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CP-1626

Metallurgical Project

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FINAL REPORT

DETERMINATION OF NEUTRON DENSITY WITH BISMUTH FOILS

PROBLEM ASSIGNMENT 131-X21P

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Introduction

It is usual to determine the density of neutrons by using foils of metals which become radioactive as a result of bombardment in the region under investigation. From the rate of disintegration of the newly formed radioactive element the number of neutrons absorbed can be calculated. By use of the capture cross section of the element of which the foil is composed the density of the neutrons can then be computed.

Certain difficulties are encountered in such a procedure. The principal of these is that most elements are converted into radioactive substances which emit beta rays as a result of the capture of neutrons. Therefore, the number of disintegrations must be evaluated by determining the rate of emission of the nuclear beta rays at various intervals after exposure to neutrons. This in turn usually requires the use of thin-walled Geiger-Muller counters in a definitely determinable and reproducible geometry. Furthermore, the efficiency of these counters for the beta rays must be determined and maintained constant. Therefore, the equipment must be recalibrated at frequent intervals which requires considerable time and care to yield reliable results.

When the use of radioactive foils is extended to neutron densities of the order expected at W an additional problem arises from the fact that most elements suitable for such foils have a capture cross section inconveniently large. Even for very short exposures of the smallest practicable amounts from the standpoint of accurate determinations results in activities too large for measurement by usual methods.

Therefore, for detector foils at W an element would be preferable which has a low capture cross section for pile neutrons and which decays by the emission of alpha radiation. This is particularly advantageous in view of the fact that procedures are already available for counting alpha particles with considerable accuracy and in a definitely known and reproducible geometry. In the use of an element emitting alpha rays no uncertainty regarding the effects of conversion electrons and gamma radiation is encountered.

Fortunately, in the reaction

$$_{83}Bi^{209} + n \rightarrow {}_{83}Bi^{210} + \gamma$$

we have at hand a convenient means of securing an alpha-emitting detector foil with a low capture cross-section. $_{83}Bi^{210}$ is the well-known RaE of the radium series which disintegrates by emission of a beta ray to produce $_{84}Po^{210}$ which decays by emitting alpha rays. The capture cross sec-

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tion for bismuth is σ — 0.016 x 10^{-24} cm. Furthermore, the decay constants of RaE and Po are quite accurately known from many measurements on the natural form of these elements. The values are

$$\lambda_1 \text{ (RaE)} - 1.61 \times 10^{-6} \text{ sec}^{-1}$$
$$\lambda_2 \text{ (Po)} - 5.57 \times 10^{-8} \text{ sec}^{-1}$$

The corresponding half value periods are

RaE — 4.975 days
Po — 139.5 days

These periods offer an additional convenience in that for the short times of exposure required at W to activate bismuth the exposure time can be neglected in the expression for the exponential growth and decay of the RaE and Po, which simplifies the computations.

Therefore, by counting the alpha particles from a bismuth foil of known weight after exposure to neutrons the density of the neutrons which produced this activity can be calculated. The least accurately known value which enters into this evaluation is the capture cross-section of bismuth for pile neutrons. This value may be improved by future measurements. It is of importance only for absolute measurements of neutron density so that relative values can be measured with considerable accuracy on the basis of known data.

Application of Exponential Laws

In using bismuth as a monitor the observations consist in counting the alpha particles at definite intervals after exposure to neutrons for a known weight of bismuth in a foil sufficiently thin that practically all particles emitted in the forward direction are recorded. From the decay constant this gives the number of Po atoms present in the foil at these intervals. By application of the usual exponential laws for radioactive traces formations we have for t — time after bombardment

$$N_2' (t) - \frac{A \; t_o \; \lambda_1}{\lambda_1 - \lambda_2} (e^{-\lambda_2 t} - e^{-\lambda_1 t})$$

When, as in the present case, $1 - e^{-\lambda_1 t_o} - \lambda_1 t_o$ and $1 - e^{-\lambda_2 t_o} - \lambda_2 t_o$ to within the limits of the required accuracy. In the above expression

$\lambda_1 - 1.61 \times 10^{-6}$ sec^{-1}
$\lambda_2 - 5.57 \times 10^{-8}$ sec^{-1}
$N_2'(t)$ — number of Po atoms at time t after bombardment
t_o — duration of bombardment
A — neutron flux absorbed per unit weight of Bi.

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Rewriting the equation in the form

$$\frac{N_2'(t)}{A\,t_0} = \frac{\lambda_1}{\lambda_1 - \lambda_2}\left(e^{-\lambda_2 t} - e^{-\lambda_1 t}\right)$$

the values of $N_2't/A\,t_0$ may be plotted for various values of t. This provides a ready means for computing from the values of $N_2'(t)$. the corresponding values of $A\,t_0$. Knowing the value for t_0 the values of A and $nv = \frac{A}{N}$, where N = number of atoms in A grams of bismuth, can be computed." Introducing numerical values for N we have

$$\frac{N_2'(t)}{A\,t_0} = \frac{N_2'(t)}{nv\ t_0\ 4.61 \times 10^{-5}}$$

Preparation of Bismuth Foils

For efficient use it is desirable to have layers of bismuth with a small self-absorption for the alpha particles, say not over 3 to 4 mg per cm². This also reduces the relative number of beta rays from the RaE. Foils of bismuth of this thickness are very difficult to make by rolling or extrusion and the mechanical properties of bismuth are such that they would be very fragile and therefore could not be handled easily. This difficulty can be avoided by preparing the foils as a thin deposit on aluminum foil. Fortunately bismuth is one of the metals which can be deposited readily by cathodic sputtering in air. The deposit comes down as a bright metallic mirror in very uniform layers. It adheres readily to clean aluminum. Since the aluminum can be made very thin, .004" or less, and its chief activity occurs with a 2.4 minute half life the aluminum does not interfere with the measurement of the alpha particles a few hours after bombardment. These foils can be used only once so there is no tendency to build up the long period activity in the aluminum.

