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CRITICAL DIMENSIONS OF AQUEOUS SOLUTIONS OF $U(37)O_2F_2$

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ABSTRACT

The critical dimensions of a sphere of an aqueous solution of 37-wt%-^{235}U-enriched uranyl fluoride and of several unreflected and water-reflected cylinders at a number of chemical concentrations were determined experimentally. The critical uranium concentration in the unreflected sphere was 51.93 g/liter. The neutron multiplication factor of the sphere was calculated by the DSN transport code in the S_4 approximation to be 1.0100; the relative fission-rate distribution along a diameter, also calculated by DSN, compared favorably with measurements. The results from the cylinders, although less detailed and less accurate, can be generalized for nuclear criticality safety guidance. The dimensions of equivalent spheres, infinite cylinders, and infinite slabs were estimated for selected chemical concentrations by equating bucklings.

INTRODUCTION

As a part of a continuing program at the Oak Ridge Critical Experiments Facility for the measurements of the critical dimensions of fissile materials in simple geometry, a few experiments were performed with aqueous solutions of uranium enriched to about 37% in the ^{235}U isotope [U(37)]. In one experiment the critical dimensions and the relative fission rate distribution were determined for a sphere of solution having a diameter of 69.2 cm and the results were compared with those from transport theory calculations.

Somewhat less detailed and less accurate measurements, limited to critical dimensions, were made in cylindrical geometry of a range of diameters with solutions of various concentrations, the higher of these concentrations being near that where linear critical dimensions are expected to be minimal. These data are adequate to provide guidance in nuclear criticality safety and have been generalized for that purpose.

This program of critical experiments was carried on between 1958 and 1960 and many of them were summarized by Callihan.[1] Further, these experiments are an adjunct to those with uranium of comparable ^{235}U content but with less neutron moderator present reported previously by Beck *et al.*[2] and by Cronin.[3]

EXPERIMENTAL RESULTS

Spherical Geometry

The critical concentration of a nominally unreflected 69.2-cm-diam sphere of an aqueous $U(37.0)O_2F_2$ solution[4] was determined to be 19.2 g of ^{235}U/liter. The solution was contained in aluminum of 0.32 cm wall thickness. The assembly is described in Table 1.

The critical mass, 3.335 kg of ^{235}U, is only about 80 g greater than the critical mass of ^{235}U in a solution of $U(93)O_2F_2$ contained in the same spherical vessel.[5]

The energy distribution of the neutrons within the sphere was assumed to be Maxwellian in the region of thermal energies and to be supplemented above 0.2 eV by a component inversely proportional to the energy. The proportionality factor, given in Table 2, was determined from cadmium ratios measured by ^{235}U and gold foils. Although the cadmium ratio in $U(93.2)O_2(NO_3)_2$ solution measured[6] by ^{235}U foils was 0.8 unit

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1. Dixon Callihan, "A Review of Criticality Data Obtained by Experimental Methods," Criticality Control in Chemical and Metallurgical Plant, Karlsruhe Symposium, 1961, Organization for Economic Cooperation and Development, European Nuclear Energy Agency, Paris (1961).
2. C. K. Beck, A. D. Callihan, and Raymond L. Murray, "Critical Mass Studies, Part II," K-126, Carbide and Carbon Chemicals Corp. (1948).
3. D. F. Cronin, "Critical Mass Studies, Part X," ORNL-2968, Oak Ridge National Laboratory (1960) CONFIDENTIAL
4. The ^{235}U content of the uranium, reported as 36.96 wt%, may be uncertain by as much as ± 4 % because of imprecision of analyses. This item is discussed in greater detail later. Other uranium isotopes were reported as being present in the following amounts: ^{234}U, 0.26%; ^{236}U, 0.29%; and ^{238}U, 62.49%.
5. J. K. Fox *et al.*, Neutron Phys. Div. Ann. Prog. Rept. for Period Ending Sept. 1, 1958, ORNL-2609, p. 42, Oak Ridge National Laboratory (1958).
6. R. Gwin and D. W. Magnuson, *Nucl. Sci. Eng.* **12**, 364 (1962).

Table 1. Critical Conditions of a 69.2-cm-diam Unreflected Sphere of $U(37.0)O_2F_2$ Solution.

k_{eff} (at 20°C)	1.0005[a]
Solution specific gravity	1.057
Uranium concentration (mg/g of solution)	49.13
^{235}U concentration (g/liter of solution)	19.2
Uranium concentration (g/liter of solution)	51.93
H:^{235}U Atomic Ratio	1.356×10^3
Solution Volume (liters)	173.6
^{235}U Mass (kg)	3.335

a. The reported values of the critical concentration and mass were not corrected for this slight excess reactivity.

