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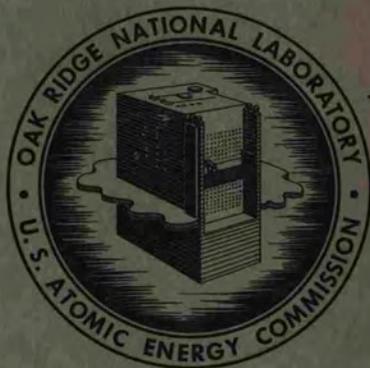
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CRITICAL MASS STUDIES  
PART VIII  
AQUEOUS SOLUTIONS OF U<sup>233</sup>

J. K. Fox  
L. W. Gilley  
E. R. Rohrer



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CRITICAL MASS STUDIES, PART VIII  
AQUEOUS SOLUTIONS OF U<sup>233</sup>

J. K. Fox, L. W. Gilley and E. R. Rohrer

Date Issued

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## ABSTRACT

A series of experiments have been performed to establish the critical parameters of aqueous solutions of uranyl nitrate and uranyl fluoride in which the uranium contained 98.7%  $U^{233}$ . Solutions were made critical in both spherical and cylindrical geometries with paraffin or water as a neutron reflector and, in two instances, with no reflector. The  $U^{233}$  concentration varied from 30 to 600 g/liter. The minimum critical mass observed was 590 g of  $U^{233}$  in the solution having an H: $U^{233}$  atomic ratio of 419 occupying a 10.4-in.-dia sphere. The minimum measured volume was 3.66 liters in a 6.7-in. equilateral cylinder containing a solution with an H: $U^{233}$  atomic ratio of 39.4. Extrapolated source neutron multiplication data indicate that a 5-in.-dia cylinder can be made critical if reflected, but a 4-in.-dia cylinder would be subcritical at all moderations. It was also found that 2.02 kg of  $U^{233}$  in an unreflected 10-in. equilateral cylinder is critical with a solution having an H: $U^{233}$  atomic ratio of 154. An unreflected sphere 12.6 in. in diameter is critical with 1.14 kg of  $U^{233}$  in a solution with an H: $U^{233}$  ratio of 381. Extension of the data to geometries other than those used experimentally was made by an empirical calculation.

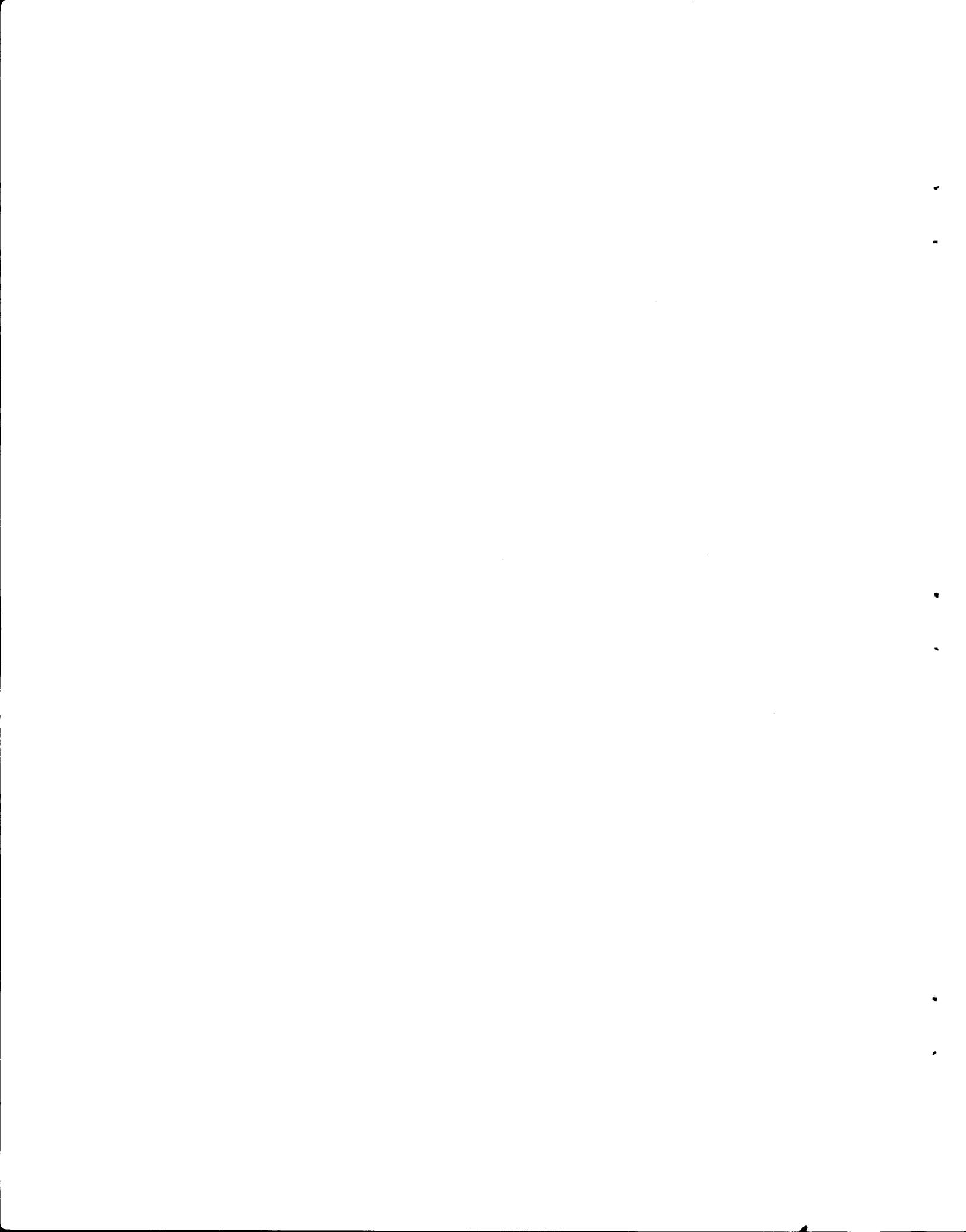
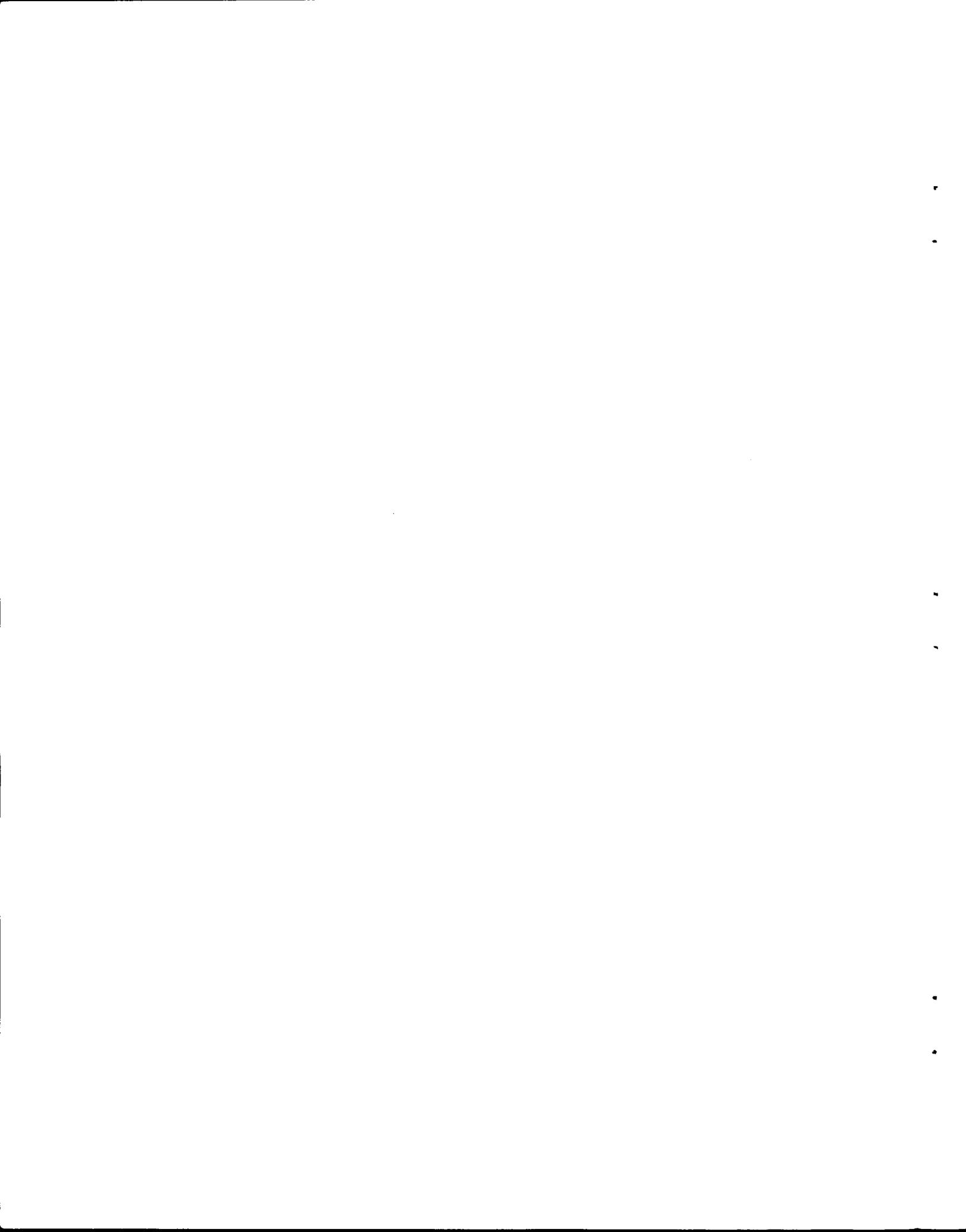


