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THE RESONANCE INTEGRAL OF ZIRCONIUM (~1% HAFNIUM) AND THE SLOWING DOWN LENGTH IN A MIXTURE (4:1 BY VOLUME) OF LIGHT WATER AND ZIRCONIUM METAL

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THE RESONANCE INTEGRAL OF ZIRCONIUM (~ 1% HAFNIUM) and THE SLOWING DOWN LENGTH IN A MIXTURE (4:1 BY VOLUME) OF LIGHT WATER AND ZIRCONIUM METAL

by

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THE RESONANCE INTEGRAL OF ZIRCONIUM (~1% HAFNIUM), THE SLOWING DOWN LENGTH IN A MIXTURE (4:1 BY VOLUME) OF LIGHT WATER AND ZIRCONIUM METAL

Abstract

Measurements have been made of the resonance integral for zirconium (~1% hafnium) and the slowing down distribution in zirconium (~1% hafnium) light water mixture containing 20.4% zirconium by volume. The resonance integral was measured in the energy range from Ra-α-Be neutrons to indium resonance. The slowing down distribution was measured to indium resonance using a fission source of neutrons. The value of the resonance integral is 23.9 cm^2, the major part of which is probably due to the hafnium impurity. The mean square slowing down length $\overline{r^2}$ = 213.97 cm^2 and τ = 35.7 cm^2.

Introduction

Inasmuch as the same apparatus was used for both the resonance integral measurements and the slowing down distribution measurement, the resonance integral experiment will be discussed somewhat in detail and the changes noted later in the description of the slowing down measurement.

The resonance integral was determined by measuring the slowing down distribution of indium resonance neutrons first in a slowing down medium (light water) and again in a mixture of the slowing down medium and zirconium metal. If a neutron source of strength Q is placed in a slowing down medium and the distribution of indium resonance neutrons measured, then Q = number of neutrons slowing down across indium resonance + the number of neutrons captured by the slowing down substances. If the same source Q is placed in a mixture of the slowing down medium and zirconium metal and the distribution again measured, then Q = number of neutrons slowing down across indium resonance + number of neutrons captured by the slowing down substance + the number of neutrons captured by the zirconium present. The number of neutrons slowing down can be determined from the measured distribution. The number captured by the slowing down material can be calculated from its resonance integral

providing its capture cross section variation with energy is known. Since the source strength is the same in both cases the resonance integral for zirconium can be computed.

$$\int_{1.4}^{\infty} \sigma_{Zr} \frac{dE}{E} = \frac{N_2 \sigma \xi}{N_{Zr}} \left[1 - 0.0038 - \frac{N_2}{N_1} (1 - 0.0038) \frac{\int_{Zr} A_s \, d\tau}{\int_{H_2O} A_s \, d\tau} \right]$$

where

$\int_{1.4}^{\infty} \sigma_{Zr} \frac{dE}{E}$ = resonance integral from 1.4 ev to ∞,

N_2 = number of molecules of water in the system during the $Zr \cdot H_2O$ measurement,

$\sigma \xi$ = slowing down power of 1 mol of H_2O,

N_{Zr} = number of atoms of zirconium in the system during the $Zr \cdot H_2O$ measurement,

N_1 = number of molecules of water in the system during the water measurement,

A_s = saturated activity of the indium foils,

$d\tau$ = volume element,

0.0038 = factor to correct for the neutron capture of the slowing down material.

Apparatus

The apparatus consists of an aluminum base plate (40 in. × 40 in. × ½ in.), a 6 in. × 6 in. elevator shaft constructed of 1 in. × 1 in. × 1/8 in. aluminum angle, an elevator made of four slotted ½ in. zirconium rods 36 in. long with

½ in. aluminum end plates, and 36 in. × 6 in. × 0.080 in. zirconium sheet. The elevator shaft which was 48 in. high was bolted to and concentric with the center of the base plate, Fig. 1. The elevator, Fig. 2, could be raised or lowered in the shaft by means of a system of pulleys and counter weights. An aluminum source holder, not shown in the figure, for a 500 mg Ra-α-Be source was bolted to the center of the base plate, the center line of the source holder being along the center line of the elevator shaft and elevator. The holder was designed so the geometric center of the source was held about 4 in. from the base plate in order to minimize undesirable boundary conditions. A hole in the bottom plate of the elevator allowed the source to extend about 3½ in. into the elevator when it was in the lowered position. The elevator, as described, was used to measure the neutron distribution in water by inserting aluminum foil holders containing cadmium covered indium foils in the various slots in the zirconium rods. For the zirconium-water measurement, zirconium sheet was drilled, machined, and stacked on the ½ in. zirconium rods. Spacing between successive sheets was maintained by four machined 5/16 in. lucite spacers, drilled to slide freely on the ½ in. zirconium rods. In Fig. 3 a few sheets are shown stacked apart from the rods to indicate the method. In this case the neutron distribution was measured by inserting cadmium covered indium foils in suitable foil holders between adjacent sheets in the elevator. Additional zirconium sheet 36 in. × 6 in. × 0.080 in. was piled layer-wise, as shown in Fig. 4, around the elevator shaft on the 40 in. × 40 in. base plate and spaced with 5/16 in. lucite strips. The sheets were oriented 90° in alternate layers to prevent the lining up of small cracks between adjacent sheets. The zirconium stack measured 30 in. × 30 in. × 22 in. with the elevator shaft in the center. The apparatus was assembled in a tank 6 ft × 6 ft × 6 ft which could be filled with water. Approximately 970 lbs of zirconium were used.

The slots in the zirconium rods of the elevator, the spacers, foil holders, and zirconium sheet in the elevator were precision machined; consequently it was possible to accurately duplicate the positioning of the foils. In the case of the water-zirconium mixture considerable difficulty was experienced in purging the system of air. The purging was done with a fast flowing water jet and the system checked frequently with underwater lights and mirrors for air entrapment. Water jets were also used to eliminate the entrapped air bebetween the sheets of the elevator as it was lowered into the shaft.