Table 2. Cadmium Ratio and Epithermal Flux Proportionality Factor in $U(37.0)O_2F_2$ Aqueous Solution.

Detector Material	Cadmium Ratio[a]	Epithermal Flux Proportionality Factor
Gold	5.97	0.368
U(93)	35.7	0.383
U(37)	34.5	--

a. The experimental uncertainty in the cadmium ratio is ± 5%.

larger than that observed in these experiments, they agree within the experimental error of ± 5%. The uncertainty in the epithermal flux is about 10% because of doubt in values of resonance integrals and of foil self-shielding corrections and because of fissions in ^{238}U. The details of the evaluation of the epithermal flux have been previously described.(6)

The neutron multiplication factor, k_{eff}, of the sphere was calculated to be 1.0100 using Carlson's[7] DSN transport code in the S_4 approximation with the 16-group cross-section set of Hansen and Roach[8] and the experimental atomic densities given in Table 3. The neutron lifetime, defined as the sum of the products of the normalized group fluxes and the average inverse group velocities, $\sum_n \Phi_n (1/v)_n$, was 65.5 μsec. The calculated[9] values of the lifetime in critical solutions of $U(93.2)O_2(NO_3)_2$ and $^{233}UO_2(NO_3)_2$ (97.7 wt% ^{233}U) in this sphere were 67.4 and 76.5 μsec, respectively, and the corresponding calculated[9] multiplication factors were 1.0082 and 1.0047.

Table 3. Atomic Densities Used in DSN Calculations of the 69.2-cm-diam Sphere of $U(37.0)O_2F_2$ Solution.

Isotope	Isotopic Analysis (wt%)	Atomic Density[a] (atoms/cm³)
^{234}U	0.26	0.348 + 18
^{235}U	36.96	49.288 + 18
^{236}U	0.29	0.386 + 18
^{238}U	62.49	82.183 + 18
H		6.570 + 22
O		3.310 + 22
F		2.640 + 20
Al (the 0.32-cm-thick container material)		6.016 + 22

a. Atomic densities are given as a number and an exponent, e.g., 1.0 + 10 means 1.0×10^{10}.

7. B. G. Carlson and G. I. Bell, Proc. Intern. Conf. Peaceful Uses Atomic Energy, 2nd Geneva, 1958 16, 353 (1959).
8. Gordon E. Hansen and William H. Roach, "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos Scientific Laboratory (1961).

D. W. Magnuson, Neutron Phys. Div. Ann. Prog. Rept. Aug. 1, 1963, ORNL-3499, Vol. I, p. 68, Oak Ridge National Laboratory (1963).

The fission rate distribution along a diameter of the sphere was calculated by the DSN transport code and was measured with a ^{235}U fission counter 0.64 cm o.d. by 2.5 cm long. The results, normalized to unity at the center of the sphere, are compared in Fig. 1 and in Table 4. The distribution between points 25 cm on each side of the center were fitted to the equation $\Phi = (A \sin BR)/BR$ to evaluate the effective buckling, B^2, and the extrapolated radius, R. The calculated and the measured distributions gave 37.47 and 36.66 cm, respectively, as values of R, corresponding to an extrapolation distance of 2.9 or 2.1 cm. These are to be compared with 2.5 ± 0.3 cm measured by foil activation in a 30.5-cm-diam cylinder of the same solution.

Comparison of the calculated radial fission rate distribution to the curve obtained from fitting the data to the above equation shows that the asymptotic flux is not accurately representative of the distribution even within a central region at least 9 cm from the boundary. The imprecision in the experimentally determined distribution does not allow a direct comparison. The extrapolation distance derived from the curve fitting is, therefore, subject to some uncertainty.

Cylindrical Geometry

A second set of experiments determined essentially only the critical dimensions of cylinders of aqueous solutions of UO_2F_2. In the course of these experiments an average value of the ^{235}U content of the uranium was determined[10] to be 37.3%. The cylinders ranged in diameter from 16.5 to 38.1 cm and the ^{235}U concentration was varied between 19 and 340 g/liter. The solution was contained in aluminum vessels of 1.6-mm wall thickness. Some cylinders were completely enclosed by an effectively infinite thickness of water and others were nominally unreflected. A few of the latter were influenced by nearby structures as is pointed out in the summary of the data appearing in Table 5.