Sputtering of Bismuth Foils

The arrangement for sputtering bismuth on aluminum foils is shown in Figure 1. A glass bell jar about 8" in diameter (F) is ground to fit a heavy iron disk (E). This joint is lubricated with stop-cock grease to permit ready removal. A glass tube J with a central wire is sealed through the iron plate as shown to provide a lead and support for the bismuth cathode which is covered on top by a glass shield (G). A connection to a Cenco mechanical pump is provided at H to evacuate the inclosure. A rectified high voltage source (2200 volts) is provided at B with the positive terminal grounded. The iron plate serves as the anode and is grounded through a 0-10 ma meter at A to read the sputtering current.

Since in sputtering the metal is detached from the cathode by bombardment with positive ions and then diffuses as an uncharged metallic vapor depositing on nearby objects it is convenient to mount the aluminum foils in a vertical plane so that four or more can be deposited simultaneously. This is accomplished by using the aluminum frame G, or box open at top and bottom, supported on the glass strips CC. The detail of this box is shown in Figure 2 where the upper view is looking down at the top and the lower is a side view. A hole 3/8" diameter with bevelling as indicated is cut in the center of each face. The aluminum foil 12 mm square are laid over these holes and clamped in position by screwing on the cover plates C as shown. This assembly is then mounted beneath the cathode as shown in Figure 1. The 3/8" diameter for the hole was chosen to give a circular deposit which could be centered on an aluminum foil 12 mm square. This dimension was in turn chosen for insertion in the 1/2" diameter foil tube at W.

The pressure of air in the sputtering jar was not measured chiefly because it lies in a region where measurement is not easy. To provide uniform conditions for deposition of bismuth and to prevent melting the bismuth cathode the following procedure was adopted. The pressure was adjusted by means of an adjustable leak so that with continuous operation of the Cenco pump a current of 3 ma was maintained steadily at a constant voltage of 2200 volts. This gave a cathode dark space of several centimeters and yielded a fairly rapid deposit.

It is of interest that under these conditions the amount of bismuth deposited, as expected, is quite accurately proportional to the time of sputtering. This was tested by depositing films for 2, 4 and 8 hours with the results shown in Figure 3. In fact with a more refined regulation of the pressure line the somewhat crude arrangement used, a pinch cock on a rubber tube, the weight of bismuth can be determined to a few percent from the time once a definite arrangement has been calibrated. The weight of all deposits used in the present work were determined by weighing of the individual foils before and after deposition.

Bombardment

The bismuth deposits were bombarded for varying periods 4 to 16 hours in slot #62 at the center of the pile at X. This position was chosen to give the maximum flux and to secure a uniform region where a number of deposits of different weights could be exposed simultaneously. The power levels were of the order of 1000 kw with a flux of the order of a $\times 10^5$ per sec per watt. Under these conditions approximately 10 hr; exposure produced optimum rates for alpha particle counting in foils weighing from 2 to 4 mg.

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Unfortunately the flux is not known sufficiently accurately in this pile to provide an accurate means of standardization of the bismuth deposits or in other words to determine the σ of bismuth for pile neutrons. Therefore these bombardments can be utilized chiefly to determine the reproducibility of the results from a set of deposits exposed simultaneously. Since changes in the pile are in constant progress it is not to be expected that foils exposed on different days would yield the same value for the flux.

The deposits were prepared in groups of 10 or 12 and mounted in a thin aluminum strip with edges crimped up to retain them. This strip was attached to a wood strip for insertion in the foil slot. Wood was selected since a heavy aluminum strip would become too active for safe handling within a reasonable time after exposure.

Counting of Alpha Particles

From 24 to 48 hours after exposure the deposits were placed in a conventional parallel plate alpha ray chamber filled with air at atmospheric pressure. The individual ion pulses were counted in the usual way by means of linear amplifier and scaling circuit. From 10,000 to 100,000 counts were recorded for each observation to give a satistical accuracy of the order of 1/2%. The weights of the foils were also measured within the same limit of accuracy.

The results of the counting are shown in Table I.

Table I

Foil No.	Wt. mg.	Alpha count per mg per sec at t — $6.26x10^5$sec	N_2't	A t_o from alpha count	A t_o from pile data
21	0.795	30.31	$1.06x10^{12}$	$1.70x10^{12}$	$1.20x10^{12}$
22	1.006	30.21			
46	2.183	29.67			
25	2.941	28.16			

The ratio of the values of At_o as computed from the alpha count and from the value of the flux in the pile and exposure time, to, combined with the value for the cross section of bismuth equals 1.4 in this instance. An average of 5 similar sets of observation yields 1.55 for this ratio. This indicates either that the cross section for bismuth is in error by about 50% or that the density of neutrons generally accepted for the pile at X, 4×10^5 sec$^-$ watt^{-1} is in error by the same factor or

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a combination of these circumstances. The general agreement of the ratios for several series of observations indicates that bismuth foils can be relied on for relative values.

The question of absolute values cannot be solved until more careful measurements of the cross section of bismuth, particularly for pile neutrons, are available. Since it is apparent that this element can be quite useful in measurements of high densities of neutron flux a redetermination of cross section for bismuth would be of value. The factor that makes bismuth useful for this purpose, low cross section, also creates difficulties in its accurate measurement. Perhaps the rate of production of polonium, determined by chemical separation and weighing, when exposed to high intensities of pile neutrons would give a better value.

Foil Holders for W

For effective use of these bismuth foils in the pile at W a holder must be available for inserting them into the pile which when removed will not constitute a serious physiological hazard. At X wood strips could be used but these would be far too active for safe use at W. A further important consideration is that it is desirable to keep the bismuth foils below 100°C to prevent oxidation. In any case they could not be permitted to reach the melting point of bismuth (271°C). Since temperatures in excess of 200° are not improbable in the foil tubes at W it is evident that water cooling is desirable.