Metal Purity

The zirconium metal was supplied by the U. S. Bureau of Mines through contract with the U. S. Naval Research Laboratory. The ore was specially selected low hafnium ore procured from the Foote Mineral Company. Analysis of the metal showed a hafnium content of ~1%, average iron 0.1%, average carbon 0.25% and boron <0.1 ppm. The metal was cold rolled into sheets 36 in. × 6 in. × 0.080 in. Before using it was annealed in a jig for straightening. The thermal neutron cross section as measured by Pomerance was 0.85 barns.

Measurements

The neutron distributions were measured using cadmium covered indium foils. The cadmium covers were 0.040 in. (~880 mg/cm^2) thick. Two sizes of indium foils were used (5.55 cm × 3.97 cm and 1 cm × 1 cm) both of which were ~75 mg/cm^2 thick. The foils were constant in weight to within 1% of the average. The small foils (1 cm × 1 cm) were used in the region close to the source and the large foils (5.55 cm × 3.97 cm) in the region far from the source. In this way geometry corrections were kept small. The two regions of the distribution were then normalized together at points where overlapping data were taken. In general the foils were activated for an hour and counted for an hour. Only two foils were activated simultaneously and they were kept sufficiently far apart to prevent interaction between foils. Two counters were used. The ratio between counters was obtained by bombarding a set of calibration foils of about the same size and weight in the standard pile. The calibration foils were bombarded daily and served to check the consistency of the counters. At least three foils were bombarded for each position measured and an average taken. In all cases only the side of the foil nearest the source was counted.

Results

The average measured saturated activity for each point was determined and corrected for a point source and a point detector according to the method

described by Hill, Roberts, and Fitch.[1] The sections of the curve obtained with different size foils were then normalized together using these corrected saturated activities.

Figure 5 is a plot of the corrected saturated activities as a function of the distance from the source for both the water and the zirconium-water measurements. The portion of the curve between 0 and about 2.5 cm was extrapolated. The integrations were performed numerically from 0 to 30 cm and a ratio of the integrals of 1.01195 obtained. Using this ratio plus the quantity of water and zirconium in the system during each measurement the resonance integral for zirconium is

$$\int_{1.4}^{\infty} \sigma_{Zr} \frac{dE}{E} = 23.9 \text{ cm}^2 .$$

Extending the measurements to distances greater than 30 cm from the source would not change the value of the resonance integral significantly. This can be seen from Fig. 6 where the value of the resonance integral is plotted as a function of the distance from the source over which the integral was determined. From Fig. 5 it can be seen that the contribution to the areas under the curve between 0 and 5 cm is quite large. Consequently the validity of the result depends a great deal on the extrapolation in this region. In addition the center line of the source is not accurately known and as a result the zero of the curve in Fig. 5 could be displaced to the right or left of its true value. Deducting the extrapolated portion of the curve, *i.e.*, considering the integrals as taken only over the measured range 2.5 cm to 30 cm, the value of the resonance integral is 20.25 cm^2. From a consideration of the statistics of the counts this value should be good to better than 5%; however the measured value plus the extrapolated value may not be good to any better than 20%.

Several resonances are known to exist in hafnium above 1.4 ev. It seems reasonable therefore to assume that the contribution from the hafnium constitutes almost all of the measured value of the resonance integral.

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(1) Hill, J. E., Roberts, L. D., and Fitch, T. E., *The Slowing Down Distribution from a Point Source of Fission Neutrons in Light Water*, ORNL 181 (Dec. 10, 1948).

Slowing Down Distribution From a Point Fission Source to Indium Resonance

In general the apparatus and methods used in the slowing down measurements were the same as that used in the measurement of the resonance integral. There were, however, some modifications made in order to use a fission source.

The apparatus was assembled, using the same system of spacing, in the 5 ft × 5 ft × 6 ft 2-S aluminum tank on the top of the thermal column of the Clinton pile. The assembly contained about 1300 lbs zirconium (ratio of volume of metal to volume of water = 0.2) in a 30 in. × 30 in. × 27 in. cube. A fission source 5.08 cm in diameter of U - Al alloy (18% U - 95% U - 235) was attached in the center of the elevator shaft to aluminum guides, which in turn attached to the 40 in. × 40 in. base plate. The bottom of the base plate was covered with 0.060 in. cadmium sheet except for a 2-1/16 in. hole directly under the source. This hole extended through the base plate. The source was mounted in such a way that a cadmium shutter 4 in. × 2¼ in. × 1/16 in. could be slid in the aluminum guides and under the source when the source was not in use. Provisions were also made to mount the source on stand-off aluminum rods about 3 in. long. With the stand-off rods in place there was about 4 in. of water between the source and the bottom of the aluminum tank. Measurements close to the source were made using this geometry. With the stand-off rods removed, *i.e.*, the source attached to the aluminum guides, there was about 3/4 in. of water between the source and the bottom of the tank. Measurements far from the source were made using this geometry. Thus the boundary conditions were kept favorable and the sections of the measured distributions were normalized together with overlapping data.

Measurements

The 5.55 cm × 3.97 cm foils and the 5.08 cm source were used throughout the measurements. The neutron distribution was measured between 0 and 40 cm. The distribution between 3 cm and 10 cm was measured with cadmium covered foils and the source ~10 cm from the bottom of the tank; between 8 cm and 30 cm with cadmium covered foils and the source ~2 cm from the bottom of the tank;

and between 26 and 40 cm using aluminum covered (135 mg/cm^2 of Al) foils with the source ~2 cm from the bottom of the tank. Beyond 30 cm the counting rate was too small for accurate results using cadmium covered foils, and since the distribution with cadmium covered foils is proportional to the distribution in aluminum covered foils over this range the aluminum covered foils were used to improve the statistics. The various sections of the curve were normalized together with overlapping data. In all cases the saturated activity averaged over both sides of the foil was used in the calculations. This was obtained by counting both sides of the foils at predetermined positions and plotting a ratio of the counts on the two sides as a function of the distance from the source. A smooth curve was drawn through these points and the ratio, as read from this curve, used to determine the average saturated activity of the remaining measured points. Two counters were used as before and only two foils were bombarded simultaneously. At least three foils and usually more were bombarded at each position and an average taken.