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## INTRODUCTION

Nuclear safety in processing plants for fissionable materials can be imposed by limiting equipment dimensions, batch sizes, and other plant variables in accordance with the results of experimental investigations of the critical parameters of the fissionable material of interest. Experiments of this kind have been performed at Hanford<sup>1</sup> with plutonium and have been in progress at Oak Ridge<sup>2</sup> with U<sup>235</sup> for some time, but the unavailability of sufficient amounts of U<sup>233</sup> has prohibited any study of this isotope. A limited experimental program became possible, however, when approximately 2.5 kg of isotopically pure U<sup>233</sup> was released to the ORNL Critical Experiments Facility in 1952. A series of critical experiments were performed in which the U<sup>233</sup> was used in aqueous solutions contained in cylindrical and spherical vessels and reflected with paraffin or water. The solution concentrations varied from 30 to 600 g of U<sup>233</sup> per liter of solution. It was not possible to make all of the attempted experiments critical because of the limited inventory of U<sup>233</sup> or the limited capacity of some of the test vessels. Source neutron multiplication measurements in these subcritical experiments were extrapolated to give estimates of the critical conditions. It was also possible to make two unreflected vessels critical.

A summary of all of these measurements and an extension of them by an empirical calculation has provided some design basis for equipment for processing the U<sup>233</sup> isotope.

### I. EXPERIMENTAL APPARATUS AND PROCEDURE

The uranium used in these experiments contained 98.7% U<sup>233</sup>, the remainder being about equal quantities of U<sup>234</sup> and U<sup>238</sup>. It was first used as an aqueous solution of uranyl nitrate (UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>) and later as a solution of the more soluble uranyl fluoride (UO<sub>2</sub>F<sub>2</sub>) in order to extend the measurements to higher chemical concentrations and to reduce the neutron absorption. The solutions were contained in cylinders having diameters between 4-1/2 and 12 in. and in spheres having diameters of 10.4 and 12.6 in. The vessels in most of the experiments had effectively infinite hydrogenous reflectors, but in two cases it was possible to achieve criticality with no reflector. The critical assemblies were of simple geometry and relatively free of extraneous neutron absorbers.

1. F. E. Kruesi, J. O. Erkman and D. D. Lanning, "Critical Mass Studies of Plutonium Solutions," HW-24512 (1952).
2. See, for example, J. K. Fox, L. W. Gilley and D. Callihan, "Critical Mass Studies, Part IX, Aqueous U<sup>235</sup> Solutions," ORNL-2367 (1958).

Since neutron moderation was one of the principal variables investigated, the chemical concentrations of the solutions were altered in the course of the experiments and H:U<sup>233</sup> atomic ratios were determined from uranium analyses. The uranyl nitrate solution was shown to contain some excess nitric acid, equivalent to a total H:U<sup>233</sup> atomic ratio of 2.66. Some of the properties of the solutions, including the results of spectrochemical analyses for metallic impurities resulting from corrosion of the equipment, are given in Appendix A.

The solution vessels were constructed of type 3S aluminum and coated with thin layers of either Heresite or Unichrome to reduce corrosion. Neither the Heresite nor the Unichrome contains any significant neutron-absorbing metallic impurities, although Unichrome is about 30 wt% chlorine and is thus a weak neutron absorber. Estimates of the effect of the Unichrome on the experimental results are noted in the recorded data.

The cylinders having diameters less than 6.7 in. were 36 in. high, while those with diameters of 6.7 to 12 in. were approximately equilateral. The cylinders of larger diameter and the spheres were mounted in an outer cylinder of sufficient capacity to provide an effectively infinite water or paraffin neutron reflector completely surrounding the vessel under study. The smaller diameter cylinders had reflectors on the sides and bottom but not on the top. Neoprene gaskets were used to seal the cylinder covers.

Immediately adjacent to the periphery of each cylinder was a water-filled annular section into which a cadmium sheet, normally suspended from a magnet, could be inserted as a safety operation. A photograph of one of the vessels without a reflector is shown in Fig. 1. A neutron source, located in the reflector region, was positioned by remote control.

The solution flowed into the cylinders or vessels from a storage manifold which consisted of five 3-in.-dia type 347 stainless steel pipes mounted vertically on a bracket and spaced on 15-in. centers with their axes coplanar as shown in Fig. 2. The five pipes were each 37 in. long and were connected by smaller pipes at both the top and the bottom. The manifold had a total capacity of 23 liters and was connected by flexible tubing and diaphragm-type valves at the top and bottom of the vessel under test, thereby forming a closed transfer system. This design, which is shown in schematic form in Fig. 3, reduced health hazards and utilized the limited inventory of U<sup>233</sup> as effectively as possible. The manifold was suspended from a rack which could be moved vertically by a remotely controlled motor. The ability to lower the manifold to remove solution from a test vessel served as an additional safety device.

The radiation detection instrumentation was conventional and included four parallel channels arranged to operate the safety devices. The detectors in three of these channels were BF<sub>3</sub>-filled ionization chambers with signal amplification by dc amplifiers or vibrating reed electrometers. The signals were observed on strip chart recorders. The fourth circuit consisted of an anthracene crystal and a photomultiplier tube. While approaching critical, the apparent neutron source multiplication was measured by boron-lined proportional counters.

