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NUCLEAR REACTIONS IN ANTIMONY WITH HIGH ENERGY PARTICLES

By

Manfred Lindner

Berkeley, California

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NUCLEAR REACTIONS IN ANTIMONY WITH HIGH ENERGY PARTICLES

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ABSTRACT

Thirty-six radioactive species have been identified as products of the spallation of antimony irradiated with high-energy particles produced in the 184-inch cyclotron at the University of California Radiation Laboratory in Berkeley. The manner in which the yields change with change in the energy of the bombarding particle was also investigated. A simple mechanism has been proposed which incorporates the concept of binding energy of neutrons, protons, and alpha particles. This mechanism leads to conclusions in satisfactory agreement with the observed results.

The radiation characteristics of nine new radioactive species are described in detail. These include two tellurium, one antimony, two tin, two palladium and two rhodium activities.

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NUCLEAR REACTIONS IN ANTIMONY WITH HIGH ENERGY PARTICLES

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INTRODUCTION

The subject matter covered by this thesis is principally concerned with the identification of those products which result from the irradiation of elements in the region of atomic number 50-51 with deuterons and helium ions produced in the 184-inch cyclotron.

In this region of the periodic table, the mass defect curve is near its minimum, i.e., the nuclei of elements in this region are more stable with respect to their individual nucleons than elements in any other section of the periodic table. It is to be expected, therefore, that disintegration products should differ in degree, if not in kind, from those produced by these same beams on nuclei representative of other regions of the periodic system. A study of such disintegration products and their mode of formation may therefore serve to aid in the understanding of the forces occurring within the nucleus.

The investigation was early handicapped by the fact that many of the neutron-deficient radioactive isotopes obtained were either unknown or, having been previously reported by other investigators, could not be assigned definite mass numbers. In several cases, mass assignments and radiation characteristics were made on previously unreported isotopes. Fortunately, the situation regarding isotopes containing an excess of neutrons, that is, radioactive nuclei which decay by emission of a beta particle, is much better; the intensive work conducted on the uranium fission products[1] has served to identify many of the beta emitters found in this region.

The first several sections, therefore, concern themselves with the elucidation of some of these unknown activities encountered upon irradiations, together with their radiation characteristics and decay constants.

The results of irradiations with high-energy deuterons and helium ions are discussed in later sections. For these bombardments antimony was generally, though by no means exclusively, used as the target material. Chemical products investigated in this case include elements from atomic numbers 52 to 39, inclusive. These are discussed in the light of the cross sections for their formation, as is also the effect of varying the energy of the bombarding particle.

Radioactive decay was followed by means of a Scott-type Geiger-Müller counter tube containing a mica window at one end, of approximately three mg/cm^2 thickness. The tube was supported on a plastic stand containing five sets of slots which served to hold the cards containing the samples to be measured. The effective geometry of the samples, when placed in these slots, was known from prior calibration with a uranium standard; when placed in the second set of slots from the top, the geometry was approximately 10%. This was a standard value used throughout all the bombardments.

The decay of electromagnetic radiations was followed by interposing a block of beryllium of about 1900 mg/cm^2 between the sample and the counter tube. This stops completely all electrons up to energies of four Mev, while absorbing only about 30% of the K x-rays of the region around Z = 50. It is for this latter reason that beryllium rather than aluminum was used. The half-thickness of the K x-rays of, say, tin, is approximately 150-200 mg/cm^2 of Al. It would therefore be difficult to distinguish the absorption of various electrons from that of the x-rays if aluminum were used as absorber.

When it was desired to follow the decay of gamma-rays alone, the x-radiation was absorbed through a layer of about 130 mg/cm^2 of lead; x-rays have a half-

thickness of about 10 mg/cm^2, hence are effectively absorbed. It was also found necessary to add a layer of several hundred milligrams of beryllium on top of the lead to stop Compton electrons ejected from the lead by interaction with any hard gamma radiation being measured; since these electrons have a counting efficiency roughly one hundred times that of gamma radiation, the former could cause serious errors in measurement of a gamma-ray if not stopped in this manner. In obtaining absorption data of electrons, beryllium absorbers were used; for x-ray absorption, sufficient beryllium to block all electrons, plus varying thicknesses of aluminum. For hard gamma radiation, sufficient aluminum to screen out the x-rays and electrons plus varying thicknesses of lead were used.

Another instrument of which frequent use was made was a crude beta-ray spectrometer which had an $H\rho$ constant of about 53 gauss-cm per milliampere of current through the field coil. Both negative and positive electrons could be distinguished, as well as the approximate shape of the energy distribution curve. It was, therefore, quite often possible to distinguish between a "line" of conversion electrons and a true beta-spectrum and to determine small amounts of β^+ activity in the presence of large amounts of negative electrons and β^- particles. From the constant of the instrument, the energy of a conversion electron or the maximum energy of either a β^- or β^+ spectrum could be approximately ascertained.

The beam intensities used in all bombardments on the 184-inch cyclotron were more or less constant. Deuteron beam currents were about one microampere, while helium ion beams were taken to be about 0.1 microampere. These values will henceforth be understood to apply to all bombardments unless specific statements are made to the contrary.

Section I

NEUTRON-DEFICIENT ISOTOPES OF PALLADIUM AND RHODIUM

When antimony was irradiated with 200 Mev deuterons, among the products formed were palladium activities. In Section V the chemical separation procedure by which a palladium fraction was obtained will be described. For the present, it suffices merely to state that two rhodium daughter activities could be removed from this palladium fraction, having half-lives of about one day and six days, the parents of which appeared to have half-lives of at least several days and ten hours, respectively.

Two such rhodium activities had previously been reported by Sullivan, Sleight, and Gladrow[2] who obtained the activities from deuterons on ruthenium. They assigned the one-day activity to $_{45}Rh^{100}$, and the six-day activity to $_{45}Rh^{101}$. If these assignments are correct, then $_{46}Pd^{100}$ has a half-life of the order of days, $_{46}Pd^{101}$ about 10 hours.

The total yield of palladium activity obtained from a bombardment of antimony was so low that it was not possible to conduct an accurate appraisal of the activities of either the palladium or its rhodium daughters. Because it was highly desirable to known both the counting efficiencies and the mass assignments of these activities, they were produced by another means.

From considerations of the simple energetics involved, it was reasoned that rhodium foil, consisting of only one natural isotope, $_{45}Rh^{103}$, when irradiated with 50 Mev deuterons from the 184-inch cyclotron should produce the following activities:

$$Rh^{103}(d,n)Pd^{104} \text{ (stable)}$$

$$Rh^{103}(d,2n)Pd^{103} \xrightarrow{17\ d\ K} Rh^{103*}$$

$$Rh^{103}(d,3n)Pd^{102} \text{ (stable)}$$

$$Rh^{103}(d,4n)Pd^{101} \longrightarrow Rh^{101} \longrightarrow Ru^{101}$$
$$Rh^{103}(d,5n)Pd^{100} \longrightarrow Rh^{100} \longrightarrow Ru^{100}$$
$$Rh^{103}(d,6n)Pd^{99} \longrightarrow Rh^{99} \longrightarrow Ru^{99}$$

Pd^{103} is known to decay by K-electron capture[3] with a half-life of 17 days, Pd^{102} is stable. Pd^{101} and Pd^{100}, unknown, should have measurable half-lives whereas Pd^{99} should be formed in lower yield; the possibility of its presence cannot, however, be discounted.

Accordingly, a strip of rhodium foil was bombarded with 50 Mev deuterons for one hour. The strip was then fused with molten potassium bisulfate[4]. After the melt had cooled it was dissolved in water. Twenty milligrams of palladium, as chloride, were added, and the solution made 0.5 N in HCl to a volume of about 20 ml. Five ml. of dimethylglyoxime--saturated ethanol were added, the resultant yellow precipitate being digested for five minutes in a hot water bath and centrifuged. The precipitate was washed, dissolved in a minimum of aqua regia, diluted to 20 ml and the procedure repeated twice in the presence of added rhodium carrier. At specified intervals of time rhodium daughter fractions were removed from the palladium fraction in the following manner. The neutralized palladium solution was made about 2 N in KNO_2. Several milligrams of rhodium carrier were added and the solution heated to boiling. The white insoluble precipitate of potassium rhodonitrite was then centrifuged, washed and filtered. A typical decay curve is shown in Fig. I-1. Obviously, two components are present, a 20-hour and a four-day activity. Table 1 summarizes the data pertinent to the decrease in yield of these activities with each successive removal of rhodium from the palladium.

Table I-1

TOTAL C/M OF 20-HOUR AND FOUR-DAY RHODIUM ACTIVITIES OBTAINED FROM PALLADIUM FRACTION, AS FUNCTION OF TIME

Hrs. between Rh Removals	20-hour Activity Total c/m through Be	4-day Activity Total c/m through Be
12.0	15,500	4680
12.0	17,600	2780
12.0	13,650	960
12.0	18,100	850
12.0	15,100	610
24.0	11,600	270
24.0	15,300	390

Although these data do not readily lend themselves to quantitative reckoning, several important conclusions may be drawn. First, the yield of four-day activity diminishes with roughly a half-life of from eight to twelve hours. The apparent "tailing" of yield was shown later to be due to small quantities of impurities of a palladium activity of similar half-life. Since the rhodium daughter fractions were never repurified from the palladium, this is not unreasonable. Secondly, the 20-hour rhodium daughter appears to grow from a parent whose half-life is of the order of many days. The remainder of the work thus became concerned with a more accurate appraisal of these relationships.

The palladium fraction early showed a positron of maximum energy of approximately 2.5 Mev which appeared to decay, from measurements of the decrease in area of the positron peak, with a half-life of ten hours. No negative electrons were found to behave similarly. In addition, this component showed up readily in the gross palladium decay curve; twenty per cent of the counting rate was found to be due to x-rays. Assuming the counting efficiency of x-rays to be about one per cent, one concludes that this period decays ninety per cent by K-electron capture, ten percent by positron emission. This was presumed to be the parent of the four-day rhodium activity.

The latter was characterized after the decay of the 20-hour component in the early rhodium daughter fractions. An absorption curve is shown in Fig. I-2. In the beta-ray spectrometer it was found that this activity consisted entirely of conversion electrons of energy 0.2 Mev.(Fig. I-3)

If the presumptions concerning the chain,

$$Pd \xrightarrow{10\text{-hr.}} Rh \xrightarrow{4.3\text{-d}} Ru$$

are correct, then, after a time sufficient for the virtually complete decay of the ten-hour palladium, further rhodium removals should -- and as has been seen, did-- result only in the presence of the 20-hour activity. A parent-daughter growth relationship on repurified palladium was obtained by following the growth of the 20-hour activity. Its parent was found to have a half-life of four days. This is shown in Fig. I-4. It will be noted that the difference between the extrapolated decay curve of the equilibrium mixture and the experimental points gives a decay curve characteristic of the daughter. Fig. I-5 extends these decay curves, includes the growth-decay curve also for the case in which no absorber was used in the counting, and shows the eventual "tailing" into the 17-day K-capture Pd^{103}. Because of the low abundance of the latter, its daughter Rh^{103*} was never detected in rhodium daughter separations.

An absorption curve of the 4-day palladium was taken immediately after separation of the one-day daughter. This is shown in Fig. I-6. No electrons could be detected. This phenomenon explains the reason that the upper curves in Fig. I-5 show a larger growth than the lower curves, for, in the case of the one-day rhodium, the beta-ray spectrometer measurements indicate the presence of both positrons and conversion electrons (Figs. I-8 and I-9).

An absorption curve for the 20-hour rhodium is shown in Fig. I-7.

Mass Assignments

The two decay chains are

$$_{46}Pd \xrightarrow{4\ d.\ K} {}_{45}Rh \xrightarrow[\beta^+, K]{20\ hr.} {}_{44}Ru \qquad \text{and}$$

$$_{46}Pd \xrightarrow[K, \beta^+]{10\ hr.} {}_{45}Rh \xrightarrow[K,\ e^-, \gamma]{4.3\ d.} {}_{44}Ru \ .$$

Table I-2 shows the present status of isotopes in this region.

Table I-2

	96	97	98	99	100	101	102	103	104	105
Ru	S		S	S	S	S	S	42 d ↓	S	↓
Rh								S	↓	↓
Pd							S	17 d. K ↑	S	S

A decay chain representing isobars of even mass number would be the only type likely to lead to a situation in which an "even-even" parent would give rise to a shorter-lived "odd-odd" daughter. The most reasonable possibility for the first chain shown above is, therefore, mass number 100. Mass number 98 can be excluded on the grounds that it would probably not have been produced with 50 Mev deuterons and that Rh^{98} would likely have a much shorter half-life.

For reasons analogous to those mentioned above, it appears that the second chain is probably of odd mass number. The two immediate possibilities are 99 and 101. Although 99 cannot be definitely excluded, it seems unlikely that it would be formed at these energies. Further if one does assign this to mass number 99, then it must still be explained why another activity which could be assigned to 101 was not found; since Pd^{103} has a half-life of 17 days, Pd^{101} probably has a shorter period and hence would be detectable. Through this reasoning, the assignment was made as mass 101 for the second chain.

Three palladium activities were thus produced in the irradiation of rhodium with 50 Mev deuterons. The cross sections, in barns, for these processes are given below in Table I-3.

Table I-3

CROSS SECTION FOR FORMATION OF Pd ISOTOPES FROM Rh + 50 Mev DEUTERONS

Pd Isotope	Reaction	σ
103	d,2n	0.0012
101	d,4n	0.12
100	d,5n	0.14

Information concerning the radiation characteristics of these activities is summarized in Table I-4. The notation conforms to that of Seaborg[13].

Table I-4

Isotope	Half-Life	Type of Radiation	Energy of Radiation, Mev	
			Particles	γ-Rays
$_{46}Pd^{101}$	8 hrs. - Pd decay 10 hrs. decr. yld Rh^{101} 10 hrs. β^+ peak decay	β^+ (10%) K (90%)	(+) 2.5 β Spect	-
$_{46}Pd^{100}$	4.0d. - Pd equilib. decay (av. 4 vals)	K,γ	None	0.09(abs.Ag) 1.8(?)(abs Pb)
$_{45}Rh^{101}$	4.28 d.(av. 15 vals)	K,e^-,γ	0.21(e^-)β spect. 0.3(e^-)abs. Be	0.35(abs.Pb)
$_{45}Rh^{100}$	19.4 hrs.(av.24 vals)	β^+(5%) K,e^-,γ (95%)	3.0(β^+) spect. 0.6(e^-) spect.	1.2(abs.Pb)

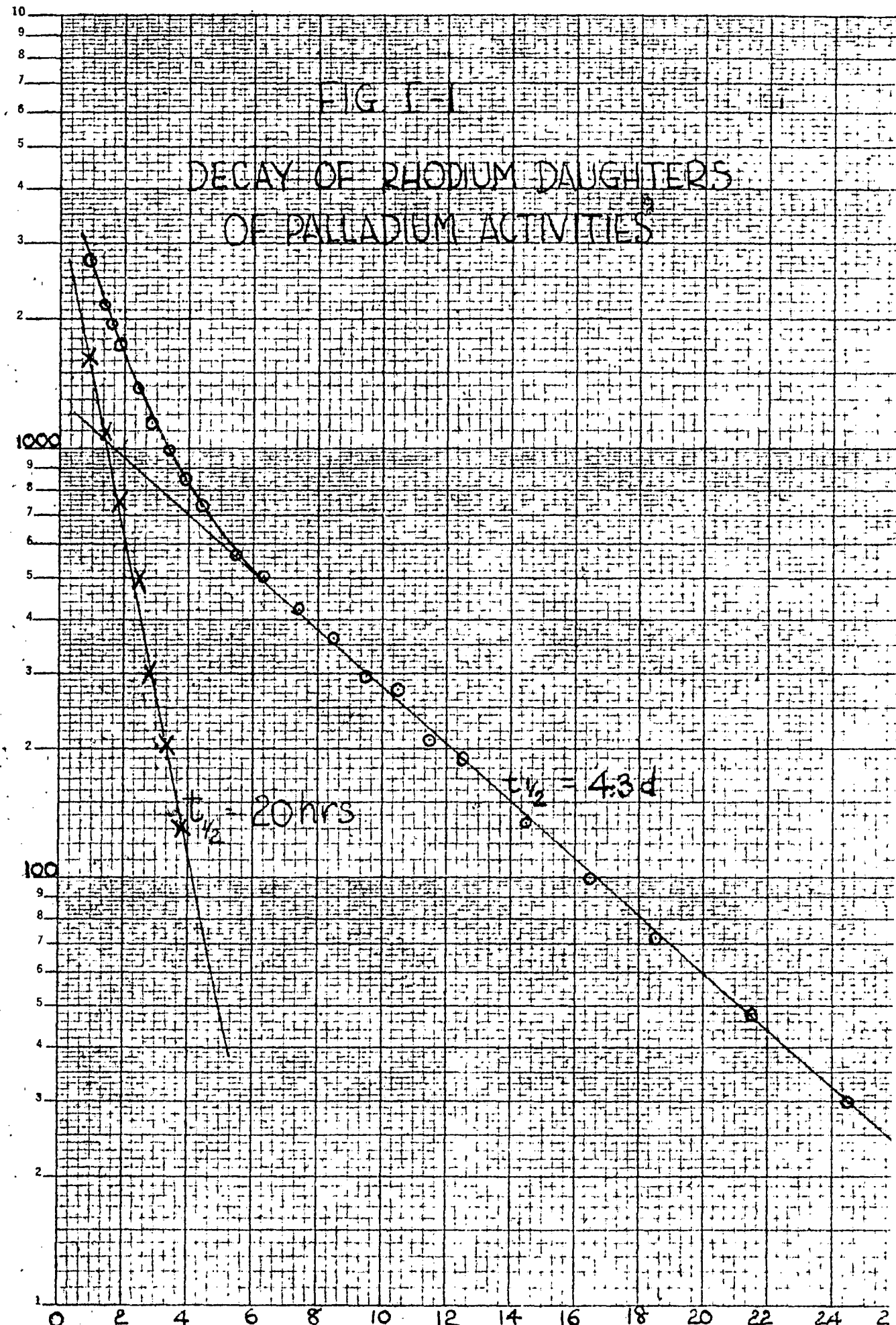


FIG I-1
DECAY OF RHODIUM DAUGHTERS
OF PALLADIUM ACTIVITIES
$t_{1/2}$ = 20 hrs
$t_{1/2}$ = 4.3 d
1000
100
0
2
4
6
8
10
12
14
16
18
20
22
24

FIGURE I-2

ABSORPTION CURVE FOR 43-DAY RHODIUM

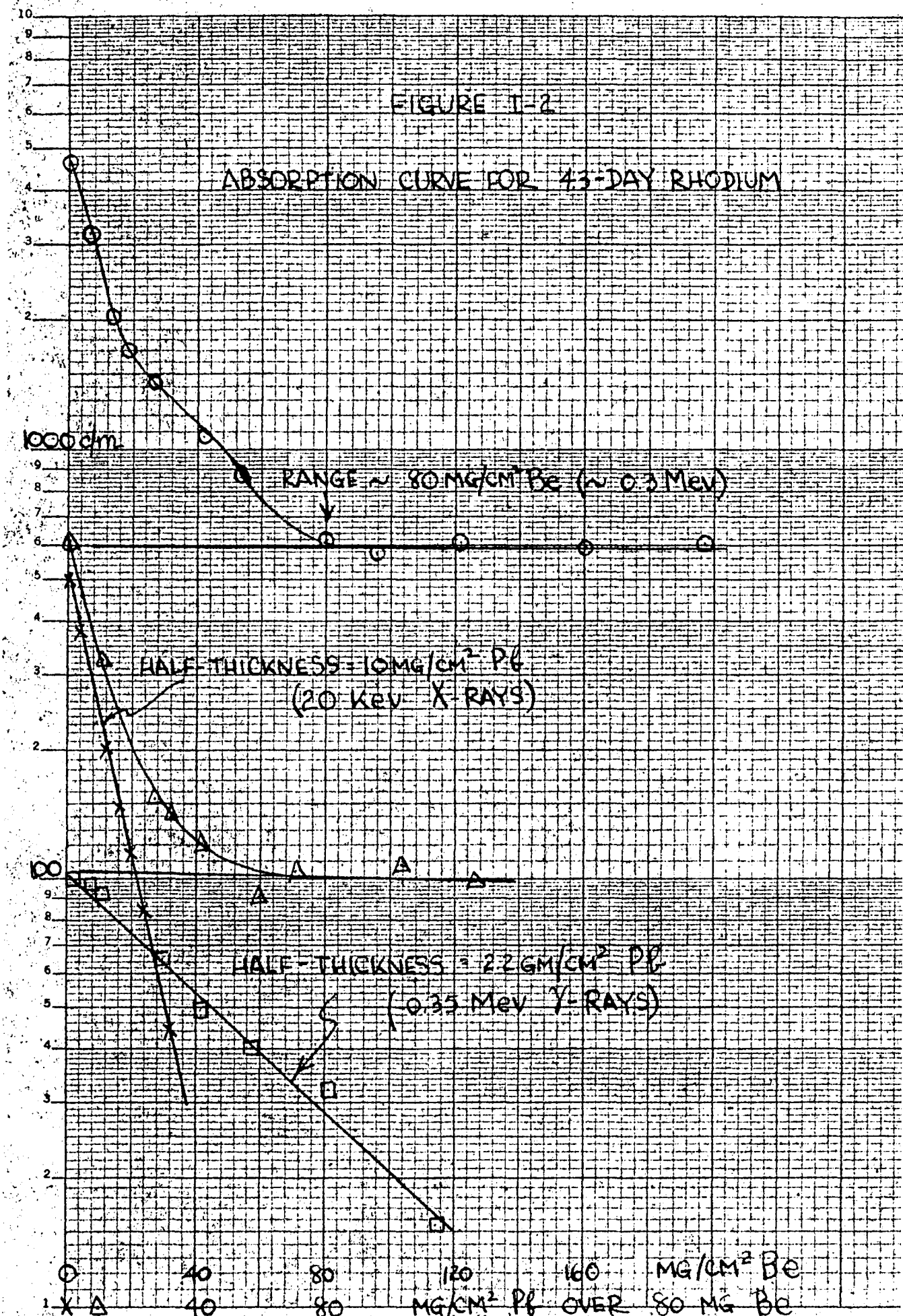


FIGURE I-3

CONVERSION ELECTRON OF 43-DAY RHODIUM

RECORDED ON BETA-RAY SPECTROMETER

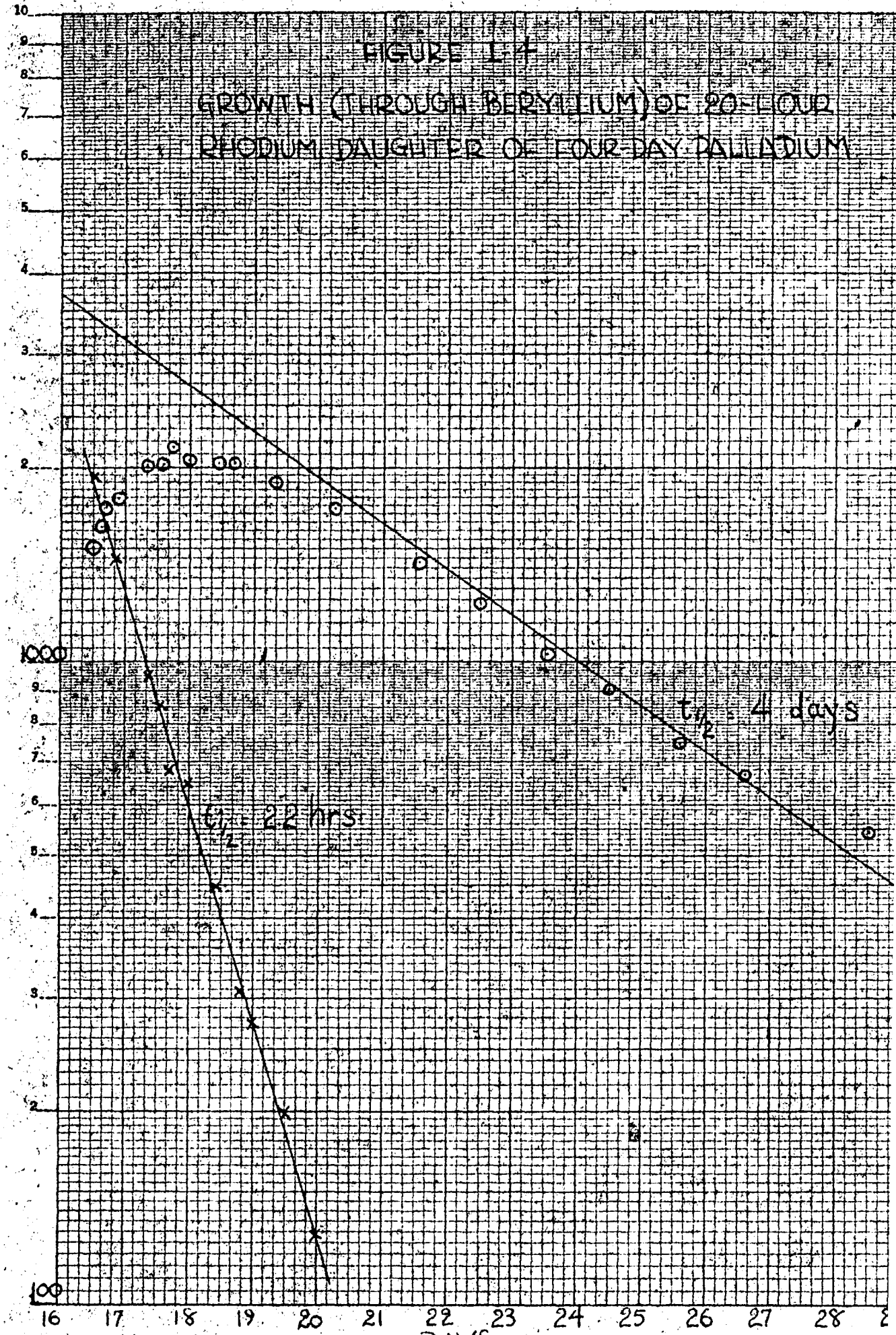


FIGURE 1-4
GROWTH (THROUGH BERYLLIUM) OF 20-HOUR RHODIUM DAUGHTER OF FOUR-DAY PALLADIUM
$t_{1/2}$ = 4 days
$t_{1/2}$ = 22 hrs.
1000
100
16
17
18
19
20
21
22
23
24
25
26
27
28
DAYS