Compensation for Pile Power Variation

An ionization chamber was placed in the bottom of the thermal column tank in an alumnium tube. The output of the ion chamber, which actuated a recorder, was used to monitor the slow neutron flux. The recorder readings were then used to normalize the foil bombardments to a constant pile power, the zero of the ion chamber being checked before and after each bombardment. This proved to be a simple and very effective means of compensating for variations in pile power from day to day.

Calculations and Results

The average saturated activity as measured at each point was corrected for geometry and normalized to one curve in the same manner as described previously. This data is plotted in Fig. 7. The values of $A_s r^2$ and $A_s r^4$ were determined and plotted as a function of the radius on a rather large scale. These curves were integrated numerically from zero to 40 cm and extrapolated

from 40 cm to infinity using the extrapolation function

$$A_s = \frac{k\ e^{-r/\lambda}}{r^2}$$

where k and λ were determined from the experimental curves at about 40 cm where the extrapolated curve joins the measured curve. Table I gives the measured and extrapolated portion of the $A_s r^4$ and $A_s r^2$ curves. The age $\tau = \frac{\overline{r}^2}{6}$ where $\overline{r}^2$ is the ratio

$$\frac{\int_0^\infty A_s r^4\ dr}{\int_0^\infty A_s r^2\ dr}.$$

From Table I, $\overline{r}^2 = 213.97$ cm^2 and $\tau = 35.7$ cm^2.

TABLE I

Measured and Extrapolated Parts of the $A_s r^2$ and $A_s r^4$ Curves

BY VOLUME	MEASURED		EXTRAPOLATED		$\overline{r}^2$	τ
	$A_s r^2$	$A_s r^4$	$A_s r^2$	$A_s r^4$		
20% Zr + 80% H_2O	42.2965×10^6	8023.88×10^6	0.4678×10^6	1126.41×10^6	213.97	35.7
100% H_2O	31.5931×10^6	5148.43×10^6	0.2445×10^6	588.63×10^6	180.19	30.03

As a means of comparison the slowing down distribution in light water was measured using exactly the same procedure. Figure 8 is a plot of the saturated activity as a function of the radius in light water. Table I gives the values of the measured and extrapolated portion of $A_s r^4$ and $A_s r^2$ curves. From the table $\overline{r}^2 = 180.19$ and $\tau = 30.03$ cm^2.

The results for light water agree well with the data of Hill et al.[2] who measured a value 30.8 cm^2. Our data normalized to theirs at 18 cm from the source is in remarkably good agreement except near the source where our distribution measures slightly higher. The reason for this is not clear unless it is due to the relatively large amount of metal in the elevator and elevator shaft. It is difficult to estimate the accuracy of the measured τ in light water and zirconium since an infinite system was not possible. However, for the geometry used the results should be in as good agreement as those of water since the procedures used were identical.

Acknowledgments

It is a pleasure to acknowledge the many helpful discussions with and beneficial suggestions of Drs. A. M. Weinberg, S. Bernstein, and L. D. Roberts. The visiting author wishes to thank the entire Physics Division of ORNL, and in particular Dr. A. H. Snell for making available the facilities to conduct these experiments.

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(2) Hill, J. E. *et al.*, *op. cit.*

