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NEUTRON AGE IN MIXTURES OF LIGHT AND HEAVY WATER

by

J. W. Wade

Experimental Physics Division

June 1956

E. I. du Pont de Nemours & Co.
Explosives Department - Atomic Energy Division
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ABSTRACT

The age for fission neutrons was determined for six mixtures of D_2O and H_2O by measuring the neutron slowing-down distributions at 1.4 ev. Similar measurements were made in a lattice of aluminum rods in D_2O. In this case, the slowing-down distributions in the directions parallel and perpendicular to the rods were different, so that the age was anisotropic.

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NEUTRON AGE IN MIXTURES OF LIGHT AND HEAVY WATER

INTRODUCTION

Measurements of the neutron age(1) in mixtures of light and heavy water were undertaken as part of a program to study the effects of contamination by light water in reactors moderated with heavy water. The only previous work in this field is a single measurement of the neutron slowing-down distribution in 99.8 per cent D_2O(2). No measurements were available for mixtures of light and heavy water, although some theoretical estimates have been made(3).

It has been predicted that streaming of fast neutrons along aluminum rods will result in an anisotropy of the slowing-down distances(4). The testing of this prediction required that measurements be made in a nonmultiplying lattice of aluminum rods in D_2O.

SUMMARY

The neutron age from fission energies to 1.4 ev was determined experimentally for six mixtures of light and heavy water ranging from 99.8 to 91.8 per cent D_2O.* The age was evaluated from the distribution of the slowing-down density of neutrons in the moderating media. The slowing-down density was determined by activating cadmium-covered indium foils with neutrons from a "point" fission source.

Over the range of water mixtures studied, the age decreased as a linear function of the H_2O concentration, within experimental error. The values of the age ranged from 109 ± 3 cm^2 in 99.8 per cent D_2O to 78 ± 3 cm^2 in 91.8 per cent D_2O.

A large anisotropy in age was found in a lattice of aluminum rods in 99.3 per cent D_2O. The age, as calculated from the neutron slowing-down density in the direction parallel to the rods, was 140 cm^2, while it was 108 cm^2 in the direction perpendicular to the rods. The latter value is equal, within experimental error, to the age for pure moderator.

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* Concentrations of D_2O, listed in this report, are in terms of mol per cent D_2O.

DISCUSSION

THEORY

The neutron age between two energies is defined as one-sixth of the mean square distance traveled by the neutrons as they are slowed down from the higher energy to the lower energy. It can be evaluated from the following relation:

$$\tau(E_o,E) = \frac{\overline{r^2}\ (E_o,E)}{6} = \frac{\int_v r^2\ q(r,E)dv}{6\int_v q(r,E)dv} \tag{1}$$

where

q = slowing-down density of neutrons at energy E, neutrons - cm^{-3}-sec^{-1}

E_o = initial energy of the neutrons

E = lower energy of the neutrons

r = distance from the source, cm

v = volume, cm^3

The neutron age between the energy of fission neutrons and 1.4 ev can be determined by activating a large number of cadmium-covered indium foils placed at various distances from a fission source. The foil activities are proportional to the slowing-down densities at 1.4 ev, as shown below.

The activity of a foil exposed at the distance r from the source of fission neutrons, A(r), can be written as

$$\Lambda(r) \sim \int_{E_{Cd}}^{E_{fission}} \Sigma_{In}(E)\Phi(E,r)dE \tag{2}$$

where

Σ_{In} (E) is the macroscopic absorption cross section of indium,

and

$\Phi(E)$ is the isotropic(5) component of the neutron flux at the energy E and at the distance r.

The integration is performed from the energy of the cadmium cutoff to the energy of fission neutrons. The activity of the foil is obtained by averaging the activities of both sides, since this average is proportional to the isotropic neutron flux(5).

For nonabsorbing or weakly absorbing media, such as those investigated in this report, Equation 2 can be written(1)

$$A(r) \sim \int_{E_{Cd}}^{E_{fission}} q \frac{\Sigma_{In}}{\xi\Sigma_s} \frac{dE}{E}$$

where

q is the slowing-down density

and

$\xi\Sigma_s$ is the slowing-down power of the moderator.

Since the cross section of indium is nearly zero except at 1.4 ev, the foil activity is proportional to the slowing-down density at 1.4 ev.

In terms of the foil activities, Equation 1 becomes now, in spherical coordinates

$$\tau(\text{fission to } 1.4 \text{ ev}) = \frac{\overline{r^2}}{6} = \frac{4\pi \int_0^\infty r^4 A(r) dr}{24\pi \int_0^\infty r^2 A(r) dr} \qquad (3)$$

DESCRIPTION OF EXPERIMENTS

Experimental Facility

The experiments were performed in the aluminum tank of an exponential facility(6). The tank is five feet in diameter and seven feet high. As shown in Figure 1, it is situated directly over the "Standard Pile" or SP(7), a small pile of enriched uranium and graphite that serves as a source of neutrons for the exponential experiment and that is similar to the Thermal Test Reactor(8).

Slowing-Down Media

Six different mixtures of light and heavy water were used in these experiments. The concentrations of heavy water in each mixture are given in Table VIII. Each value is an average from eight samples that were analyzed by a mass spectrograph.

To study a possible anisotropy of the age, a lattice of aluminum rods was placed in the moderator containing 99.3 per cent D_2O. The experimental lattice consisted of 2S aluminum rods one inch in diameter and five feet long that were hung, in clusters of four, from grid beams at the top of the exponential tank. The hexagonal lattice of the clusters within the tank is shown in Figure 2. Quatrefoil housing tubes made of 63S aluminum were used to accommodate the four rods of each cluster. The cross-sectional areas of a quatrefoil and of a set of four rods were 0.732 in^2 and 3.14 in^2 respectively. The total area of each lattice cell was 42.4 in^2. Thus, the volume ratio of aluminum to water was one to ten.

Fission Source and Detectors

The fission source for the experiments was a stack of ten foils of highly enriched uranium that was irradiated by the thermal neutrons from the SP to produce fission neutrons. The foils were one inch in diameter and 0.005 inch thick. A thin coating of lacquer was applied to each foil to prevent corrosion. The source is shown in Figure 3. Also shown in Figure 3 are the indium foils and cadmium covers that were used to measure the slowing-down distribution. The indium foils were 3/4 inch in diameter, 0.0055 inch thick, and weighed 100 mg/cm^2. They were selected from a calibrated set such that, for a given exposure, the activation was the same within ± 1/2 per cent. The cadmium covers were 0.030 inch thick.

Positioning of Source and Detectors

Figure 4 shows the aluminum framework and the foil holders that were used in the mixtures of D_2O and H_2O. The apparatus consisted of an aluminum frame five feet by five feet, and two thin-walled aluminum tubes that formed a crossbar. These tubes had a wall thickness of 0.018 inch and an outside diameter of 1/2 inch. Holes were drilled in the tubes to allow entry of the moderator. The fission source and the foils were attached to the crossbar by means of small aluminum clamps as shown in Figure 4. The frame was placed in the tank so that the fission source was 35 cm above the bottom of the tank.

Figure 5 shows the apparatus that was used to position the fission source and the foils for the measurements in the Al-D_2O lattice. The fission source was placed inside a one-inch aluminum tube with the plane of the source horizontal. The aluminum tube was placed in one of the channels of the central quatrefoil so that the fission source was approximately 25 cm from the bottom of the tank.

To make a vertical traverse in the Al-D_2O lattice, most of the cadmium-covered indium foils were placed inside the aluminum tube at various distances from the source and were separated from one another by aluminum spacers. One such aluminum spacer is shown in Figure 3. Six foils were positioned outside the aluminum tube in the moderator, approximately an inch away from the tube.

Radial traverses in the Al-D_2O lattice were taken along the diameter of the tank in the horizontal plane of the source. Figure 2 shows the path of the radial traverses through the lattice. The indium foils were placed on the aluminum arm, shown in Figure 5, with the plane of the foils perpendicular to the line joining the source and the foils.

Experimental Procedure

The slowing-down density of the neutrons at 1.4 ev was proportional to the activity of the cadmium-covered indium foils measured with the U-235 fission source in place, minus the activity observed in a similar run with the fission source removed. The traverse without the fission source measured the activity induced by neutrons from the Standard Pile, while the traverse with the fission source measured the activity induced by neutrons originating in the source as well as in the SP. Therefore, the net activity was induced by neutrons born in the fission source that had been slowed down to 1.4 ev.

The activity of the indium foils was determined with two intercalibrated GM tubes. The tubes were Tracerlab TCG-1 counters with a mica end-window. A total of at least 20,000 counts were obtained for each foil. The activity of a foil was taken to be the average of the activities of both sides.

The cadmium-covered foils were spaced at least 8 cm apart to avoid shadowing of one foil by another. They were placed at a minimum distance of 25 cm above the graphite pedestal at the bottom of the tank to eliminate the effect of neutrons moderated in the graphite.