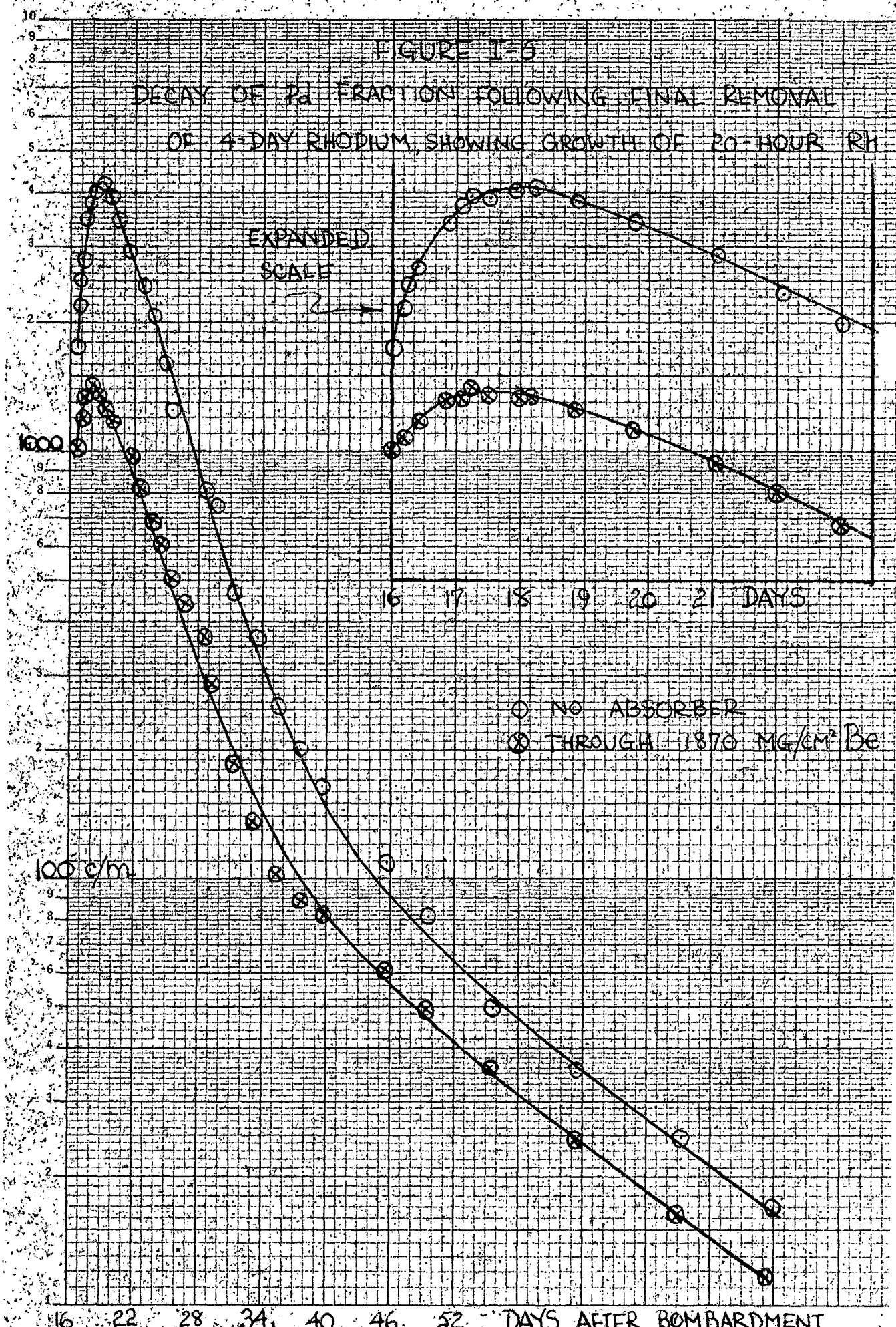


FIGURE I-6
DECAY OF Pd FRACTION FOLLOWING FINAL REMOVAL
OF 4-DAY RHODIUM, SHOWING GROWTH OF 20-HOUR Rh
EXPANDED
SCALE
2
1
1000
100 c/m
16 17 18 19 20 21 DAYS
NO ABSORBER
THROUGH 1870 MG/CM² Be
16 22 28 34 40 46 52 DAYS AFTER BOMBARDMENT

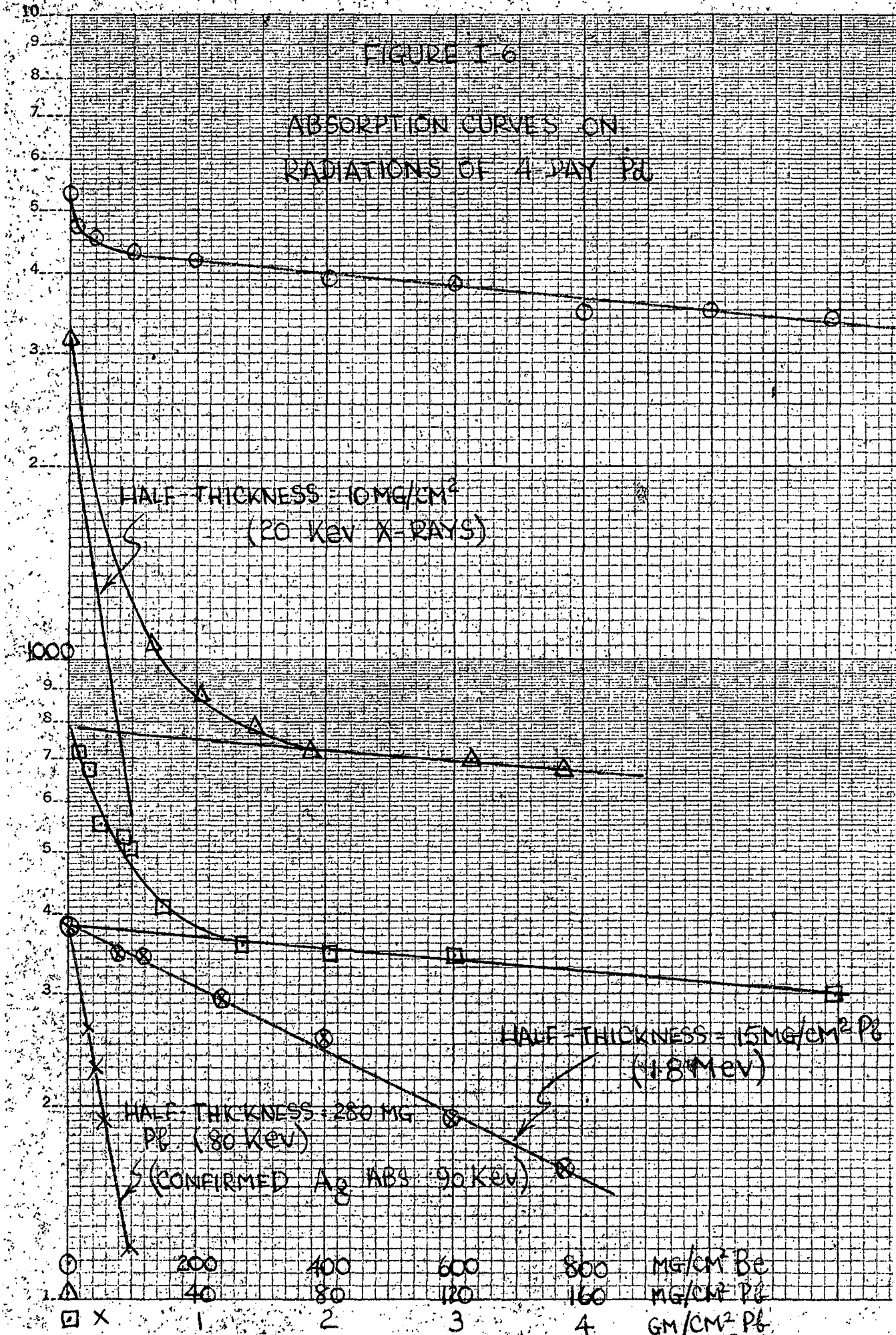


FIGURE I-6
ABSORPTION CURVES ON
RADIATIONS OF 4 DAY Pb
HALF-THICKNESS = 10 MG/CM²
(20 Kev X-RAYS)
HALF-THICKNESS = 15 MG/CM² Pb
(1.8 MeV)
HALF-THICKNESS = 280 MG
Pb (80 Kev)
(CONFIRMED Ag ABS 90 Kev)
200
400
600
800
MG/CM² Be
40
80
120
160
MG/CM² Pb
1
2
3
4
GM/CM² Pb

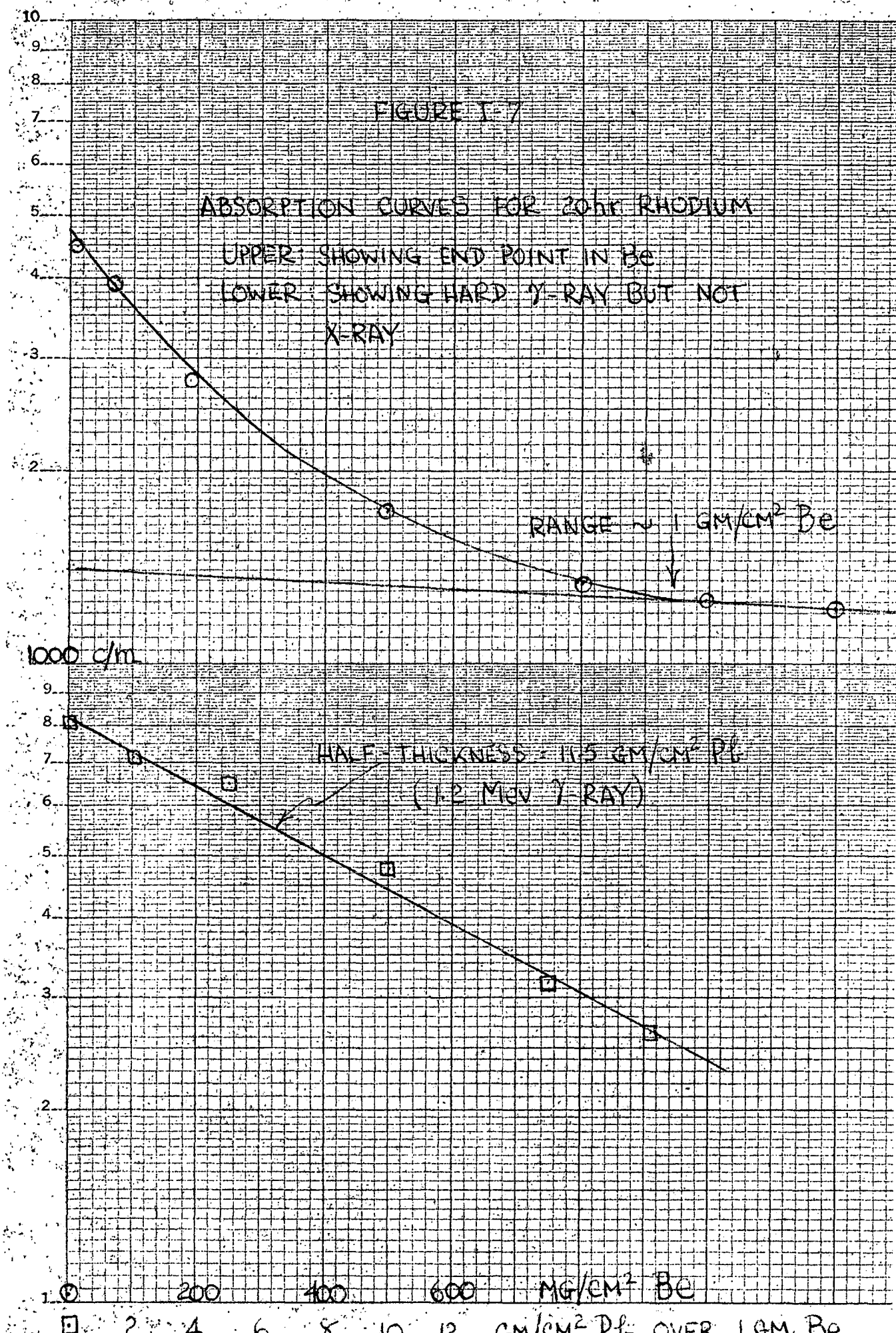


FIGURE I-7
ABSORPTION CURVES FOR 20hr RHODIUM
UPPER: SHOWING END POINT IN Be
LOWER: SHOWING HARD γ-RAY BUT NOT X-RAY
RANGE ~ 1 GM/CM² Be
1000 c/m
HALF-THICKNESS = 11.5 GM/CM² Pb
(1.2 MEV γ-RAY)
200
400
600
MG/CM² Be
2
4
6
8
10
12
GM/CM² Pb OVER 1 GM Be

FIGURE I-8

BETA-RAY SPECTROMETER RESULTS FOR ELECTRON ASSOCIATED WITH 20-HOUR RHODIUM

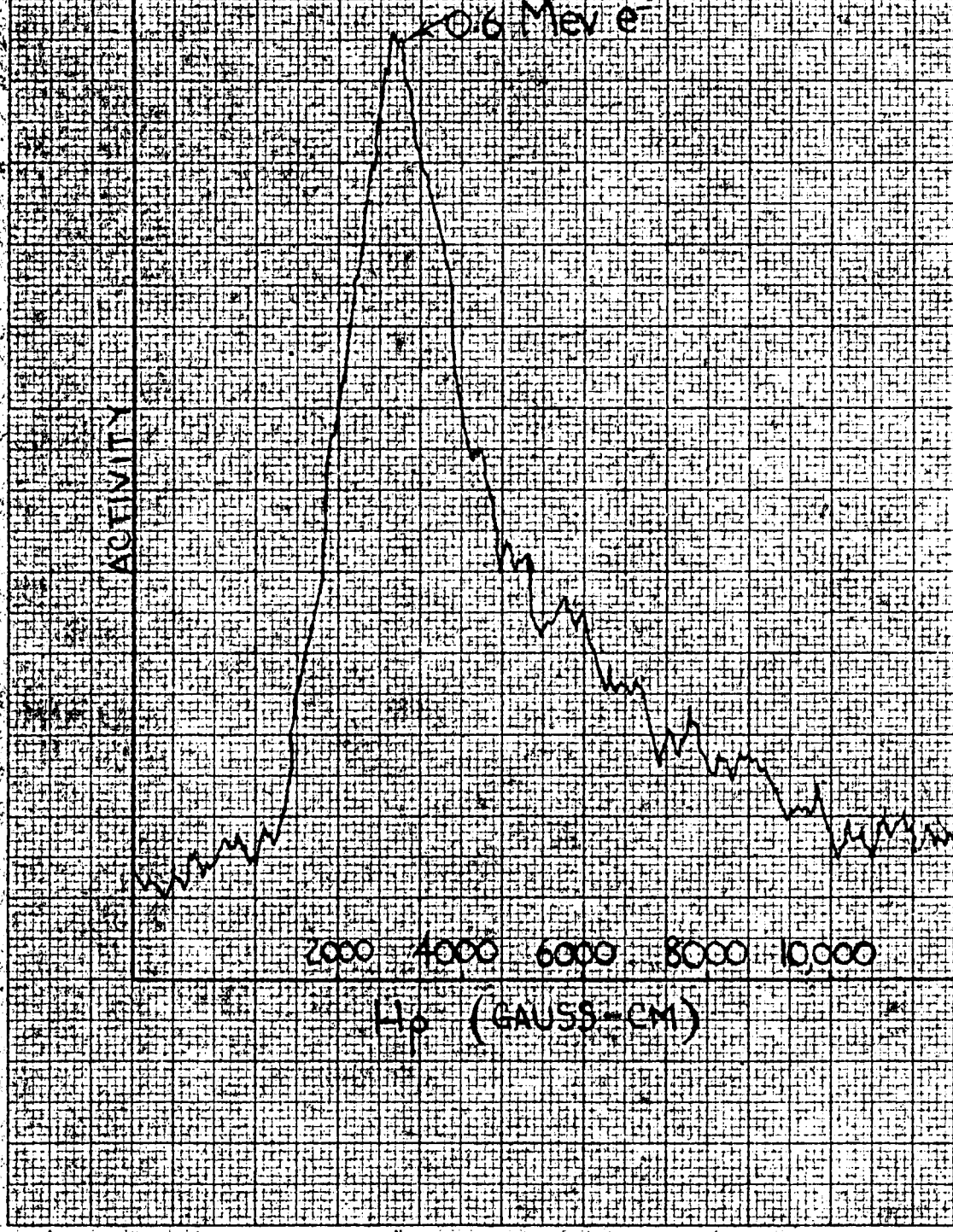
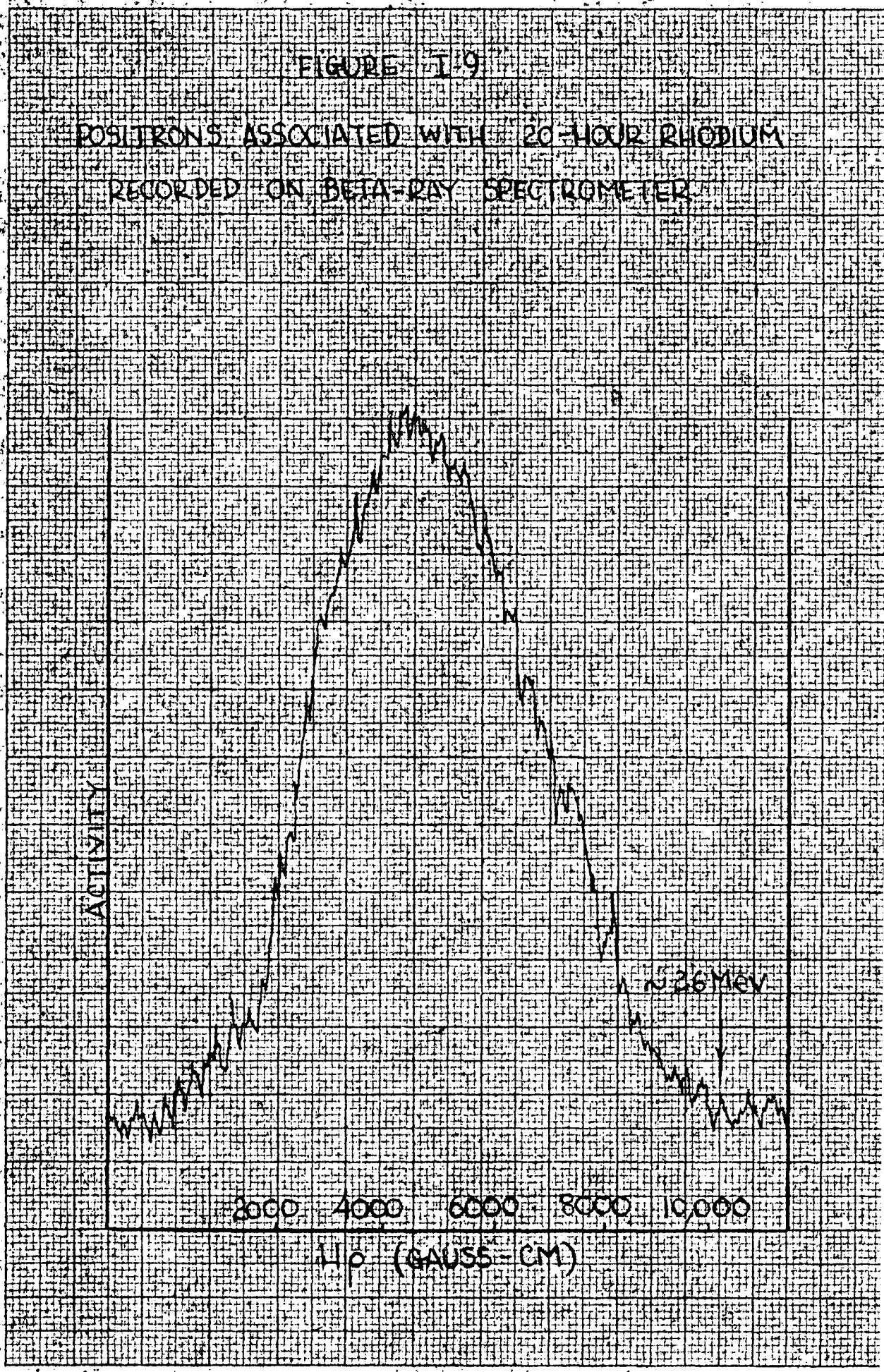


FIGURE I-9

POSITRONS ASSOCIATED WITH 20-HOUR RHODIUM

RECORDED ON BETA-RAY SPECTROMETER



Section II

PREVIOUSLY UNKNOWN ISOTOPES OF TELLURIUM AND THEIR ANTIMONY DAUGHTERS

Bombardment of antimony with 200 Mev deuterons has led to the production of a number of new activities, the elucidation of which we shall be concerned with in this section. A typical decay curve for the tellurium fraction obtained from such a bombardment is given in Fig. II-1. The presence of a 2.6-hour and a 6.0-day period is prominent.

A beryllium absorption curve for the 6.0-day period is shown in Fig. II-2, together with a Feather analysis[5] while Fig. II-3 illustrates the typical result of the $H\rho$ curves obtained for this activity. Evidently a positron of approximately 3 Mev energy accounts for nearly all of the activity of the six-day tellurium component, while the contribution from negative electrons is essentially negligible. It was found that this positron activity was due entirely to a 3.5-minute antimony daughter which could be removed from the tellurium in the following manner: SO_2 gas was passed through a hot solution 2 N in HCl containing the tellurium activity and several milligrams of Sb carrier until precipitation as Te metal was complete. The supernatant was boiled and H_2S passed through the solution. The resultant Sb_2S_3 was filtered and counted. In this manner, the total lapse of time from the separation to the time of counting seldom exceeded nine minutes. A typical decay curve for this daughter activity is shown in Fig. II-4. This period is probably identical to that reported by Risser, Lark-Horowitz and Smith[6] who obtained a three-minute period by bombarding indium with alpha particles. They reasoned that the most likely mass assignment was either 116 or 118.