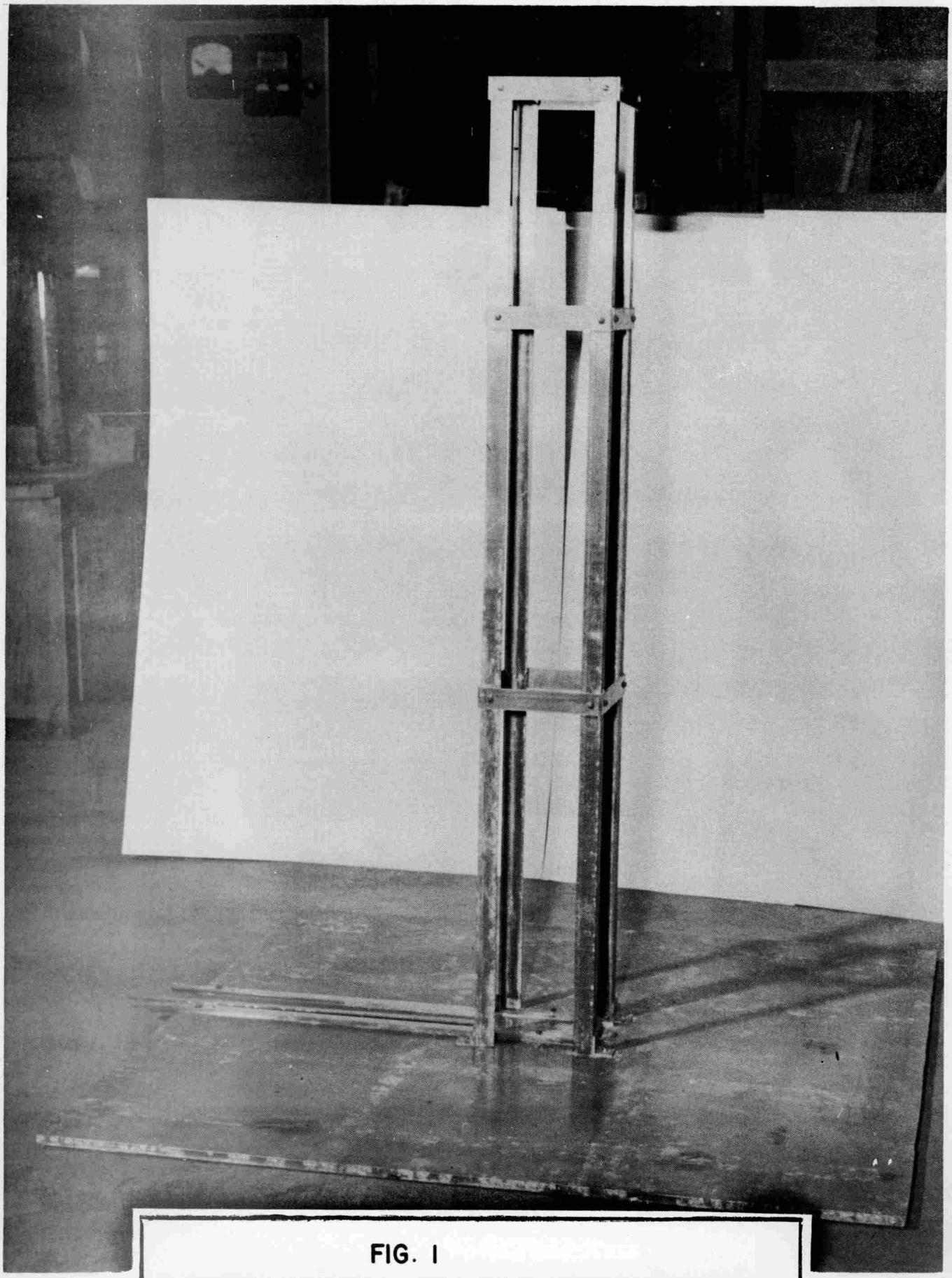

FIG. 1
BASE PLATE AND ELEVATOR SHAFT

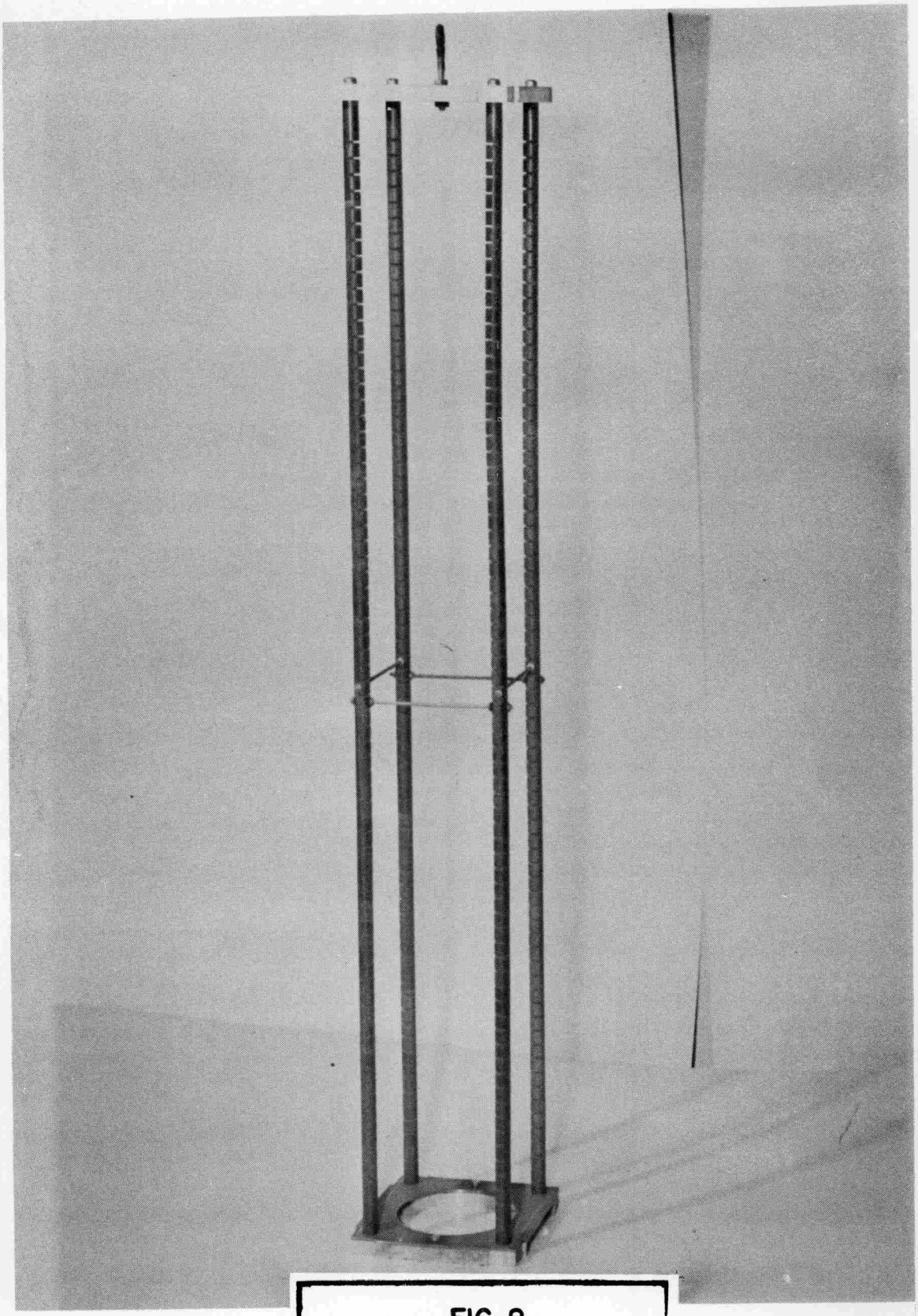

FIG. 2
ELEVATOR