Lead and bismuth both have low capture cross sections and produce negligible gamma radiation as a result of neutron bombardment. Therefore, an alloy of these two metals formed into tubing to fit the W holes should be satisfactory. Accordingly a commercial purity alloy of 20% Bi and 80% lead, having quite good mechanical properties, has been prepared. This alloy melts at about 240°C, is readily malleable, and sufficiently rigid when extruded as a pipe 1/2" O.D. and approximately 1/16" wall thickness. Samples have been exposed in the pile at X and found to show a gamma radiation decaying as shown in Figure 5. This radiation is relatively soft as well as of low intensity. A lead filter 1/2" thick reduces it practically to zero as indicated in Figure 6. Under these conditions protection must be provided for personnel only from the relatively intense beta radiation present immediately after removal from the pile. This can be accomplished by an ordinary iron pipe shield. After a few hours the beta rays will have decayed sufficiently to permit removal of the bismuth for counting.

In Figure 5 a drawing of a model foil holder constructed from this alloy. It is designed to fit in the 1/2" W foil tube. To protect the bismuth foils the tube is cut to the diameter at intervals and a flat portion soldered in place to provide a level area 1/2" wide and 2 or 3 inches long to receive the bismuth foils. A solder of half and half bismuth and lead is very useful for this purpose since it melts at about 15?°C

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and parts can be soldered together using an ordinary soldering iron. An inner tube 1/4" O.D. flattened throughout its length is inserted to near theffar end of the outer tube to provide water cooling throughout the length of the assembly. Small stirrups of the alloy are soldered at each end of the flattened area of the large tube to permit foil holders to be inserted and removed readily. It is obvious that the flat sections can be provided at any desired interval along the tube to provide for simultaneous measurement at various distances from the center of the pile.

Figure 7 is a photograph of the model showing a bismuth foil in position and unmounted foils beside it. To hold the foils a thin strip of aluminum, .004", is crimped up at the edges to retain the square aluminum foi,s which carry the circular deposit of bismuth. Ears are provided at each end to go under the stirrups provided to receive them.

In conclusion it may be well to point out that on the basis of the gamma ray measurementsacomplete pipe 25 feet long of the Pb-Bi alloy tube would have a gamma ray activity of approximately 50 mc immediately on removal from the pile for an exposure of nvt — 2.55×10^{16}. This is an approximate exposure to activate the bismuth foils to a maximum usable activity. Since no short period is evident in the decay curve the same behavior could be expected at W.

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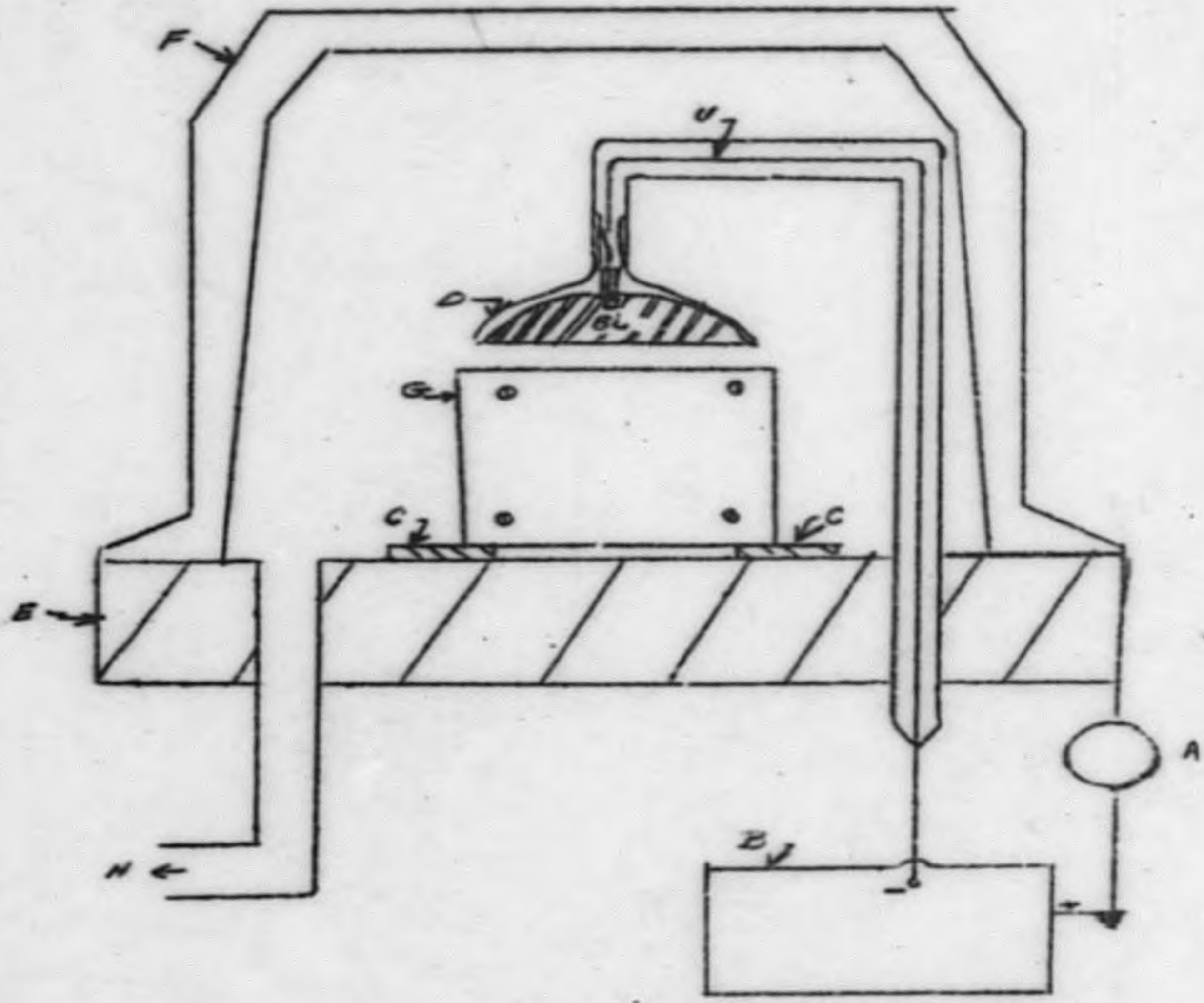

Legend

A 0-10 ma.