Since the foils that were placed along the radius of the tank were closer to the SP than the foils that were placed vertically, the background activity caused by neutrons from the SP, relative to the total activity, was higher for the radial traverses than for the vertical traverses.

To avoid poor counting statistics, the foils were placed in the Al-D_2O mixture at distances less than 45 cm from the source in the radial direction, and less than 70 cm in the vertical direction. For the same reason, in the D_2O-H_2O mixtures, the foils were placed at distances less than 50 cm from the source.

Evaluation of the Data

The integrals in Equation 3 were evaluated from the slowing-down density. The age τ, as defined in Equation 3, is the age measured by means of point detectors and a point fission source. In fitting the experimental results to these formulae, geometric corrections were made to the foil activities to account for the finite size of source and detectors. The formulae for the geometric corrections have been calculated previously(9) and are listed in the Appendix. The geometric corrections to the foil activities never exceeded 0.5 per cent and were less than 0.3 per cent for distances greater than 20 cm from the source.

Corrections were also made for the activation of the indium by neutrons of energies greater than 1.4 ev. The correction for the high energy activation was determined by a technique also described in the Appendix. The corrections for high energy activation were always less than 15 per cent, and were less than 0.5 per cent for distances greater than 25 cm from the source.

To evaluate τ from the corrected data, the product Ar^2 was plotted against r, where A is the corrected foil activity and r is the distance from the source to the detector. A smooth curve was drawn through the experimental points. This curve was used to construct the curve of Ar^4 versus r.

In order to evaluate the integrals of Ar^2 and Ar^4 from zero to infinity, the experimental curves of Ar^2 were extrapolated to infinity by assuming(9) that the slowing-down density was proportional to $\frac{e^{-r/\lambda}}{r^2}$ at distances greater than 30 cm from the source. The parameter λ was determined from the slope of the curve of $\ln Ar^2$ versus r, at distances greater than 30 cm from the source. The portion of the area under the curve of Ar^4 versus r beyond the last experimental point varied from eight to fifteen per cent of the total area, whereas the area under the extrapolated portion of the curve of Ar^2 versus r was less than one per cent of the total area.

After the curves were extrapolated, the integrals in Equation 2 were evaluated by measuring the area under the curves with a planimeter.

Figures 6, 7, and 8 show the development of a typical set of curves, in the case of 94.0 per cent D_2O. Similar sets of curves were drawn for each of the other measurements. Figure 6 is a plot of $\ln Ar^2$ versus r from which the parameter λ was determined, and Figures 7 and 8 are plots of Ar^2 and Ar^4 versus r. The ratio of the areas under the curves of Ar^4 and Ar^2 from $r = 0$ to $r = \infty$ was 518 cm². This result is the experimental value of $\overline{r^2}$. The neutron age is, by definition, one-sixth of $\overline{r^2}$ and therefore equals 86 cm², for 94.0 per cent D_2O.

RESULTS

Tables I through VII give the results of the foil measurements for the different slowing-down media. These results are also shown graphically in Figures 9 and 10, which are plots of $\ln Ar^2$ versus r. The curves in Figure 9 were normalized so that the foil activity at $r = 0$ was approximately the same for each mixture.

The values of τ calculated from these data are given in Table VIII. The errors in τ listed in the table were estimated from the uncertainty with which the curves of Ar^4 and Ar^2 versus r were extrapolated beyond the last experimental point. The parameter λ, determined from curves of $\ln Ar^2$ versus r, is listed in Column 4 of Table VIII. The fraction of the area under the Ar^4 curves that were evaluated by extrapolation is listed in Column 5.

A previous measurement(2) of the neutron age in D_2O gave a result of 106 cm² for 99.8 per cent D_2O. This result agrees with the present result within the experimental error. The variation in age as a function of light water contamination is shown in Figure 11. Over this small range of moderator purities, the decrease in τ as a function of purity is very nearly linear. The slope of this line shows that one per cent of H_2O decreases τ by 4 cm^2.

Experiments in the lattice of aluminum and heavy water showed that the age measured parallel to the aluminum rods was 140 cm^2 and that the age measured perpendicular to the rods was 108 cm^2. Figure 10 compares the curves of $\ln Ar^2$ versus r that were obtained in the vertical and radial directions. The difference in the foil activities caused by the anisotropy in the age is noticeable at distances greater than 20 cm from the source and becomes more apparent as the distance increases. This anisotropy results from the streaming of fast neutrons along the aluminum rods. The value of 108 cm^2 for the age measured in the direction perpendicular to the aluminum rods equals the value of the age for pure moderator within the precision of the measurements.

Since the radius of the tank was 76 cm while its height was 185 cm, it was necessary to make sure that the differences in the slowing-down distributions in the radial and vertical directions were not influenced by leakage of neutrons in the radial direction. An auxiliary experiment in 99.0 per cent D_2O showed that, with all aluminum removed, the slowing-down distributions were the same in both the radial and vertical directions. Therefore, the radial boundary, 76 cm from the source, did not affect the radial measurements in the mixture of D_2O and aluminum.

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APPENDIX

CORRECTION OF FOIL ACTIVITIES FOR FINITE SIZE OF SOURCE AND DETECTORS

To account for the finite size of the foils and the source, geometric corrections must be applied to the activity of the indium foils. Such corrections have been published previously(9) for various shapes of foils and sources. For a plane source of radius a_1 and a plane detector of radius a_2, the correction for the activity of the foil is

$$A(r_o) - A_m(r_o) = -\left(\frac{a_1^2 + a_2^2}{4r_o}\right)\left(\frac{dA}{dr}\right)_{r_o}$$

where

$A(r_o)$ = activity induced by a point source in a point detector at r_o

$A_m(r_o)$ = measured activity in the finite foil at r_o

r_o = distance between the center of the fission source and the foil

Since this correction was small, the derivative $\left(\frac{dA}{dr}\right)_{r_o}$, was taken to be the slope of the experimental curve of A_m versus r.

The correction factor for a source with a radius of 1.0 inch and foils with radii of 0.750 inch is shown in Figure 12. In the range of the measurements that were used to calculate τ, the correction was less than 0.5 per cent.

CORRECTIONS FOR ACTIVITY INDUCED IN THE FOILS BY HIGH ENERGY NEUTRONS

The correction for the activation of the cadmium-covered indium foils by high energy neutrons was determined by experiment, by means of a technique that has been described previously(10). The technique consists of comparing the activity of indium foils covered by cadmium with the activity of foils covered by both cadmium and indium. Since the resonances of In-115 above 1.4 ev are weak compared to the resonance at 1.4 ev, the self-shielding of the high energy resonances is small compared to that of the 1.4-ev resonance. Consequently, covering the foils with indium will have little effect on the activation by neutrons of higher energy, but will decrease the activation at 1.4 ev by a large factor.

At distances less than $\sqrt{\tau}$ from the fission source, the total activation of cadmium-covered indium foils is

$$A_{total} = A(1.4 \text{ ev}) + A(\text{high energy}) \qquad (1)$$

At distances of two or three times $\sqrt{\tau}$ from the source, the activation by high energy neutrons is negligible so that the activation is

$$A_{total} = A(1.4 \text{ ev}) \qquad (2)$$

With the foils covered with both indium and cadmium, Equations 1 and 2 can be written

$$A'_{total} = g_o A(1.4 \text{ ev}) + A(\text{high energy}) \qquad (1a)$$

$$A'_{total} = g_o A(1.4 \text{ ev}) \qquad (2a)$$

where g_o is the self-shielding factor for the foil at the 1.4-ev indium resonance. It is independent of r. Equations 2 and 2a may be used to obtain g_o and then Equations 1 and 1a may be solved simultaneously to obtain A(high energy).

The table below lists the results of experiments with cadmium plus indium covers and with cadmium covers alone.

r, cm	A_{Cd}	$A_{Cd + In}$	$\frac{A_{Cd + In}}{A_{Cd}}$
4.4	513,300	228,470	0.445
14.0	305,020	128,180	0.420
31.5	57,200	22,390	0.392
42.0	13,650	5,194	0.381

Figure 13 is a plot of the ratio of the indium foil activities when covered with indium plus cadmium to the activities when covered with cadmium. At distances greater than 25 cm from the fission source this ratio equals g_o. From Figure 13, g_o equals 0.38. The resulting correction to the foil activity is plotted also in Figure 13 and is less than 0.5 per cent for distances greater than 25 cm from the source.