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10. This value is the average of 19 isotopic analyses ranging between 35.32% and 38.48% and is indicative of the precision of the measurements.

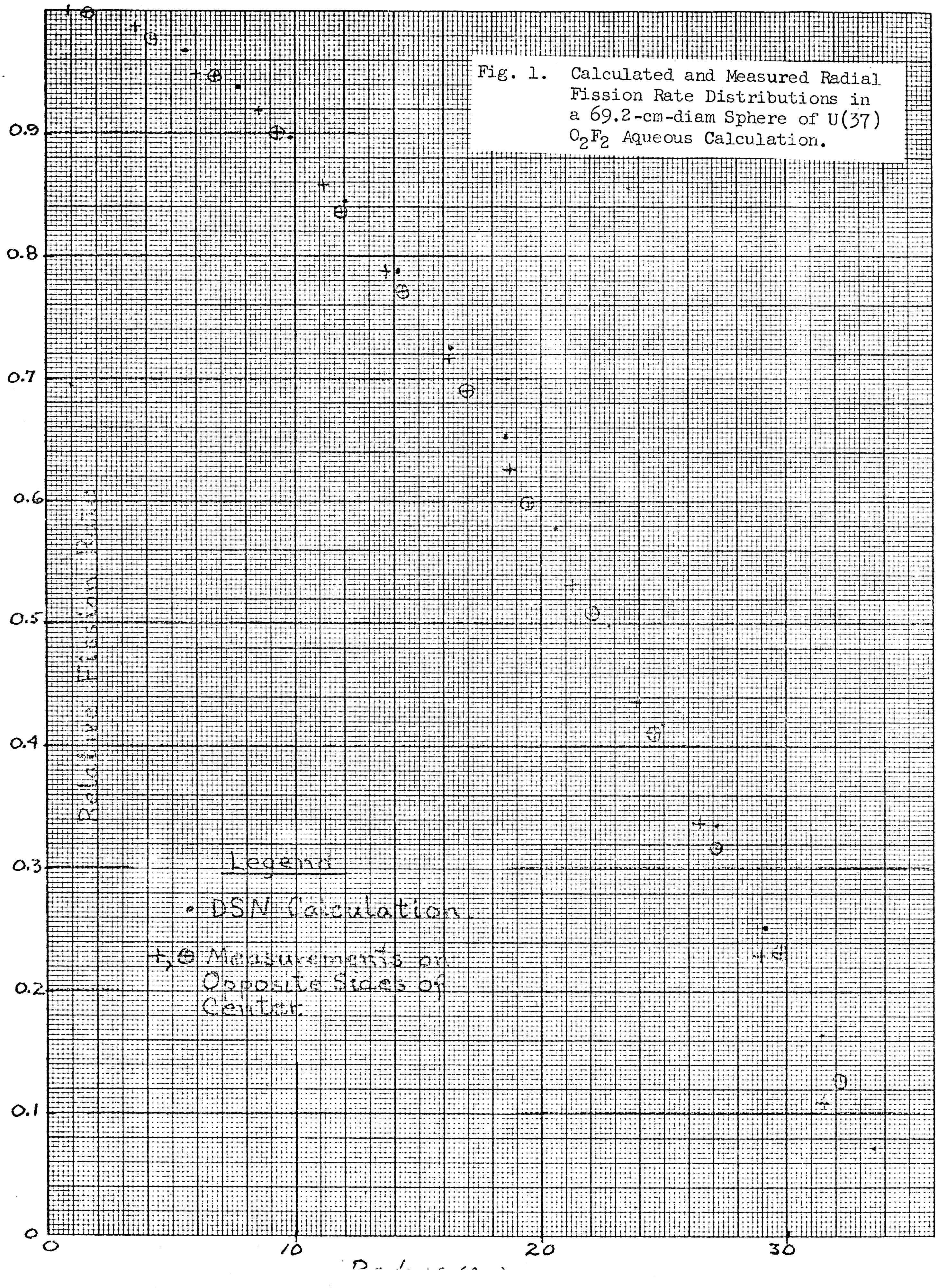

Fig. 1. Calculated and Measured Radial Fission Rate Distributions in a 69.2-cm-diam Sphere of U(37) O_2F_2 Aqueous Calculation.