B Power Pack
0-2000 volts

C Glass Plates

D Glass Container
for Cathode

E Steel Plate

F Bell Jar

G Aluminum Box

H To Vacuum Pump
Outlet

J Copper Wire

Figure 1

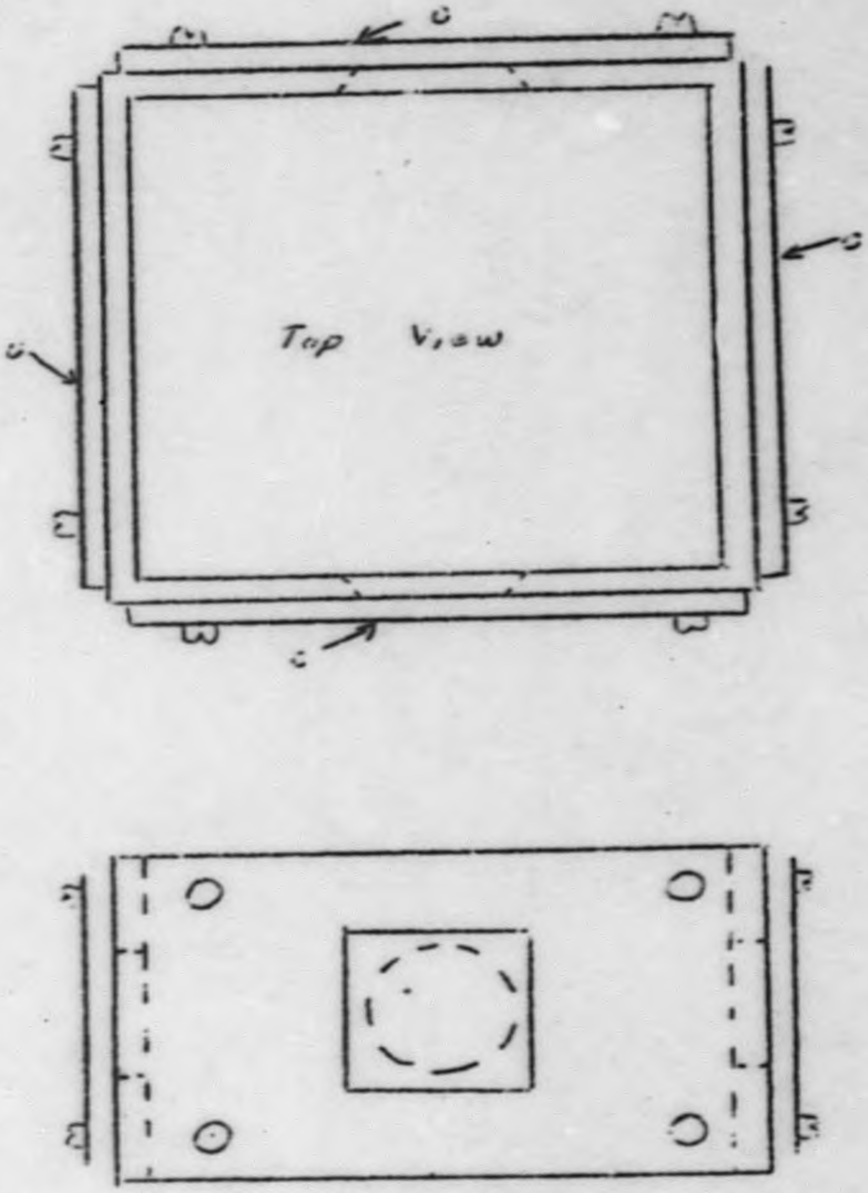