The half-life of the parent of this 3.5-minute activity was corroborated by measurement of the decrease in yield of the 3.5-minute antimony activity obtained in a separation of tellurium and its antimony daughters. This is shown in the

upper curve of Fig. II-7. Not only does the yield of the 3.5-minute antimony decrease with a six-day half-life, but within experimental error, virtually all of the six-day tellurium activity is accounted for by this antimony daughter. It is therefore evident that the six-day tellurium parent must decay by electron capture. Whether the latter was accompanied by gamma emission and some subsequent conversion was not determined.

Because of the rapidity with which the separation of the 3.5-minute antimony daughter had to be made from the tellurium parent, repurification of the antimony was not feasible and there always occurred a "tailing" due to tellurium impurities remaining in the antimony supernatant. In general, to increase accuracy in determination of the amount of 3.5-minute activity associated with the tellurium, three to five Te-Sb separations were made on the same Te sample within several hours so that an average yield could be taken. Invariably, however, the background "tail" obtained in the first antimony sample removed from the tellurium was decidedly higher than that in subsequent antimony samples removed within several hours. Further investigation of this abnormally high background showed that the counting rate was only slightly affected by nearly two gm/cm^2 of beryllium absorber, but addition of 130 mg/cm^2 of lead absorbs virtually all of the activity. This is shown in Table II-1 below:

Table II-1

CHARACTERISTICS OF HIGH BACKGROUND RADIATION REMOVED FROM TELLURIUM WITH THE 3.5-MINUTE ANTIMONY ACTIVITY

Absorber, mg/cm^2	c/m
-----	5290
1870 Be	3740
130 Pb	10

Evidently then, this activity must be another antimony daughter formed from tellurium, consisting entirely of x-rays, no hard gamma radiation, and no electrons detectable with the apparatus used. Separation and repurification of antimony daughters from a very large tellurium sample provided sufficient activity to follow closely the decay of the period, and also to run a critical absorption curve of the x-radiation in the antimony sample.

The period of this antimony activity proved to be 39 hours, and is shown in Fig. II-5. The upper line shows that the use of 1870 mg/cm^2 of beryllium has only slight effect upon the counting rate. This activity is undoubtedly that reported by Coleman and Pool[7] who detected the period and assigned to it the mass number 119.

Table II-2 shows the energy of the K-x-rays of antimony and tin, while Table II-3 shows the "K-edge" for the elements cadmium, silver, and palladium.

Table II-2

ENERGY, IN Kev, OF THE K-x-rays of Sb AND Sn

	K_α	K_β	K_γ
Sb	26.2	-	30.4
Sn	25.1	28.5	29.2

Table II-3

K-EDGES, IN Kev, OF Cd, Ag, AND Pd

Element	E (Kev)
Cd	26.7
Ag	25.6
Pd	24.4

If the 39-hour period is due to an isomeric transition, the K-x-rays should be those of antimony, and will hence be critically absorbed by silver and palladium, but not by cadmium. If, on the other hand, the decay process is one of K-electron capture, then the emitted x-rays should be those of tin, and hence should be critically absorbed by palladium but not by cadmium or silver. The experimental critical absorption measurements leave no doubt but that the latter is the case and that the mode of decay is indeed K-electron capture. This critical absorption curve is shown in Fig. II-8.

Thus far, nothing has been said of the parent of the 39-hour period. It could not be observed directly in the decay of the tellurium fraction because the counting rate of this isobaric pair never represents more than several percent of the total counting rate of the tellurium fraction. Rough determination of the parent tellurium period by measuring the decrease in yield of the 39-hour antimony period indicated a half-life perhaps slightly less than that of the six-day-3.5-minute isobaric pair. Assuming then a half-life of 4.8 days, the theoretical equation for the total counting rate in a sample of hypothetically pure 4.8-day tellurium in which no 39-hour antimony daughter is originally present, is given:

$$\text{Counting rate} = k\,(0.0985\,e^{-0.144t} - 0.0596e^{-0.414t})$$

Graphically, this is represented in Fig. II-6. It will be noted that parent-daughter equilibrium is not essentially attained until about eight days after the zero of time. Thus, with this rough guide, the half-life of the tellurium parent of the 39-hour antimony was determined quite accurately in the following manner: A large fraction of the total tellurium initially separated from the antimony bombarded with 200 Mev deuterons was set aside for about 10 days. After this time identical aliquots were taken every several days, and the antimony activity removed from these aliquots. In this manner, volumetric errors were minimized and each aliquot was treated only once for the removal of the 39-hour daughter.

The results are illustrated by the lower curve of Fig. II-7. The half-life of the tellurium parent is thus 4.5 days, presumably decaying by electron capture.

Mass Assignments

The two decay chains may be represented by

(1) $Te \xrightarrow{6.0d\ K} Sb \xrightarrow{3.5\ min\ \beta^+} Sn$

(2) $Te \xrightarrow{4.5\ d\ K} Sb \xrightarrow{39\ hr.\ K} Sn$

Again, reference to the present configuration in the periodic table proves helpful (Table II-4).

Table II-4

	116	117	118	119	120	121	122	123
Sn	S	S	S	S	S		S	
Sb					↑	S	↓	S
Te					S	↑	S	S

The first isobaric pair must be less than 119 since Fisser, Clark-Horowitz, and Smith first prepared the antimony isobar by bombarding indium with alpha particles. Their assignment to mass number 116 or 118 is consistent with the assignment of the isobaric pair to mass 118. The half-lives of six days and 3.5 minutes suggest an "even-even" isotope decaying to an "odd-odd". Mass 116 is probably not correct since the removal from stability might be expected to lead to an antimony daughter of extremeley short half-life since even Sb^{120} has a period of only 17 minutes. This pair will thus be given the tentative assignment of mass 118.

Because of the similarity in half-lives, the 4.5-day-39-hour pair undoubtedly represents a transition from an "even-odd" to an "odd-even" isotope. Most likely possibilities are 119 and 117. But if the 6-day tellurium is 118, then it is not

likely that Te^{117} would have a period almost identical to 118. On the other hand, it is far more likely that Te^{119} (odd-even) and Te^{118} (even-even) have similar periods. The 4.5-day-39-hour pair were thus assigned to mass 119. This is in agreement with the 39-hour period assignment made by Coleman and Pool.

These assignments were corroborated by bombarding antimony with 40 Mev deuterons. Since the reactions are

$$Sb^{121}(d,5n)Te^{118} \qquad Sb^{121}(d,4n)Te^{119}$$
$$Sb^{123}(d,7n)Te^{118} \qquad Sb^{123}(d,6n)Te^{119}$$

it was reasoned that at lower energies the yield of Te^{119} relative to Te^{118} should oe enhanced. That this is the fact is seen from Table II-5 which lists the cross sections found for the formation of the isotopes Te^{118} and Te^{119} at 200 Mev and at 40 Mev.

Table II-5

CROSS SECTION FOR FORMATION OF Te^{118} AND Te^{119} FROM ANTIMONY WITH DEUTERONS

	200 Mev	40 Mev
Te^{119}	0.021	0.068
Te^{118}	0.017	0.0016

The experiment at 40 Mev made it possible to obtain an absorption curve for the electromagnetic radiation from Te^{119} after the 3.5-minute positrons were absorbed out, for through beryllium absorber, the contribution from the mass 118 pair is negligible in comparison with that from the 119 pair. An absorption curve is given in Fig. II-9. It is seen to consist of characteristic x-rays and a gamma ray of 1.6 Mev.

Typical results of the beta-ray spectrometer measurements of a tellurium fraction at 40 Mev are given in Fig. II-10. The positrons are those of the 6.0-day Te^{118}. The conversion electrons must belong to Te^{119} since these could not be

detected in bombardments at 200 Mev (cf. Fig. II-3), and their presence in bombardments at 40 Mev is correlated with the relatively large amount of Te^{119} known to be present by the presence of its Sb daughter. Furthermore, the area of this conversion electron peak appears to decrease with time with a period nearly like that of the 6.0-day positron. The presence of two conversion electrons is seen to be a possibility. The tailing of the conversion lines into what appears to be a high-energy β^- particle is probably due to secondary Compton electrons scattered into the counting chamber by the highly abundant 1.6 Mev gamma rays.

Table II-6 summarizes the properties of the two new tellurium isotopes and those of their antimony daughters.

Table II-6

Isotope	Half-life	Type of Radiation	Energy of Radiation, Mev	
			Particle	γ-rays
Te^{119}	4.5 d.	K, e^-, γ	0.2, 0.5	1.6
Te^{118}	6.0 d	K	-	-
Sb^{119}	39 hr	K	-	No γ
Sb^{118}	3.5 min	β^+	2.5	-

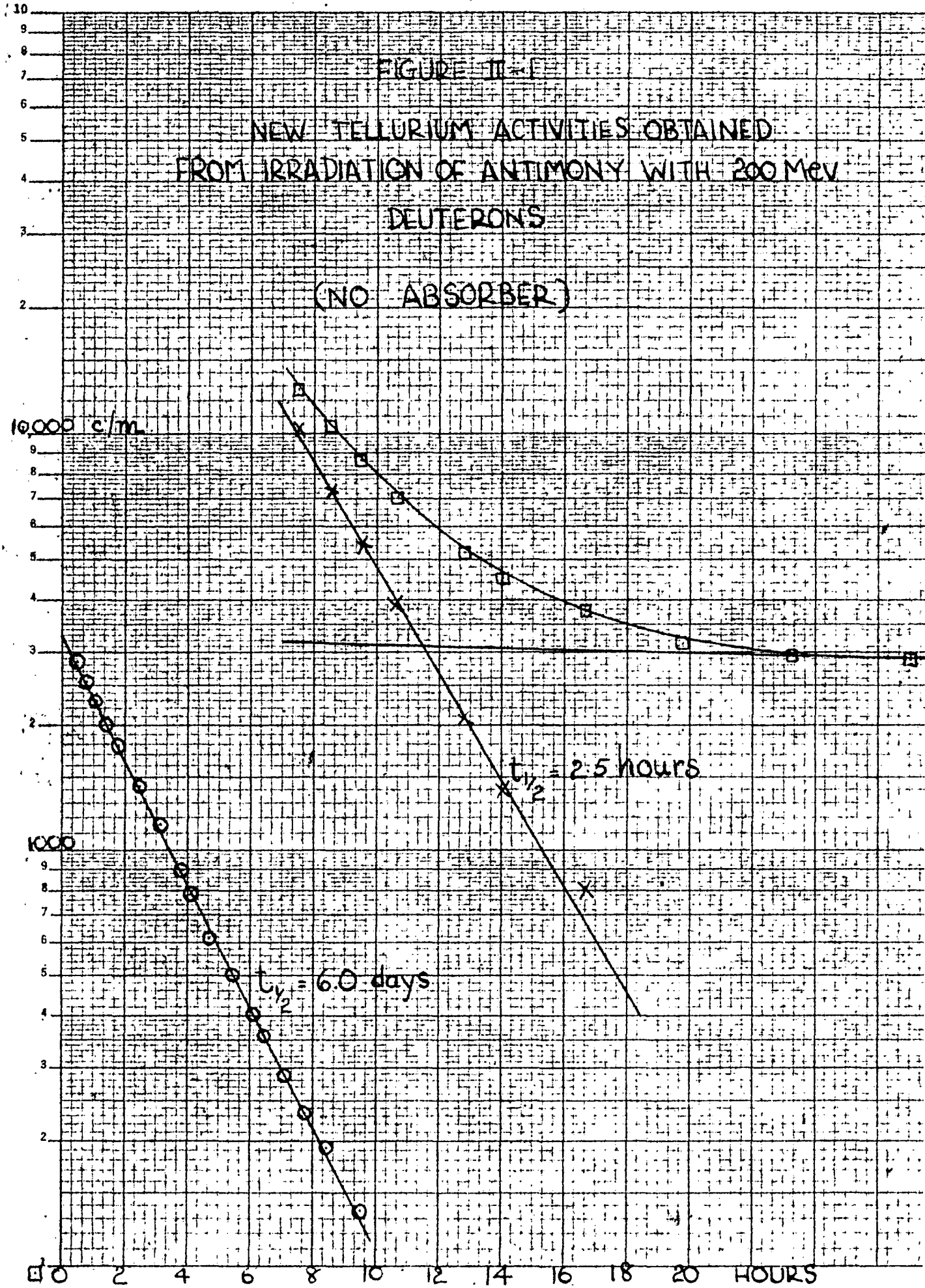


FIGURE III-1
NEW TELLURIUM ACTIVITIES OBTAINED FROM IRRADIATION OF ANTIMONY WITH 200 MeV DEUTERONS
(NO ABSORBER)
10,000 c/m
1000
$t_{1/2}$ = 2.5 hours
$t_{1/2}$ = 6.0 days
0 2 4 6 8 10 12 14 16 18 20 HOURS
0 6 12 18 24 30 DAYS

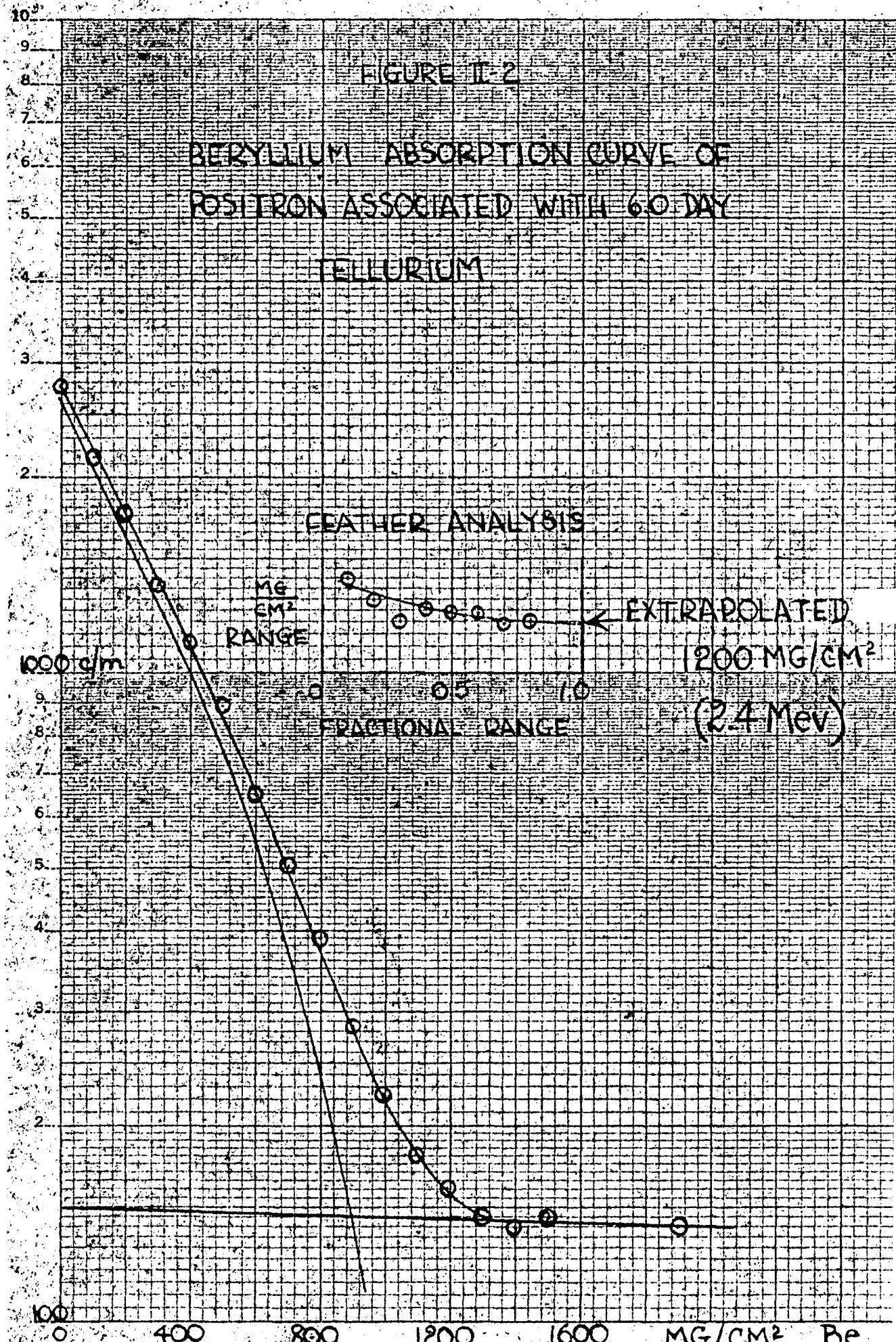


FIGURE II-2
BERYLLIUM ABSORPTION CURVE OF
POSITRON ASSOCIATED WITH 6.0 DAY
TELLURIUM
FEATHER ANALYSIS
MG/CM²
RANGE
EXTRAPOLATED
1200 MG/CM²
(2.4 MeV)
1000 c/m
0
0.5
1.0
FRACTIONAL RANGE
100
0
400
800
1200
1600
MG/CM² Be

FIGURE II-3 (a)

NEGATIVE PARTICLES ASSOCIATED WITH 6.0-DAY Te OBTAINED FROM 200 Mev DEUTERONS ON ANTIMONY.

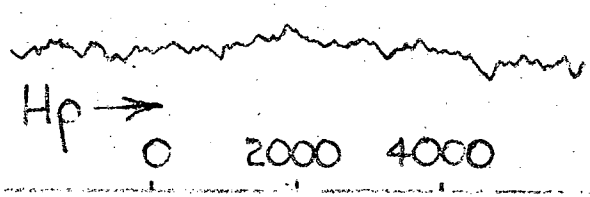


FIGURE II-3 (b)

POSITRONS ASSOCIATED WITH 6.0-DAY TELLURIUM (200 Mev D^+)

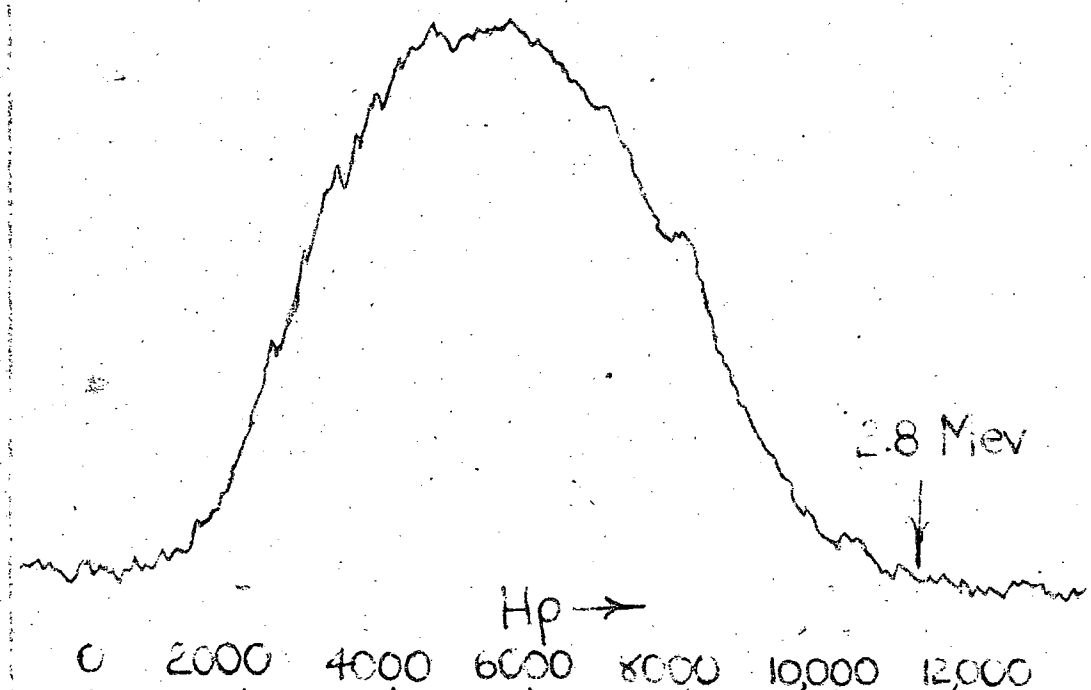


FIGURE II-4

DECAY OF ANTIMONY DAUGHTER OF 6.0 DAY TELLURIUM

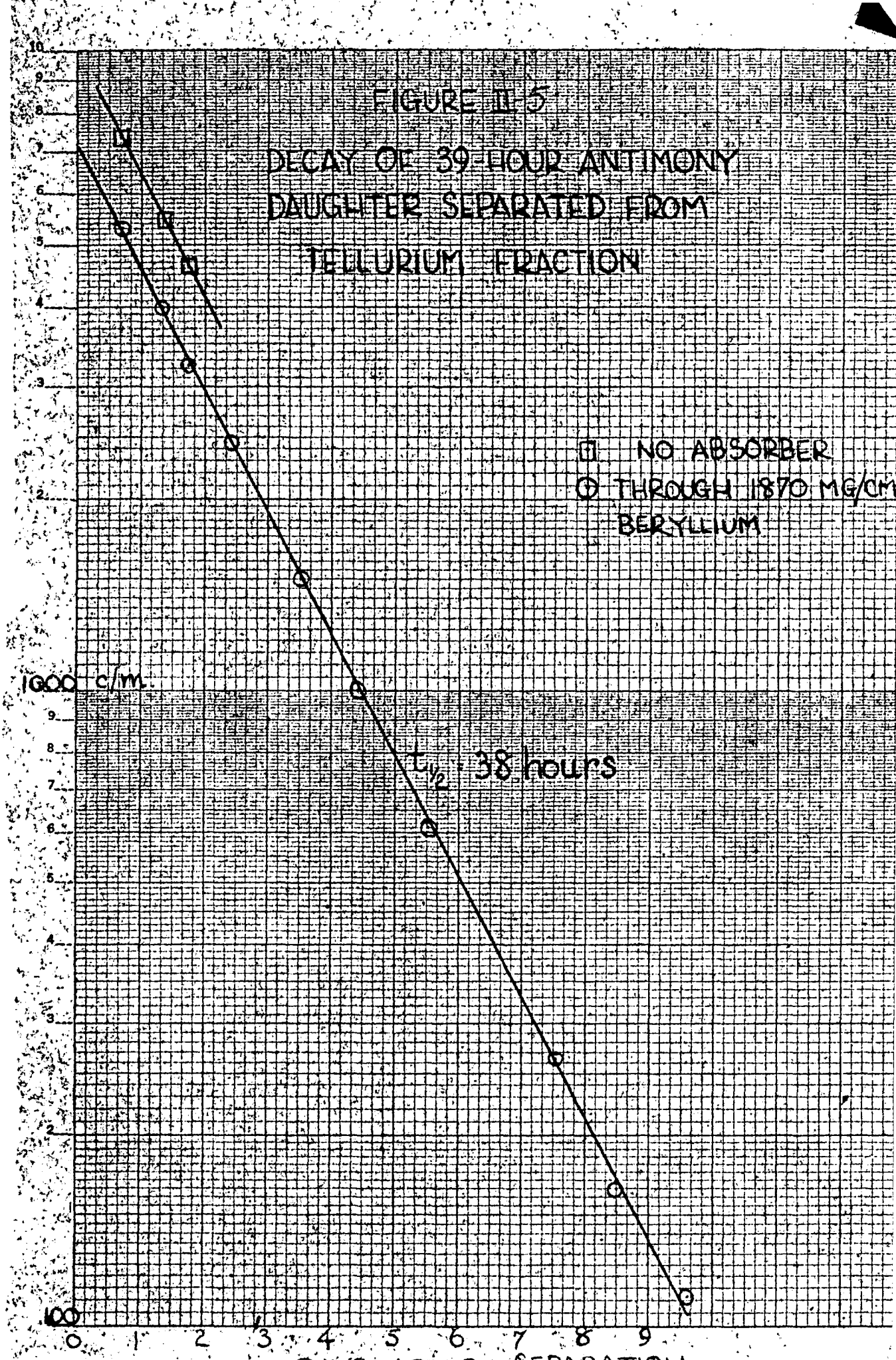


FIGURE II-5
DECAY OF 39-HOUR ANTIMONY DAUGHTER SEPARATED FROM TELLURIUM FRACTION
□ NO ABSORBER
○ THROUGH 1870 MG/CM
BERYLLIUM
1000 c/m
$t_{1/2}$ = 38 hours
100
0 1 2 3 4 5 6 7 8 9