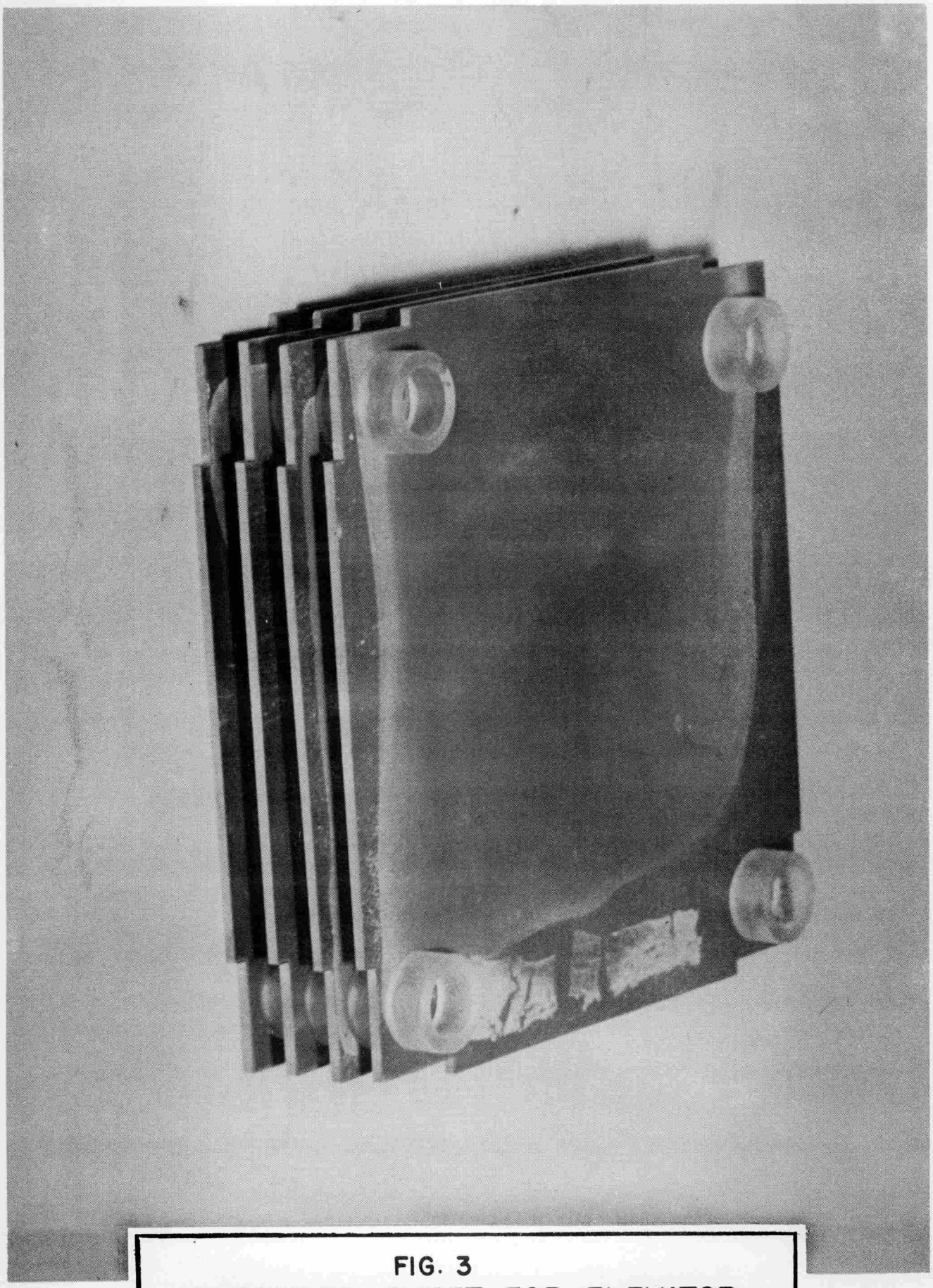

FIG. 3
ZIRCONIUM SHEET FOR ELEVATOR

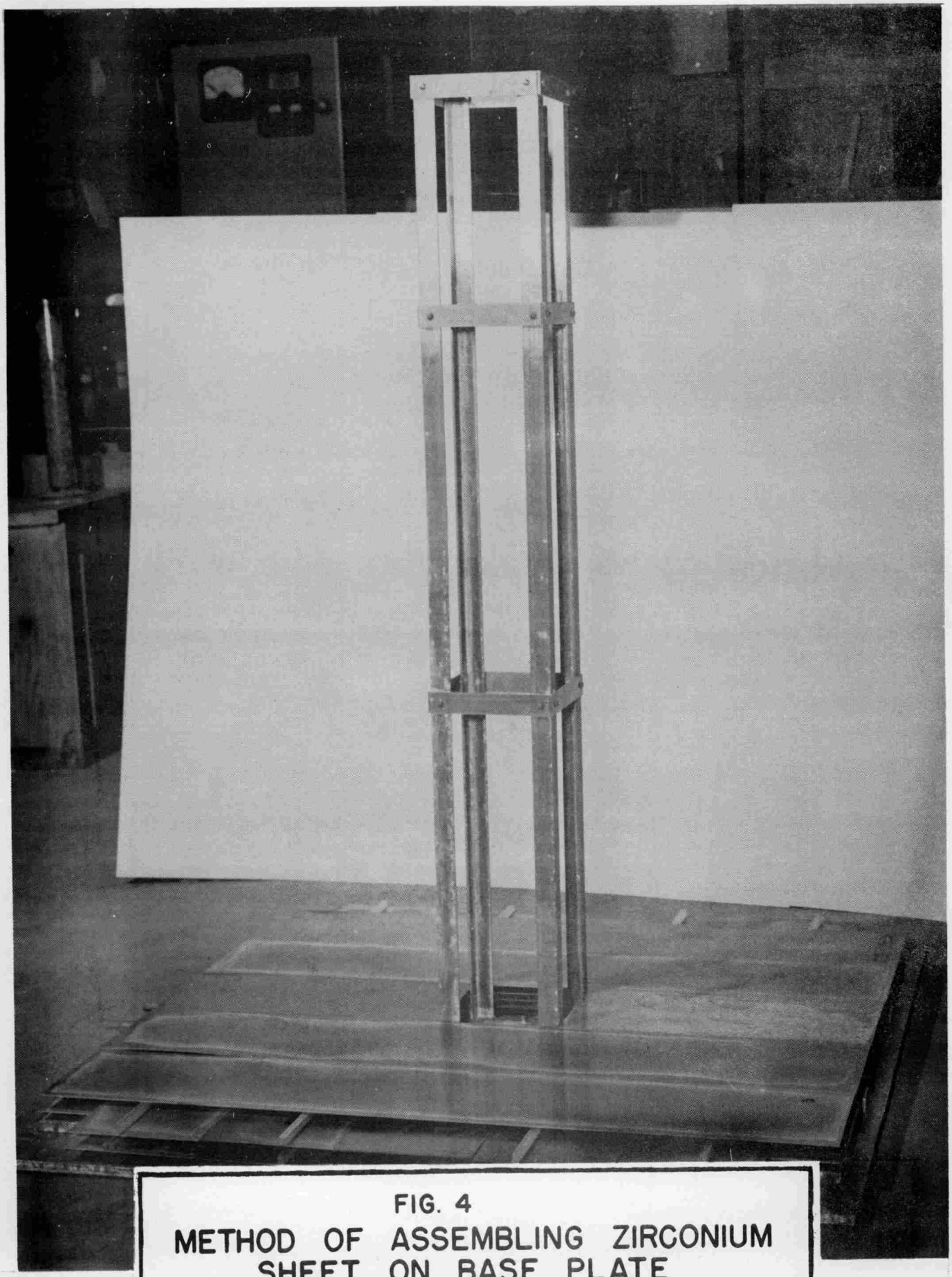

FIG. 4
