of radius r_1.

Maximum bucklings and minimum critical masses for fuel tubes are compared with the respective maximum bucklings and minimum critical masses for fuel tubes are compared with the respective maximum bucklings and minimum critical masses for solid rods of the *same enrichment and effective surface-to-volume ratio* in Tables 2 and 3. Values for optimum size rods are also listed for comparison. pared, the solid rods with the same effective surface-to-volume ratios have bucklings equal to or greater than those measured for tubular elements; and, with one exception, critical masses are less than those of the corresponding tubular elements.

Exponential experiments have been performed with 2.6 wt. % U-235 enriched UO_2 tubes in light water at Hanford (*23*). The tubes were made up from 0.705 in. O.D., 0.323 in. I.D. UO_2 pellets mounted in plastic tubes. The uranium oxide density was 10.43 g./cc. The results of the experiments are presented in Table 4.

THE PROBLEM OF LARGE RODS OR BILLETS

Questions concerning the criticality of large rods, slabs, and billets frequently arise in connection with fuel element fabrication, as in the extrusion process in which fuel tubes are extruded from large billets.

Criticality data are generally lacking for slightly enriched uranium fuel rods greater than about 2 in. diameter. The only data available are a series of exponential experiments with 3 in. diameter rods of 3.0 wt.% U-235 enrichment made at the Savannah River Laboratory (*24*). These measurements corre-

TABLE 2. COMPARISON BETWEEN BUCKLINGS OF TUBES AND SOLID RODS

Enrichment, wt. % U^{235}	Tubes				Solid rods with same S_{eff}/V ratio as measured tubes			Solid rods for maximum buckling		
	O.D., in.	I.D., in.	Measured buckling, 10^{-6}, $cm.^{-2}$	Vw/Vu	O.D., in.	Calculated buckling, 10^{-6}, $cm.^{-2}$	Vw/Vu	O.D., in.	Calculated buckling, 10^{-6}, $cm.^{-2}$	Vw/Vu
0.95	1.36	0.49	2,850	(1.7)	0.95	2,900	(1.8)	0.81	3,000	(1.9)
1.002	2.34	1.79	2,550	(2.4)	0.55	3,400	(2.5)	0.78	3,600	(2.1)
1.007	1.66	0.94	2,950	(1.8)	0.74	3,500	(2.2)	0.78	3,600	(2.1)
(1.007 solid rods)	(1.66)		(2,750)	(1.3)						
1.25	1.34	0.50	5,200	(1.7)	0.92	5,850	(2.0)	0.71	6,000	(2.3)
1.44	1.34	0.48	6,090	(1.9)	0.95	7,400	(2.0)	0.67	7,600	(2.4)
(1.44 solid rods)	(1.34)		(5,800)	(1.6)						
1.47	1.39	0.46	6,400	(1.8)	0.99	7,500	(1.9)	0.66	7,800	(2.5)
1.60	1.39	0.46	7,000	(1.8)	0.99	8,500	(2.0)	0.64	8,900	(2.5)

Water-to-uranium volume ratios given in parentheses.

TABLE 3. COMPARISON BETWEEN CRITICAL MASSES OF TUBES AND SOLID RODS

Enrichment, wt. % U^{235}	Measured tubes				Solid rods with same S_{eff}/V ratio as measured tubes			Solid rods for minimum critical mass		
	O.D., in.	I.D., in.	Estimated MCM from experiments, lb.	Vw/Vu	Diameter, in.	Calculated MCM, lb.	Vw/Vu	Diameter, in.	Calculated MCM, lb.	Vw/Vu
0.95	1.36	0.49	7,676	(1.9)	0.96	7,872	(2.0)	0.80	6,615	(2.2)
1.002	2.34	1.79	8,110	(2.6)	0.55	5,938	(2.9)	0.66	4,190	(2.7)
1.007	1.66	0.94	7,000	(2.2)	0.74	4,983	(2.5)	0.66	4,190	(2.7)
(1.007 solid rods)	(1.66)		(10,000)	(1.4)						
1.25	1.34	0.50	2,750	(2.2)	0.92	2,043	(2.5)	0.42	1,544	(3.6)
1.44	1.37	0.48	2,000	(2.1)	0.95	1,347	(2.5)	0.34	904	(4.2)
(1.44 solid rods	1.34		(2,300)	(1.9)						
1.47	1.39	0.46	1,840	(2.1)	0.99	1,317	(2.5)	0.33	882	(4.3)
1.60	1.39	0.46	1,520	(2.2)	0.99	1,016	(2.6)	0.30	684	(4.6)

Water-to-uranium volume ratios given in parentheses.

late reasonably well with the Hanford measurements at 3.06% U-235 at the smaller rod diameters of 0.175, 0.60, and 0.925 in.