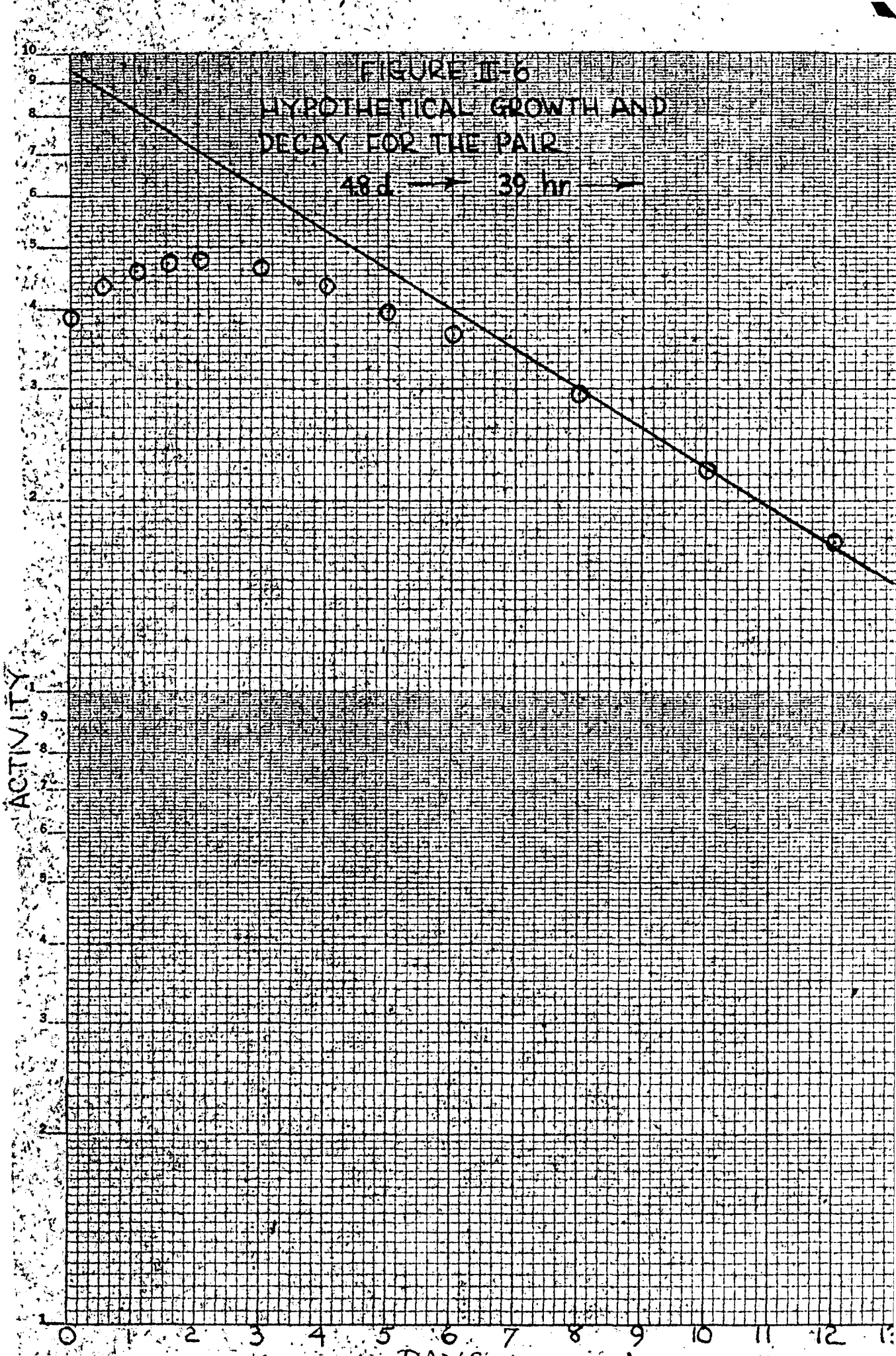


FIGURE III-6
HYPOTHETICAL GROWTH AND
DECAY FOR THE PAIR
4.8 d → 39 hr →
ACTIVITY
DAYS
0 1 2 3 4 5 6 7 8 9 10 11 12

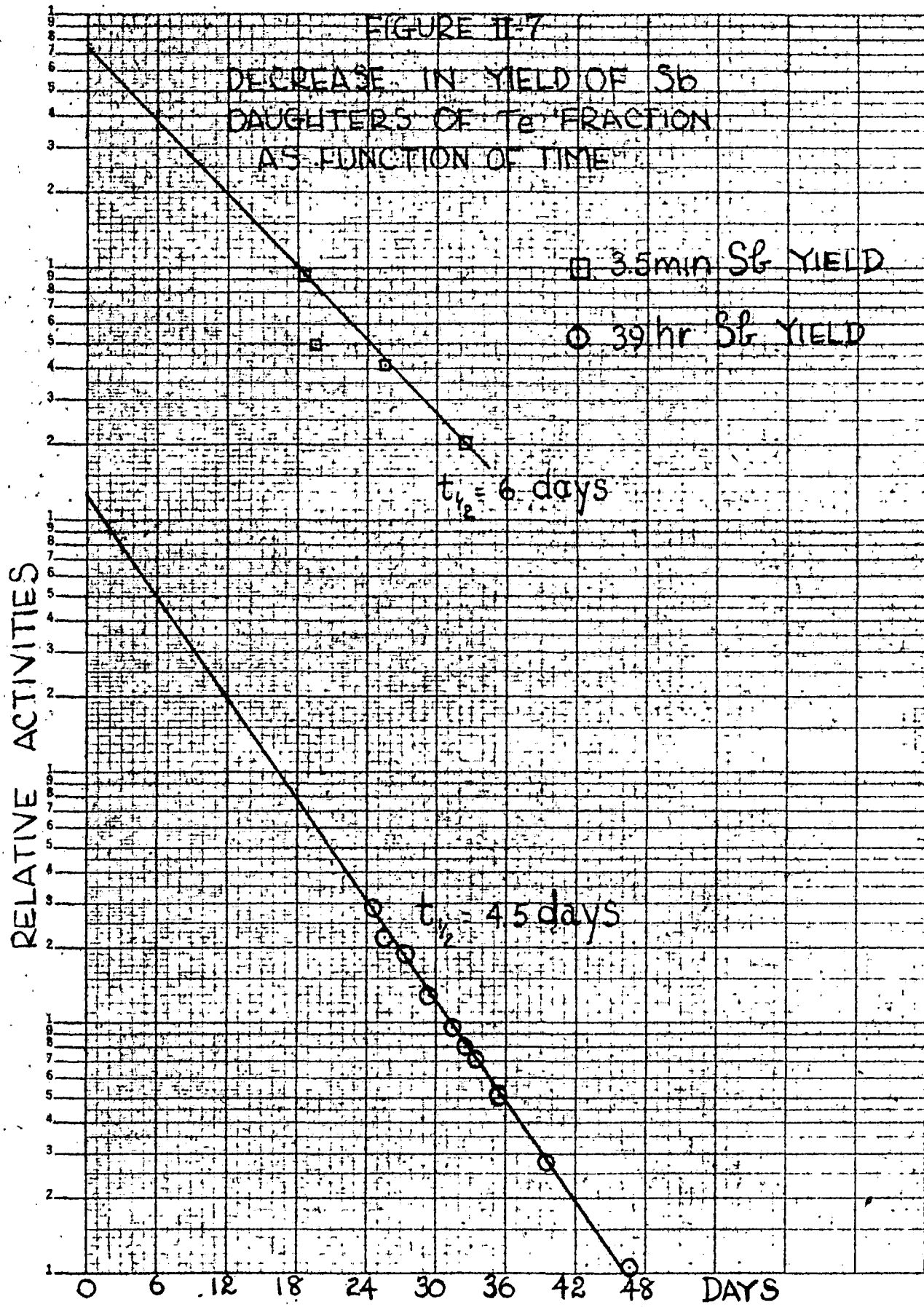


FIGURE II-7
DECREASE IN YIELD OF Sb DAUGHTERS OF Te FRACTION AS FUNCTION OF TIME
3.5min Sb YIELD
39 hr Sb YIELD
$t_{1/2}$ = 6 days
$t_{1/2}$ = 4.5 days
RELATIVE ACTIVITIES
0
6
12
18
24
30
36
42
48
DAYS

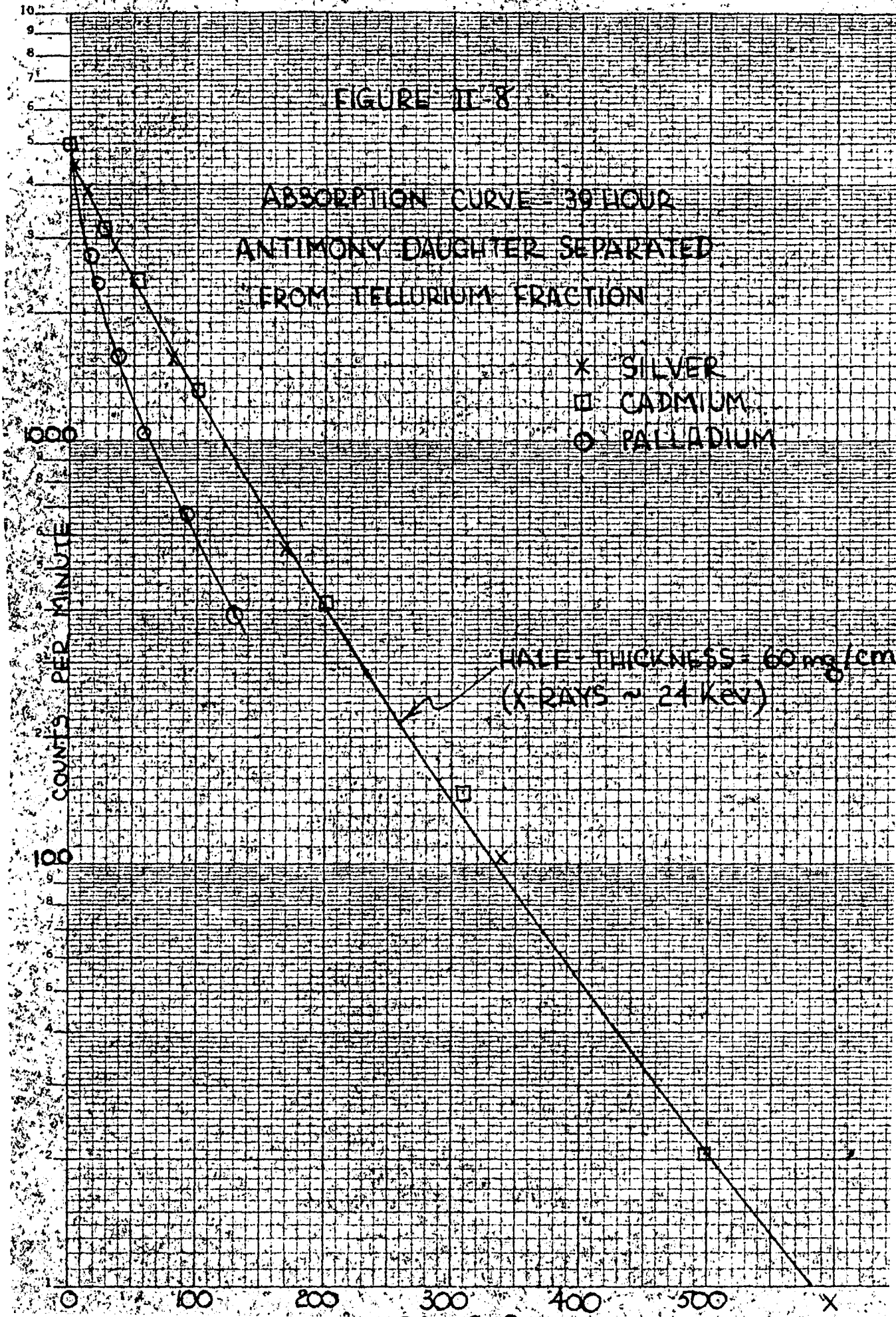


FIGURE II-8
ABSORPTION CURVE - 39 HOUR
ANTIMONY DAUGHTER SEPARATED
FROM TELLURIUM FRACTION
X SILVER
□ CADMIUM
○ PALLADIUM
HALF-THICKNESS = 60 mg/cm²
(X-RAYS ~ 24 Kev)
COUNTS PER MINUTE
1000
100
0
100
200
300
400
500
X

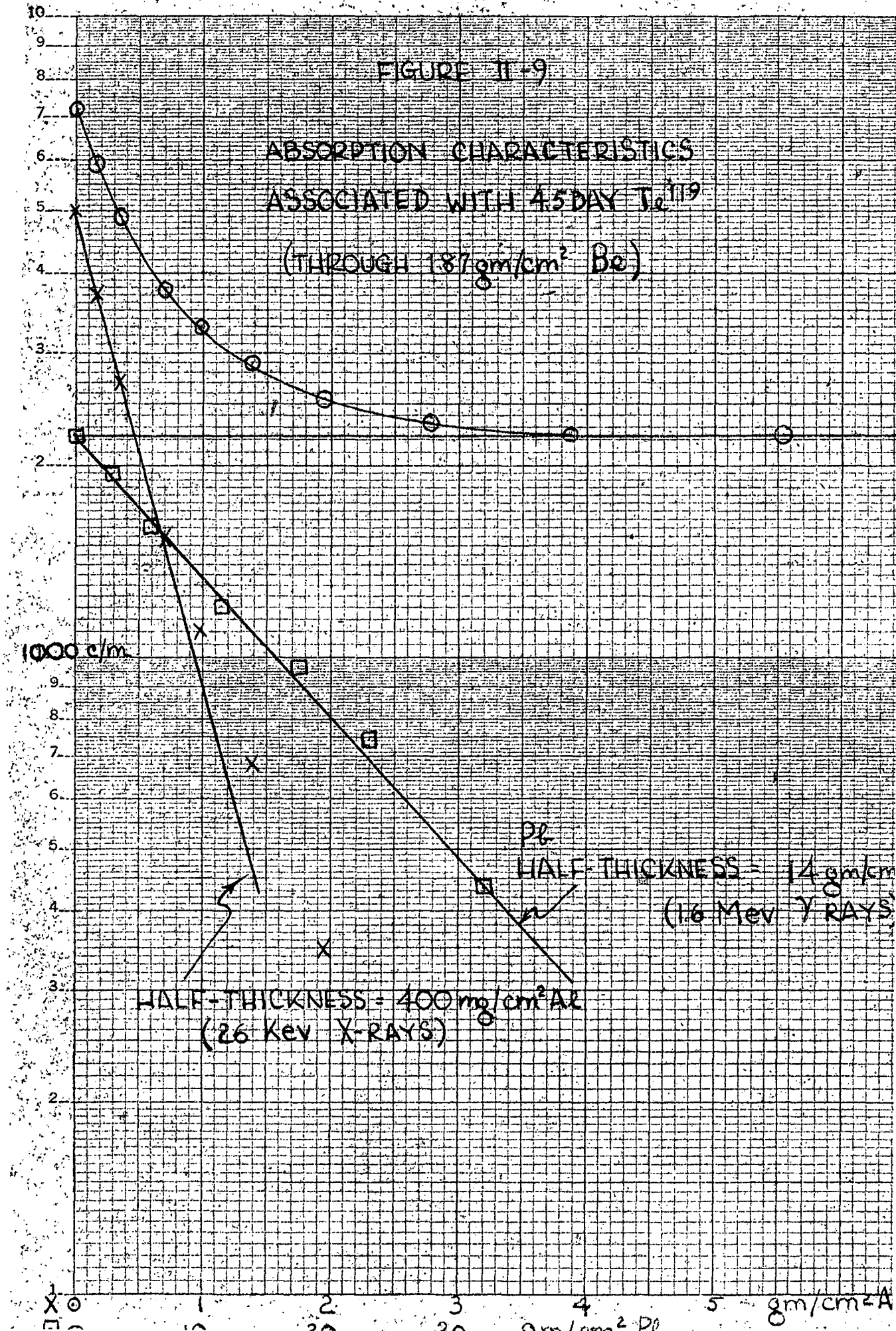


FIGURE II-9
ABSORPTION CHARACTERISTICS
ASSOCIATED WITH 45 DAY Te^{119}
(THROUGH 1.87 gm/cm² Be)
1000 c/m
Pb
HALF-THICKNESS = 14 gm/cm²
(1.6 Mev γ RAYS)
HALF-THICKNESS = 400 mg/cm² Al
(26 Kev X-RAYS)
gm/cm² Al
gm/cm² Pb

FIGURE II-10(a)

ELECTRONS ASSOCIATED WITH THE 45-DAY TELLURIUM FROM 40 Mev DEUTERONS ON ANTIMONY

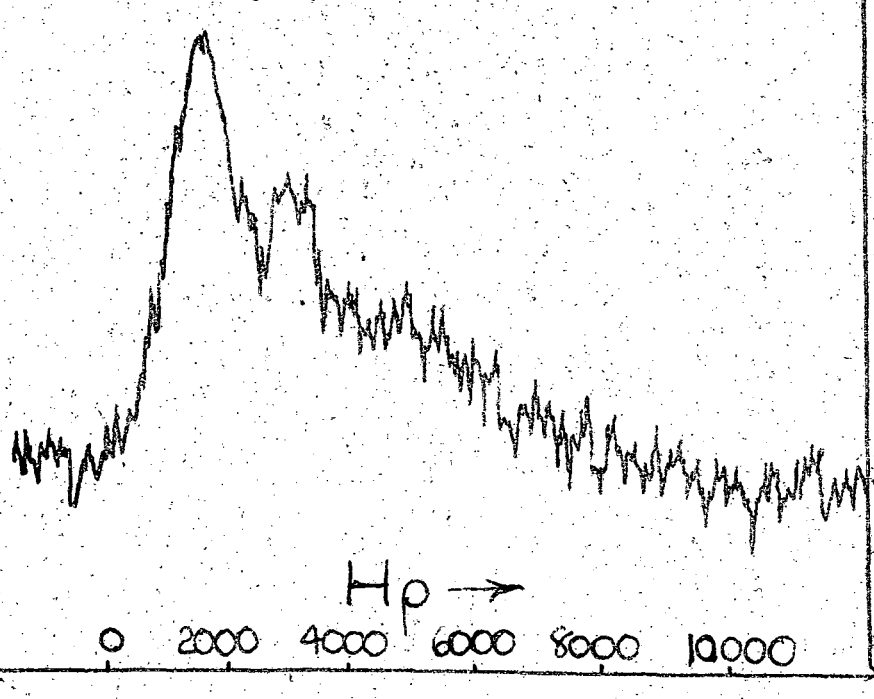
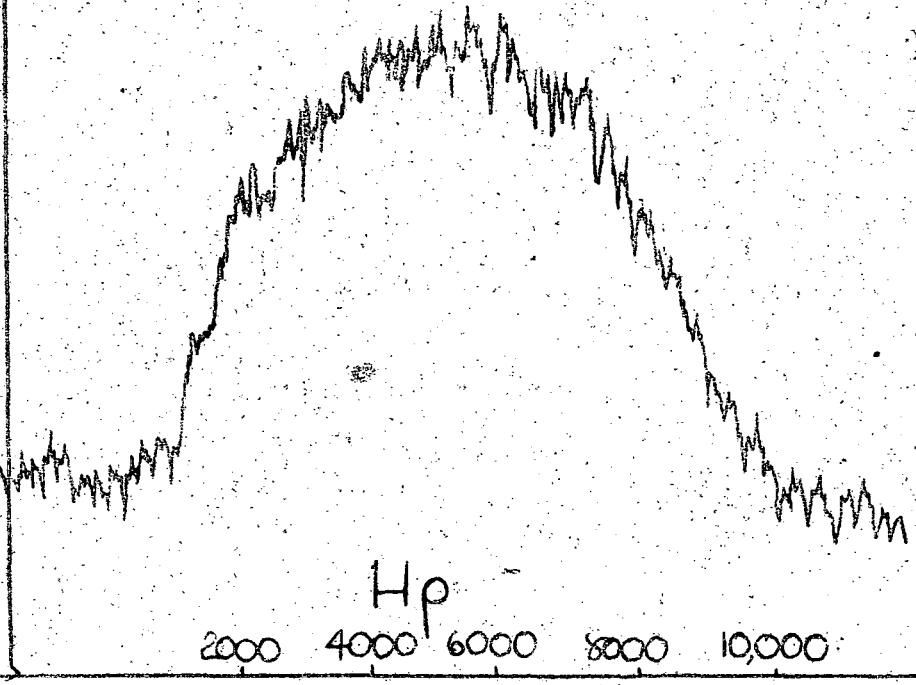


FIGURE II-10(b)

POSITRONS ASSOCIATED WITH THE 6.0-DAY TELLURIUM (40 Mev D+)



Section III

ISOTOPES OF ANTIMONY AND TIN PRODUCED BY BOMBARDMENT OF Sn^{120} WITH 19 Mev DEUTERONS

Among the several activities which were formed in the antimony fraction as a result of the irradiation of antimony metal with 200 Mev deuterons was a period of approximately six days which was prominent in the decay followed through 1.87 gm/cm^2 of beryllium. It was also detectable when no absorber was used but under the latter conditions the presence of the 2.8-day Sb^{122} β^- emitter nearly masked the six-day activity. It was nevertheless possible to show that the period in question consisted of x-rays, very hard gamma radiation, and some conversion electrons. Inspection of Table III-1 reveals that this activity is probably an isomer of an already known isotope. The most likely possibilities are:

a) An isomer of the 2.8-day β^- emitter Sb^{122}, but decaying by electron capture to Sn^{122}.

b) An isomer of the 17-minute β^+ emitter Sb^{120}.

Table III-1

	118	119	120	121	122	123	124
Sn	S	S	S		S		S
Sb	3.5 min β^+	39 hr K	17 min β^+	S	2.8 d β^-	S	
Te	6.0 d K	4.5 d K	S		S		S

In order to determine which, if either, of the above postulates be so, a sample of 95% isotopically pure Sn^{120} ‡ metal foil, obtained through the courtesy of the Atomic Energy Commission from Oak Ridge, Tennessee, was irradiated

‡ Isotopic analysis of this sample is as follows:

Isotope	% abund.	Isotope	% abund.	Isotope	% abund.
112	1.1	116	0.4 ± 0.1	119	1.3 ± 0.5
114	0.2	117	1.1 ± 0.5	120	95.4 ± 1.0
115	0.1	118	0.8 ± 0.1	122	0.2 ± 0.1
				124	$0.2 (+0.1) (-0.3)$

with 19 Mev deuterons in the Berkeley 60-inch cyclotron. Reactions expected would be:

$Sn^{120}(d,n)Sb^{121}$(stable)

$Sn^{120}(d,2n)\ Sb^{120} \xrightarrow{17\ \text{min}\ \beta^+}$

$Sn^{120}(d,3n)Sb^{119} \xrightarrow{39\ \text{hr. K}}$

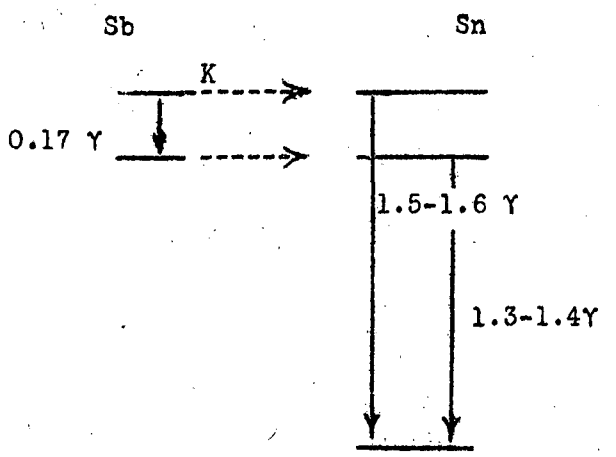
$Sn^{120}(d,p)Sn^{121} \xrightarrow{\beta^-} Sb^{121}$

It should thus be possible to detect the 39-hr K-capture activity due to Sb^{119} and the 17-minute β^+. If the six-day isomer should show up, it is probably an isomer of the 17-minute activity or of stable Sb^{121}. If it were an isomer of Sb^{122}, then it could not be reached by this bombardment. In addition, it should be possible to identify the isotope Sn^{121}, whose identity was not known.