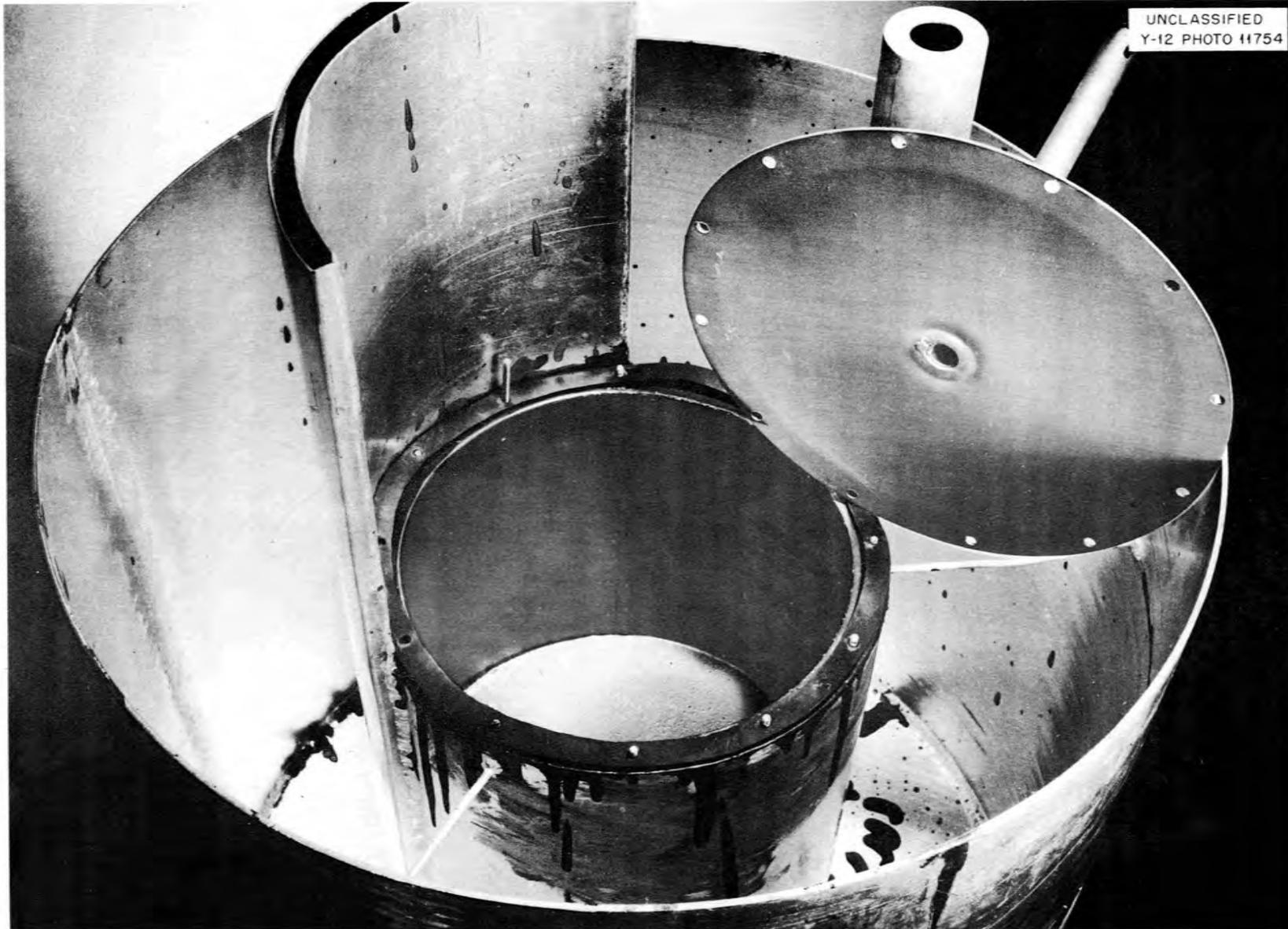


Fig. 1. Typical Containing Vessel for Experiments with Aqueous Solutions of  $U^{233}$ .  $U^{233}$  solution was contained in the inner cylinder and surrounded by a paraffin or water reflector.

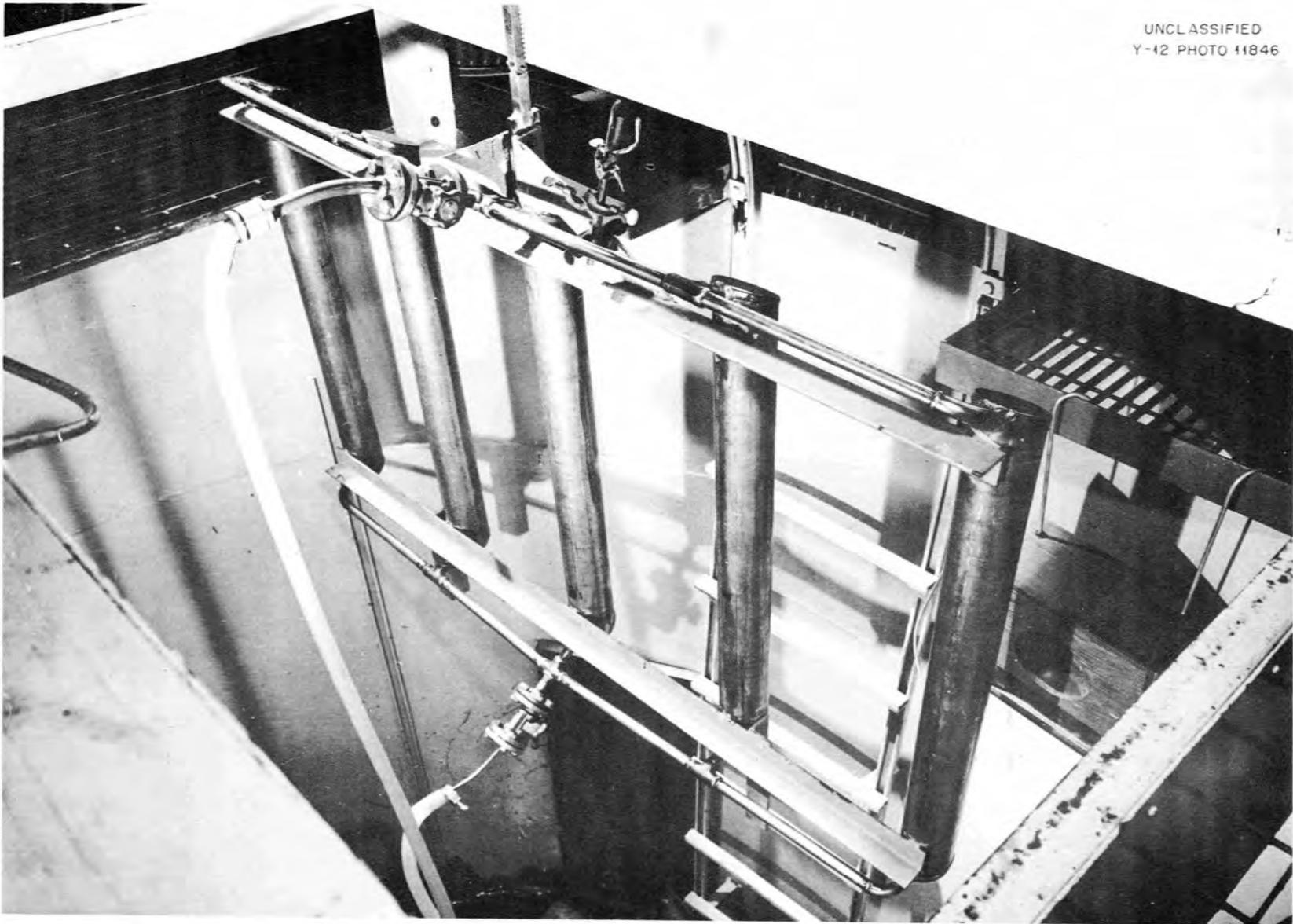


Fig. 2. Storage Manifold for Aqueous Solutions of  $U^{233}$ . The five pipes are 3 in. in diameter, 37 in. long, and mounted 15 in. apart.

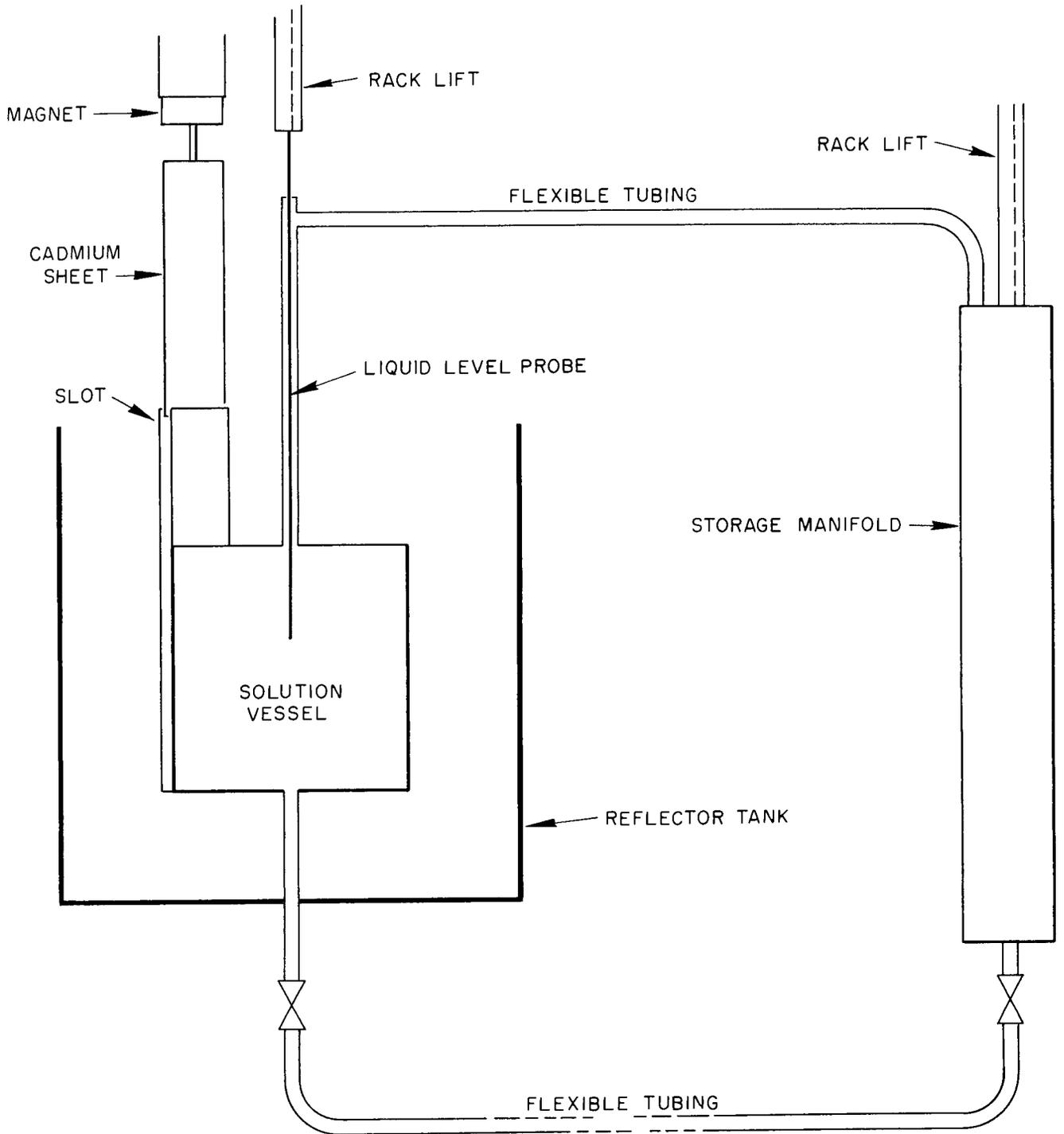


Fig. 3. Arrangement of Solution Vessel and Storage Manifold for Critical Experiments with Aqueous Solutions of  $U^{233}$