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TABLE I

MEASUREMENTS IN AL-D_2O LATTICE

D_2O Purity = 99.3%

Placement of Foil

Angle from Vertical Line through Source, degrees	Foil Placed Inside of	Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
0	Al	2.4	267,461	1.50
0	Al	4.4	251,440	4.96
0	Al	9.4	219,807	19.58
0	Al	11.9	182,919	26.07
0	Al	14.0	160,250	31.31
10	D_2O	15.8	144,217	36.11
0	Al	21.4	84,909	39.02
0	Al	21.5	84,759	39.23
0	Al	24.4	62,405	37.27
0	Al	26.0	56,222	37.93
8	D_2O	31.1	27,531	26.70
0	Al	31.5	28,313	28.12
0	Al	34.0	20,811	24.02
0	Al	34.1	20,341	23.59
0	Al	37.0	13,042	17.84
0	Al	37.0	13,442	18.39
0	Al	42.1	6,809	12.04
5	D_2O	43.1	5,474	10.15
5	D_2O	47.0	2,634	5.82
3	D_2O	50.7	1,644	4.22
0	Al	59.5	600	2.13
2	D_2O	63.8	243	0.99
0	Al	64.5	294	1.22
0	Al	72.1	804	0.42
0	Al	76.1	80	0.46
45	D_2O	19.9	96,694	38.15
		31.1	24,940	24.17
90	D_2O	6.1	273,632	10.15
90	D_2O	7.9	253,109	15.86
270	D_2O	14.0	169,153	34.05
90	D_2O	17.7	117,324	36.67
90	D_2O	23.9	62,883	35.80
270	D_2O	31.8	21,766	22.03
90	D_2O	35.5	14,286	18.04
270	D_2O	35.5	15,567	19.66
270	D_2O	42.2	3,530	6.29
90	D_2O	43.0	2,684	4.97

TABLE II

MEASUREMENTS IN 99.8 PER CENT D_2O

Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.7	270,540	12.0
10.7	216,830	24.9
13.6	154,140	28.3
18.6	101,420	35.2
20.6	88,660	37.8
25.6	47,500	31.0
29.1	27,890	23.5
35.0	11,370	13.9
39.6	5,417	8.5
45.1	3,224	4.5
50.0	868	2.2

TABLE III

MEASUREMENTS IN 99.0 PER CENT D_2O

Distance From Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.2	382,900	14.66
10.2	301,200	31.07
13.4	217,600	39.23
19.6	125,400	48.18
20.5	110,100	46.26
24.9	59,600	37.01
29.7	31,110	27.38
30.5	28,300	26.29
34.1	16,010	18.67
38.8	6,910	10.38
40.0	4,072	7.54
49.9	928	2.31
50.0	1,056	2.64

TABLE IV

MEASUREMENTS IN 98.4 PER CENT D_2O

Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.5	329,030	14.0
10.8	238,190	27.7
14.5	165,630	34.9
18.7	106,810	37.5
20.6	82,320	35.1
24.9	45,940	28.5
29.1	24,960	21.2
35.0	11,050	13.5
39.7	5,270	8.3
45.1	2,447	5.0
50.1	1,372	3.4

TABLE V

MEASUREMENTS IN 95.3 PER CENT D_2O

Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.7	1,029,500	46.8
10.8	739,350	86.1
14.4	515,580	107.5
18.8	282,940	104.8
20.6	230,640	97.4
24.9	118,950	73.9
29.1	65,740	55.5
35.0	21,900	26.8
39.4	11,240	17.4
45.2	4,010	8.2

TABLE VI

MEASUREMENTS IN 94.0 PER CENT D_2O

Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.7	1,375,700	62.5
10.9	998,300	118.0
14.3	666,000	136.0
18.8	379,590	134.3
20.7	289,110	124.0
25.0	153,600	96.0
29.0	78,200	65.8
35.1	29,230	36.0
39.7	13,540	21.3
45.2	4,859	9.9
50.0	1,844	4.6

TABLE VII

MEASUREMENTS IN 91.8 PER CENT D_2O

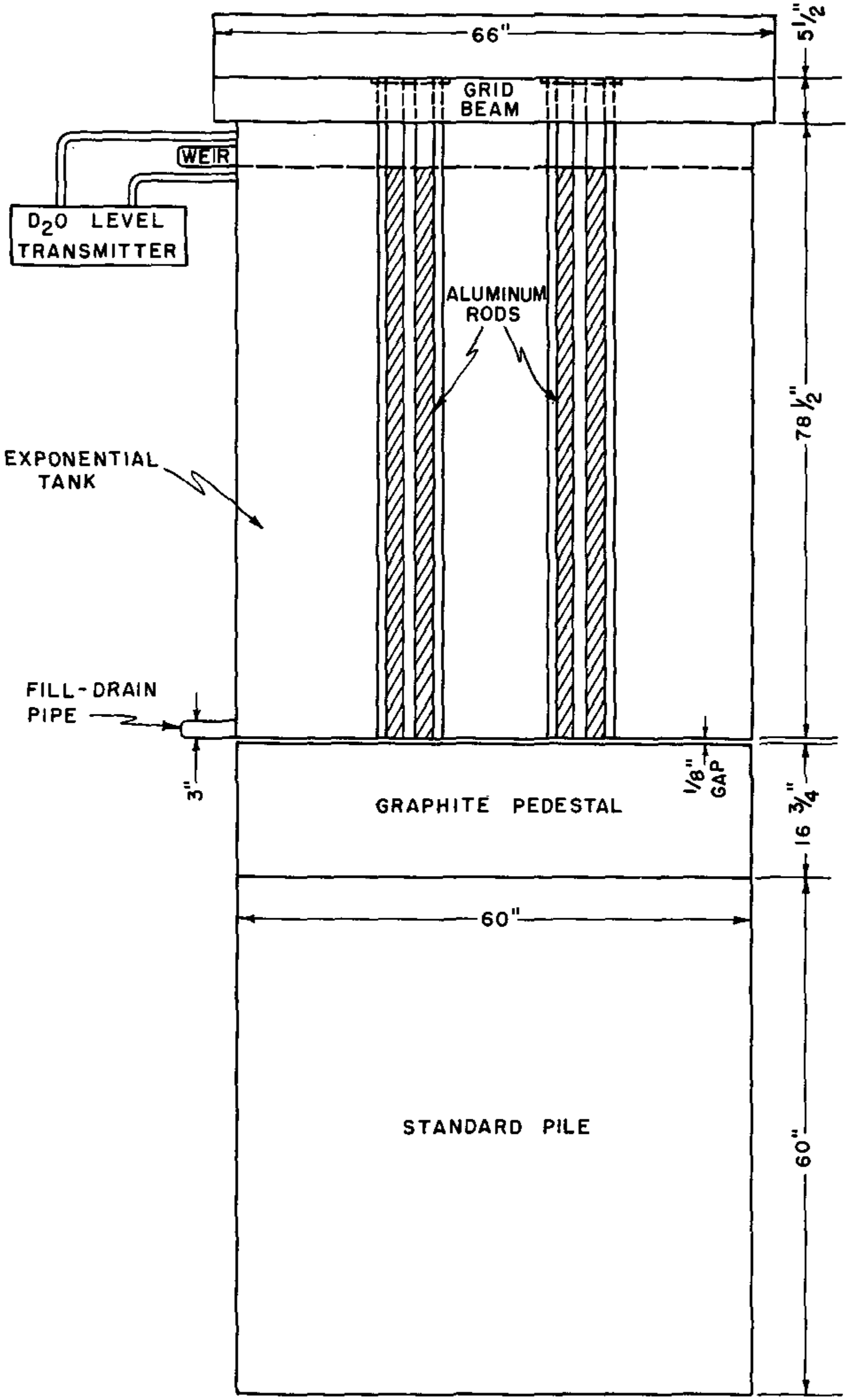
Distance from Source r, cm	$A_{corrected}$, Relative cts/min	$Ar^2 \times 10^{-6}$
6.7	1,055,020	47.9
10.8	732,240	85.3
14.4	473,570	98.7
18.8	261,760	92.6
20.6	201,640	85.8
25.0	98,800	61.8
29.1	49,110	41.7
35.0	17,700	21.7
39.6	7,616	12.0
45.2	2,030	4.1

TABLE VIII

SUMMARY OF AGE MEASUREMENTS

	Slowing-Down Media, Per Cent D_2O	τ, cm^2	Estimated Error cm^2	λ, cm	Extrapolated Portion of Area, Per Cent
A.	In Water				
	99.8	109	± 3	8.7	13
	99.0	107	± 5	8.7	14
	98.4	106	± 3	9.3	15
	95.3	93	± 3	9.0	13
	94.0	86	± 3	7.5	8
	91.8	78	± 3	6.7	9
B.	In Al-D_2O with 99.3 Per Cent D_2O				
	Parallel to aluminum rods	140	± 3	10.2	4
	Perpendicular to aluminum rods	108	± 5	7.8	20