Table 4. Calculated and Measured Radial Fission Rate Distributions in a 69.2-cm-diam Sphere of $U(37.0)O_2F_2$ Solution.

Calculated		Measured	
Radial Position (cm)	Relative Fission Density	Radial Position (cm)	Relative Fission Density
0.1	1.000	- 32.1	0.128
1.5	0.998	- 29.6	0.233
3.4	0.988	- 27.0	0.316
5.6	0.967	- 24.5	0.411
7.7	0.936	- 22.0	0.507
9.8	0.895	- 19.4	0.597
12.0	0.845	- 16.9	0.689
14.1	0.787	- 14.3	0.770
16.3	0.722	- 11.8	0.835
18.4	0.651	- 9.3	0.901
20.5	0.576	- 6.7	0.946
22.7	0.497	- 4.2	0.977
24.8	0.416	- 1.6	0.998
27.0	0.335	+ 0.9	1.000
29.1	0.252	3.5	0.984
31.3	0.166	6.0	0.948
33.4	0.072	8.5	0.917
34.6	0.018	11.1	0.858
		13.6	0.787
		16.2	0.715
		18.7	0.624
		21.2	0.531
		23.8	0.425
		26.3	0.337
		28.9	0.230
		31.4	0.110

Table 5. Experimental Critical Conditions of Cylinder of U(37.3)O_2F_2 Solution in Aluminum Containers.

	Unreflected Critical Dimensions			Water-Reflected Critical Dimensions		
$H:^{235}U$ Atomic Ratio	Height (cm)	Volume (liters)	Mass (kg of ^{235}U)	Height (cm)	Volume (liters)	Mass (kg of ^{235}U)
			16.5 cm Diameter			
67.8	-	-	-	>75[a]	-	-
			17.8 cm Diameter			
65.0	-	-	-	76.4	19.0	6.47
71.0	-	-	-	72.6	18.0	5.71
98.4	-	-	-	67.3	16.7	3.99
			20.3 cm Diameter			
67.8	-	-	-	32.8	10.6	3.51
98.4	-	-	-	31.6	10.2	2.44
			24.1 cm Diameter			
98.4	>165[a]	-	-	-	-	-
			25.4 cm Diameter			
76.9	-	-	-	18.5	9.4	2.80
98.4	75.6[b]	38.3[b]	9.14[b]	17.9	9.1	2.17
108.4	77.9	39.5	8.68	-	-	-
113.4	77.3	39.2	8.28	-	-	-
116.4	77.1	39.1	8.07	-	-	-
123.5	77.3	39.1	7.65	-	-	-
123.5	72.8[b]	36.9[b]	7.21[b]	-	-	-
126.5	77.4	39.2	7.51	-	-	-
129.5	77.6	39.3	7.36	-	-	-
130.4	78.2	39.6	7.33	-	-	-
			30.5 cm Diameter			
65.0	32.0	23.3	7.94	-	-	-
76.9	30.5	22.2	6.66	13.9	10.2	3.04
98.4	29.2[b]	21.3[b]	5.07[b]	13.9	10.1	2.41
			38.1 cm Diameter			
76.9	21.3	24.3	7.27	10.8	12.3	3.68

a. Apparently these cylinders will be subcritical at infinite height.
b. In these experiments the cylinder was mounted at the center of a 6-ft-square steel tank and the critical dimensions are uncertain because of the neutrons reflected by the steel.

An extrapolation distance for unreflected volumes, 2.5 ± 0.3 cm, was obtained from a radial neutron flux traverse measured in the 30.5-cm-diam cylinder with gold foils 0.005 cm thick and 0.80 cm in diameter. An extrapolation distance for the water-reflected volumes, 5.0 cm, was derived from the measured reflector savings, 2.5 cm.

Of value in the safety assessment of processes with fissile materials are the observations that an unreflected cylinder 24.1 cm (9.5 in.) in diameter and 165 cm high was subcritical at several concentrations corresponding to values of H:^{235}U between 98.4 and 108.4 and that a water-reflected cylinder 16.5 cm (6.5 in.) in diameter and 75 cm high at an H:^{235}U atomic ratio of 67.8 was also subcritical. It was concluded from apparent source-neutron multiplication measurements that cylindrical volumes of neither of these diameters can be made critical with aqueous solutions of U(37.3) under the specified reflector conditions.