In the uranyl nitrate solution experiments, the liquid level in a vessel was determined from an empirical calibration curve relating the vertical position of the manifold to the areas of the manifold and the solution vessel. In the uranyl fluoride solution experiments the position of the liquid level in a vessel was indicated by a selsyn connected to a motor-driven electrical contact inserted through a rubber diaphragm across the tube connected to the top of the vessel.

The procedure in these experiments was similar to that used previously<sup>2</sup> with solutions of  $U^{235}$ , although the smaller delayed neutron yield of  $U^{233}$  required a more careful approach to critical. An effort was made in each case to adjust the concentration in order to have the system critical when full.

## II. EXPERIMENTAL RESULTS

The critical data obtained in these experiments are tabulated in Appendix B along with estimates of the critical parameters derived from the source neutron multiplication curves of those assemblies which could not be made critical because of limitations imposed by the available quantity of  $U^{233}$  or by the capacities of the containers. The results from critical assemblies and from the extrapolations in which there is greatest confidence are summarized in Figs. 4, 5, and 6.

Figure 4 is a plot of the critical mass as a function of the critical volume of reflected vessels. The data for uranyl nitrate and uranyl fluoride solutions contained in approximately equilateral cylinders can be represented by the same curve for the more dilute solutions, but, in the more concentrated range, the effect of the nitrate ion on the critical dimensions is measurable. This difference in critical masses at higher concentrations may be due to the lower hydrogen density in the uranyl nitrate solution. The  $U^{233}$  density in the two solutions is shown as a function of the H: $U^{233}$  ratio in Fig. A-1 in Appendix A.

The critical volumes and critical masses measured in spheres and essentially equilateral cylinders, each surrounded by a neutron reflector, are plotted in Figs. 5 and 6, respectively, as functions of the concentration of the solutions expressed as the H: $U^{233}$  atomic ratio. The curves in each figure have been drawn through the points describing the cylinders.

The two experiments without neutron reflectors used a 10-in.-dia equilateral cylinder and a 12.6-in.-dia sphere. The critical data for these two unreflected vessels are included in Appendix B.

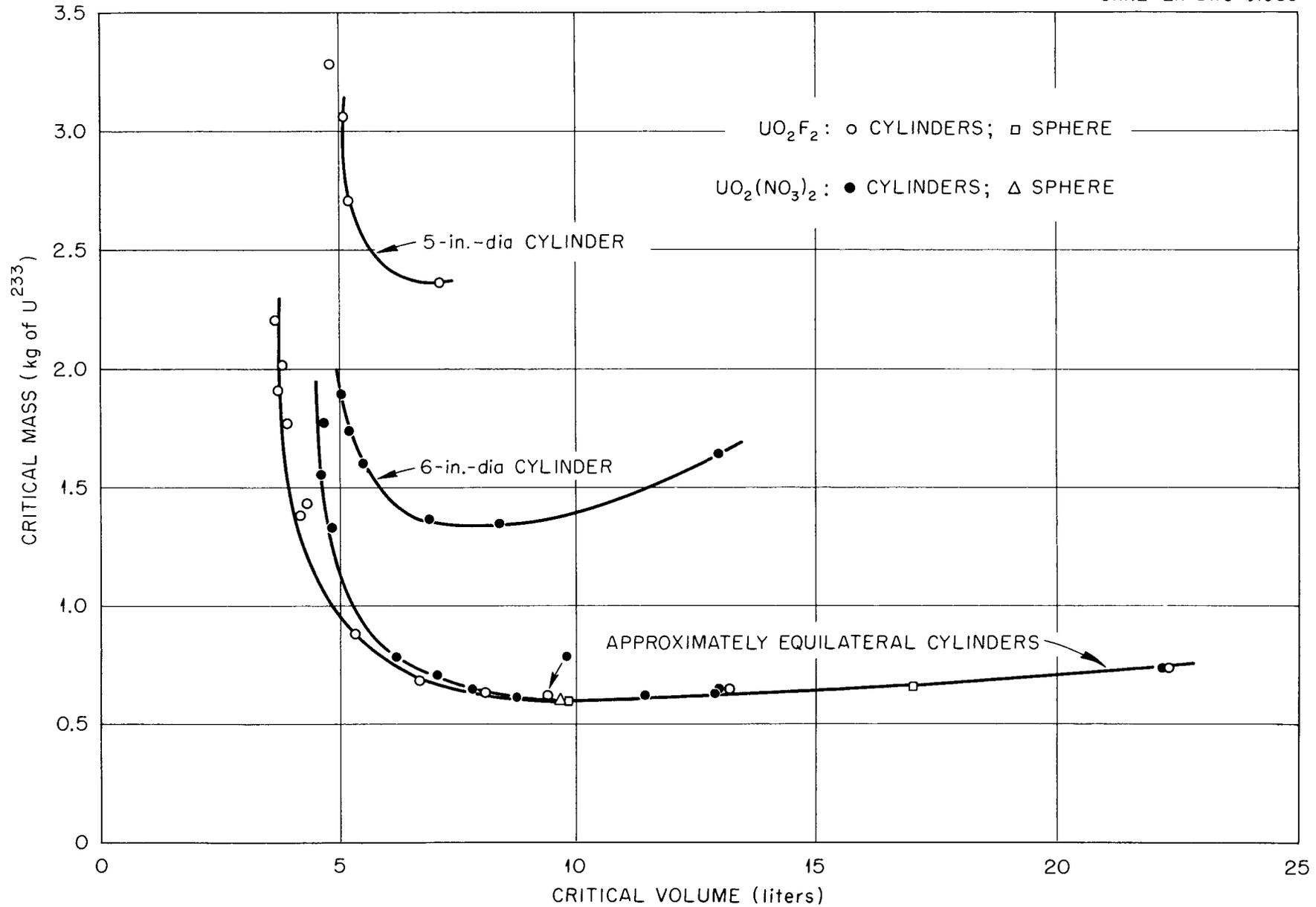


Fig. 4. Critical Mass as a Function of Critical Volume of Reflected Vessels Containing Aqueous Solutions of U<sup>233</sup>.