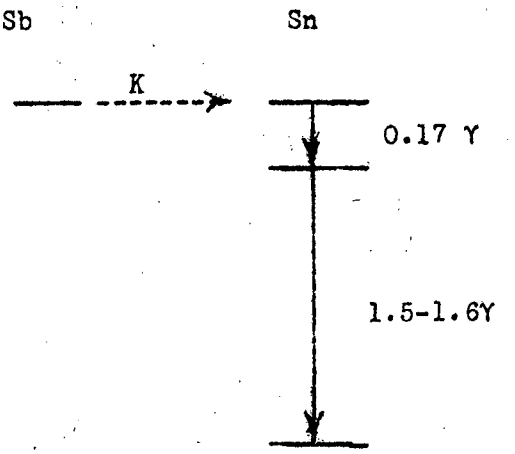
The chemical procedure employed consisted in dissolving the tin in hot concentrated HCl. A platinum wire was kept in contact with the foil to prevent polarization by occluded hydrogen gas. 20 mg of Sb carrier was added and the solution made to 20 ml of 2 N HCl. This was placed in a hot water bath and H_2S passed through the solution for not more than five minutes. The Sb_2S_3 was centrifuged. The supernatant was decanted, boiled to expel the H_2S, and several milligrams of antimony carrier added. H_2S was again passed through the solution and the entire procedure repeated. The final supernatant was then taken to be free of antimony activity. The acidity was reduced to 0.3 N and H_2S passed through, the Sn^{120} S being centrifuged out and dissolved in HCl.

The original Sb_2S_3 precipitate was dissolved in HCl, boiled to expel the H_2S, and diluted to 2 N in HCl. Several milligrams of ordinary Sn carrier were added and the solution placed in a hot water bath. Treatment with H_2S again precipitated the Sb_2S_3, essentially free of tin activities. The activities obtained in the antimony fraction were the 17-minute activity confirmed as positrons in the beta-ray spectrometer, a 5.7-day activity and the 39-hour x-ray emitter. The

decay curves are shown in Figs. III-1, III-2, and III-3. The 5.7-day activity had decay characteristics in agreement with those observed as the six-day antimony in bombardments of antimony with high-energy deuterons in that there is a high abundance of x-rays and very hard γ-rays, and some conversion electrons. The complete absorption curve is shown in Fig. III-4 while the beta-ray distribution curve is shown in Fig. III-9. The prominent radiations are a 1.5 Mev γ-ray, a similar abundance of x-rays characteristic of this region of the periodic table, and conversion electrons of roughly 150 kilovolts energy. The x-rays of this activity were shown to be those of tin by the use of the critical absorbers cadmium, silver and palladium. The critical absorption curve is shown in Fig. III-5. Because of the high gamma background, the results of this critical absorption are not as striking as those obtained for Sb^{119} described in Section II, but the conclusion is nonetheless the same. It is not altogether clear why the gamma ray corresponding to this conversion line was not readily observed. The sharp upsweep at the beginning of the lead absorption curve for the 1.5 Mev γ-ray could have a half-thickness of 150-300 mg/cm^2 Pb, a more exact value not being possible. This could correspond to a γ-ray of approximately the requisite energy, and thus account for the apparent anomaly. Consequently it is not possible with these data to know with any degree of certainty the exact decay mechanism occurring. To do this it would be necessary to employ a large spectrometer and coincidence methods for the gamma radiations. Nevertheless several possible decay mechanisms could be postulated. These are shown below:



SCHEME (a)



SCHEME (b)

In scheme (a), the two hard Y rays postulated would not be differentiated by absorption methods but rather would require careful measurement of secondary electrons.

Mass Assignment Referring again to Table III-1, almost by a process of elimination it is apparent that the most likely mass number of the 5.7 day antimony electron capture isotope is 120, for, as predicted, Sb^{119} showed up, as well as the 17 minute β^+ emitter Sb^{120}.

The tin fraction from the bombardment of Sn^{120} with 19 Mev deuterons yielded a single β^- activity of 28-hour half-life. The decay curve is shown in Fig. III-6. A beryllium absorption curve, illustrated in Fig. III-7, indicates that no gamma radiation is associated with this. The maximum energy thus obtained by a Feather analysis is 0.4 Mev, in approximate agreement with the value of 0.45 Mev obtained from the $H\rho$ value on the beta-ray spectrometer shown in Fig. III-8.

Since only one β^- activity could possibly have been produced by this bombardment, namely

$$Sn^{120}(d,p)Sn^{121}$$

the mass assignment of the 28-hour tin is obviously 121, and is probably the 26-hour tin activity reported by Livingood and Seaborg[8].

Cross sections. The cross-sections for the various radioactive species produced in this bombardment are given below in Table III-2. Because the half-life of the 17-minute period was short compared to the time of bombardment, it was necessary to take this into account in calculating the cross section.

Table III-2

CROSS-SECTION FOR FORMATION OF ISOTOPES FROM 19-MEV DEUTERONS ON Sn^{120}

Elem. & Mass	$T_{1/2}$	Produced by	(barns)
Sb^{120}	5.7 d K	$Sn^{120}(d,2n)$	0.14
Sb^{120}	17 min β^+	(d,2n)	0.11
Sb^{119}	39 hr K	(d,3n)	0.044
Sn^{121}		(d,p)	0.018

Table III-3 summarizes the decay characteristics of the two activities thus found:

Table III-3

Isotope	Half-life	Type of Radiation	Energy of Radiation, Mev	
			Particles	γ-rays
Sb^{120}	5.7 days	K,e^-,γ		1.5, 0.2
Sn^{121}	28 hours	β^-	0.4-0.45	no γ

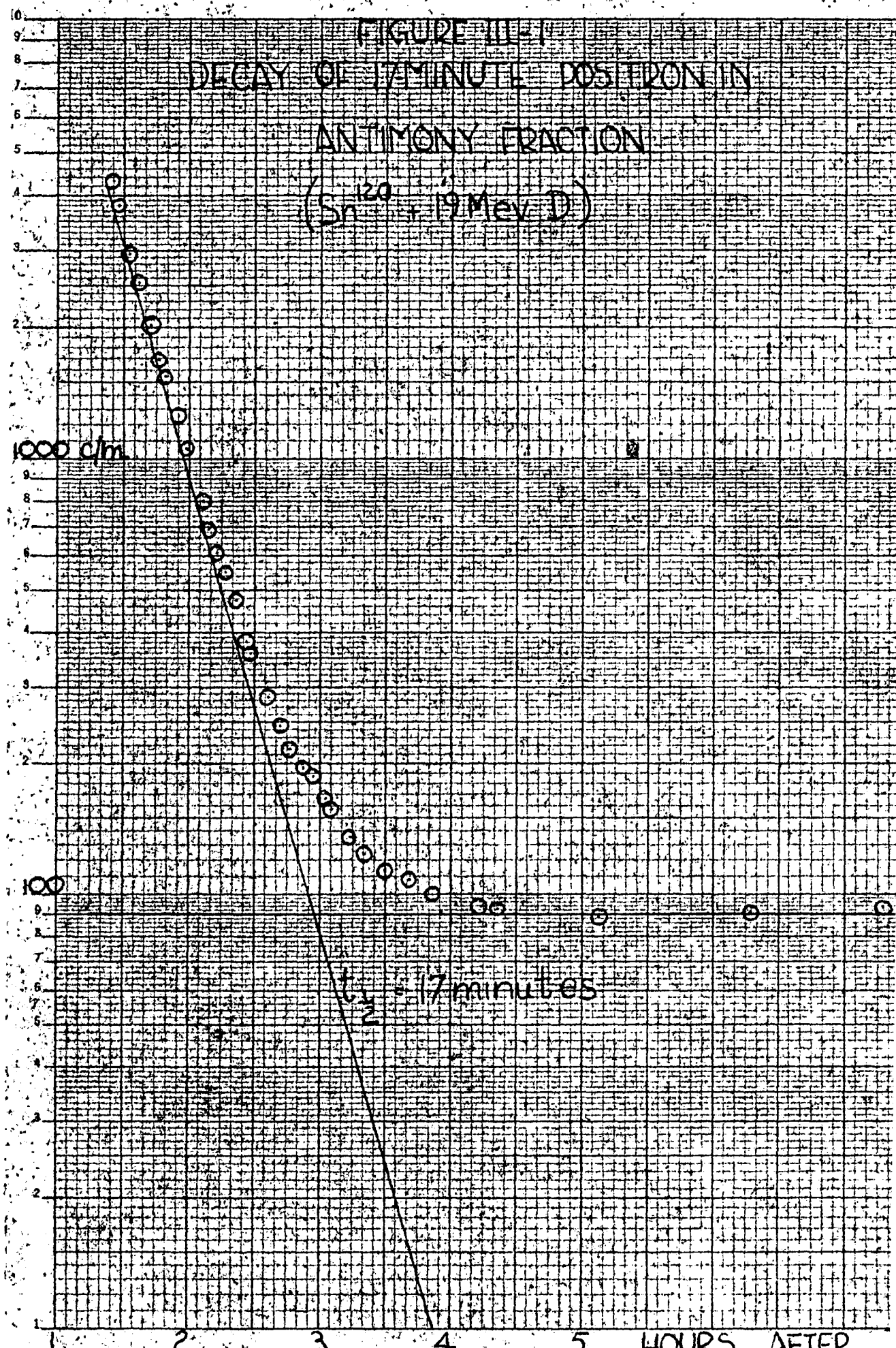


FIGURE III-1
DECAY OF 17 MINUTE POSITRON IN
ANTIMONY FRACTION
(Sn^{120} + 19 Mev D)
1000 c/m.
100
$t_{\frac{1}{2}}$ = 17 minutes
1
2
3
4
5
HOURS AFTER

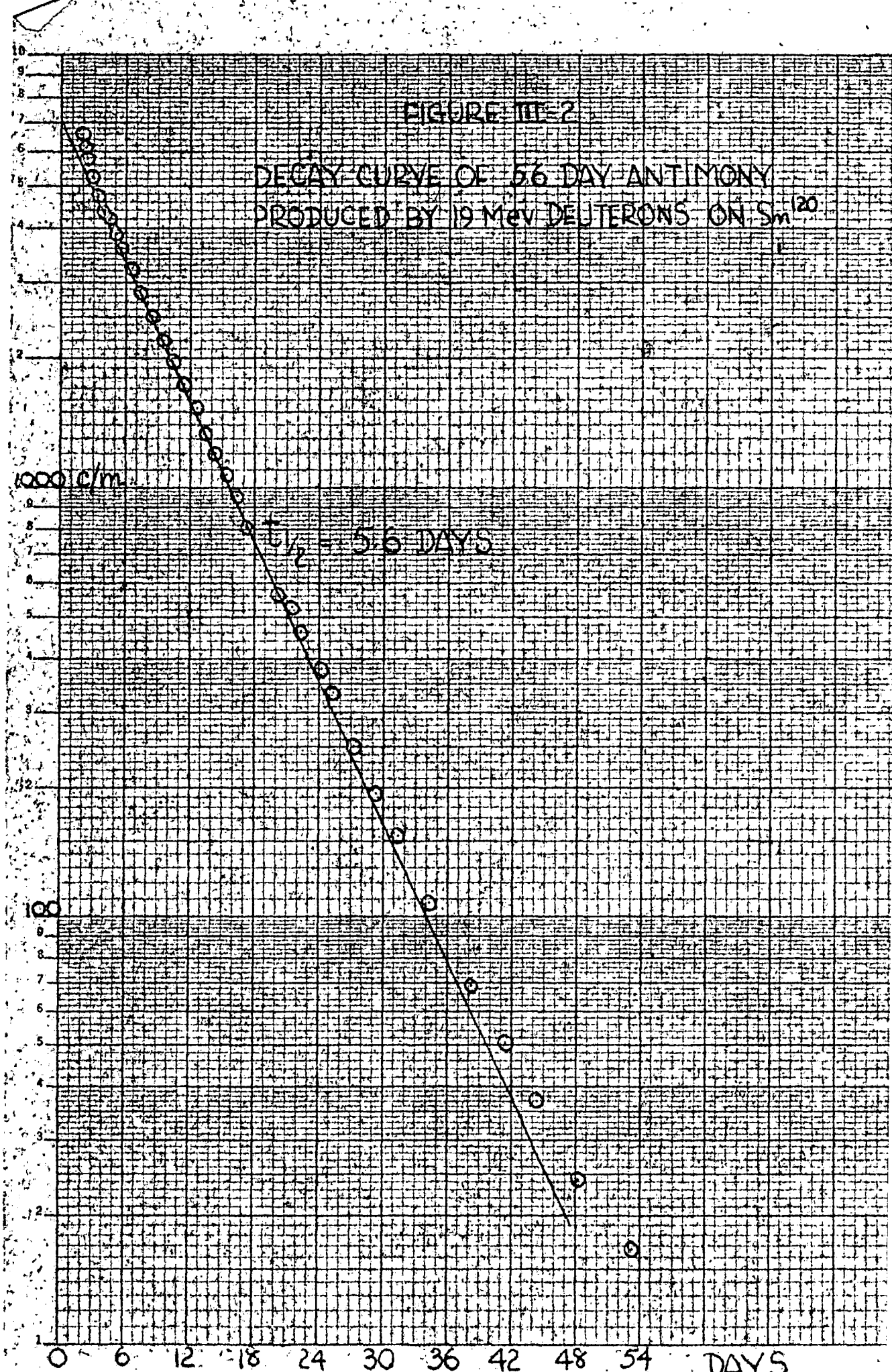


FIGURE III-2
DECAY CURVE OF 56 DAY ANTIMONY
PRODUCED BY 19 Mev DEUTERONS ON Sm^{120}
1000 c/m.
100
$t_{1/2}$ = 56 DAYS
0
6
12
18
24
30
36
42
48
54
DAYS

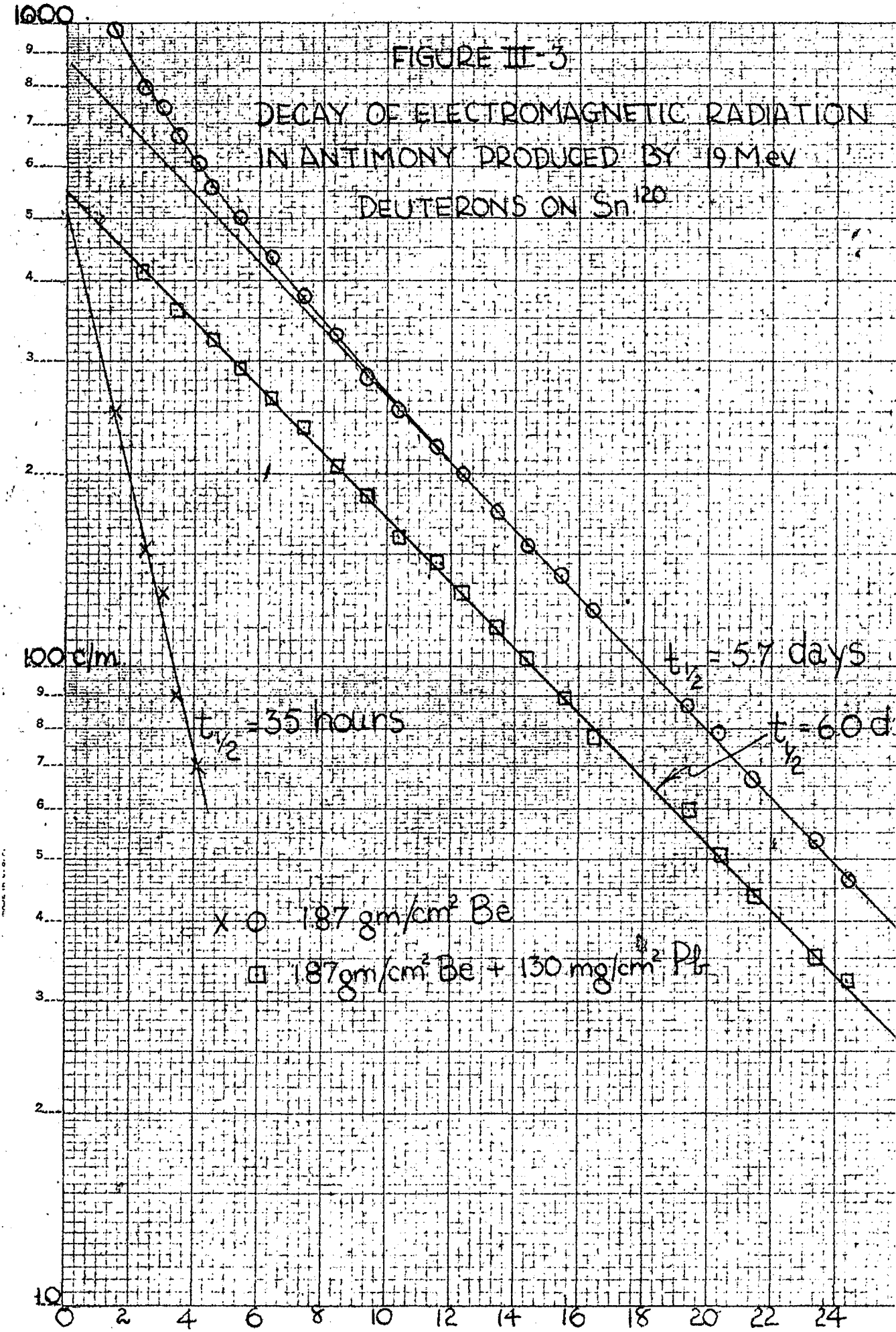


FIGURE III-3
DECAY OF ELECTROMAGNETIC RADIATION IN ANTIMONY PRODUCED BY 19 Mev DEUTERONS ON Sn^{120}
1000
100 c/m
10
$t_{1/2}$ = 5.7 days
$t_{1/2}$ = 60 d
$t_{1/2}$ = 35 hours
× ○ 187 gm/cm² Be
□ 187 gm/cm² Be + 130 mg/cm² Pb
0 2 4 6 8 10 12 14 16 18 20 22 24

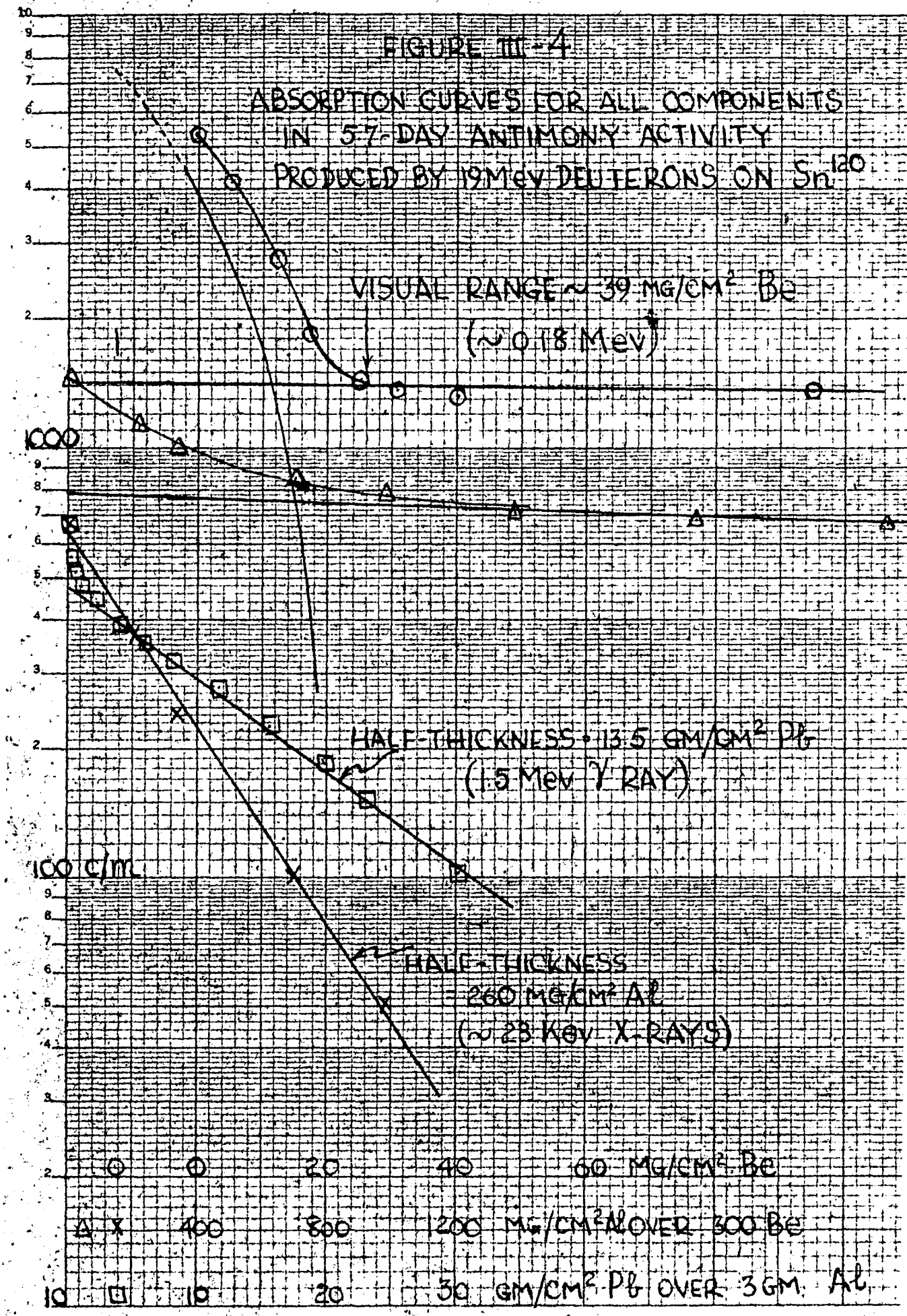


FIGURE III-4
ABSORPTION CURVES FOR ALL COMPONENTS IN 57-DAY ANTIMONY ACTIVITY PRODUCED BY 19 Mev DEUTERONS ON Sn^{120}
VISUAL RANGE ~ 39 MG/CM² Be
(~ 0.18 Mev)
1000
HALF-THICKNESS = 13.5 GM/CM² Pb
(1.5 Mev γ RAY)
100 c/m
HALF-THICKNESS
= 260 MG/CM² Al
(~ 23 Kev X-RAYS)
20
40
60 MG/CM² Be
400
800
1200 MG/CM² OVER 300 Be
10
20
30 GM/CM² Pb OVER 3 GM. Al