METHOD OF ASSEMBLING ZIRCONIUM SHEET ON BASE PLATE

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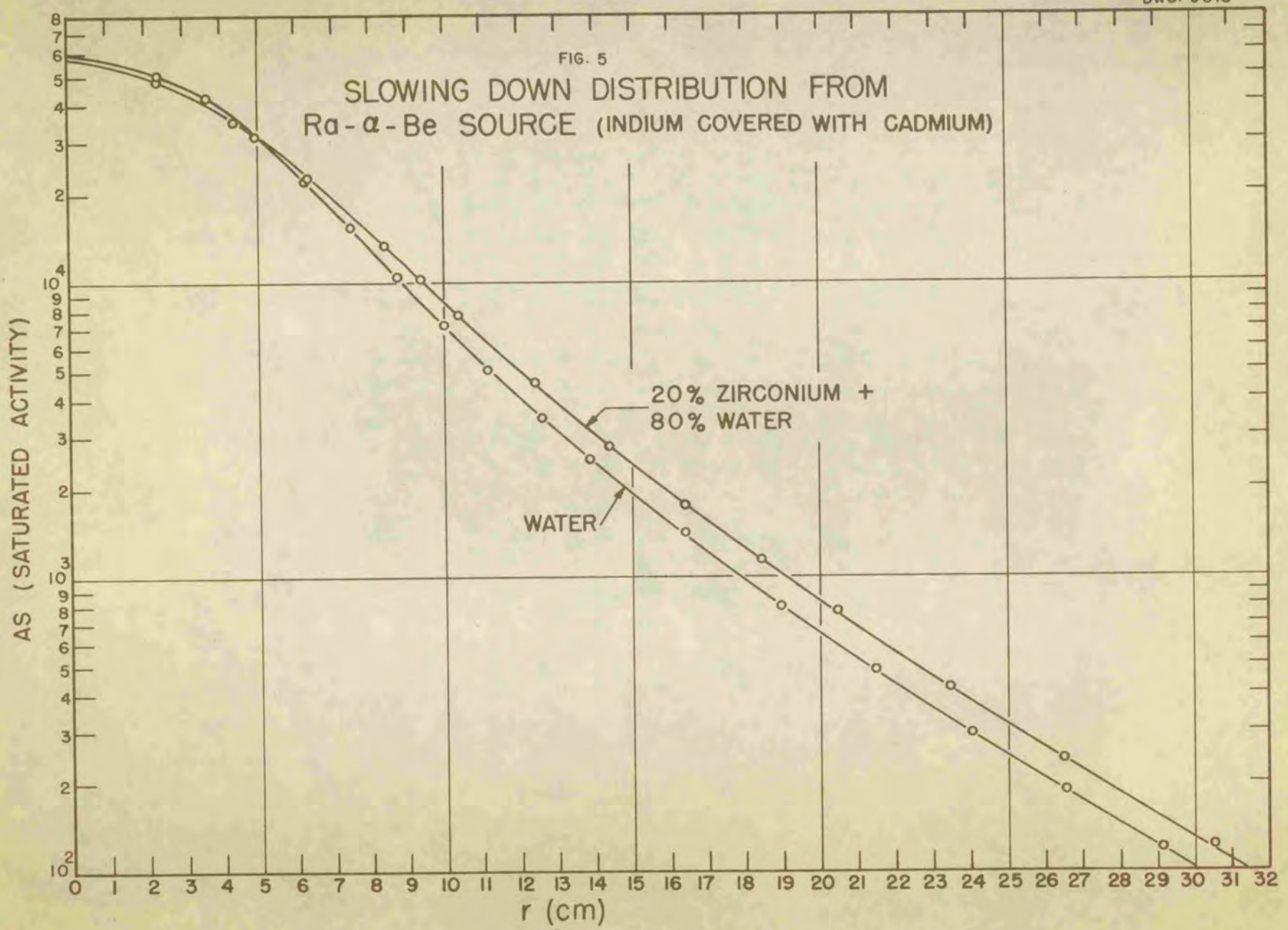