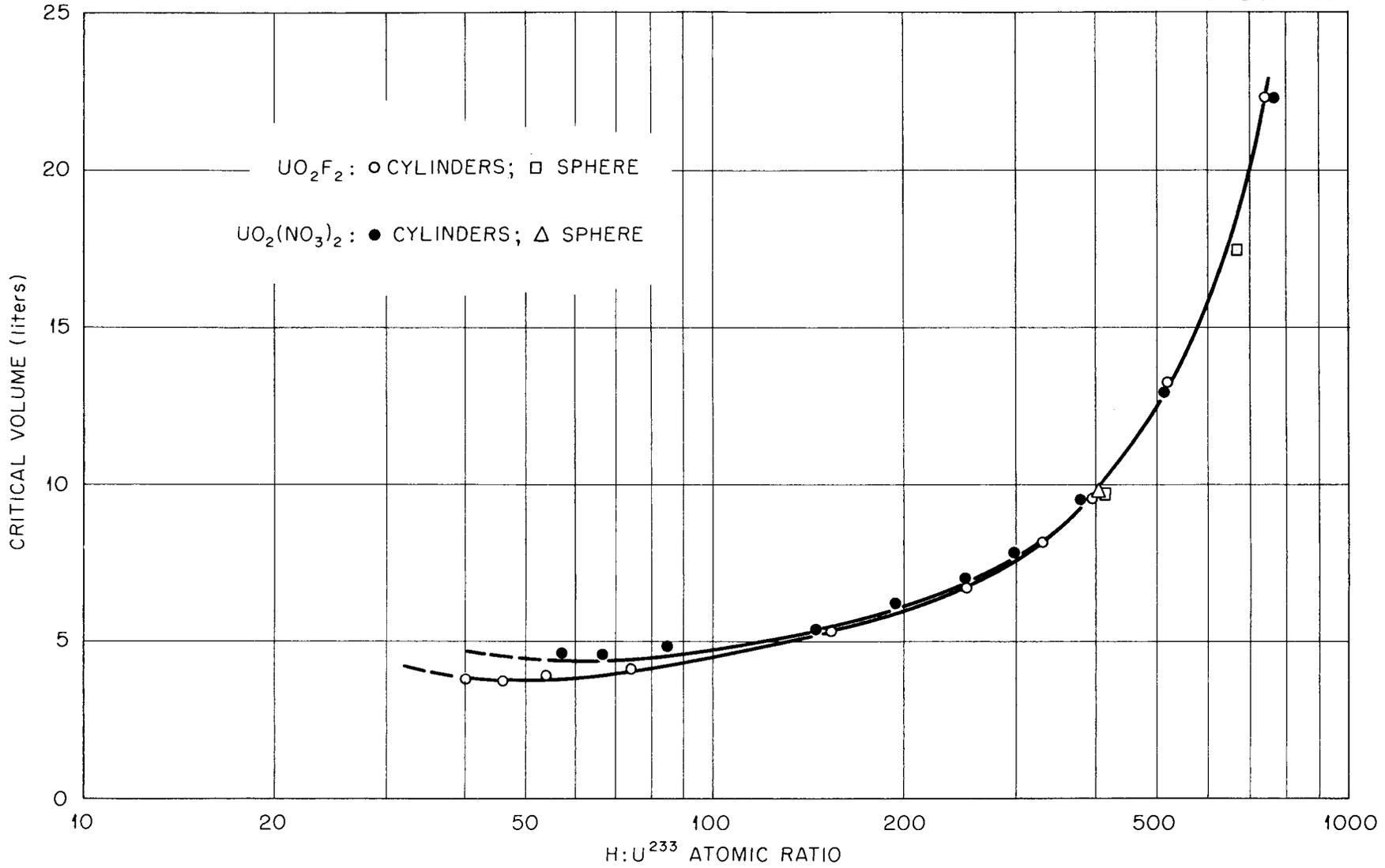


Fig. 5. Critical Volume as a Function of the H:U<sup>233</sup> Atomic Ratios of Solutions Contained in Reflected Equilateral Cylinders and Spheres

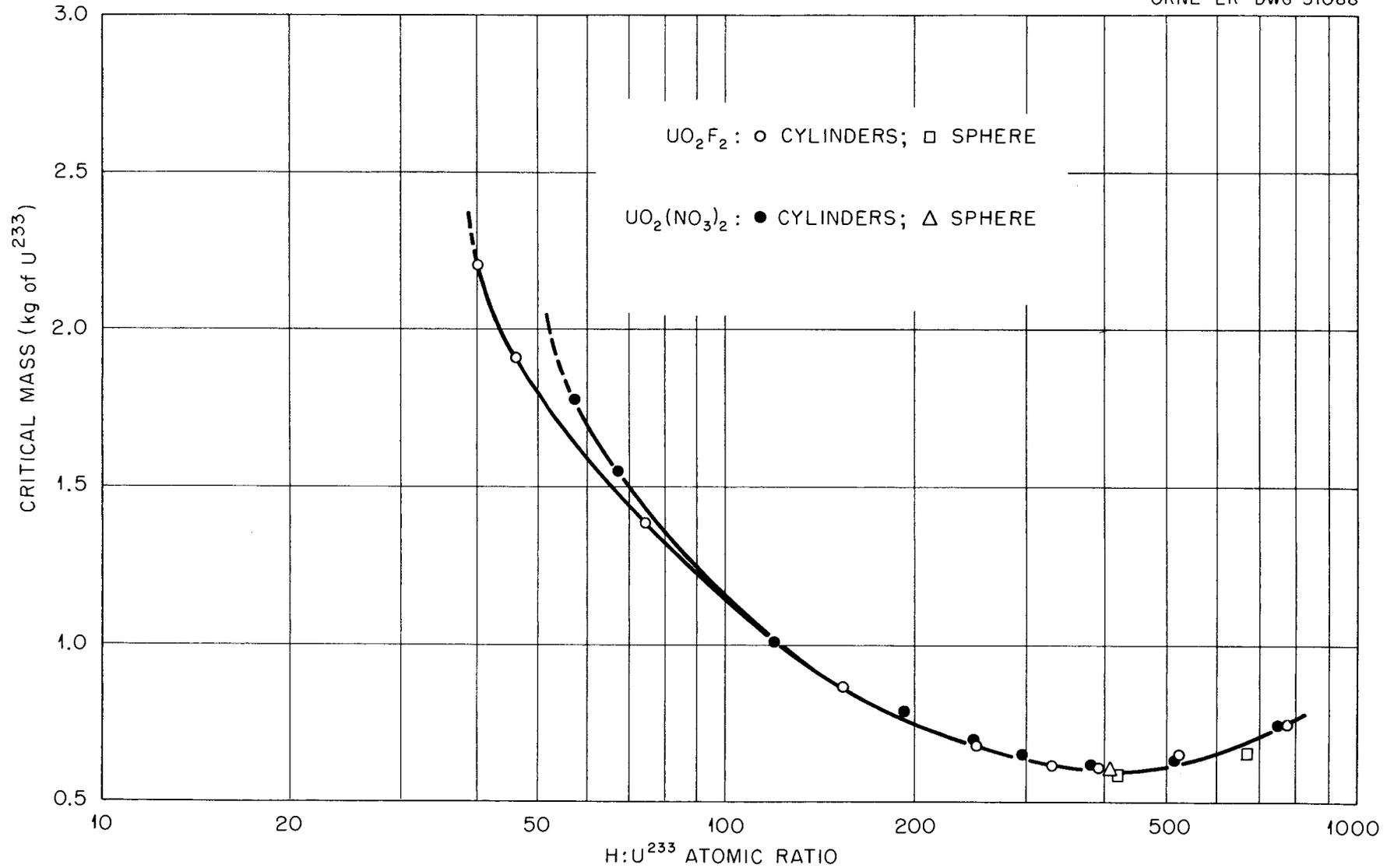


Fig. 6. Critical Mass as a Function of the H:U<sup>233</sup> Atomic Ratios of Solutions Contained in Reflected Equilateral Cylinders and Spheres

## III. ACCURACY OF THE MEASUREMENTS

The results of these experiments are subject to the usual errors in uranium analyses and solution densities and in the calibration of the vessel capacities, all estimated to be  $\pm 0.5\%$ . There are, however, two additional sources of error which broaden the uncertainty. In the experiments in which uranyl nitrate solutions were used the indirect method of measuring the height, and hence the volume, of the solution in the vessels makes the results less certain than those from later experiments with uranyl fluoride in which the position of the liquid surface was measured directly.

An error in the experiments with the uranyl fluoride solutions is a bias resulting from the properties of a corrosion-inhibiting coating material with which one spherical and three cylindrical vessels were lined. This coating was a polyvinyl chloride plastic, Unichrome, which is about 30 wt% chlorine. It is difficult to evaluate the increase in critical mass due to the absorption of neutrons by the chlorine because the effect is a function of geometry. It is noted in Table B-1<sup>2</sup>, however, that the critical volume at a particular concentration ( $H:U^{233} = 74.1$ ) in a 14-cm-dia cylinder lined with Unichrome is about the same as that in a 12.7-cm-dia cylinder lined with a phenol base plastic, Heresite, whereas it would be expected to be somewhat less. After the discovery of the chlorine impurity, one definitive experiment was performed to determine the effect of a Unichrome liner on the critical concentration of a  $U^{235}$  uranyl fluoride solution in a sphere 32 cm in diameter. A 2% decrease in the concentration was observed upon removal of the Unichrome.

In summarizing the question of accuracy, it is believed that the results from critical experiments with uranyl fluoride in Heresite-lined vessels are good to  $\pm 1\%$ ; data from the same vessels with uranyl nitrate have an uncertainty of no more than  $\pm 3\%$ . The critical mass measured in the 32-cm-dia sphere is high by about 2% because of a bias. The masses in the 14- and 17-cm-dia cylinders are too high by an amount not well known but probably less than 10%.

Extrapolations have been made of the source neutron multiplication curves obtained in those tests which could not be made critical because of inventory or geometric limitations. The critical parameters obtained in this manner are given in the tabulated results with the maxima from which the extrapolations were made. The accuracy of these values is strongly dependent upon the length of the extrapolation.