FIGURE III-5

CRITICAL ABSORPTION CURVES OF THE X-RAYS FROM 57-DAY ANTIMONY OBTAINED FROM 19 Mev DEUTERONS ON Sn^{120}

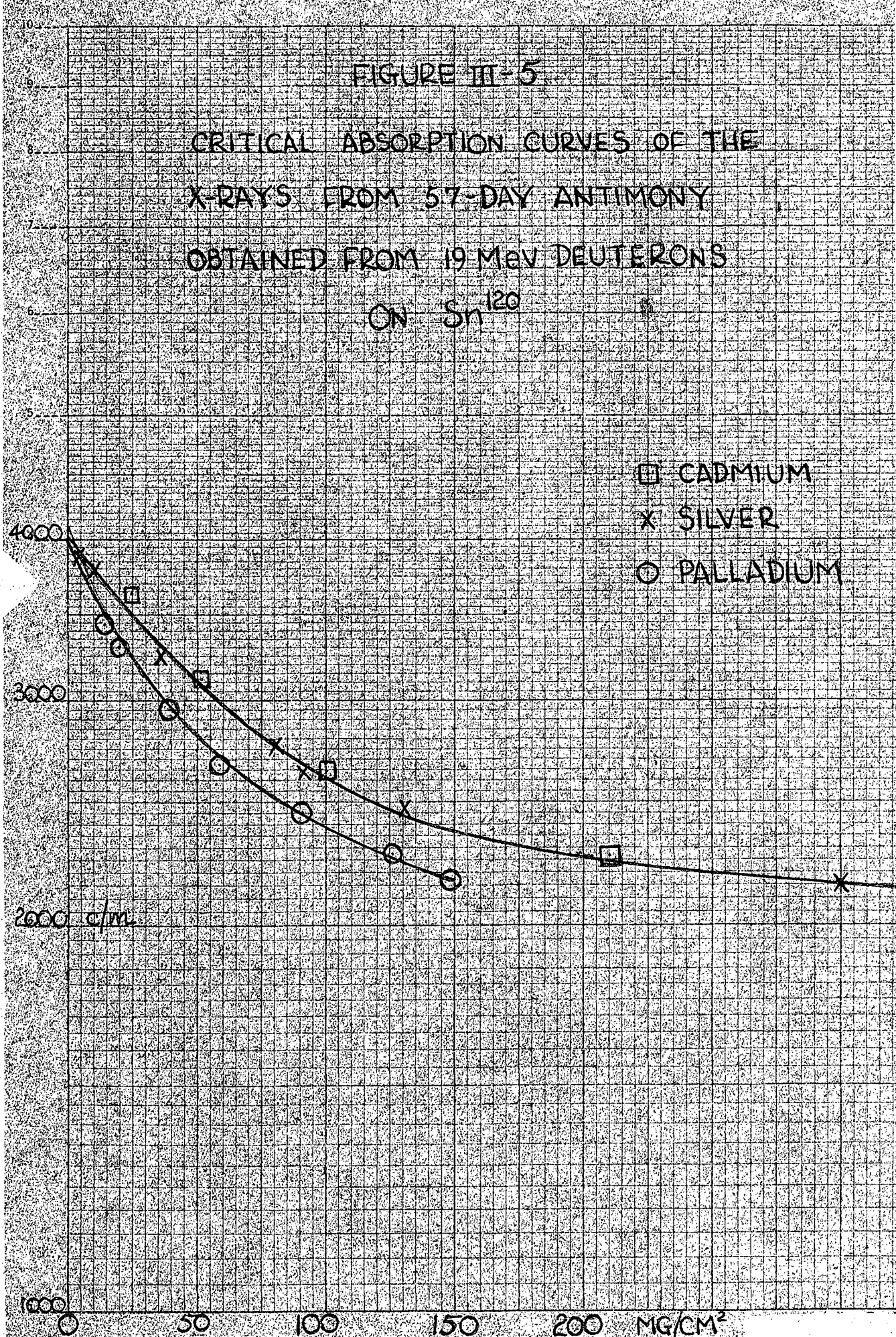


FIGURE III-6

DECAY CURVE OF Sn^{121} PRODUCED BY 19 MeV DEUTERONS ON Sn^{120}

$Sn^{120}(d,p)Sn^{121}$

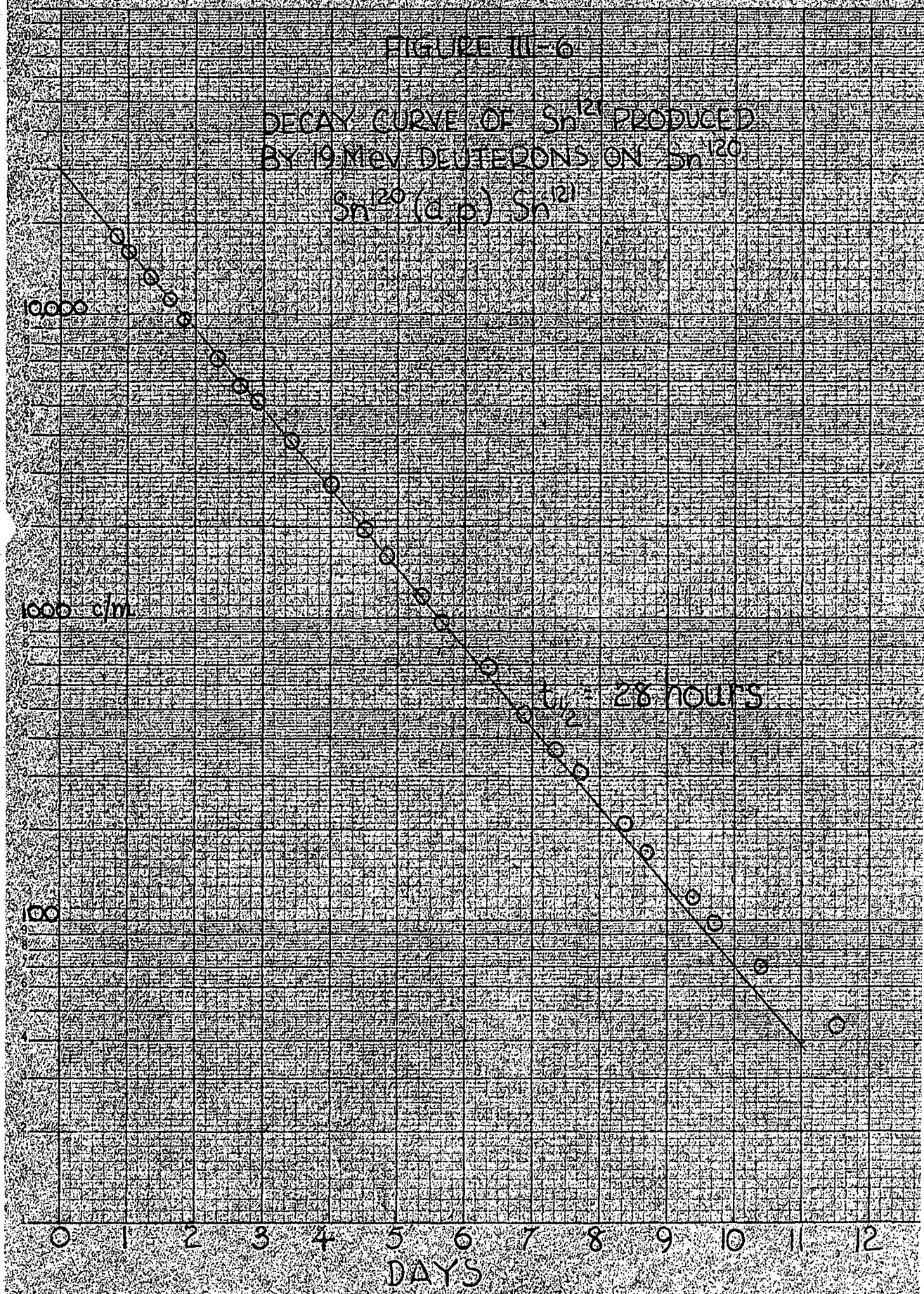


FIGURE III-7

BERYLLIUM ABSORPTION CURVE OF 28-HOUR Sn^113 ACTIVITY OBTAINED FROM DEUTERONS OF 19 MeV ON Sn^120

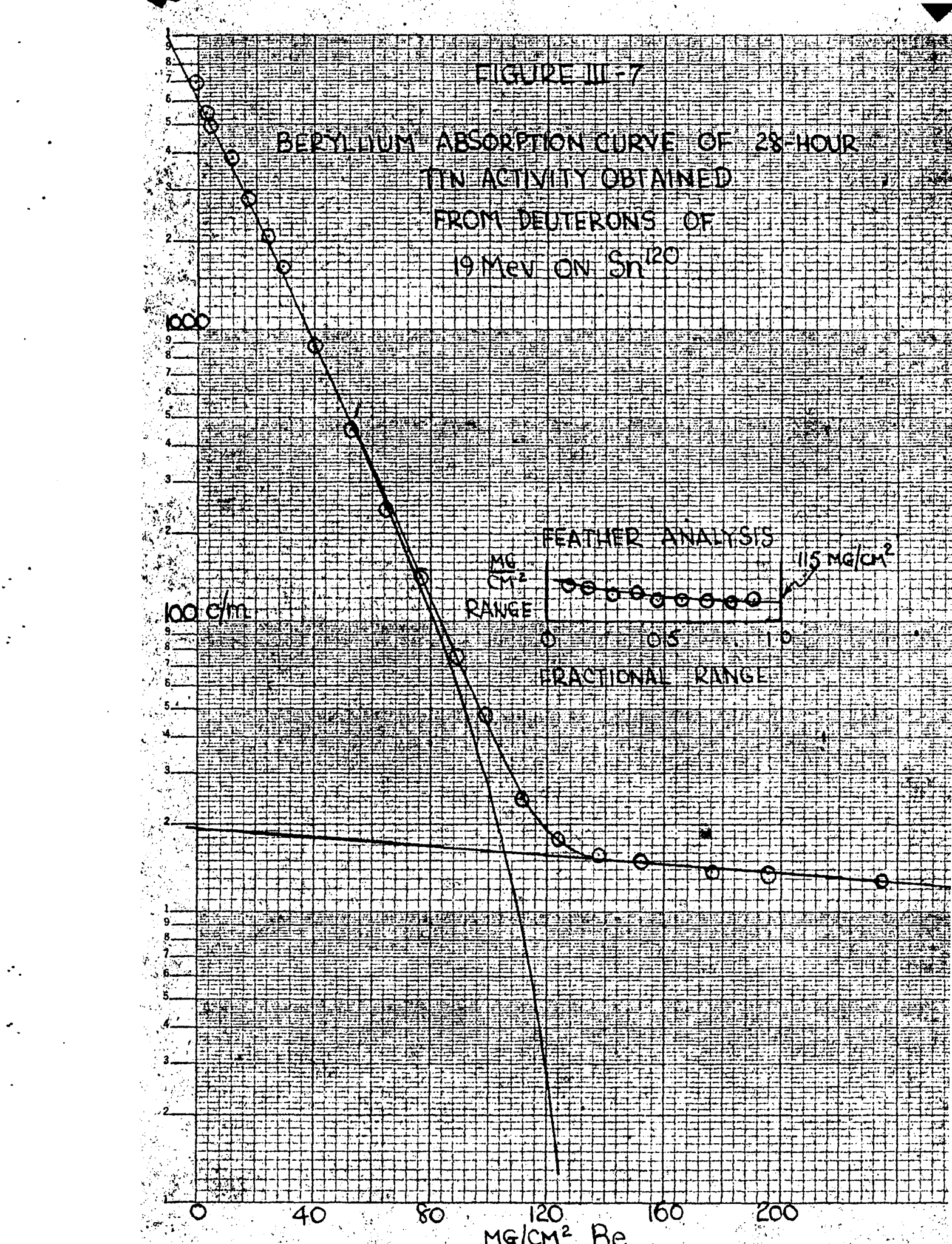


FIGURE III-8

β^- DISTRIBUTION CURVE FOR 28-HOUR Tin RECORDED ON BETA RAY SPECTROMETER

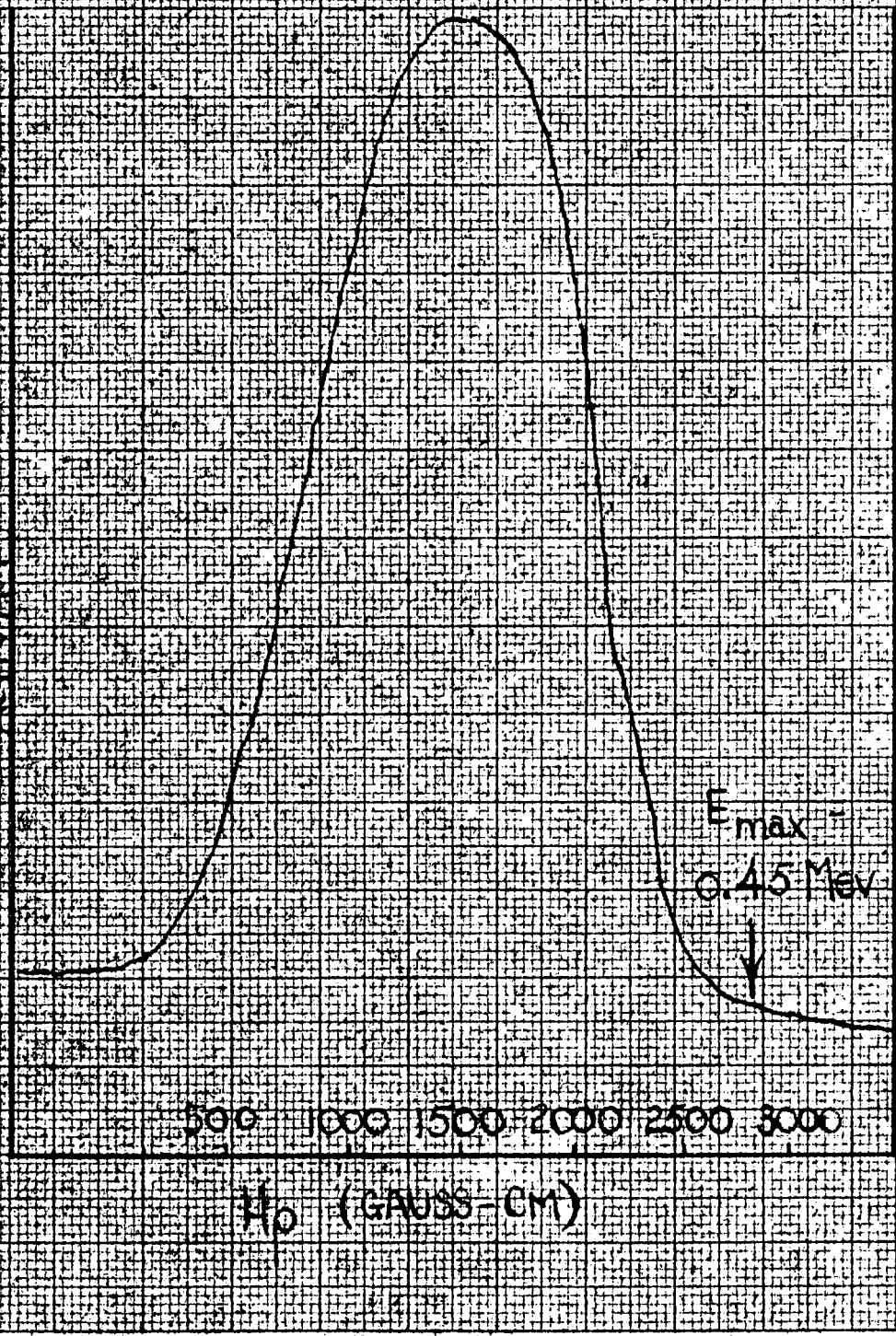


FIGURE III-9

CONVERSION ELECTRON OF 5.7-DAY ANTIMONY

RECORDED ON BETA-RAY SPECTROMETER

Section IV

PREVIOUSLY UNREPORTED ISOTOPES OF TIN FORMED IN BOMBARDMENTS OF ANTIMONY METAL WITH 200-MEV DEUTERONS

This chapter will be devoted to the elucidation of several tin isotopes isolated from the mixture of radioactive products formed from the bombardment of antimony with 200-Mev deuterons.

Among the activities encountered were a 4.5 hr period showing initial growth, the 28-hour β^- emitter described in Section III, a 14-day activity, and the 110-day Sn^{113} (9) which decays by electron capture to an excited state of In^{113}.

4.5-hour Sn. The initial stages in the decay of the Sn fraction showing the growth and decay associated with the 4.5-hour activity is shown in Fig. IV-1. The difference between the extrapolated decay curve after establishment of equilibrium and the growth curve indicates the growth of a daughter of approximately 75 minutes half-life. After equilibrium had been established, an absorption curve, shown in Fig. IV-2, was obtained, indicating the presence of a 2.2-Mev beta particle. A distribution curve obtained from the low-resolution beta-ray spectrometer showed that these particles were positrons of energy 2.5 Mev, and that at this time, the positron accounted for approximately 90% of the activity of the sample. It was subsequently shown that the 4.5-hour activity, at equilibrium, likewise accounted for 90% of the activity of the sample. Hence the growth cannot be associated with one of the longer-lived tin isotopes present, and the growth, the positron, and the 4.5-hour activity are indeed inter-related.

It was then found that the 2.5-Mev positron belonged to an indium daughter of approximately 70-minutes half-life which could be separated from the tin. The identity of the particle was corroborated with both the beta-ray spectrometer and by a beryllium absorption curve. These are shown in Figs. IV-3 and IV-4, respectively. A typical decay curve for this indium daughter is shown in Fig. IV-5.

As a further check, it was found that the yield of the 70-minute activity isolated from tin, decreased with roughly a four-hour half-life, until finally the only activity obtained from a Sn-In separation was that of the 105-minute In^{113*}.

The simple chemical procedure employed for the separation of the indium and tin consisted of the precipitation of $In(OH)_3$ with NaOH, the tin remaining in solution.

Mass Assignment. Barnes[9] observed a 65-minute indium activity, produced by irradiation of cadmium with protons, which decayed by the emission of 1.6-Mev positrons. This he assigned to In^{110}. More recently, Tendam and Bradt[10] showed that the excitation curve for the production of this isotope by irradiating silver with alpha particles corresponded to that for an (α,n) reaction, and is hence either In^{112} or In^{110}.

Ghoshal[11] has confirmed the assignment made by Tendam and Bradt. In view of the similarity in properties between the 70-minute β^+ daughter and the 65-minute isotope reported by the above investigators, it might seem that the 4.5-hour tin activity be Sn^{110} decaying by electron capture (since no positrons other than those of the In daughter were found). There are, however, several objections to this conclusion. In the first place, inspection of Table IV-1 suggests that Sn^{110}, two mass numbers below the last stable tin isotope, may be a long-lived "even-even" isotope, decaying to a much shorter-lived "odd-odd" indium daughter. Several examples of the phenomenon exist[12], although no general rule can be stated. The fact that the half-lives of the isobaric pair are of the same order

Table IV-1

	108	109	110	111	112	113	114	115
Cd	S	↑	S	S	S	S	S	↓
In						S	↓	S
Sn					S	↑	S	S

of magnitude suggests an odd mass number for the pair. A similar situation was encountered in Section II in two tellurium-antimony isobaric relationships in which the comparison of the half-lives of the parent and daughter aided in the assignment of mass numbers.

The second objection to the assignment of the even mass number 110 is the fact that the maximum energy of 1.6 Mev of the positron reported by Barnes[9] is in serious disagreement with that found for the equilibrium mixture of the tin and indium, and for the 70-minute indium above (cf. Fig. IV-2 and IV-3) as a result of beryllium absorption measurements. The values of 2.2 Mev are in fair agreement with the value of 2.5 Mev obtained from the end point of the crude beta-ray distribution curve shown in Fig. IV-3. Hence, unless the value of 1.6 Mev of Barnes is in serious error, it seems probable that the 65-minute indium positron of Barnes, and Tendam and Bradt is different from the 70-minute daughter of 4.5-hour tin in the present treatment. This would seem to suggest that the latter pair could be assigned either to mass number 111 or 109. Since In^{111} has been reported by Tendam and Bradt[10] definitely to be the 2.7-day K capture isotope, it is possible that the mass be 109. It could still be 111 in which the indium daughter is an unrelated isomer of the 2.7-day activity. If the mass number be 109, it remains to be explained why another tin activity was not found which could be assigned to Sn^{111}.

14-day IT Sn. The decay curve of the tin fraction illustrating the existence of the fourteen-day component will be deferred until a discussion of spallation reaction products. However, a complete absorption curve is shown in Fig. IV-6. This, together with the conversion line illustrated by Fig. IV-7 from beta-ray spectrometric measurements indicates that electrons of approximately 150 Kev, x-rays characteristic of the tin region, and a γ-ray corresponding to the

150-Kev electrons are all prominent. Thus, the γ-ray is about 80% converted, assuming a one-percent counting efficiency for the γ-ray and 100% efficiency for the electrons. Fig. IV-8 is a critical absorption curve for the x-rays. This indicates that the x-rays are those of tin, and that the decay process is an isomeric transition from an excited state. Although adequate proof will be deferred until later, the mass number of this activity is almost certainly high, for the activity was obtained in relatively high yield over a deuteron energy range from 50 Mev to 200 Mev. This was found to be quite characteristic of masses in the region of 120 ± 2 for any Z, whereas, for example, the yield of Sn^{113} decreased very markedly from 200 Mev to 50 Mev. Hence the present period represents an isomeric transition with a high conversion factor, to some stable tin isotope, probably of high mass number.

Table IV-2 summarizes the data thus found concerning these tin activities.

Table IV-2

Isotope	Half-life	Type of Radiation	Energy of Radiation, Mev.	
			Particles	γ-rays
$_{50}Sn$	14 d	IT, e^-, γ		0.175
$_{50}Sn^{110?}$	4.5 hr	K	?	?

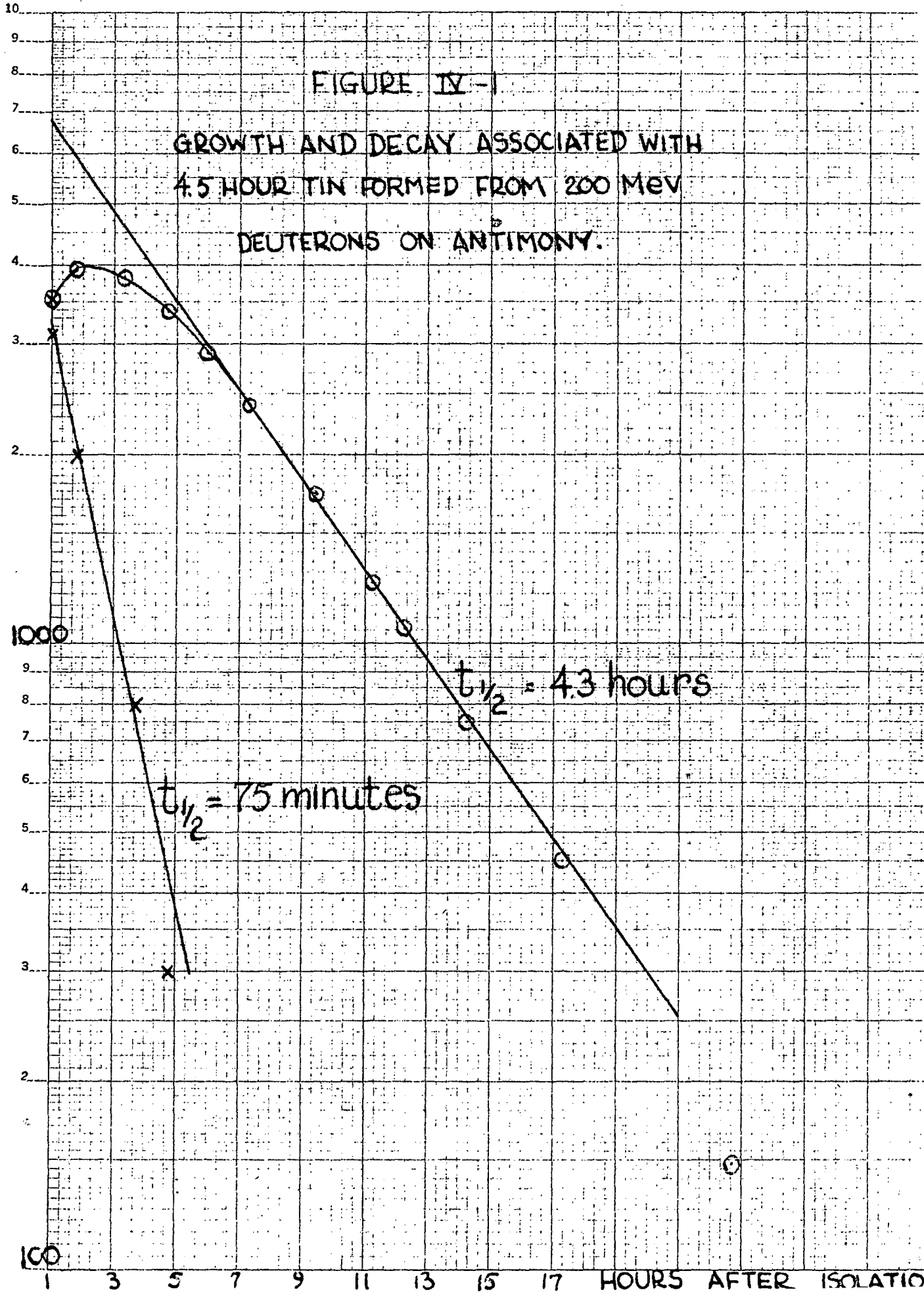
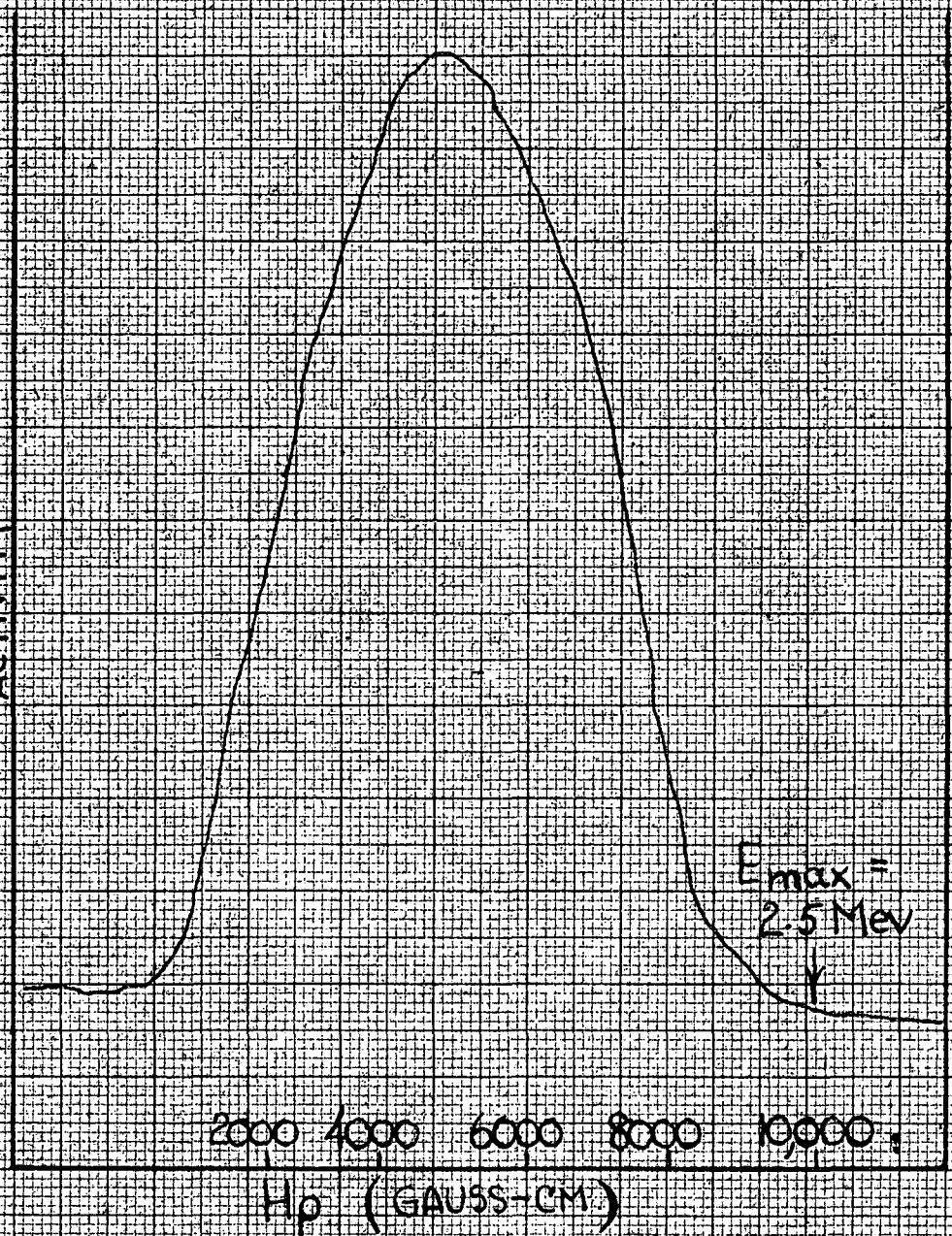