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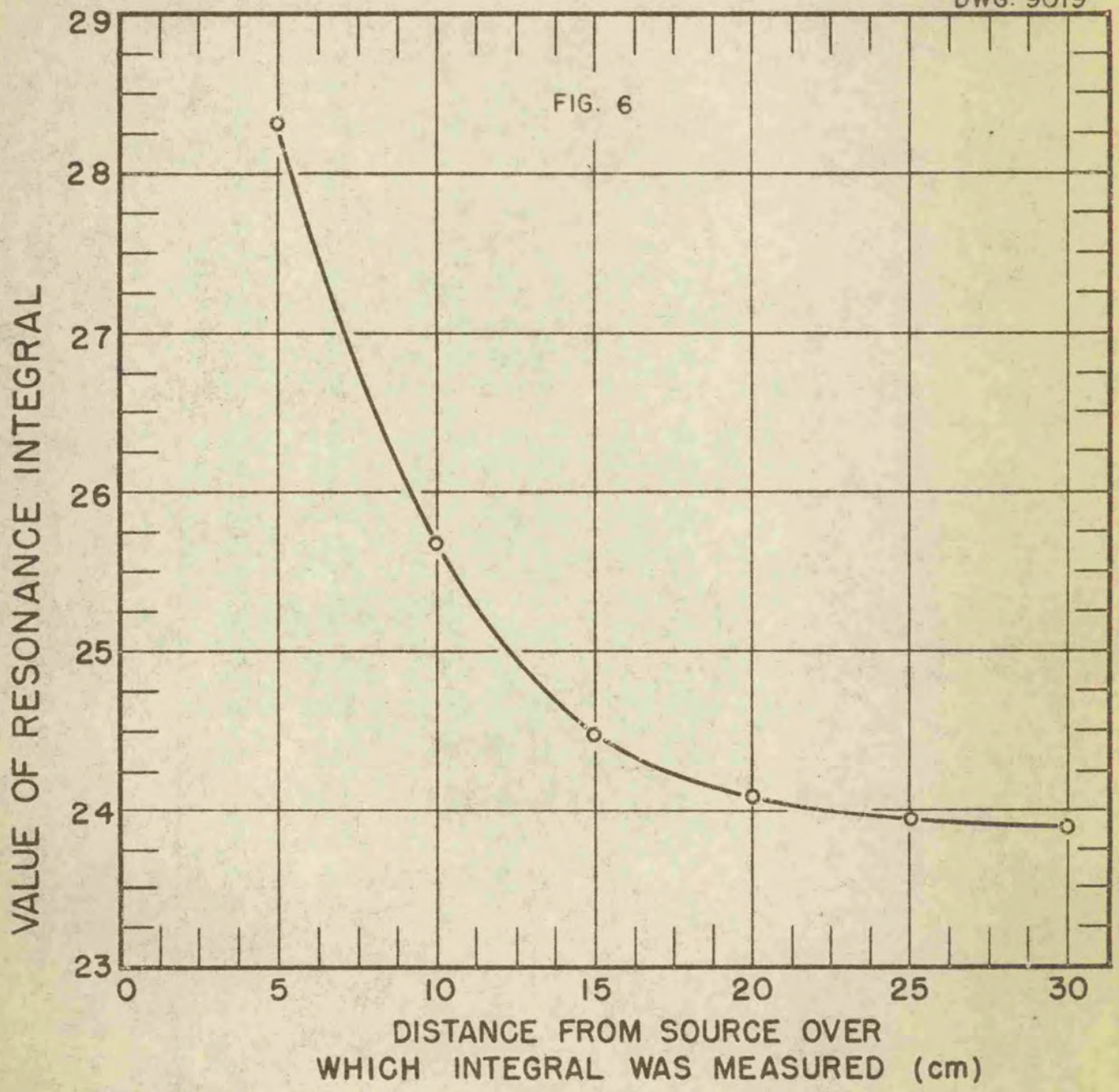

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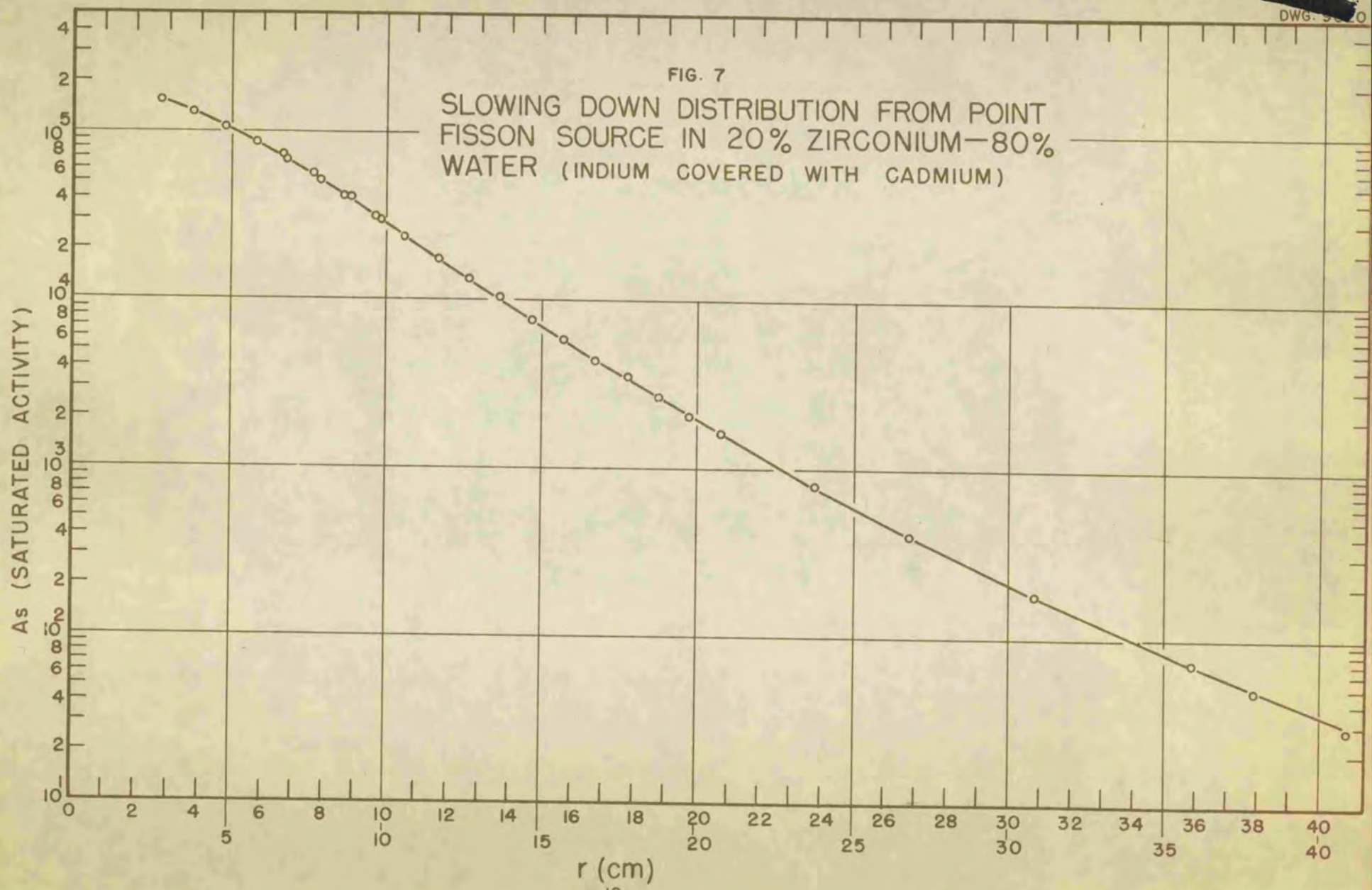