FIGURE 1



SIMPLIFIED ELEVATION OF THE EXPONENTIAL AND STANDARD PILE

FIGURE 2

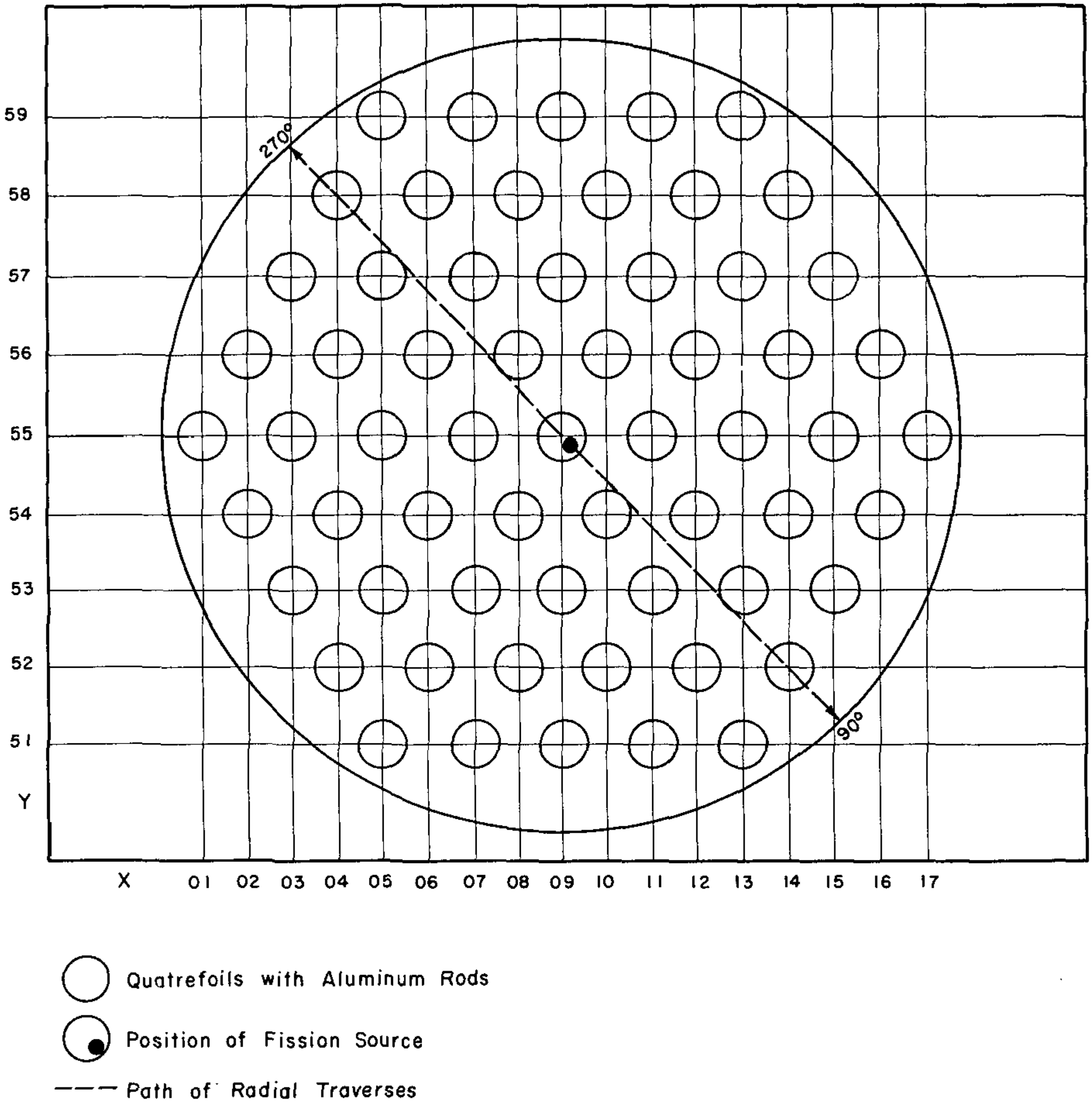
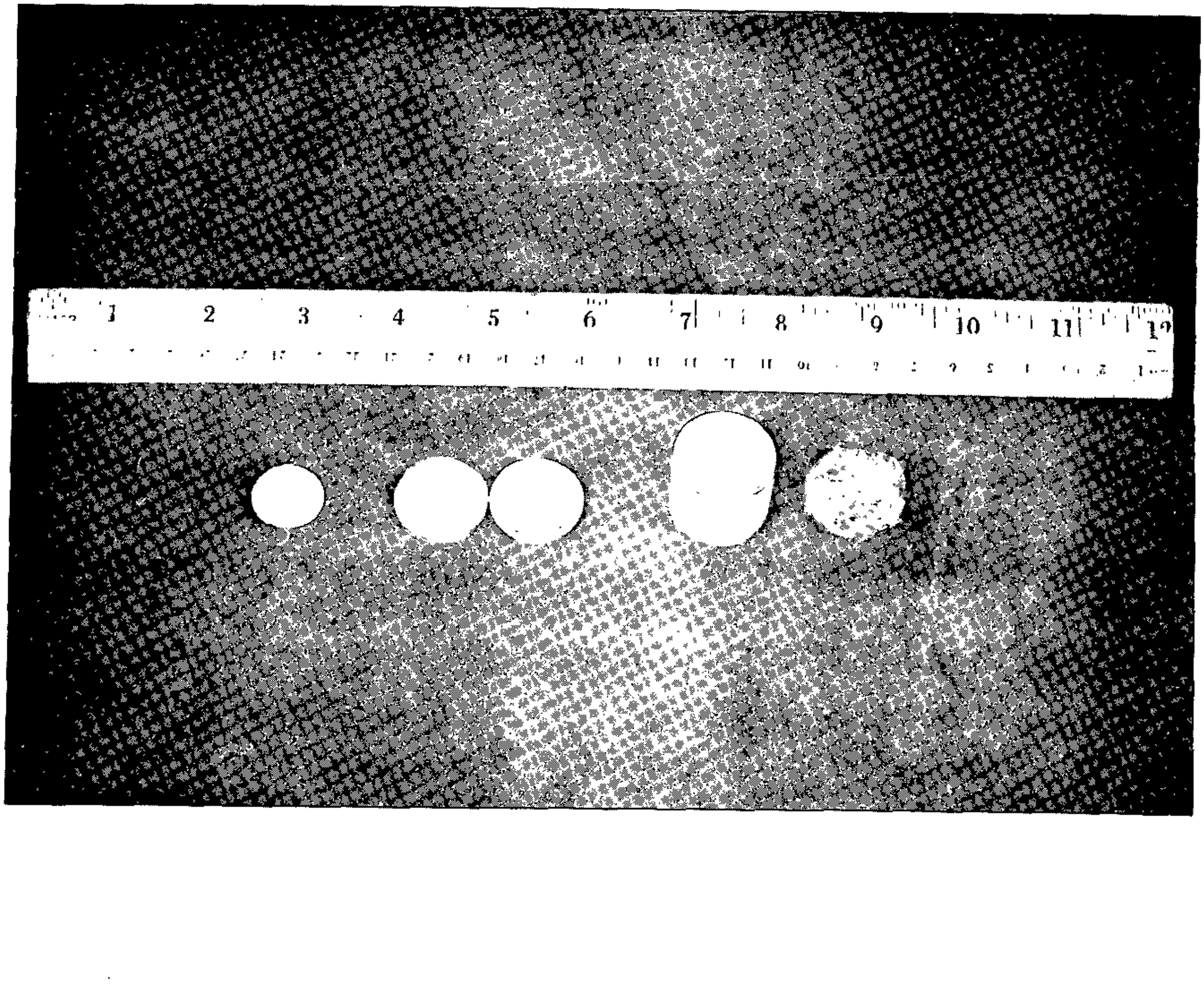