FIGURE IV-1
GROWTH AND DECAY ASSOCIATED WITH
4.5 HOUR TIN FORMED FROM 200 Mev
DEUTERONS ON ANTIMONY.
$t_{1/2}$ = 4.3 hours
$t_{1/2}$ = 75 minutes
10
9
8
7
6
5
4
3
2
1000
100
1
3
5
7
9
11
13
15
17
HOURS AFTER ISOLATIO

FIGURE V-3

DISTRIBUTION CURVE FOR POSITRON OF 70-MIN. INDIUM ACTIVITY DAUGHTER OF 4.5 HOUR TIN PERIOD



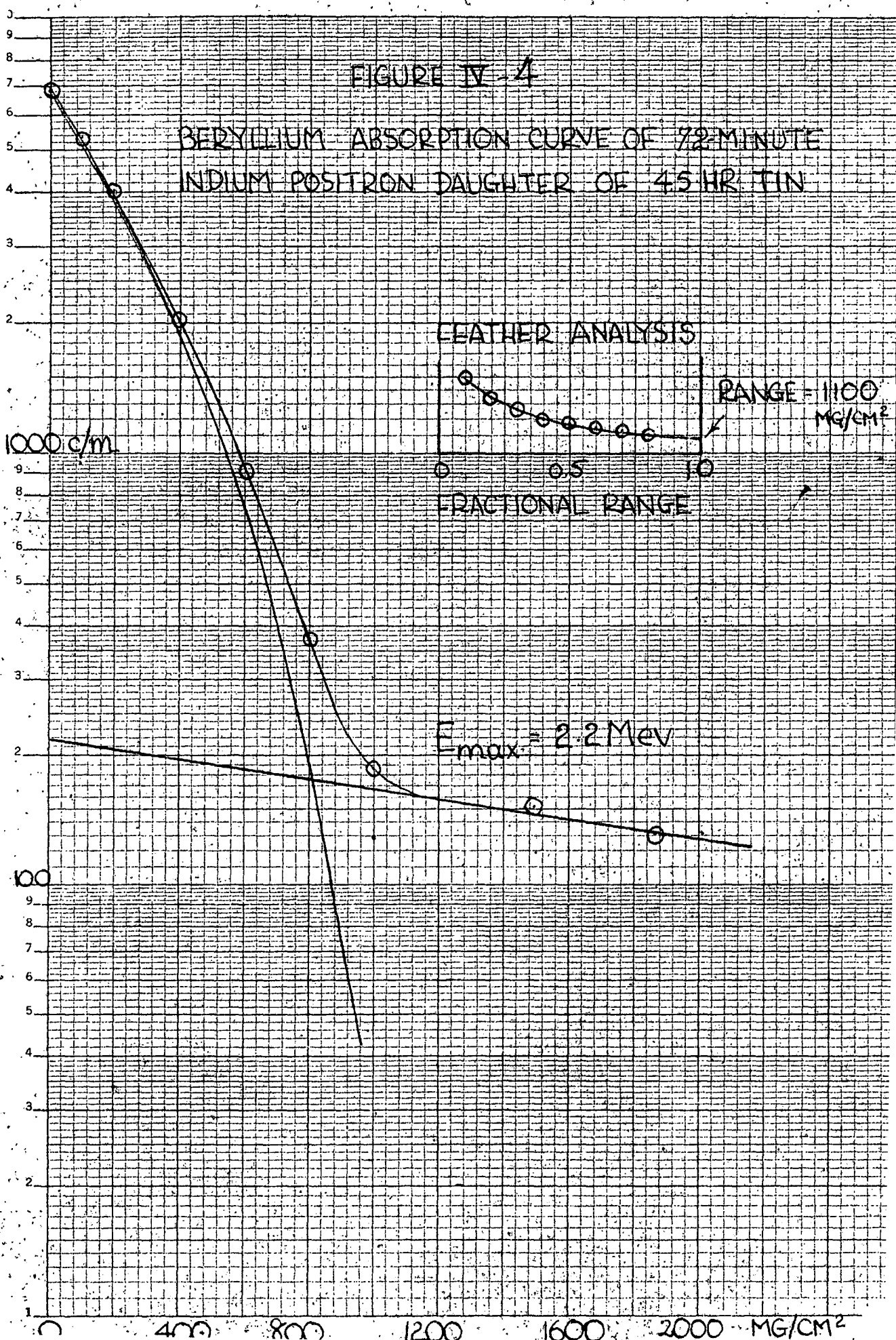


FIGURE IV-4
BERYLLIUM ABSORPTION CURVE OF 72-MINUTE INDIUM POSITRON DAUGHTER OF 45 HR TIN
FEATHER ANALYSIS
RANGE = 1100 MG/CM²
1000 c/m
0
0.5
1.0
FRACTIONAL RANGE
E-max = 2.2 Mev
100
0
400
800
1200
1600
2000
MG/CM²

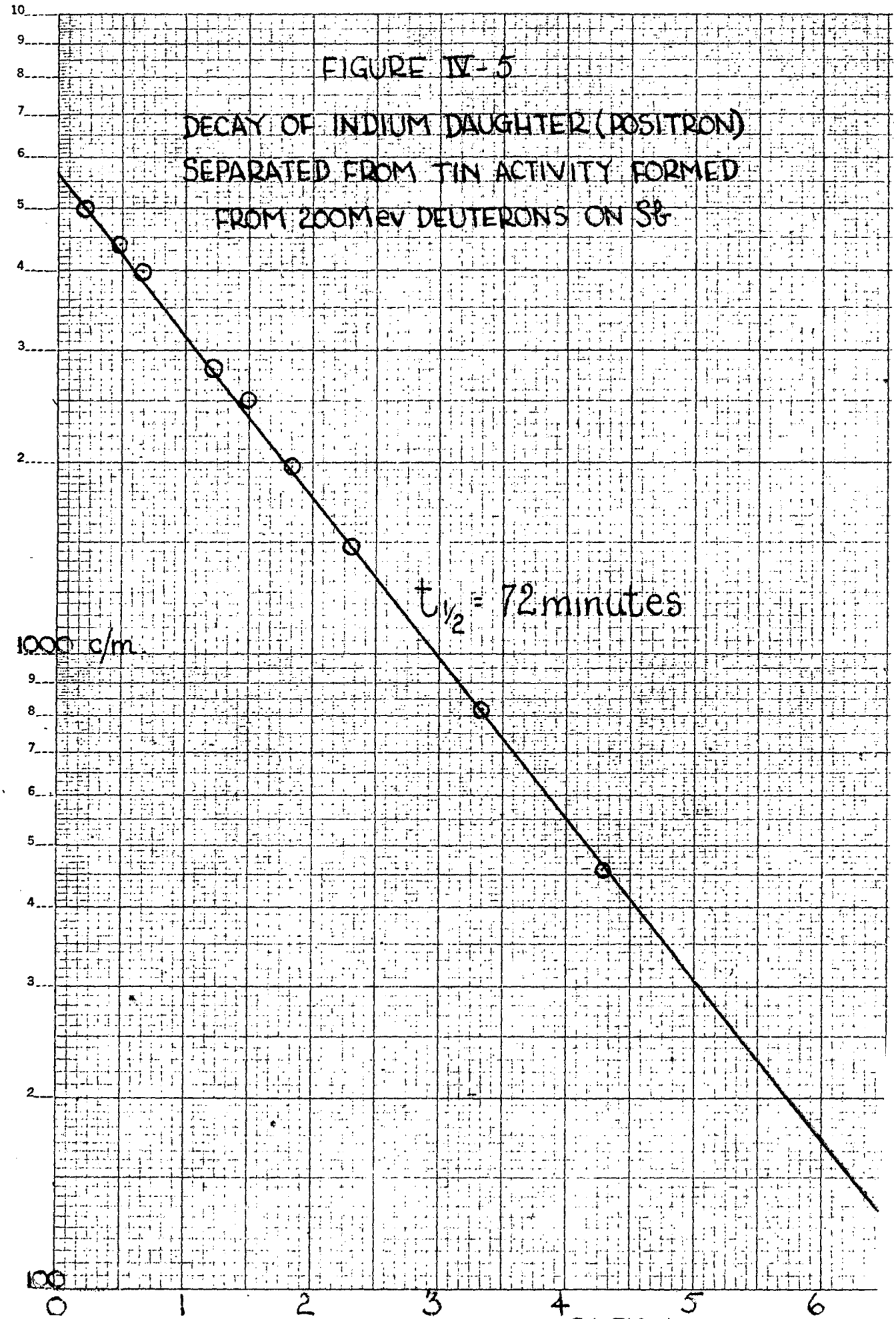


FIGURE IV-5
DECAY OF INDIUM DAUGHTER (POSITRON)
SEPARATED FROM TIN ACTIVITY FORMED
FROM 200Mev DEUTERONS ON Sb
$t_{1/2}$ = 72 minutes
1000 c/m
100
0
1
2
3
4
5
6

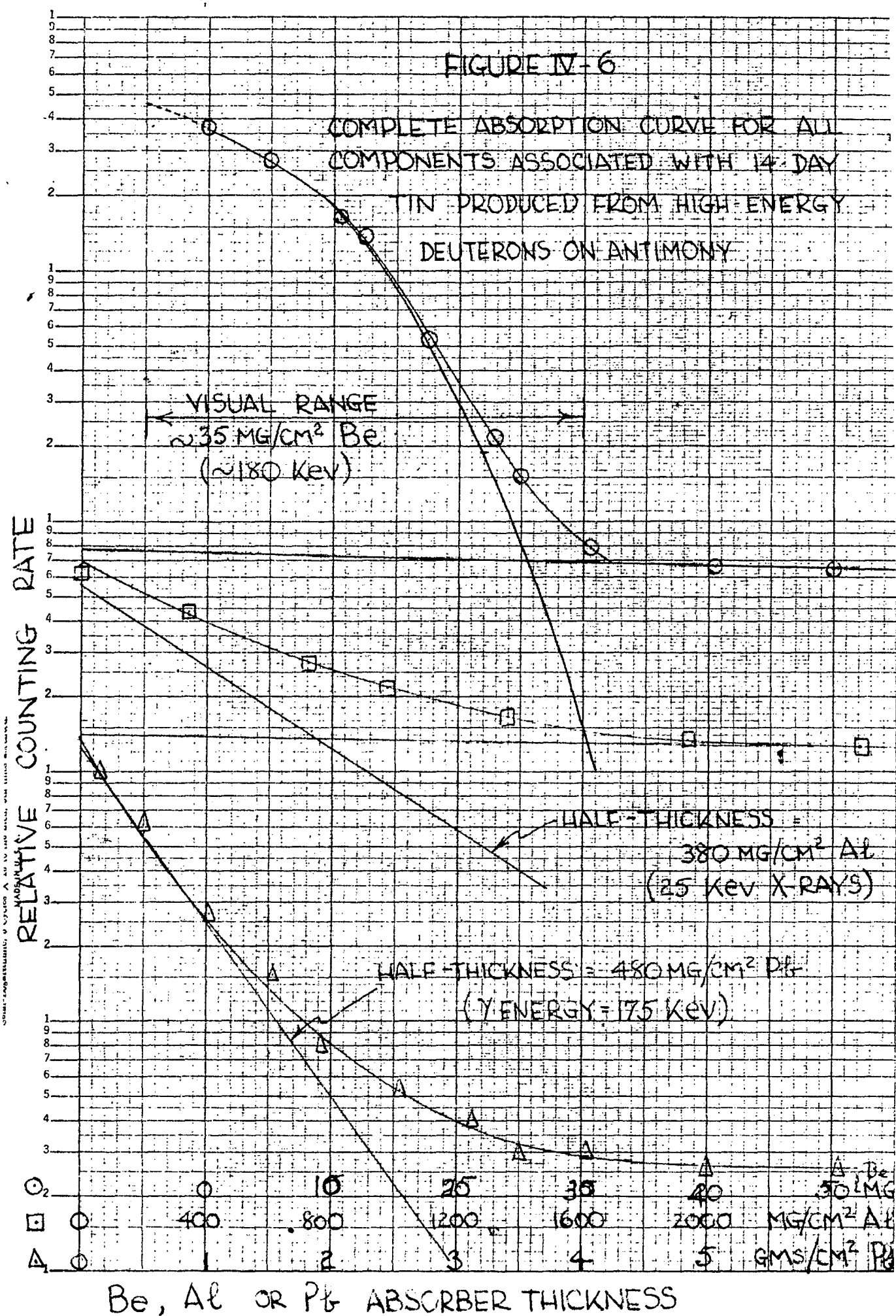
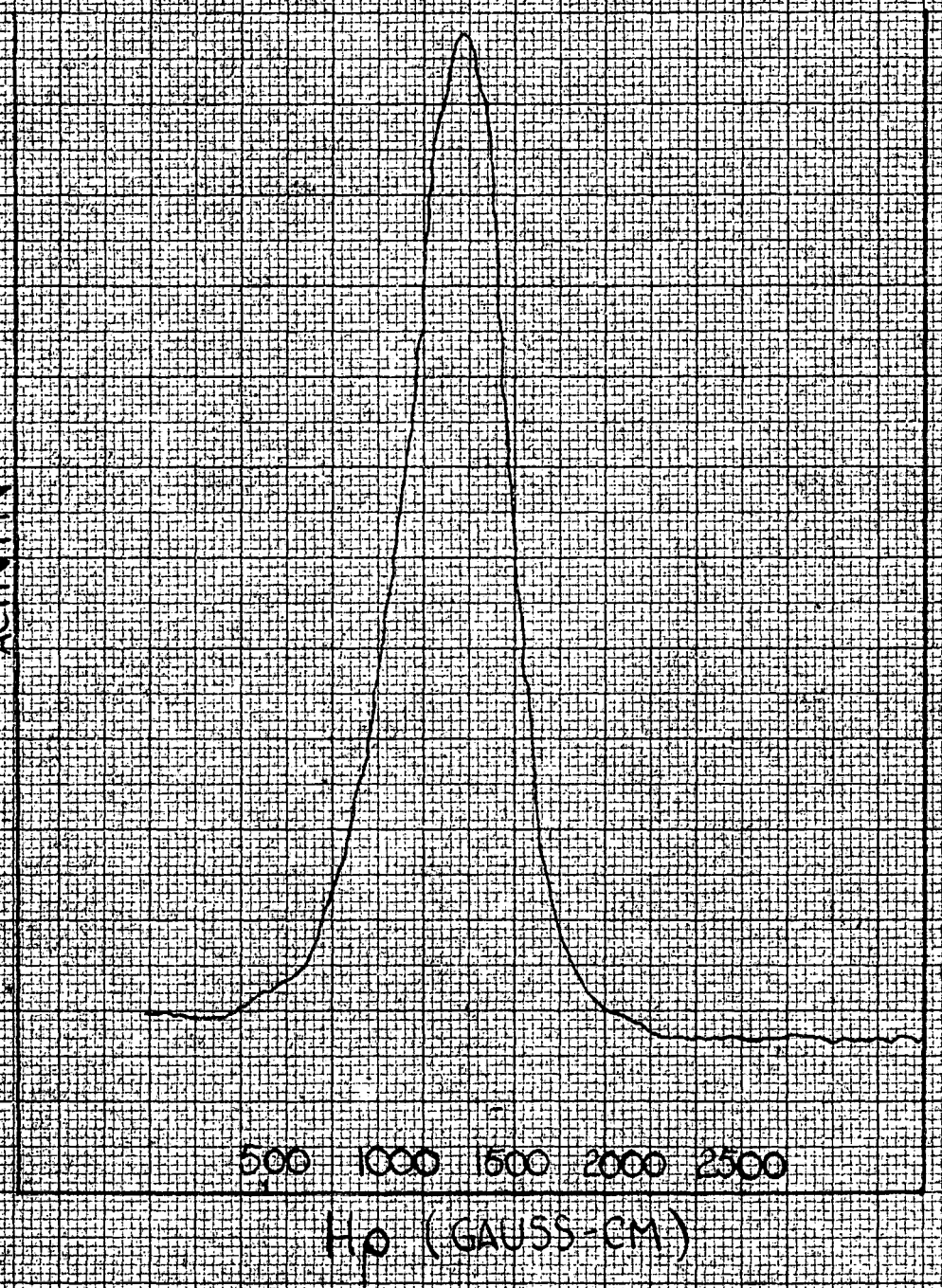


FIGURE IV-6
COMPLETE ABSORPTION CURVE FOR ALL COMPONENTS ASSOCIATED WITH 14-DAY TIN PRODUCED FROM HIGH-ENERGY DEUTERONS ON ANTIMONY
VISUAL RANGE
≈35 MG/CM² Be
(≈180 Kev)
HALF-THICKNESS = 380 MG/CM² Al
(25 Kev X-RAYS)
HALF-THICKNESS = 480 MG/CM² Pb
(γ ENERGY = 175 Kev)
RELATIVE COUNTING RATE
Be, Al OR Pb ABSORBER THICKNESS
MG/CM² Al
GMS/CM² Pb

FIGURE IV-7

CONVERSION ELECTRON ASSOCIATED WITH 14-DAY ISOMERIC TIN

RECORDED ON BETA-RAY SPECTROMETER



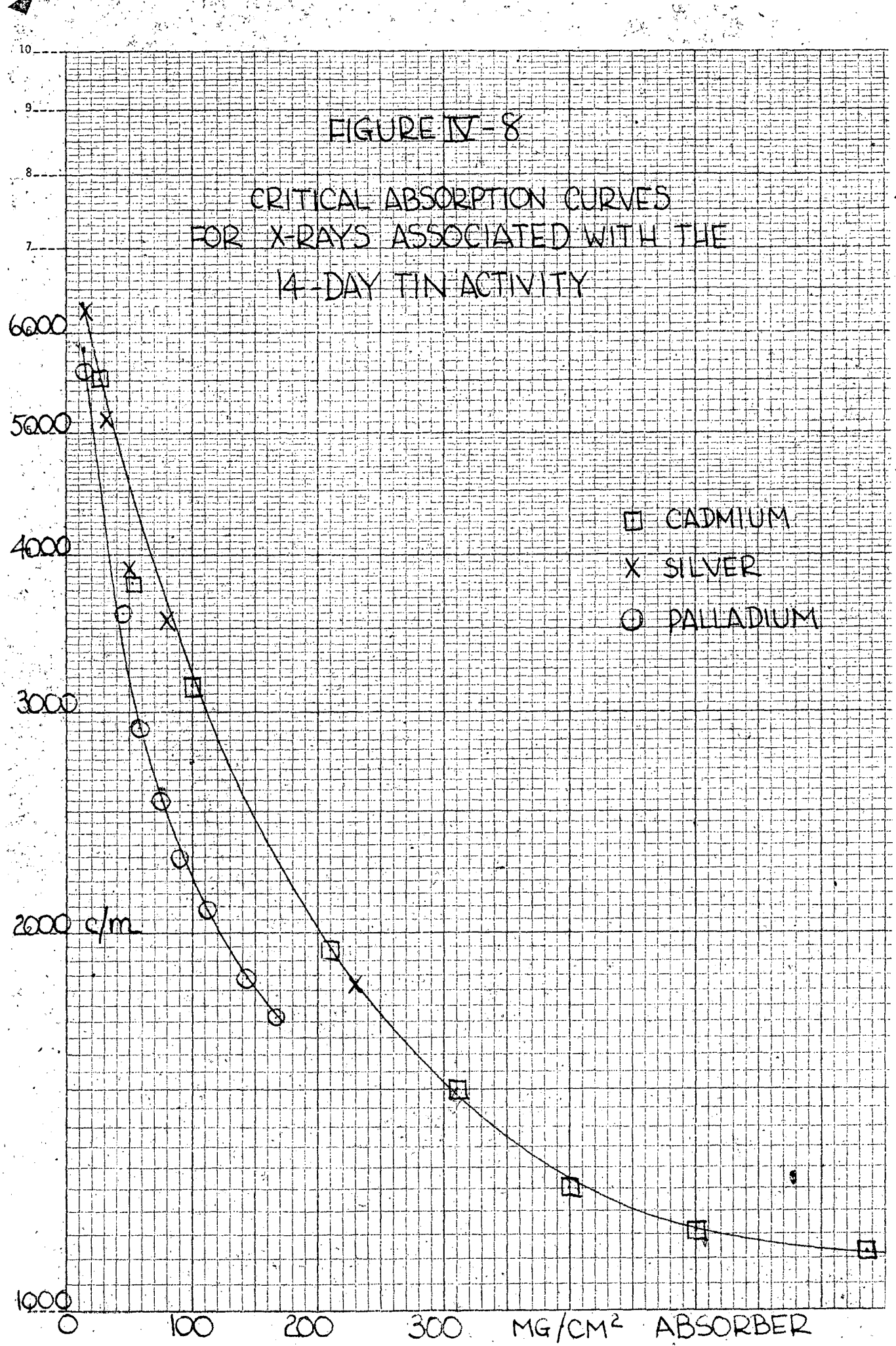


FIGURE IV-8
CRITICAL ABSORPTION CURVES
FOR X-RAYS ASSOCIATED WITH THE
14-DAY TIN ACTIVITY
CADMIUM
SILVER
PALLADIUM
6000
5000
4000
3000
2000 c/m
1000
0
100
200
300
MG/CM² ABSORBER

Section V

THE SPALLATION PRODUCTS OF ANTIMONY IRRADIATED WITH HIGH ENERGY PARTICLES OF THE 184" CYCLOTRON

Previous to the operation of the 184" cyclotron, targets exposed to the beams of the world's cyclotrons would, in general, contain radioactive isotopes of two, or at most, three elements (an exception to this rule is the fission reaction). For example, antimony, if irradiated with 20-Mev deuterons would be expected to produce tellurium, antimony and perhaps tin activities as a result of (d,xn), (d,pxn) and (d,αxn) reactions, respectively. In general x is an integer varying from one to three. Similar reactions would come about as the result of bombardments with protons or alpha particles. Thus one would expect product nuclei of two, or, at most, three atomic numbers to be formed. The abundant literature pertinent to such nuclear reactions is so vast that reference is given only to Seaborg's "Table of Isotopes"[13] in which an extensive literature survey of the field of artificially produced radioactivities is given. From this one can verify the reaction types briefly mentioned above. Irradiation with 200-Mev deuterons and 400-Mev alpha particles from the 184" cyclotron has, in general, given rise to so large a number of radioactive products that rather elaborate chemical procedures had to be worked out in order to identify these products. Thus it has been found in the case of antimony targets that activities were produced which differ in mass from the target atoms by 36 units, and in atomic number by 15 units. Of course, a multitude of isotopes in between these limits was also formed.