Estimates were made of the critical dimensions of spheres, infinitely long cylinders, and infinitely large slabs at several uranium concentrations. The dimensions were obtained from selected critical conditions in Table 5 by the usual method of equated bucklings with extrapolation distances of 5.0 and 2.5 cm for water-reflected and unreflected volumes, respectively. In instances of uncertainty, the smallest values are quoted in Table 6.

Table 6. Dimensions of Critical Spheres, Infinite Cylinders and Infinite Slabs of $U(37)O_2F_2$ Aqueous Solution Estimated by Equating Bucklings.[a]

$H:{}^{235}U$	Sphere Radius (cm)	Sphere Volume (liters)	Sphere Mass (kg of ^{235}U)	Infinite Cylinder Diameter (cm)	Infinite Slab Thickness (cm)
			Unreflected		
65.0	17.1	21.1	7.18	25.1	14.6
76.9	16.8	19.9	5.96	24.6	14.3
108.4	16.8	19.9	4.38	24.6	14.3
123.5	16.8	19.9	3.89	24.6	14.3
126.5	16.8	19.9	3.81	24.6	14.3
130.4	16.8	19.9	3.69	24.6	14.3
1.36×10^3	34.6	174	3.34	51.8	32.1
			Water Reflected		
65.0	12.8	8.70	2.97	17.2	7.76
71.0	12.7	8.63	2.73	17.1	7.72
76.9	12.3	7.79	2.34	16.5	7.33
98.4	12.7	8.58	2.06	17.1	7.72

a. Based on extrapolation distances of 5.0 and 2.5 cm for water-reflected and unreflected volumes, respectively.

Miscellaneous Experiments

Additional experiments, somewhat less basically important and with results not as accurately determined as those described earlier, are reported in this section for the guidance they may afford in specialized problems of nuclear criticality.

Two spherical vessels, constructed of 0.16-cm-thick aluminum, contained a critical volume of solution when only partially filled. The vessels were either nominally unreflected, although the presence of the walls of a surrounding tank a few feet distant undoubtedly had some effect, or were completely water reflected. The results are summarized as follows:

$H:{}^{235}U$	Reflector	Sphere Diameter (cm)	Sphere Capacity (liters)	Critical Dimensions Height[a] (cm)	Critical Dimensions Volume (liters)	Critical Dimensions Mass (kg of ^{235}U)
64.4	None	36.7	25.74	28.1	22.2	7.59
64.4	Water	28.9	14.35	22.1	12.1	4.13
70.7	Water	28.9	14.35	21.6	11.9	3.77

a. Measured from the lower pole to the surface of the solution.

In another set of experiments with a completely water-reflected 17.8-cm-diam cylinder of solution, aluminum of several thicknesses was inserted between the top of the solution and the adjacent reflector. The resultant critical dimensions were observed. The $H:{}^{235}U$ atomic ratio within the solution was 70.7.

Aluminum Thickness (cm)	Critical Dimensions Height (cm)	Critical Dimensions Volume (liters)	Critical Dimensions Mass (kg of ^{235}U)
0[a]	72.0	17.90	5.69
1.91	72.8	18.08	5.75
3.81	73.2	18.19	5.78
4.45	72.7	18.06	5.74

a. These values agree with an entry in Table 5 to within the precision of the measurements.

In still another experiment, the wall thickness of a stainless steel container, separating a cylindrical volume of solution from its lateral water reflector, was increased. The diameter of the cylinder of solution was 30.5 cm and its concentration corresponded to an $H:{}^{235}U$ atomic ratio of 66.4. The resulting critical dimensions are noted.

Thickness of Steel (cm)	Critical Dimensions Height (cm)	Volume (liters)	Mass (kg of ^{235}U)
0.16	15.1	10.9	3.62
0.48	15.8	11.5	3.83
0.80	16.2	11.8	3.92
1.11	16.2	11.8	3.92
1.43	16.2	11.8	3.92
1.75	16.1	11.7	3.89
4.29	15.5	11.2	3.72

ACKNOWLEDGEMENTS

The measurements and calculations on the largest sphere of solution reported were done by D. W. Magnuson, now of the Oak Ridge Gaseous Diffusion Plant, who also collated the other data. The measurement of the cylindrical volumes of solution was made by D. F. Cronin, now of United Nuclear Corporation; J. K. Fox, now of Kerr-McGee Corporation; L. W. Gilley of the Oak Ridge National Laboratory; and R. K. Reedy, Jr., of the Oak Ridge Critical Experiments Facility.

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