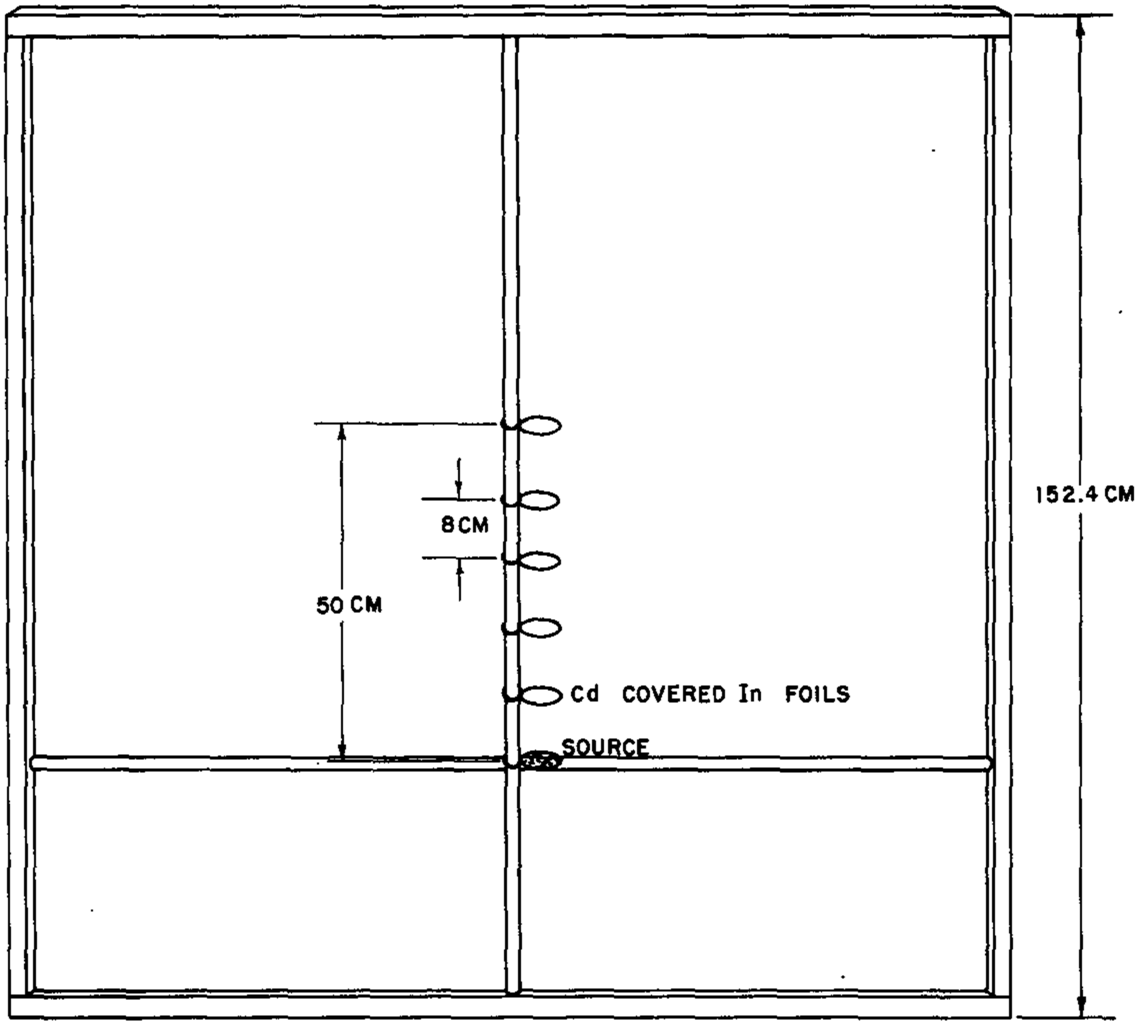
DIAGRAM OF Al - D_2O LATTICE

FIGURE 3



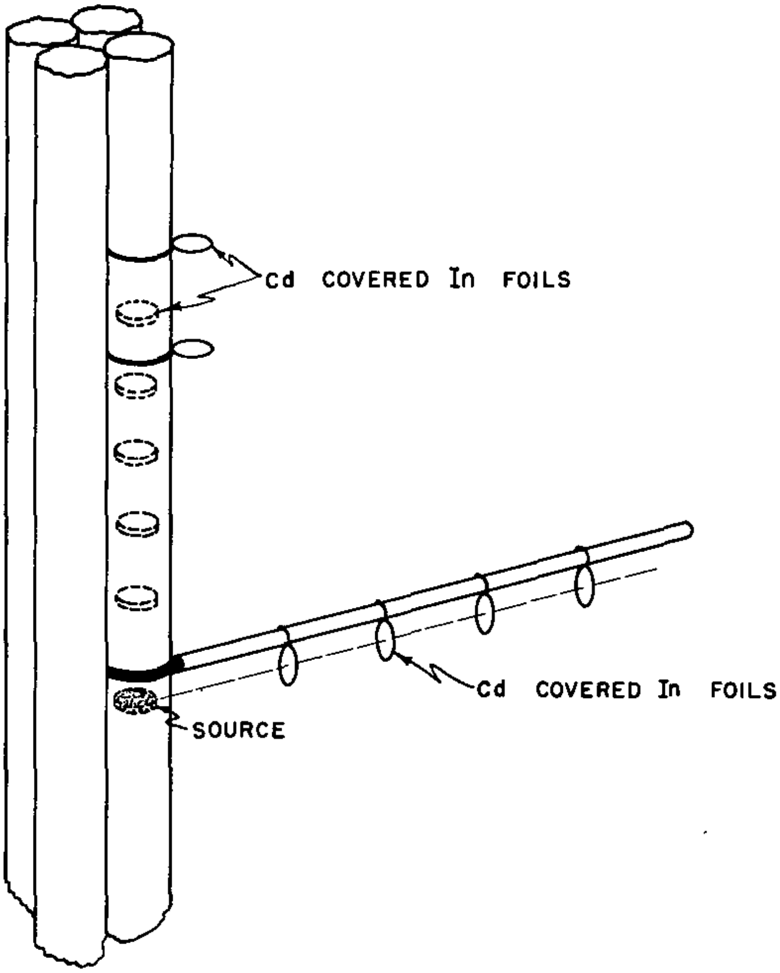
INDIUM FOIL, CADMIUM BOX, ALUMINUM FOIL HOLDER, AND FISSION SOURCE

FIGURE 4

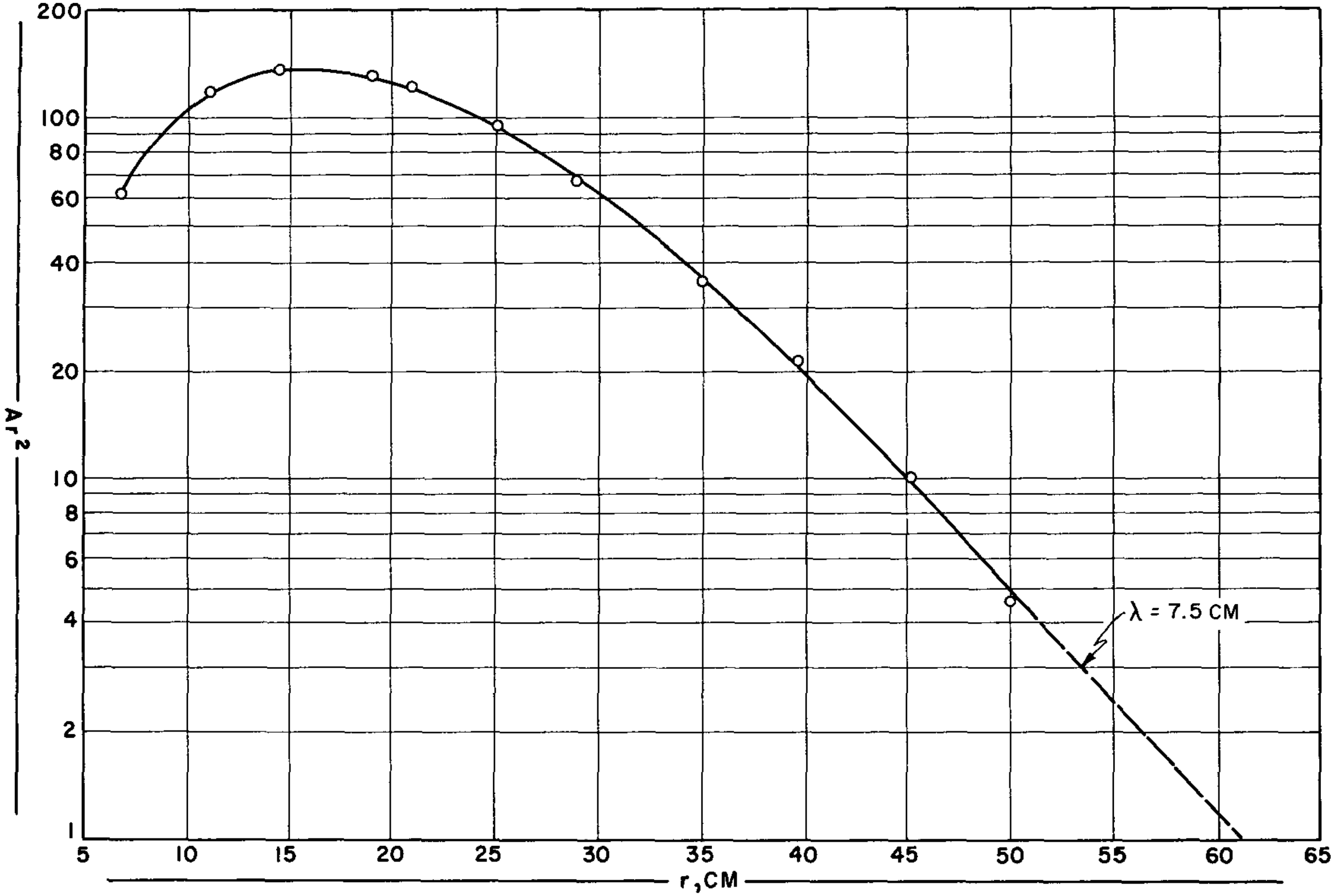


EXPERIMENTAL ASSEMBLY FOR MEASUREMENTS IN THE H_2O-D_2O MIXTURES