## Appendix A

## COMPOSITION OF URANIUM SOLUTIONS

Table A-1. U<sup>233</sup> Concentrations in Solutions

H:U <sup>233</sup> Atomic Ratio	Solution Density (g/cc)	U <sup>233</sup> Concentration	
		g per g of Solution	g per cc of Solution
In UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> Solution with N:U <sup>233</sup> = 2.66			
42.2	1.697	0.289	0.490
57.5	1.543	0.247	0.381
67.0	1.480	0.227	0.336
84.4	1.394	0.197	0.275
120	1.287	0.154	0.198
145	1.238	0.135	0.167
151	1.232	0.130	0.160
193	1.185	0.107	0.127
213	1.165	0.100	0.117
247	1.145	0.088	0.101
297	1.121	0.075	0.084
356	1.101	0.064	0.070
379	1.093	0.061	0.067
394	1.090	0.058	0.063
405	1.087	0.057	0.062
461	1.077	0.051	0.055
514	1.069	0.046	0.049
582	1.061	0.041	0.044
630	1.056	0.038	0.040
757	1.046	0.032	0.033
In UO <sub>2</sub> F <sub>2</sub> Solution			
34.2	1.801	0.380	0.684
39.4	1.707	0.352	0.600
45.2	1.625	0.327	0.531
45.9	1.604	0.324	0.519
47.9	1.592	0.316	0.503
53.7	1.530	0.295	0.451
74.1	1.388	0.239	0.332
153	1.199	0.138	0.165
154	1.198	0.138	0.165
250	1.121	0.091	0.102
329	1.090	0.071	0.078
381	1.079	0.062	0.067
390	1.076	0.061	0.066
396	1.075	0.060	0.065
419	1.071	0.057	0.061
426	1.070	0.056	0.060
522	1.059	0.047	0.049
663	1.043	0.037	0.039
775	1.035	0.032	0.033

Table A-2. Isotopic Composition of Uranium in Solutions

Isotope	wt%	
	In $\text{UO}_2(\text{NO}_3)_2$ Solution	In $\text{UO}_2\text{F}_2$ Solution
U <sup>233</sup>	98.7	98.7
U <sup>234</sup>	0.50	0.54
U <sup>235</sup>	0.01	0.04
U <sup>238</sup>	0.79	0.72

Table A-3. Principal Impurities\* in Solutions

Element	ppm			
	In $\text{UO}_2(\text{NO}_3)_2$ Solution		In $\text{UO}_2\text{F}_2$ Solution	
	Before Use	After Use	Before Use	After Use
Al	15	50	400	8000
Ca	300	1500		
Cr	10	15	8	250
Fe	20	300	40	2500
Mg	10	125	0	55
Mo			0	200
Na			650	100
Ni	20	70	0	250
Sn			0	225
Th	150			

\* Determined by spectrographic analysis.

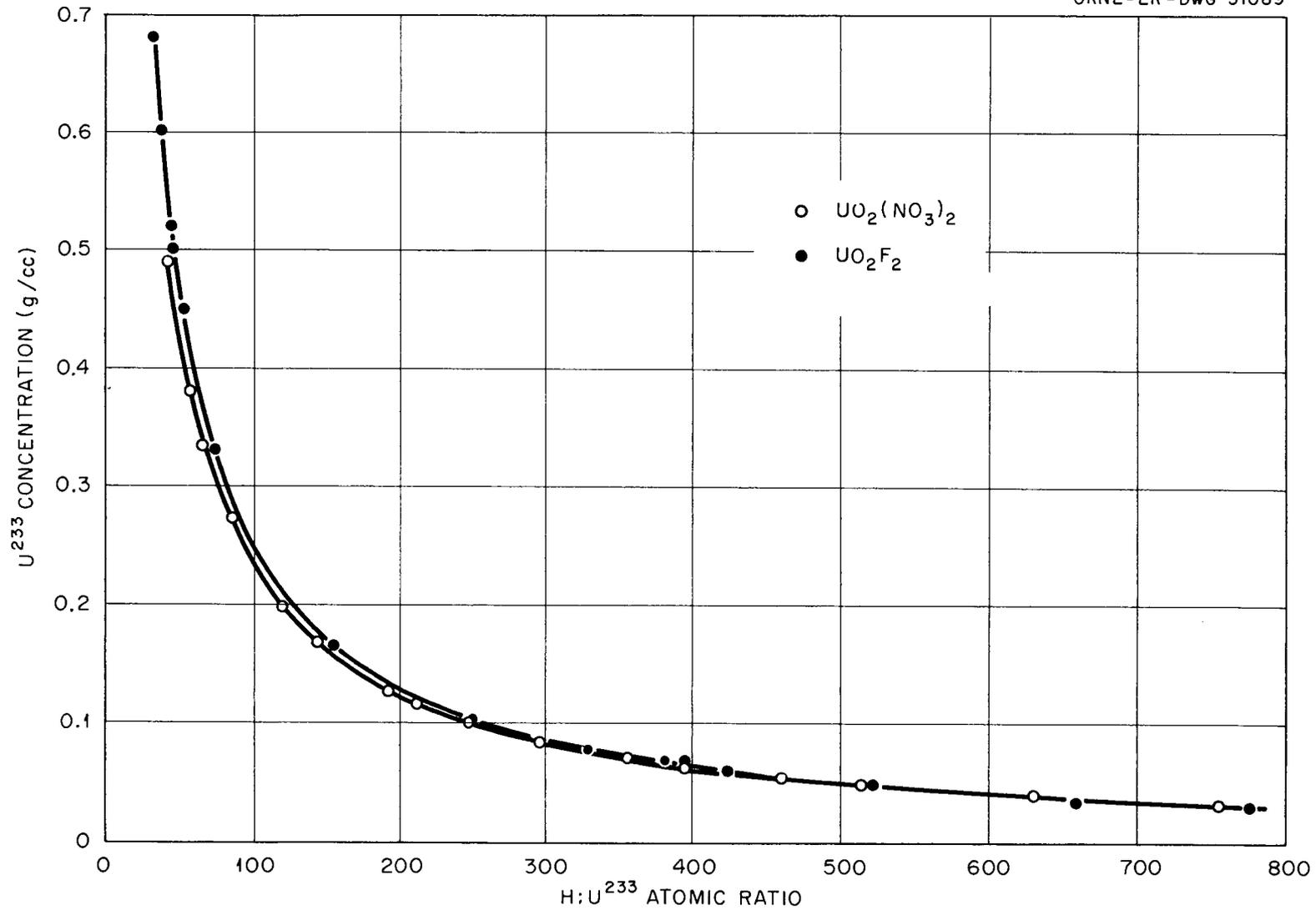


Fig. A-1.  $U^{233}$  Concentration in Aqueous Solutions of Uranyl Nitrate and Uranyl Fluoride as a Function of the H: $U^{233}$  Atomic Ratio.

Appendix B

TABULATIONS OF CRITICAL DATA

Table B-1. Critical Parameters of Uranyl Nitrate Solutions

(Numbers in parentheses represent extrapolated values derived from source neutron multiplication curves of subcritical assemblies.)