By comparing these data, it is apparent that the maximum buckling for a given enrichment is a slowly varying function of rod diameter. For example, in the case of the 3% enriched uranium, the buckling for a 0.6 in. diameter rod is about 15,400 times 10^{-6} cm.$^{-2}$, whereas for a rod diameter five times larger (a 3-in. diameter rod), the optimum

TABLE 4. BUCKLINGS AND ESTIMATED CRITICAL MASSES FOR 2.6 WT.% U-235 ENRICHED UO_2 TUBES IN LIGHT WATER

Lattice spacing, cm.	H_2O/UO_2 volume ratio	Material buckling, 10^{-6}, cm.$^{-2}$	Estimated critical mass (Spherical geometry) kg. U^{235}
	Central cavity filled with water		
2.540	1.808	10,131	4.79
3.048	3.043	10,421	3.40
3.556	4.504	8,559	3.72
	Central cavity empty		
2.540	1.542	9,644	5.12
3.048	2.778	10,084	3.59
3.556	4.237	8,467	3.79

buckling is still approximately 10,000 times 10^{-6} cm.$^{-2}$ (*24*). As the rod size increases further, the buckling is finally reduced to zero, and the critical mass becomes infinite.

An attempt has been made to estimate the largest diameter rods which can be made critical in a water lattice as a function of enrichment. The results are shown in Figure 14. For 3% enriched uranium, the diameter is about 15 in.; for 1% the diameter appears to be about 4 in. For about 6% enriched uranium, the rod diameter for zero bucklings would be infinite. In the case of natural uranium, if criticality can be achieved at all in a water lattice, the rod diameter would be about 1 in. for zero buckling.

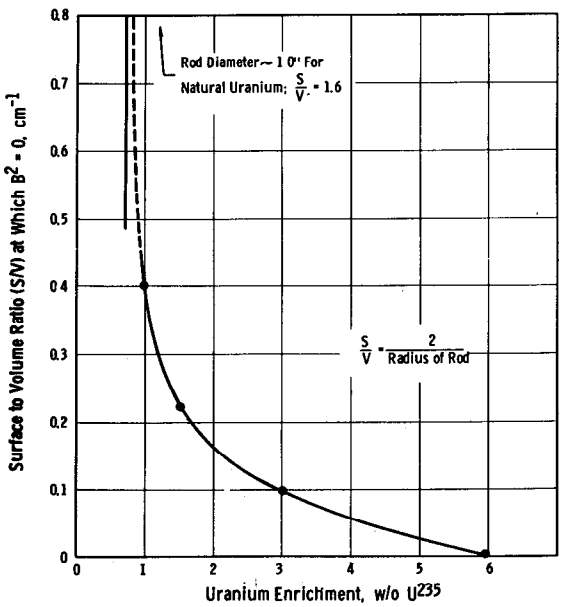

Fig. 14. Estimated surface-to-volume ratios of large rods (billets) which result in zero bucklings and infinite critical mass as a function of U-235 enrichment.

Figure 14 illustrates the necessity for making nuclear safety reviews in operations involving large billets. The limit at approximately 6% represents an entirely fast system, whereas the limit for natural uranium would be a thermal system. Unless water can be excluded, rather large billet sizes are indicated for zero buckling as the enrichment is increased.

PLUTONIUM-URANIUM SYSTEMS

ALWAYS SAFE RATIO OF PLUTONIUM-URANIUM

A parameter of interest to reprocessors of slightly enriched uranium is the limiting enrichment of plutonium in natural uranium. This value has not been measured, but has been calculated by L. L. Carter using Monte Carlo methods (*7*). The minimum Pu-239 enrichment necessary for k_∞ to be unity was calculated for aqueous homogeneous solutions of natural UO_3 (0.712 wt.% U-235) at optimum water moderation. Calculations were performed for 0, 5, 10, and 20% Pu-240. In the absence of Pu-240, the minimum critical enrichment of Pu-239 in natural UO_3 is 0.199 wt.%; and the relative Pu-239 to U-235 atom worth is 1.66. The relative worth implies that adding one atom of plutonium to natural uranium would be the equivalent of adding about 1.7 atoms of U-235. Other results are shown in Table 5. It is interesting to note that the limiting enrichment for k_∞ to be unity is strongly dependent on the Pu-240 content of the plutonium.