FIGURE 5

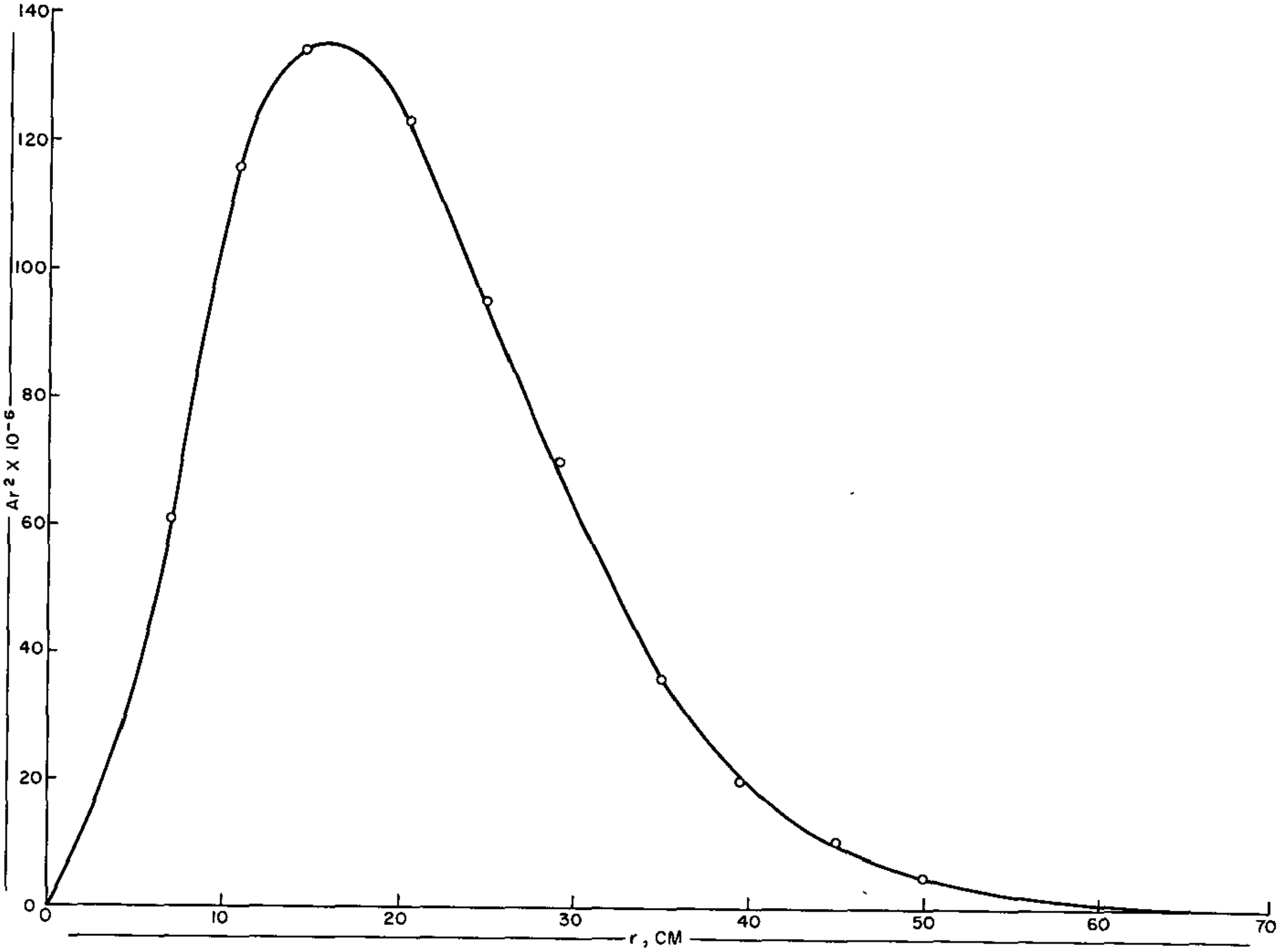


EXPERIMENTAL ASSEMBLY FOR RADIAL AND VERTICAL TRAVERSES IN THE Al - D_2O LATTICE



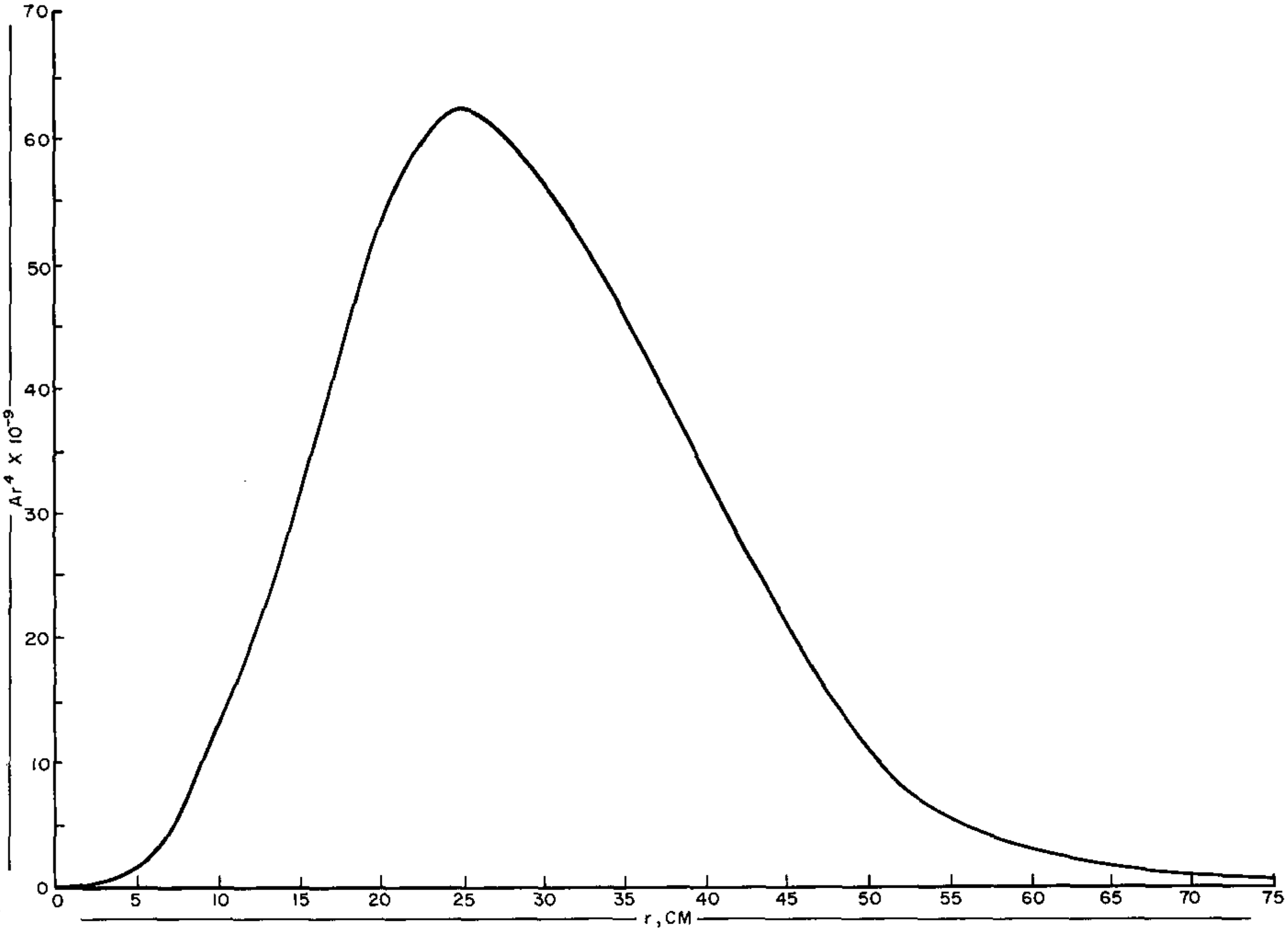
ln Ar^2 VS. r FOR 94.0 PER CENT D_2O

FIGURE 7