FIG. 7

SLOWING DOWN DISTRIBUTION FROM POINT FISSON SOURCE IN 20% ZIRCONIUM—80% WATER (INDIUM COVERED WITH CADMIUM)

[redacted]

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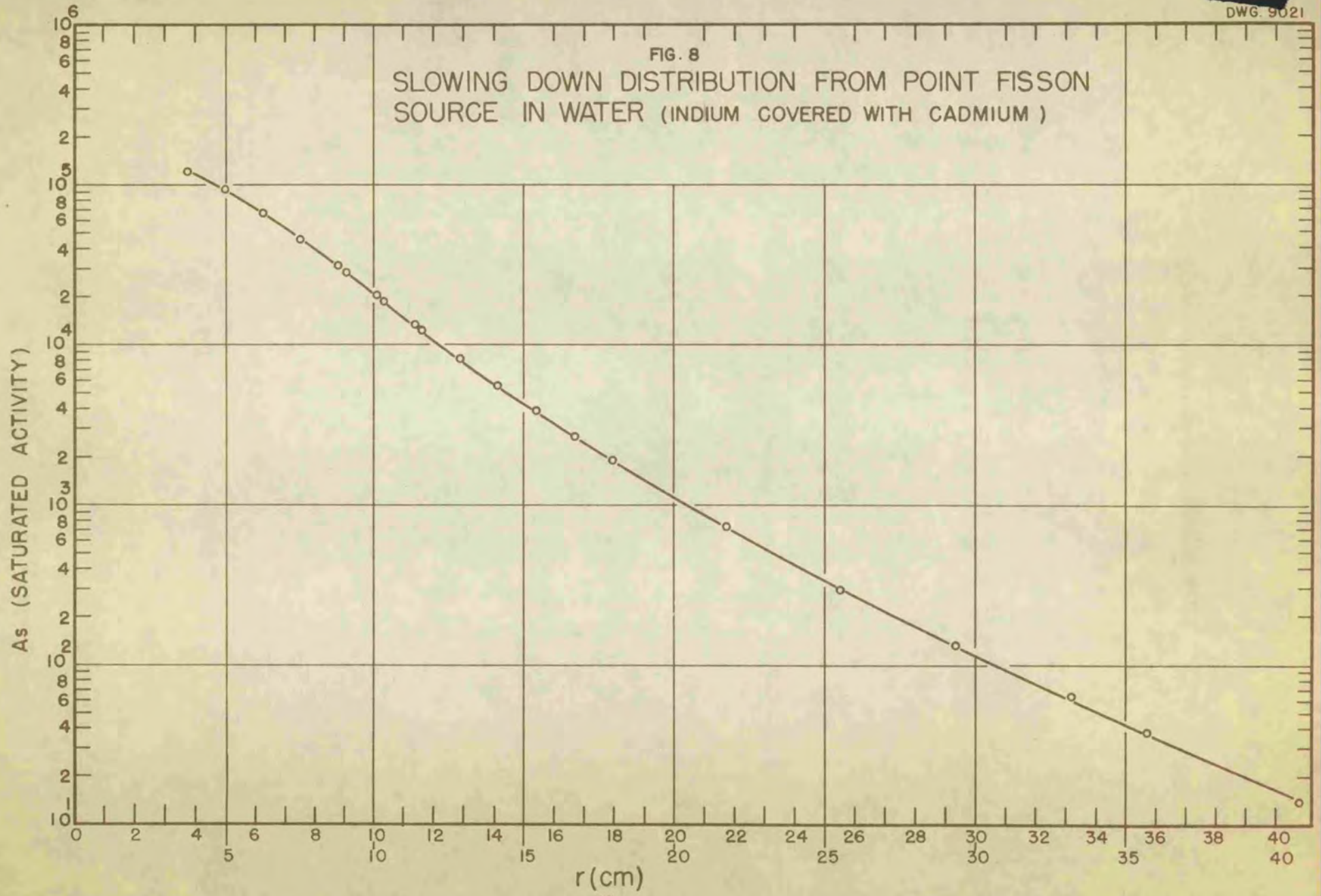

FIG. 8

SLOWING DOWN DISTRIBUTION FROM POINT FISSON SOURCE IN WATER (INDIUM COVERED WITH CADMIUM)