HETEROGENEOUS SYSTEMS OF UO_2-PUO_2 FUEL RODS

Critical approach measurements have been performed by Schmid et al. with UO_2-PuO_2 rods containing 1.5 wt.% PuO_2 in light water at Hanford (*25*), and measurements with UO_2 rods of 2.0 wt.% PuO_2 are in progress.

The 1.5 wt.% PuO_2-UO_2rods were 0.318 in. diam-

TABLE 5

% Pu-240	Critical plutonium concentration in natural uranium, wt.%	Relative Pu-atom worth Pu-239 Atom U-235 Atom
0	0.199	1.66
5	0.244	1.36
10	0.297	1.11
20	0.468	0.71

eter and 48.5 in. long, and clad with 0.028 in. Zircaloy-2. The PuO_2-UO_2 density was 9.65 g./cc.; the uranium enrichment was 0.2 wt.% U-235; and the isotopic content of the plutonium was 91.41% Pu-239, 7.83% Pu-240, 0.73 Pu-241, and 0.03% Pu-242 by weight. The number of rods for a critical loading was determined by the extrapolation of neutron multiplication data obtained with as much as 96% of the critical mass. The results of the measurements are given in Table 6.

TABLE 6. EXPERIMENTAL RESULTS FOR 1.5 WT.% PUO_2-UO_2 RODS IN LIGHT WATER

V_W/V_F	No. of rods for criticality	Buckling, 10^{-6} cm.$^{-2}$	Extrapolation length, cm.
1.10	1,487	4,800	8.40
1.56	829	6,510	7.99
2.71	484	7,850	7.27
3.79	420	7,490	6.93
5.14	452	6,090	6.75
5.58	488	5,520	6.64

CRITICALITY SAFETY

The criticality data that have been presented provide a basis for establishing safe processing limits for slightly enriched (less than 5% U-235) uranium systems. It has been pointed out that unmoderated systems are infinitely safe. Homogeneous systems below 1.03% U-235 and all natural uranium systems are, likewise, considered safe. The conditions for criticality are determined primarily by the heterogeneity of the uranium system and the degree of moderation, with a very wide range of limits resulting therefrom.

In operations involving a significant amount of fissionable material, sufficient safety features must be incorporated to prevent a criticality accident. Minimum requirements are that at least two unlikely, independent, and concurrent changes must occur in one or more of the conditions specified as essential to criticality safety, before a criticality accident is possible. In other words, process controls must be such that no single error or mishap can cause criticality. This policy is generally carried out through the use of process hazards reviews, approved criticality safety specifications, and routine audits to assure compliance to specifications.

It is not always necessary to base nuclear safety limits on theoretical *minimum* conditions, which in practice might not be possible to achieve. In actual plant operations, for example, allowance may be given for the actual size of uranium rods being fabricated or stored, rather than assuming the more conservative basis of optimum rod size. On the other hand, care and thoroughness must be exercised in evaluating plant operating conditions to take into account *all* credible changes that affect accidental criticality safety, particularly potential changes in geometry and moderation.

Safe limits are obtained by reducing critical values by a factor for safety commensurate to the reliability of the criticality data. Wherever possible, safe limits are based directly on measured data. When measured data are not available, which is often the case, then computed critical values are used, based on calculational methods that have been confirmed by comparisons with measured critical values. For critical parameters that have been confirmed, the safety factors used are as follows [see also the *Nuclear Safety Guide* (*26*)].