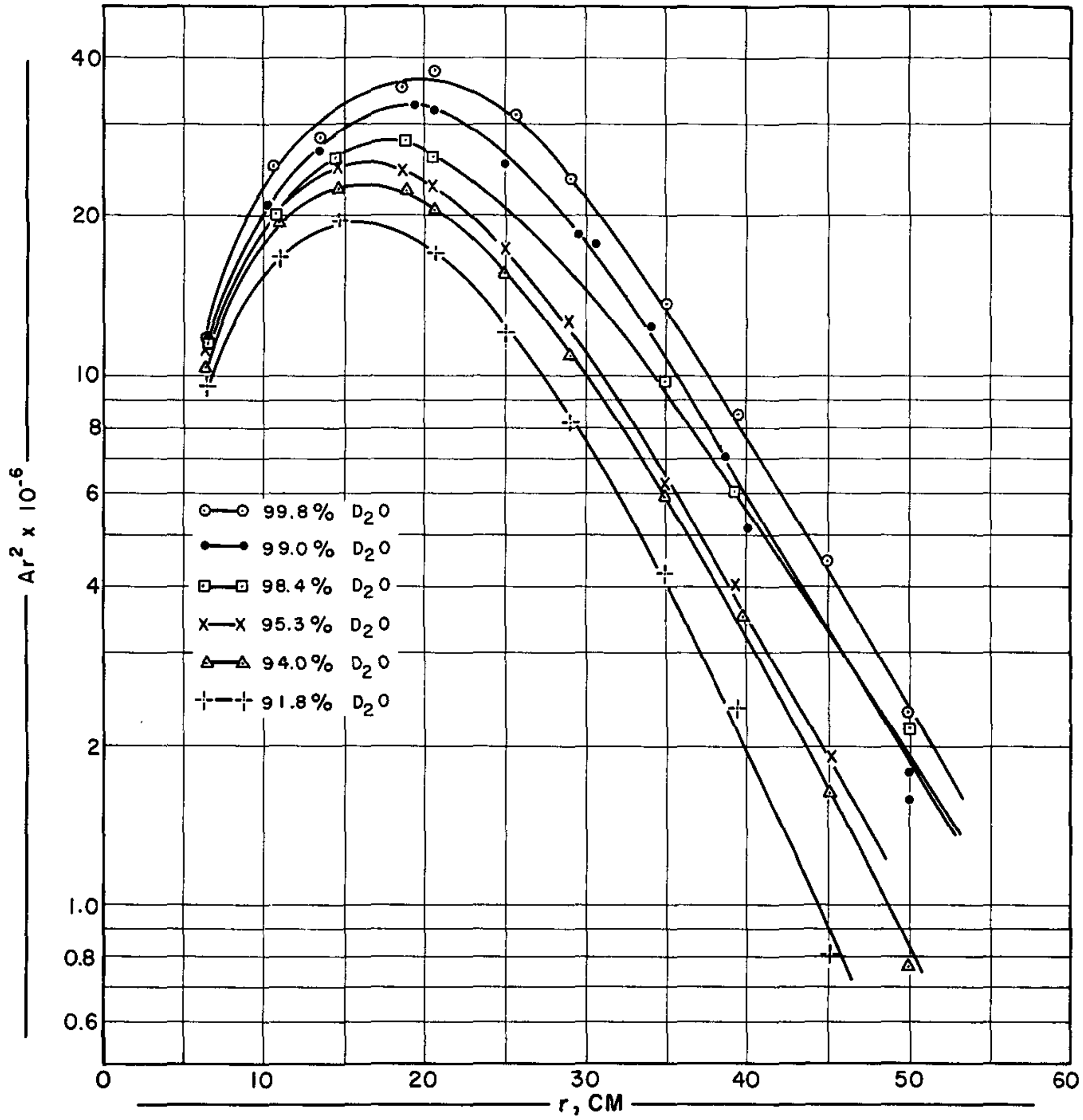
Ar^2 VS. r FOR 94.0 PER CENT D_2O

FIGURE 8



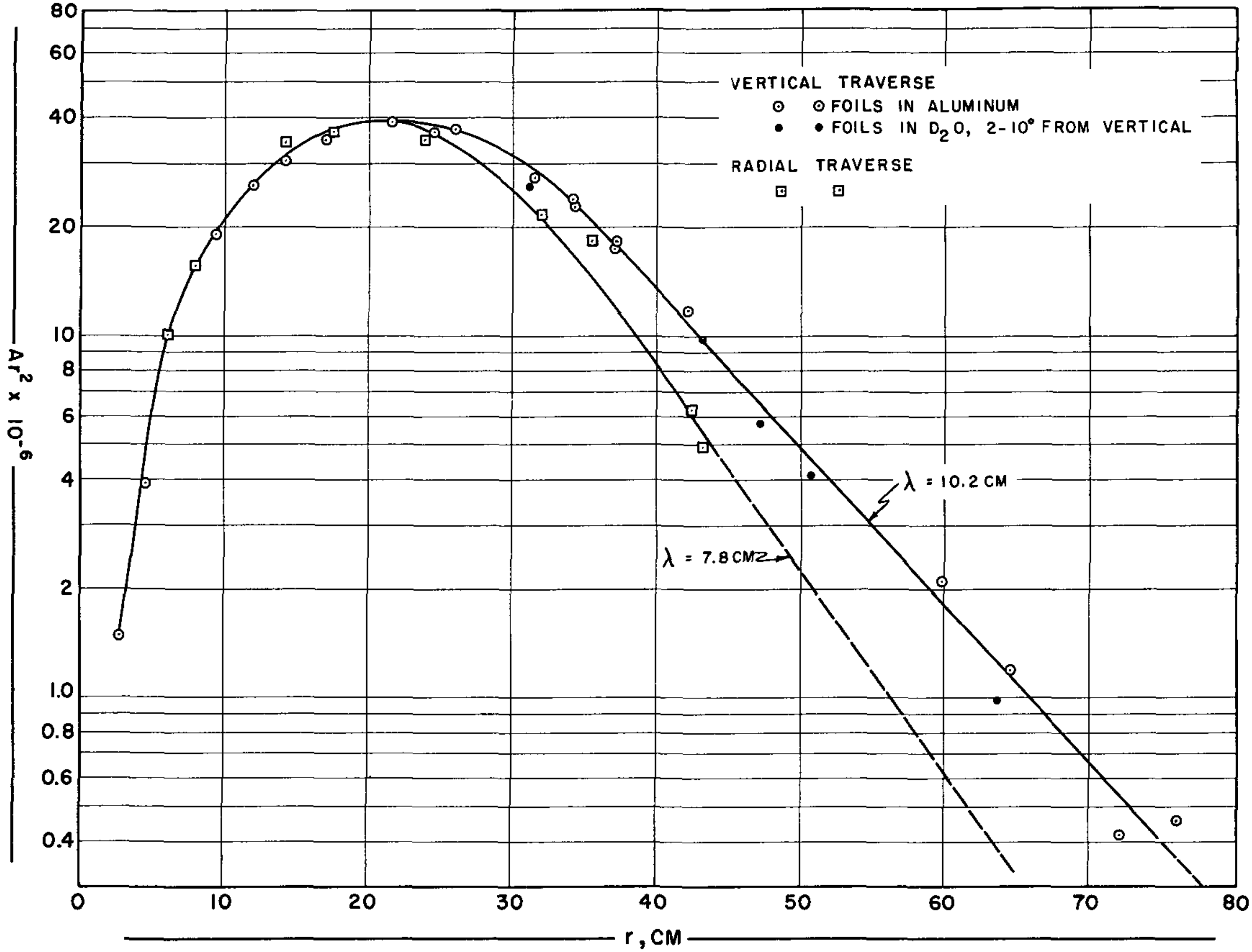
Ar^4 VS. r FOR 94.0 PER CENT D_2O

FIGURE 9



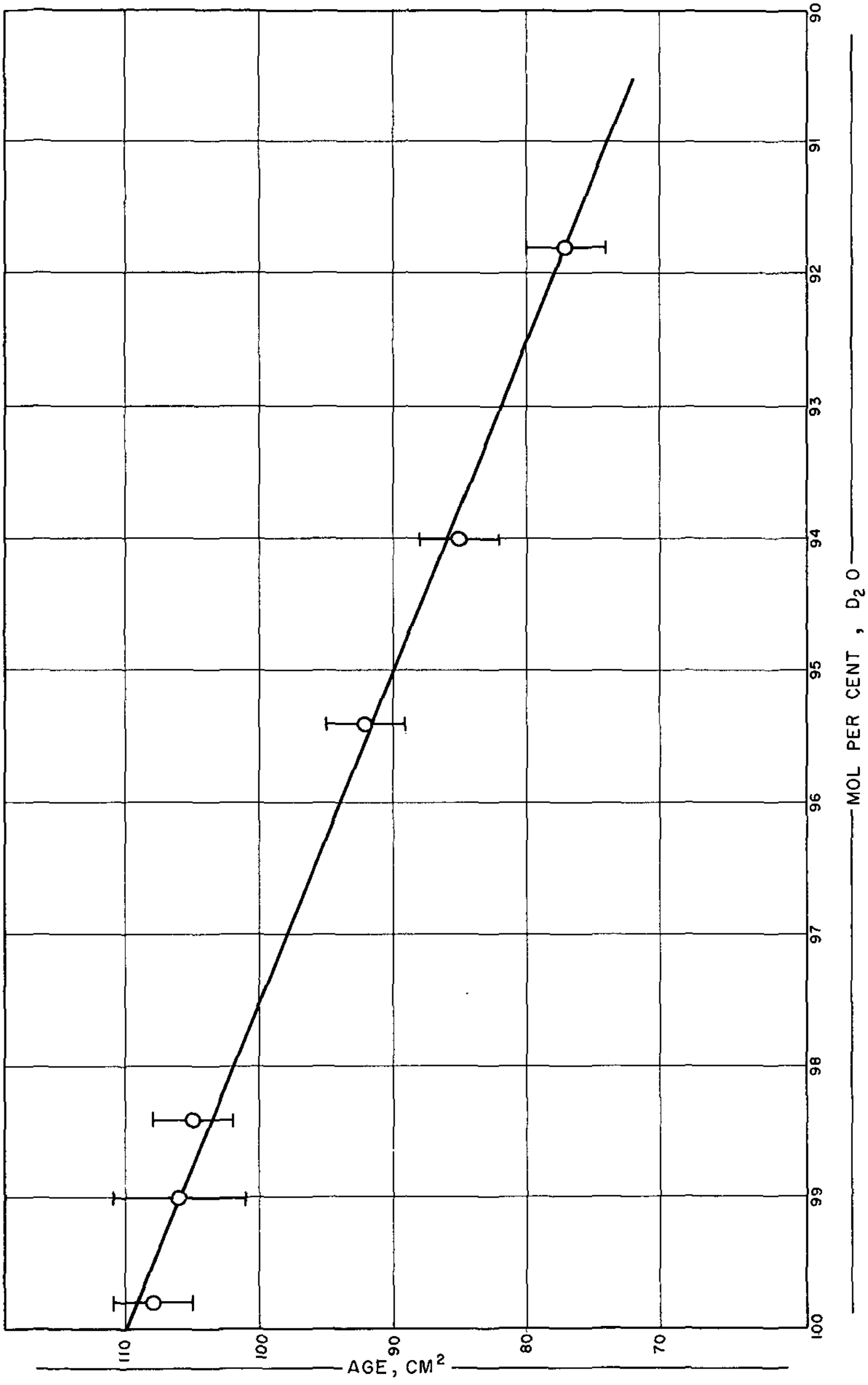
ln Ar^2 VS. r — $H_2O - D_2O$ MIXTURES

FIGURE 10



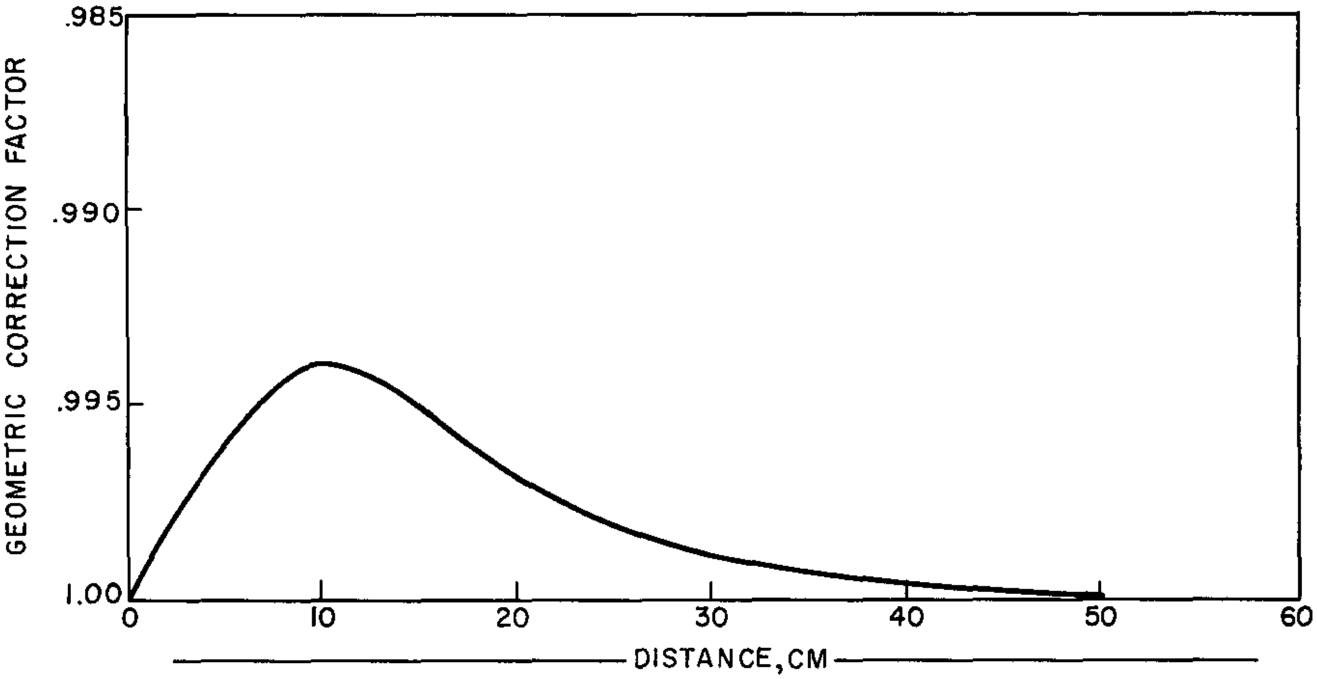