Fig 2

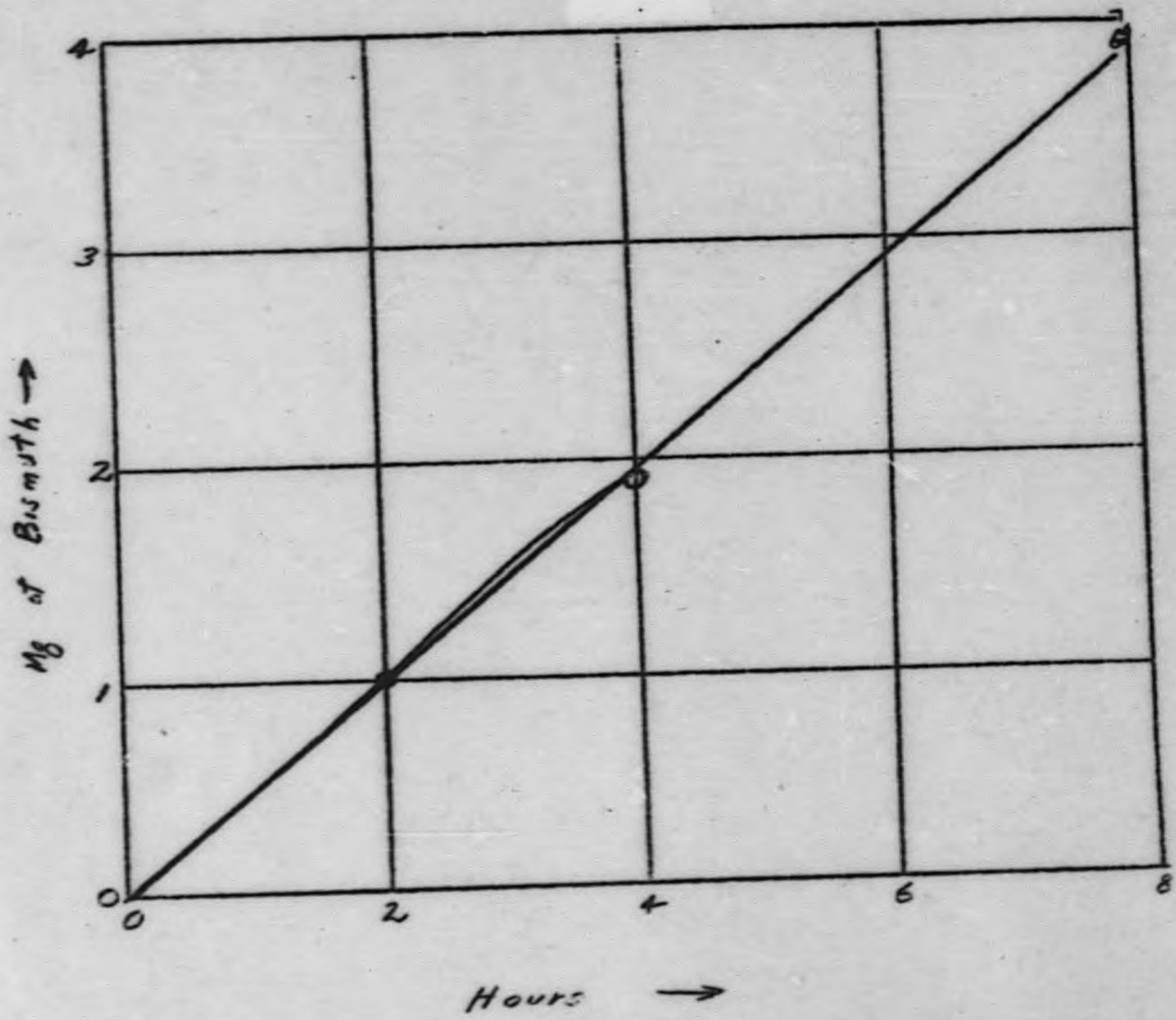

Fig. 3

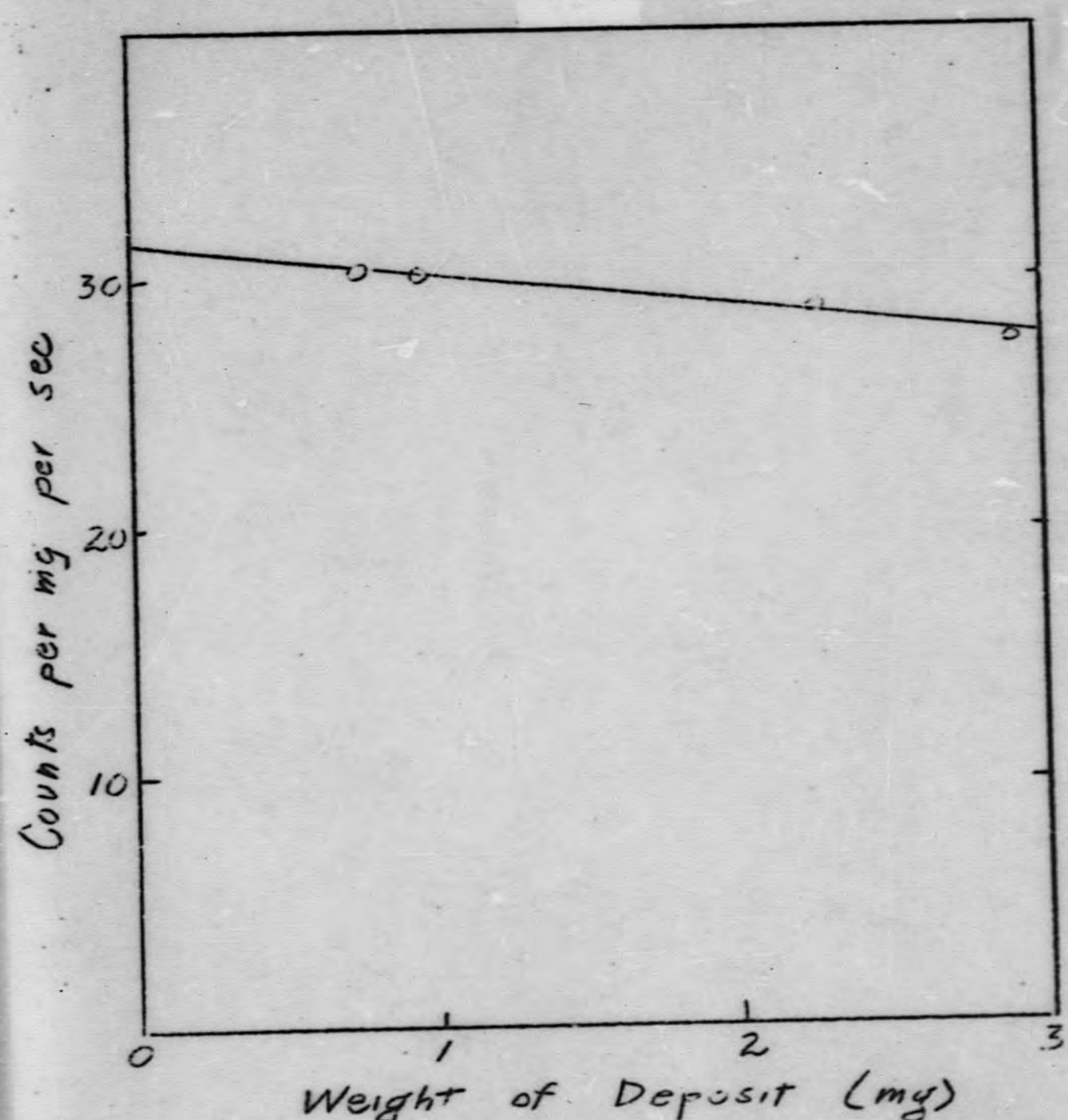

Fig. 4

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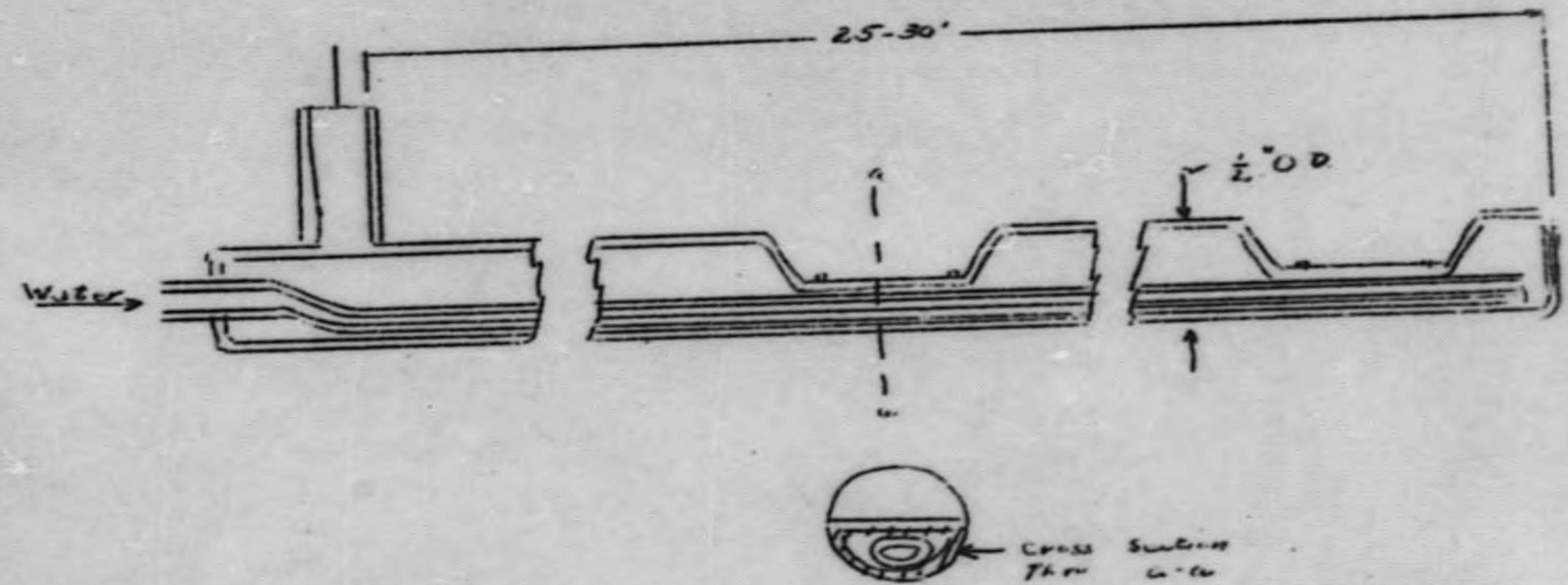