H:U <sup>233</sup> Atomic Ratio <sup>a</sup>	Reflector	Critical Parameters			Maximum Values of Subcritical Assemblies		
		Height (cm)	Volume (liters)	Mass (kg of U <sup>233</sup> )	Height (cm)	Volume (liters)	Mass (kg of U <sup>233</sup> )
Cylinder 12.7 cm (5.0 in.) in Diameter <sup>b</sup>							
57.5	Paraffin	c			51	6.40	2.44
67.0	"	c			59	7.40	2.49
84.4	"	c			61	7.65	2.10
145	"	c			55	6.90	1.15
Cylinder 15.1 cm (6.0 in.) in Diameter <sup>b</sup>							
57.5	Paraffin	27.9	5.00	1.91			
67.0	"	29.0	5.20	1.75			
84.4	"	30.7	5.50	1.51			
120	"	(38.5 ± 0.5)	(6.9 ± 0.1)	(1.37 ± 0.02)	36.8	6.60	1.30
151	"	(46.8 ± 0.5)	(8.4 ± 0.1)	(1.34 ± 0.02)	45.4	8.15	1.31
193	"	(73 ± 2)	(13.0 ± 0.4)	(1.65 ± 0.08)	55.4	9.80	1.24
Cylinder 19.1 cm (7.5 in.) in Diameter							
57.5	Paraffin	16.3	4.65	1.77			
67.0	"	16.2	4.60	1.55			
145	"	18.6	5.30	0.89			
Cylinder 20.5 cm (8.0 in.) in Diameter							
42.2	Paraffin	(16.1 ± 0.2)	(5.30 ± 0.06)	(2.6 ± 0.03)	14.0	4.60	2.25
57.5	"	14.4	4.75	1.81			
84.4	"	14.7	4.85	1.33			
120	"	16.4	5.40	1.07			
145	"	16.7	5.51	0.92			
151	"	16.7	5.51	0.88			
193	"	18.8	6.20	0.79			
213	"	19.3	6.37	0.75			
247	"	(21.2 ± 0.3)	(7.0 ± 0.1)	(0.70 ± 0.01)	19.9	6.56	0.66
Cylinder 21.5 cm (8.5 in.) in Diameter							
247	Paraffin	19.4	7.00	0.70			
297	"	21.5	7.78	0.65			
Cylinder 22.9 cm (9.0 in.) in Diameter							
356	Paraffin	21.3	8.75	0.62			
379	"	22.9	9.39	0.62			
Cylinder 25.5 cm (10.0 in.) in Diameter							
394	Paraffin	19.3	9.82	0.63			
461	"	22.5	11.45	0.63			
514	"	(25.2 ± 0.1)	(12.9 ± 0.05)	(0.63 ± 0.03)	25.1	12.77	0.63
514	Water	(25.5 ± 0.1)	(13.0 ± 0.05)	(0.64 ± 0.03)	25.1	12.77	0.63
Cylinder 30.5 cm (12.0 in.) in Diameter							
582	Paraffin	21.1	15.40	0.68			
630	"	23.8	17.40	0.70			
757	"	30.4	22.20	0.75			
Sphere 26.6 cm (10.4 in.) in Diameter							
405	Water	Full	9.66	0.60			

a. The compositions of the solutions are given in Appendix A.

b. No reflector on the top surface.

c. Apparently this assembly cannot be made critical at any height.

Table B-2. Critical Parameters of Uranyl Fluoride Solutions

(Numbers in parentheses represent extrapolated values derived from source neutron multiplication curves for subcritical assemblies.)

H:U <sup>233</sup> Atomic Ratio <sup>a</sup>	Reflector	Critical Parameters			Maximum Values of Subcritical Assemblies		
		Height (cm)	Volume (liters)	Mass (kg of U <sup>233</sup> )	Height (cm)	Volume (liters)	Mass (kg of U <sup>233</sup> )
Cylinder 11.2 cm (4.5 in.) in Diameter <sup>b,c</sup>							
34.2	Paraffin	d			29.9	2.95	2.02
39.4	"	d			34.9	3.43	2.07
45.9	"	d			42.6	4.19	2.18
53.7	"	d			49.0	4.82	2.18
74.1	"	d			68.5	6.76	2.24
Cylinder 12.7 cm (5.0 in.) in Diameter <sup>b</sup>							
34.2	Paraffin	(38 ± 2)	(4.8 ± 0.25)	(3.3 ± 0.2)	23.8	2.99	2.05
39.4	"	(41 ± 2)	(5.1 ± 0.25)	(3.1 ± 0.2)	27.6	3.46	2.08
45.9	"	(41 ± 1)	(5.2 ± 0.1)	(2.7 ± 0.1)	32.4	4.07	2.11
74.1	"	(56.5 ± 0.5)	(7.1 ± 0.06)	(2.36 ± 0.02)	53.3	6.70	2.22
Cylinder 13.7 cm (5.4 in.) in Diameter <sup>b,c</sup>							
74.1	Paraffin	(48.7 ± 0.5)	(7.13 ± 0.06)	(2.37 ± 0.02)	46.3	6.77	2.25
Cylinder 15.1 cm (6.0 in.) in Diameter <sup>b,c</sup>							
74.1	Paraffin	24.0	4.31	1.43			
Cylinder 16.7 cm (6.6 in.) in Diameter <sup>c</sup>							
34.2	Paraffin	(20 ± 1)	(4.4 ± 0.2)	(3.0 ± 0.15)	13.5	2.94	2.01
39.4	"	(16.7 ± 0.2)	(3.66 ± 0.04)	(2.20 ± 0.03)	16.3	3.55	2.13
45.2	"	(17.4 ± 0.2)	(3.79 ± 0.04)	(2.01 ± 0.03)	16.9	3.69	1.96
45.9	"	16.9	3.67	1.91			
47.9	"	(17.7 ± 0.2)	(3.85 ± 0.04)	(1.94 ± 0.03)	16.9	3.69	1.86
53.7	"	(18.0 ± 0.3)	(3.93 ± 0.06)	(1.77 ± 0.03)	16.9	3.69	1.66
74.1	"	(19.1 ± 0.4)	(4.15 ± 0.09)	(1.38 ± 0.04)	16.9	3.69	1.23
Cylinder 19.1 cm (7.5 in.) in Diameter							
154	Paraffin	18.4	5.25	0.87			
Cylinder 20.5 cm (8.0 in.) in Diameter							
250	Paraffin	(20.2 ± 0.05)	(6.66 ± 0.02)	(0.68 ± 0.02)	20.1	6.63	0.68
Cylinder 21.5 cm (8.5 in.) in Diameter							
329	Paraffin	(22.2 ± 0.1)	(8.04 ± 0.04)	(0.63 ± 0.05)	21.6	7.80	0.61
Cylinder 22.9 cm (9.0 in.) in Diameter							
396	Paraffin	(23.1 ± 0.1)	(9.47 ± 0.04)	(0.61 ± 0.05)	22.6	9.26	0.60
Cylinder 25.5 cm (10.0 in.) in Diameter							
522	Water	(25.9 ± 0.1)	(13.18 ± 0.05)	(0.65 ± 0.05)	25.6	13.05	0.64
154	None	(24.0 ± 0.05)	(12.22 ± 0.03)	(2.02 ± 0.05)	23.8	12.15	2.01
Cylinder 30.5 cm (12.0 in.) in Diameter							
775	Paraffin	30.5	22.28	0.74			
Sphere 26.6 cm (10.4 in.) in Diameter							
426	Water		(9.80 ± 0.10)	(0.59 ± 0.06)	Full	9.66	0.58
419	"	e	9.62	0.59			
390	"	f	9.28	0.61			
Sphere 31.9 cm (12.6 in.) in Diameter							
663	Water	Full	17.02	0.66			
381	None	e	16.98	1.14			

- a. The compositions of the solutions are given in Appendix A.
- b. No reflector on the top surface.
- c. Vessels coated with Unichrome; masses about 2% high because of impurities.
- d. Apparently this assembly cannot be made critical at any height with the absence of a top reflector and the presence of Unichrome.
- e. There was a 40-cm<sup>3</sup> void above the critical solution.
- f. There was a 380-cm<sup>3</sup> void above the critical solution.

## Appendix C

### ESTIMATION OF CRITICAL PARAMETERS FOR UNREFLECTED SYSTEMS

The results from experiments performed almost exclusively with equilateral cylinders have been extrapolated to other cylindrical geometries using the method developed and applied by Bell<sup>3</sup> to the extensive data on the critical parameters of U<sup>235</sup> solutions.<sup>4</sup> It is assumed that the age-theory critical equation can be made to fit experimental data from reflected and unreflected critical assemblies by proper choice of the extrapolation distances. In this analysis the reactivity is given by:

$$k_{\text{eff}} = \frac{\eta f e^{-\gamma B^2}}{(1 + L_{\text{th}}^2 B^2)} = \frac{\eta}{1 + \frac{\sigma_a(\text{H})}{\sigma_a(\text{U}^{235})} C} \frac{e^{-\gamma B^2}}{(1 + L_{\text{th}}^2 B^2)}$$

where

$$L_{\text{th}}^2 = \frac{L_0^2}{1 + \frac{\sigma_a(\text{U}^{235})}{\sigma_a(\text{H})C}},$$

$L_0$  = diffusion length of thermal neutrons in water,

$C$  = H:U<sup>235</sup> atomic ratio.

The constants used for the U<sup>235</sup> solutions are as follows:

$$L_0^2 = 8.2 \text{ cm}^2,$$

$$\frac{\sigma_a(\text{H})}{\sigma_a(\text{U}^{235})} = 0.328/575,$$

$$\gamma = 27 \text{ cm}^2 \text{ (except as noted below);}$$

$$\eta = 2.33.$$

Values for the extrapolation distances for reflected and unreflected cylinders were derived from a comparison between calculated and experimental values of critical volumes of equilateral cylinders. These values are 8.25 cm and 4.95 cm for reflected and unreflected systems, respectively. The axial and radial extrapolation distances were assumed equal in calculating the buckling. It was found necessary to apply a multiplying factor (1.16) to the ratio of the thermal absorption cross sections of hydrogen and U<sup>235</sup> in order to have both  $\gamma$  and the extrapolation distance as constants and still fit the uranyl fluoride data in the high concentration region. It was also necessary to arbitrarily vary the neutron age with concentration, as shown in Fig. C-1, to obtain agreement with the data from the nitrate experiments in the region of low moderation.