ln Ar^2 VS. r - ALUMINUM - D_2O LATTICE

FIGURE 11



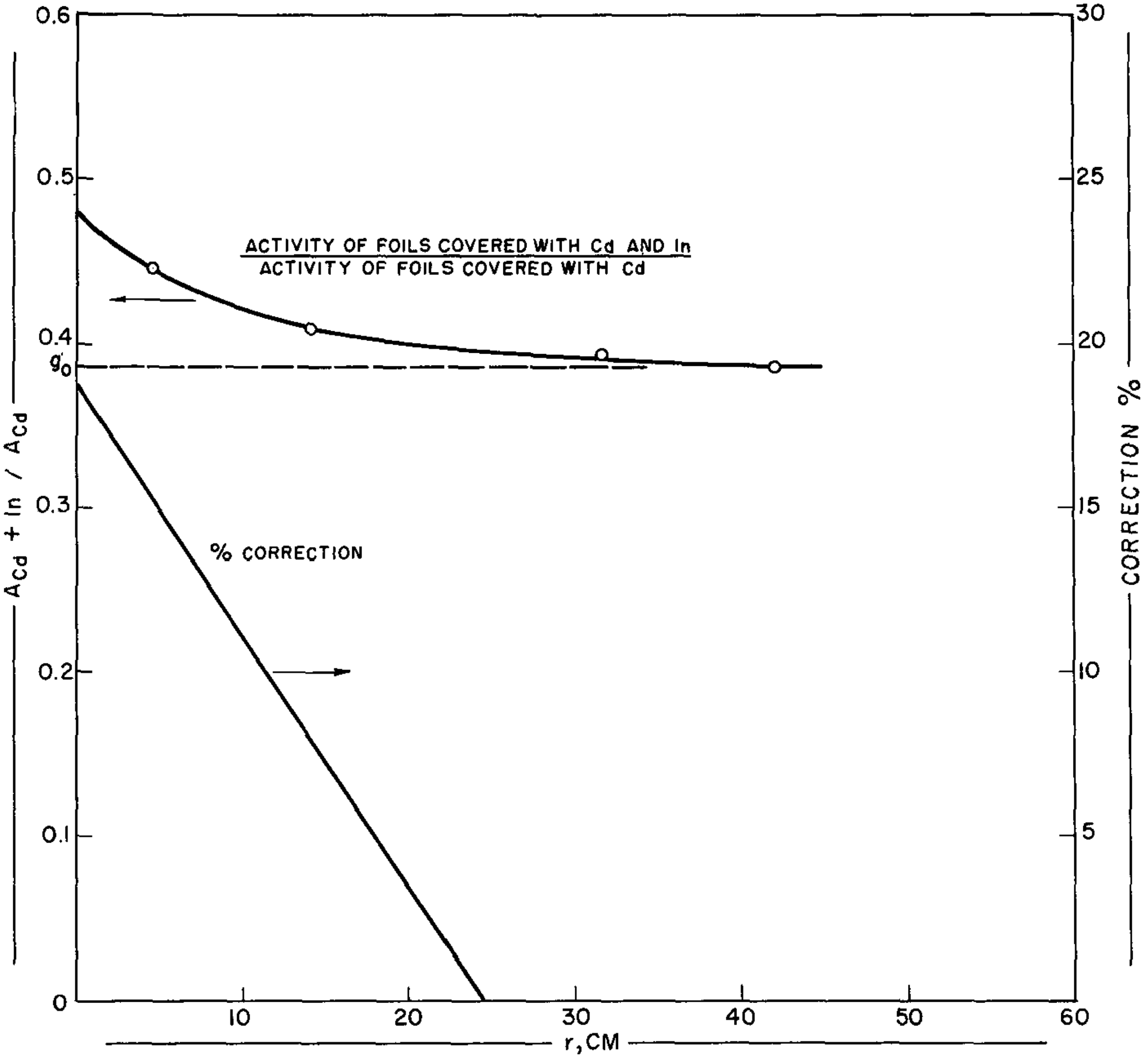
AGE TO INDIUM RESONANCE VS. MODERATOR PURITY

FIGURE 12



GEOMETRIC CORRECTION FACTOR VS. DISTANCE

FIGURE 13



CORRECTION OF FOIL ACTIVITY FOR HIGH ENERGY NEUTRONS VS. r