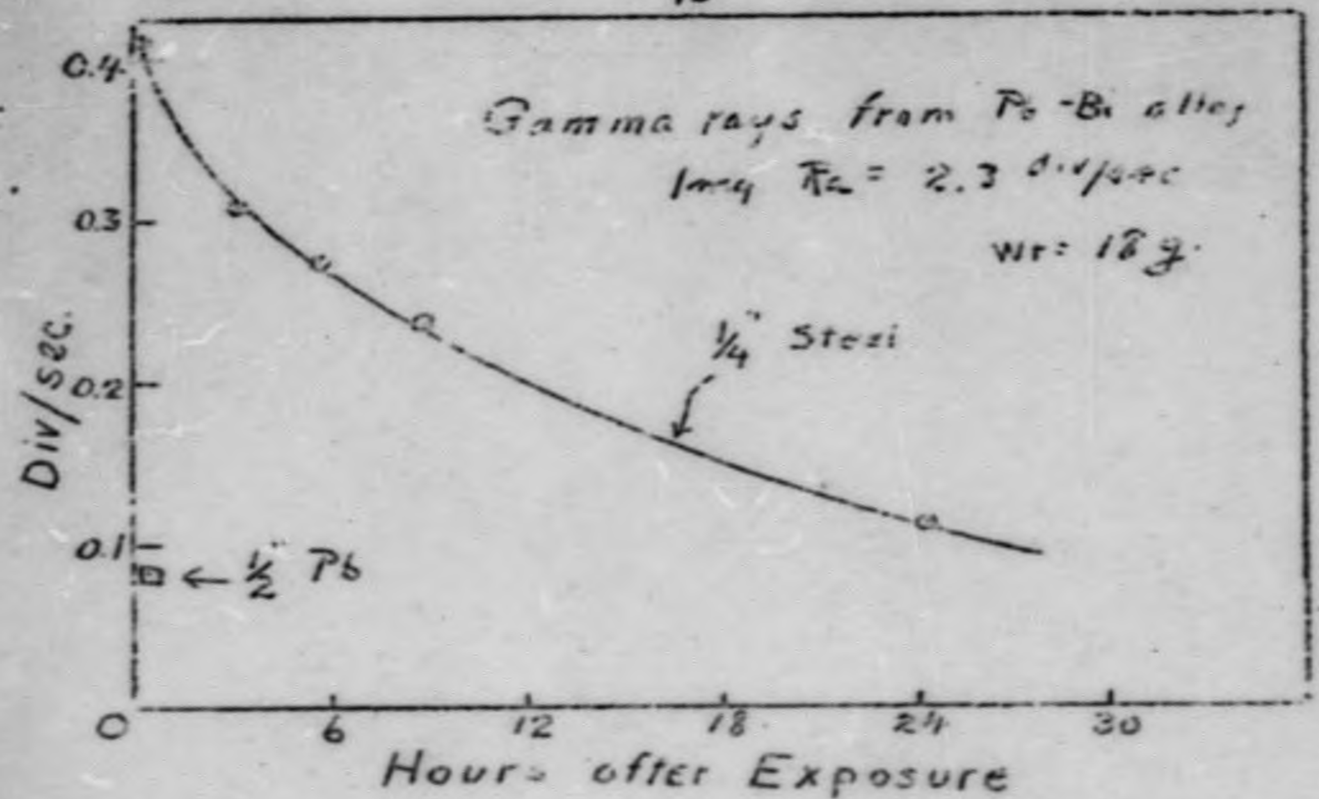


Fig 5


Gamma rays from Pb-Bi alloy
1 mg Ra = 2.3 div/sec
wt: 18 g.
1/4" Steel
1/2" Pb
Div/sec.
0.4
0.3
0.2
0.1
0
6
12
18
24
30
Hours after Exposure