	Maximum allowable fraction of critical value
Mass	0.45
Volume	0.75
Slab thickness	0.85
Cylinder diameter	0.85

Safe mass, volume, cylinder diameter, and slab thickness limits for slightly enriched uranium solutions and fuel elements are given in the *Nuclear Safety Guide* (*26*). These limits are based on optimum conditions of moderation, reflection, geometry, and fuel rod diameter. A parameter not in the guide that is useful for controlling uranium fuel element storage arrays is *safe mass per unit area* (M/A). This parameter is a function of fuel rod diameter and the density of uranium in the storage lattice. Critical values are obtained by multiplying the critical slab thickness by the density of uranium in the storage lattice. Critical values are obtained by multiplying the critical slab thickness by the density of uranium in the lattice at that slab thickness. The safe M/A parameter permits special consideration to arrays with low uranium density; thus making it possible to stack the fuel higher than permitted under the slab thickness limit. One must be certain, however, that the area selected as a basis for assessment is small enough to avoid groupings of fuel elements within the area that may be near a critical mass; and that the safe M/A cannot accidentally be exceeded anywhere in the array. Figure 15 presents calculated minimum *critical* M/A values for 1, 2, 3, and 5% U-235 enriched uranium rods in light water as a function of rod diameter. Safe M/A values are obtained by multiplying the critical values by 0.75.

FUEL ELEMENT DISSOLVING—SAFE ANNULAR DISSOLVER

An example of a safe annular dissolver for slightly enriched uranium is shown in Figure 16.

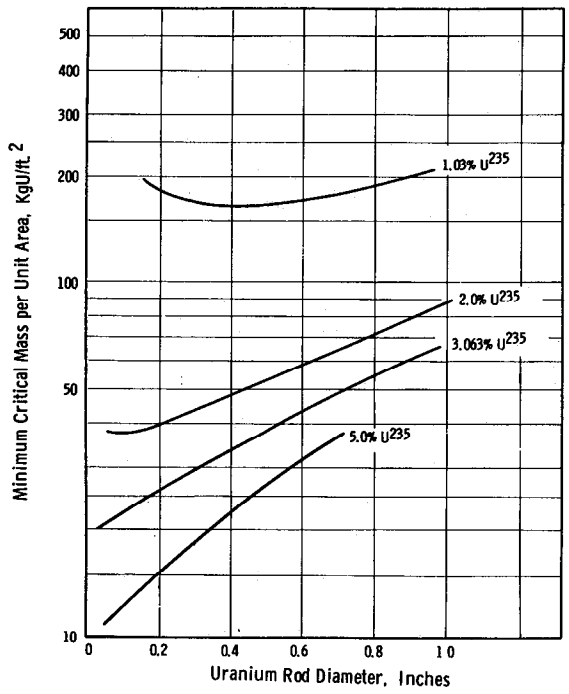

Fig. 15. Estimated minimum critical mass per unit area for 1, 2, 3, and 5 wt. % U-235 enriched uranium rods for water as a function of rod diameter.

This design is based on the critical dimensions of an infinite slab reflected on one side with concrete and on the other with water. The interior contains a layer of concrete 6 to 7 in. in thickness; the concrete (thickness of two layers) isolates one side from another. Thus the safe thickness of the solution part of the dissolver may be based on infinite critical slab thickness with a safety factor applied for uncertainties in the critical data for the infinite slab, etc. The advantage of this design should be apparent when one compares it to the number and space requirements for several cylindrical or vertical slab dissolvers with the same total volume, each safe by geometry and isolated from another.

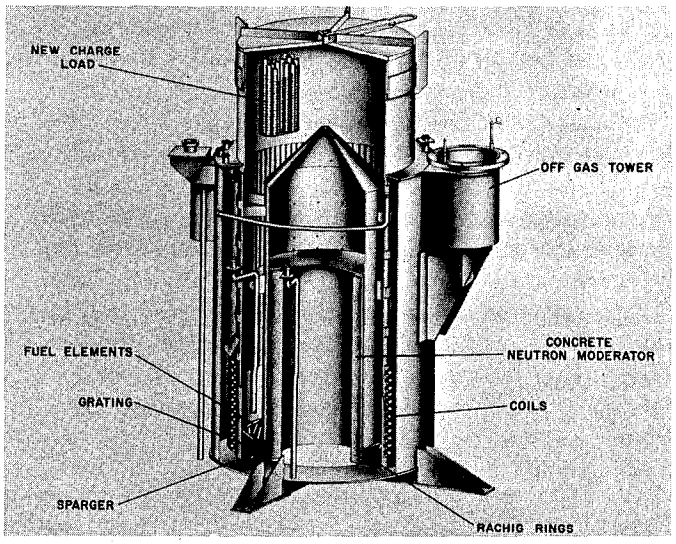

Fig. 16. Annular dissolver.

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