3. G. I. Bell, "A Method of Calculating Critical Masses of Proton Moderated Assemblies," LA-1548 (1953).

4. C. K. Beck, A. D. Callihan, J. W. Morfitt, and R. L. Murray, "Critical Mass Studies, Part III," K-343 (1949).

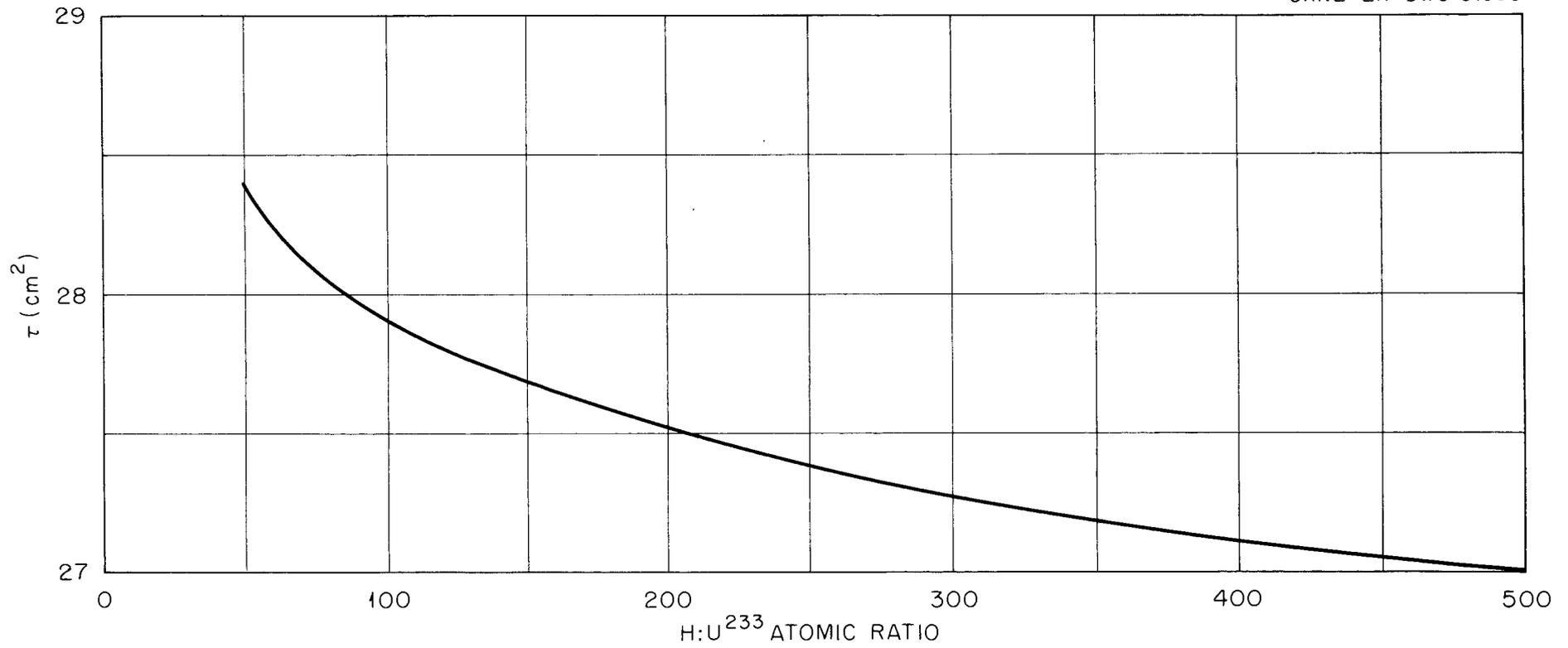


Fig. C-1. Variation of Neutron Age ( $\tau$ ) with the H: $\text{U}^{233}$  Atomic Ratio in Uranyl Nitrate Solutions

Since only two experimental determinations were made with unreflected vessels, one cylinder and one sphere, the extrapolation length adjusted to fit these points is highly uncertain. Similar comparisons using data for aqueous solutions of  $U^{235}$  in spheres indicate that the extrapolation length is always higher than for cylinders unless one applies a factor (0.965) to  $\pi^2$  in the sphere buckling equation. If this factor is used, the extrapolation distance from the unreflected  $U^{235}$  sphere experiment agrees with that found with the cylinder. Figure C-2 is a plot of the calculated values of critical masses and volumes of unreflected cylinders using 4.95 cm for the extrapolation distance. These curves are only very rough approximations of these parameters since the supporting data are very limited.

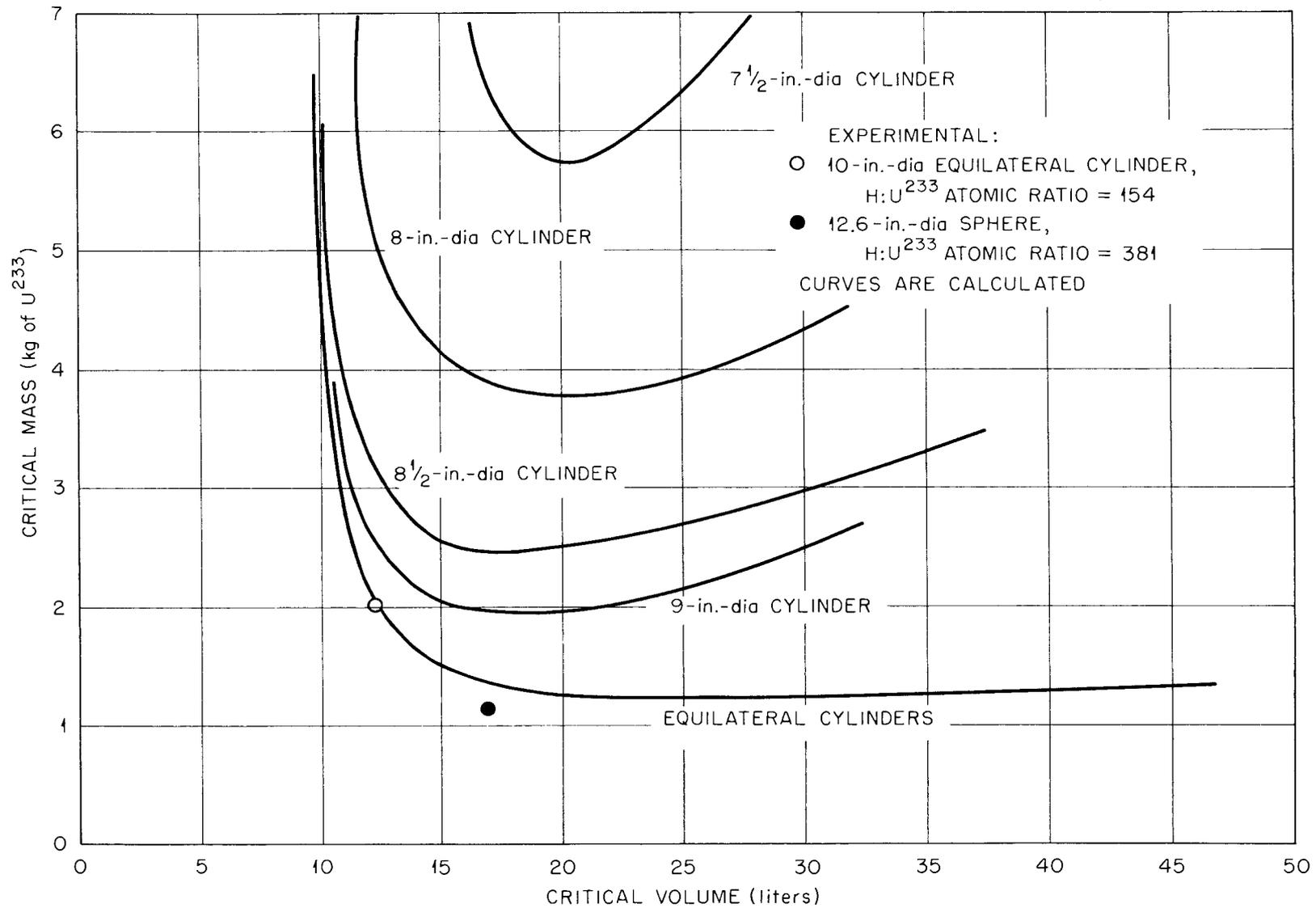


Fig. C-2. Estimated Critical Parameters of Unreflected Vessels Containing Aqueous Solutions of  $U^{233}$

## Appendix D

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