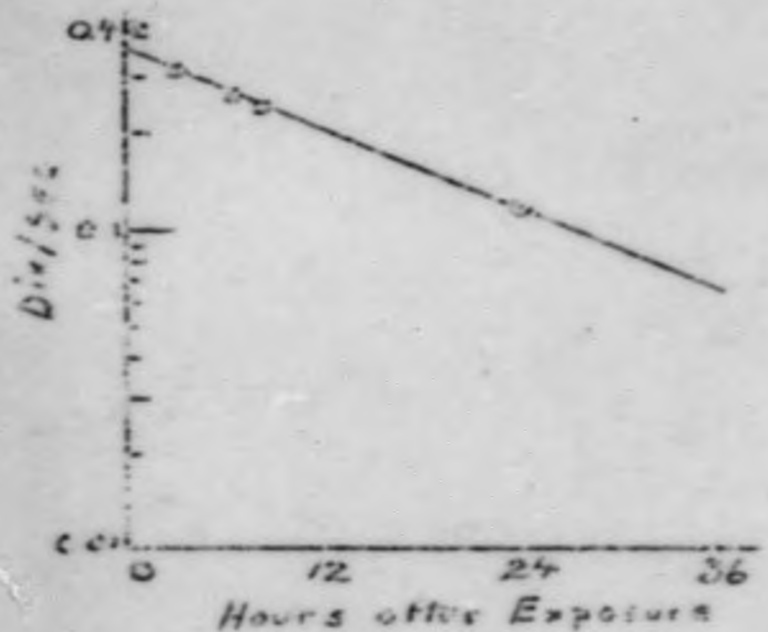

Div/sec
0
12
24
36
Hours after Exposure

Fig 6

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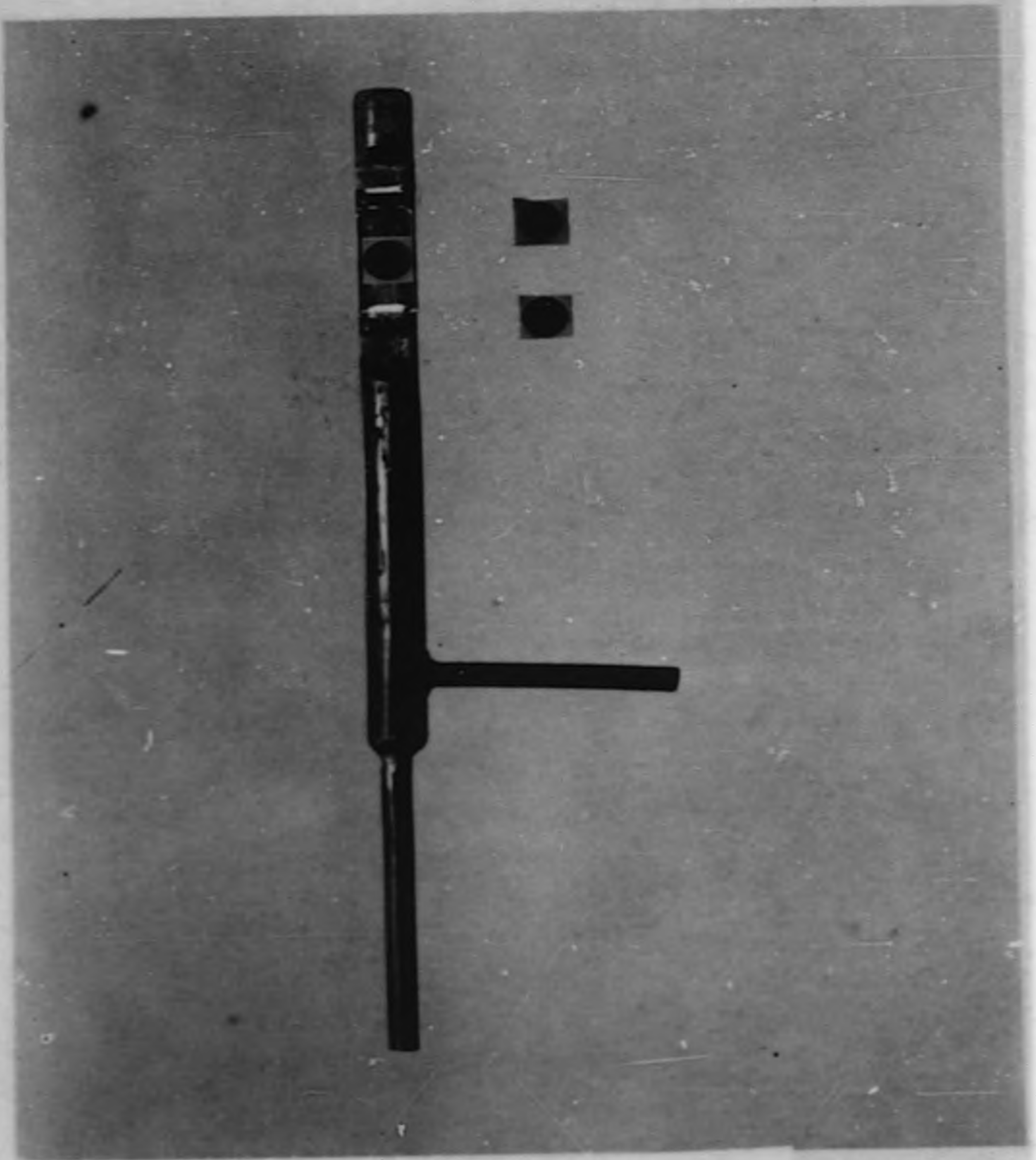

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Addendum for
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(A-2295)

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In a recent report (CP-1626) describing an exploration of the usefulness of bismuth as a monitor for the X pile it has been shown by the author that the capture cross section of bismuth for pile neutrons is approximately 50% larger than the usually accepted value of 0.016 x 10^{-24} cm. Accurate determination of the value was not possible under conditions existing at the time the bismuth foils were exposed in the pile at X.

However, it is readily possible to determine the cadmium ratio for bismuth by exposing foils protected by cadmium and uncovered foils simultaneously to the same intensity of neutrons in the pile. Since information on this ratio is as yet lacking and does throw some light on the origin of the discrepancy in the value for the cross section, a few measurements have made of this ratio. Following the method outlined in the previous report, the ratio of captures were determined by counting the polonium alpha particles from each of two sets of foils; one protected by cadmium, the other bare, when exposed to pile neutrons.

This was also done in the pile at X in slot #62 which traverses the center of the pile. The distribution of neutron intensity in this slot is symmetrical with respect to the center and changes slowly over a considerable length near the center. Therefore, the two sets of foils were mounted on a wood slat and were separated a distance of 18 inches from each other. The wood slat was then pushed into the pile until the midpoint between the sets of foils was at the center of the pile. This gave equal intensities of neutrons and prevented any marked disturbance of the neutron intensity at the position of the bare foils by the presence of the cadmium covering the other set.

To improve the accuracy of the measurement and permit the elimination of the effect of the self-absorption of alpha particles in the foils, which varied from about 1 to 5 mg. per sq. cm., several foils of different weights and the same area were included in each set. The alpha particles from these foils were counted in a usual alpha-ray chamber with linear amplifier. Five sets of foils were exposed and counted in this way. The total exposure for each set varied between 12 and 24 hours at approximately 1000 kw.

A typical alpha-ray counting curve is shown in Figure 1 where the counts per mg. per sec. are shown in relation to the weight of the foil for two sets of foils. The upper curve is for bare, the lower for the cadmium covered, bismuth. The summary of the five sets is given in Table 1.

Series No.	Cadmium / No Cadmium
1	0.230
2	0.244
3	0.210
4	0.223
5	0.256
Mean	0.233

These results show that about 23% of the captures in bismuth for pile neutrons are due to those with energies sufficient to pass through 0.5 mm

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cadmium and, therefore, are in excess of thermal. The value of this ratio does not account for the magnitude of the discrepancy observed in the value for the cross section when assuming that the value of 0.016 is based entirely on neutrons with thermal energies. The results reported here show that epithermal capture does exist and indicates its magnitude.

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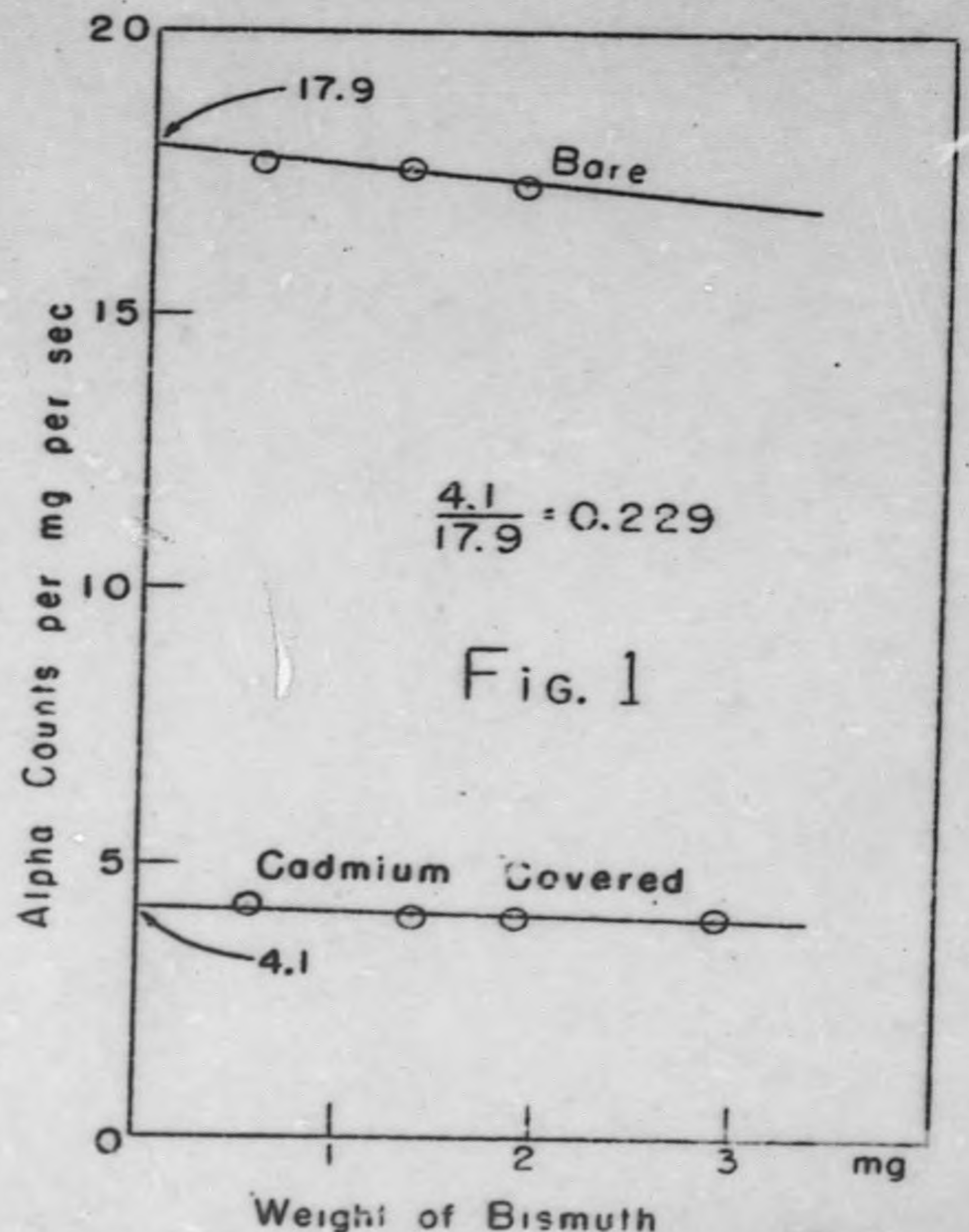

$\frac{4.1}{17.9} = 0.229$

Fig. 1

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END