The elements listed below include those which were identified from the complex mixture of spallation products of high-energy deuterons and alpha particles of the 184" cyclotron: $_{39}Y$, $_{40}Zr$, $_{41}Cb$, $_{42}Mo$, $_{43}Tc$, $_{44}Ru$, $_{45}Rh$, $_{46}Pd$, $_{47}Ag$, $_{48}Cd$, $_{49}In$, $_{50}Sn$, $_{51}Sb$, and $_{52}Te$. Zr, Cb, and Tc were not investigated, and rhodium proved

too difficult to remove from the complex mixture. However, the remaining elements, Y, Mo, Ru, Pd, Ag, Cd, In, Sn, Sb, and Te were investigated. Since many of the products of lower atomic number (Y, Mo, etc.) were found to be formed in exceedingly low yield, it was necessary to effect complete separation of a given element followed by several repurification steps. A known weight of carrier (usually twenty milligrams in the form of a solution of a readily soluble salt) was always added to the gross mixture of activities before chemical separations were made. This procedure insures against the adsorption of any tracer activity on foreign precipitates. The following procedure was found to work satisfactorily for the separation of those elements listed in table I except for Zr, Cb and Tc.

The dimensions of the metallic antimony targets were about 1 mm x 2 mm x 30 mm, weighing about 0.45 grams.

The procedures given below serve to isolate a given element in a state free from any contaminating activities. In many instances, the procedures were not suitable to the determination of periods of the order of one-hour half-life, due to the time involved in separation and purification. In such instances, it was usually necessary that initial efforts be concentrated in isolation of such a short-lived activity or that the chemical procedure be shortened if the separation and decontamination from other elements is not seriously impaired.

The following is a qualitative description of the scheme of separation and the nature of the activities found, listed by elements. The fractions were investigated over a range of energies varying from 400-Mev helium ions to 50-Mev deuterons. The discussion will attempt also to describe the results of various bombarding energies.

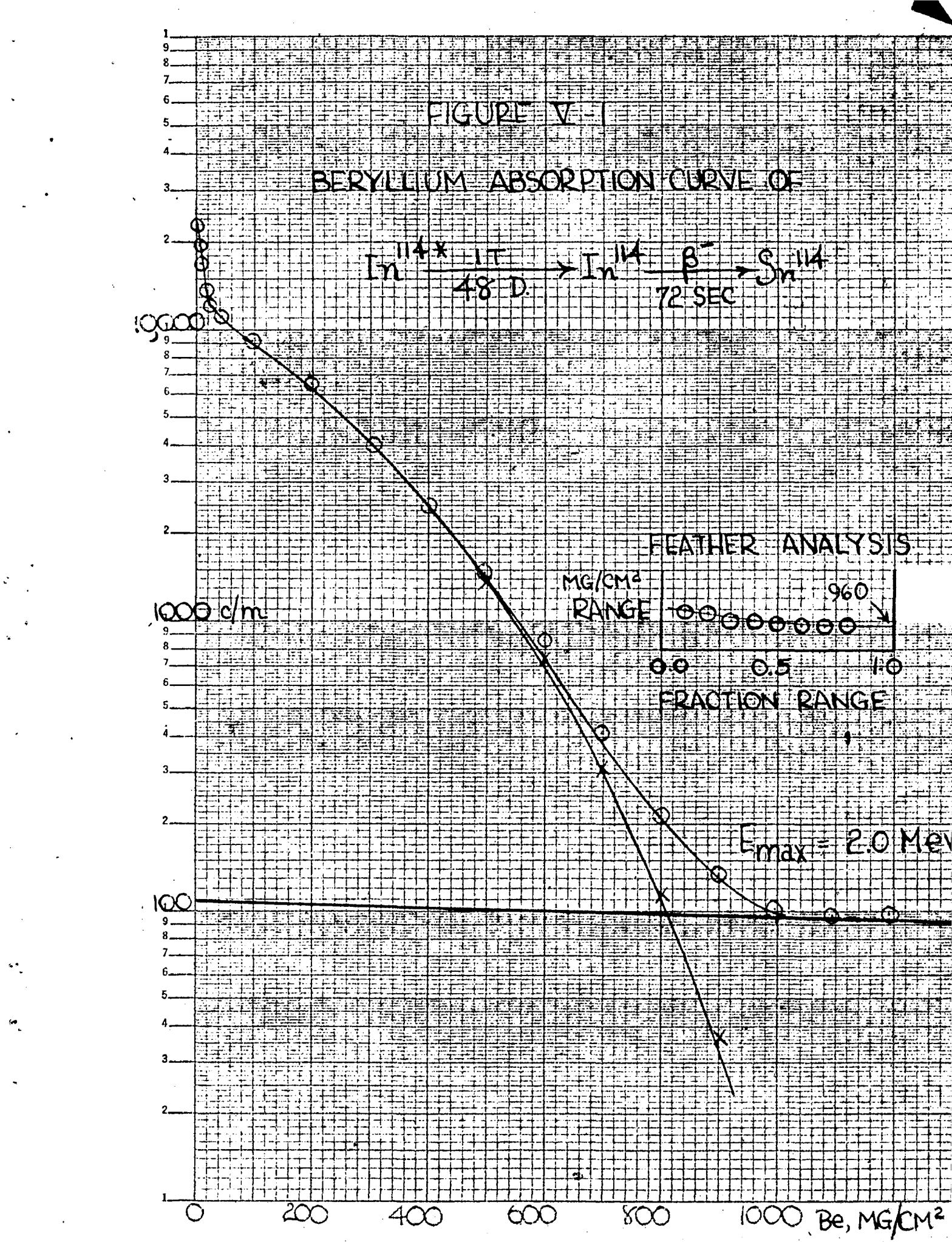
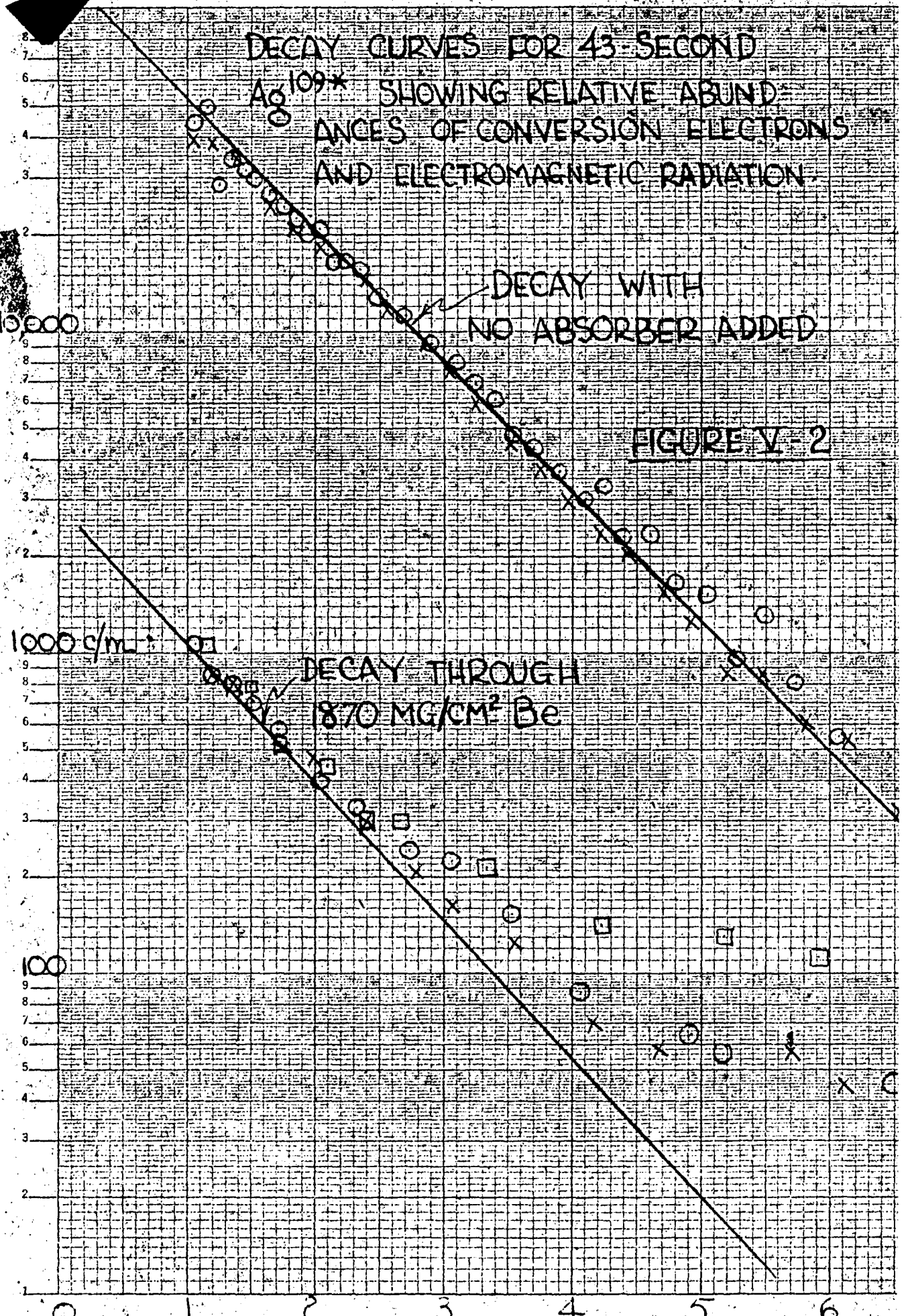
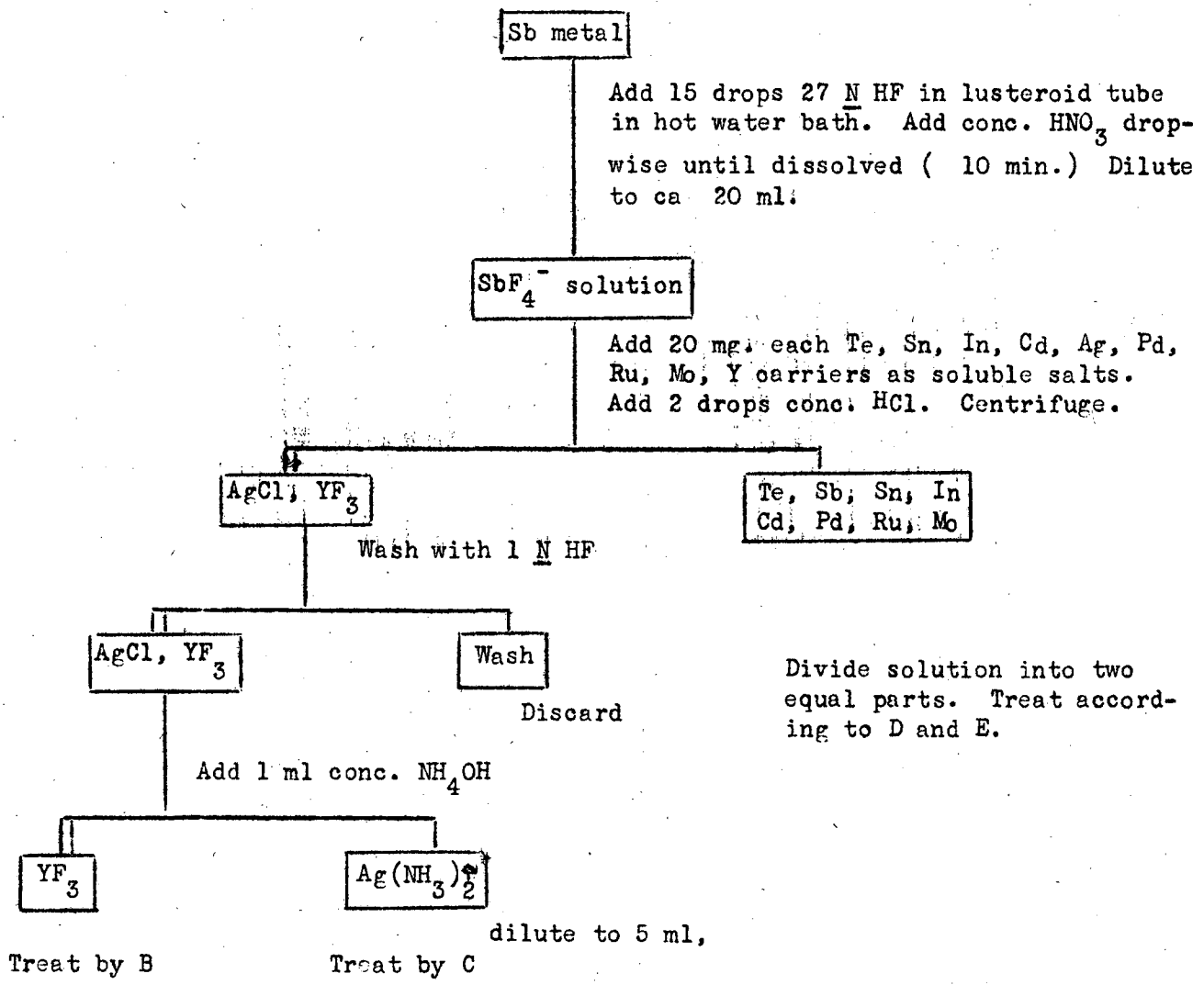


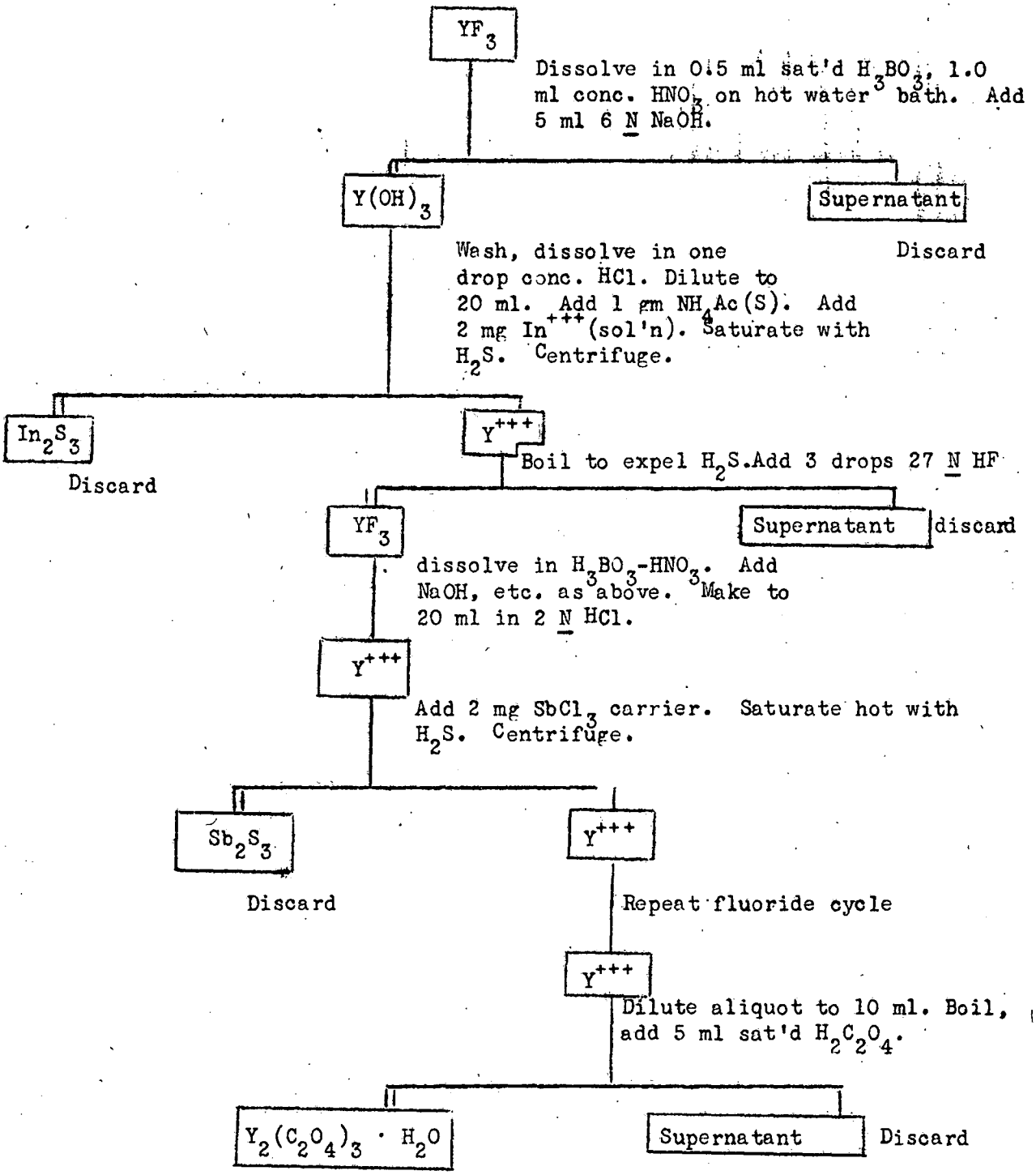
FIGURE V - I
BERYLLIUM ABSORPTION CURVE OF
$In^{114*} \xrightarrow[48\ D.]{IT} In^{114} \xrightarrow[72\ SEC]{\beta^-} Sn^{114}$
10,000
1000 c/m
100
FEATHER ANALYSIS
MG/CM²
RANGE
960
0.0
0.5
1.0
FRACTION RANGE
E_{max} = 2.0 Mev
0
200
400
600
800
1000
Be, MG/CM²


DECAY CURVES FOR 43-SECOND
Ag^{109*} SHOWING RELATIVE ABUNDANCES OF CONVERSION ELECTRONS AND ELECTROMAGNETIC RADIATION
DECAY WITH NO ABSORBER ADDED
FIGURE V-2
10,000 c/m
1000 c/m
DECAY THROUGH 870 MG/CM² Be
100
0
1
2
3
4
5
6

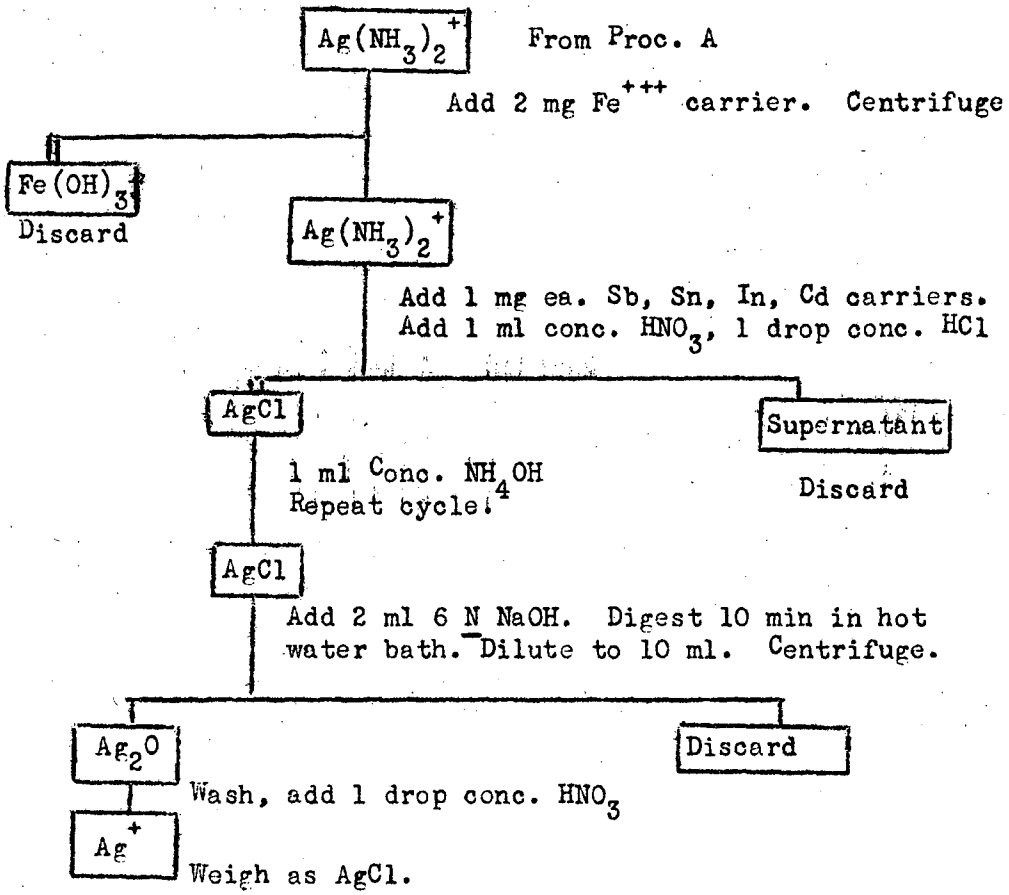
Procedure A. Dissolution of Target; Precipitation of Silver and Yttrium.



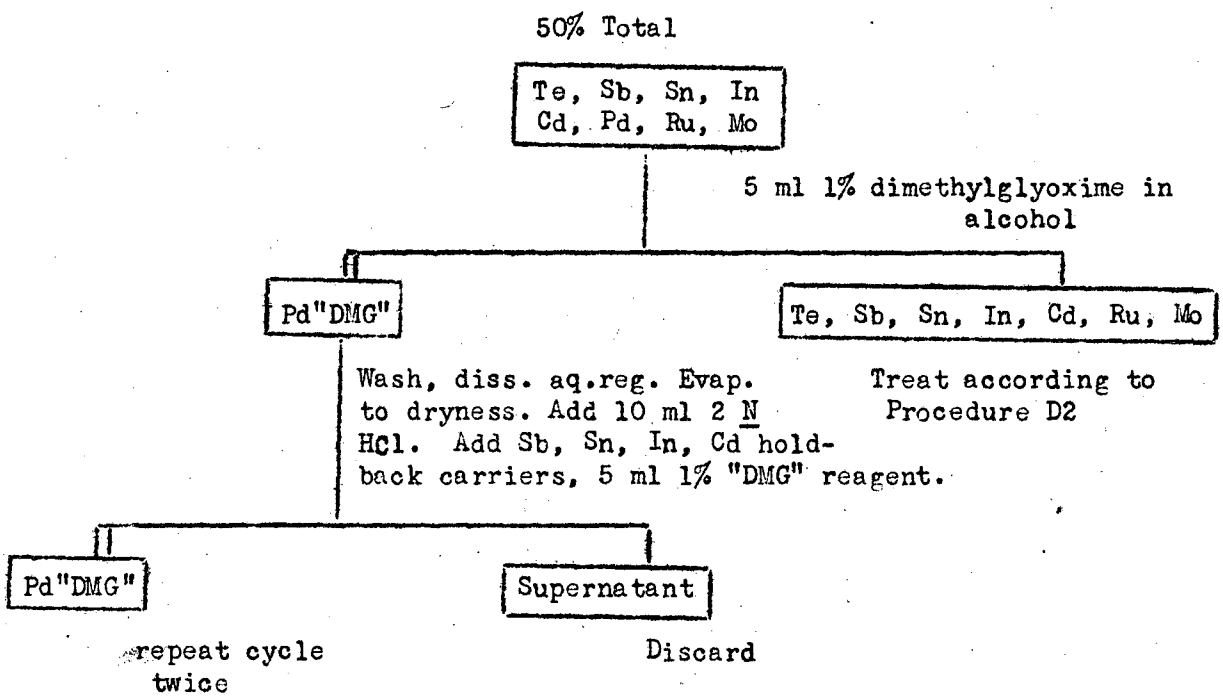
Procedure B - Isolation and Purification of Yttrium.



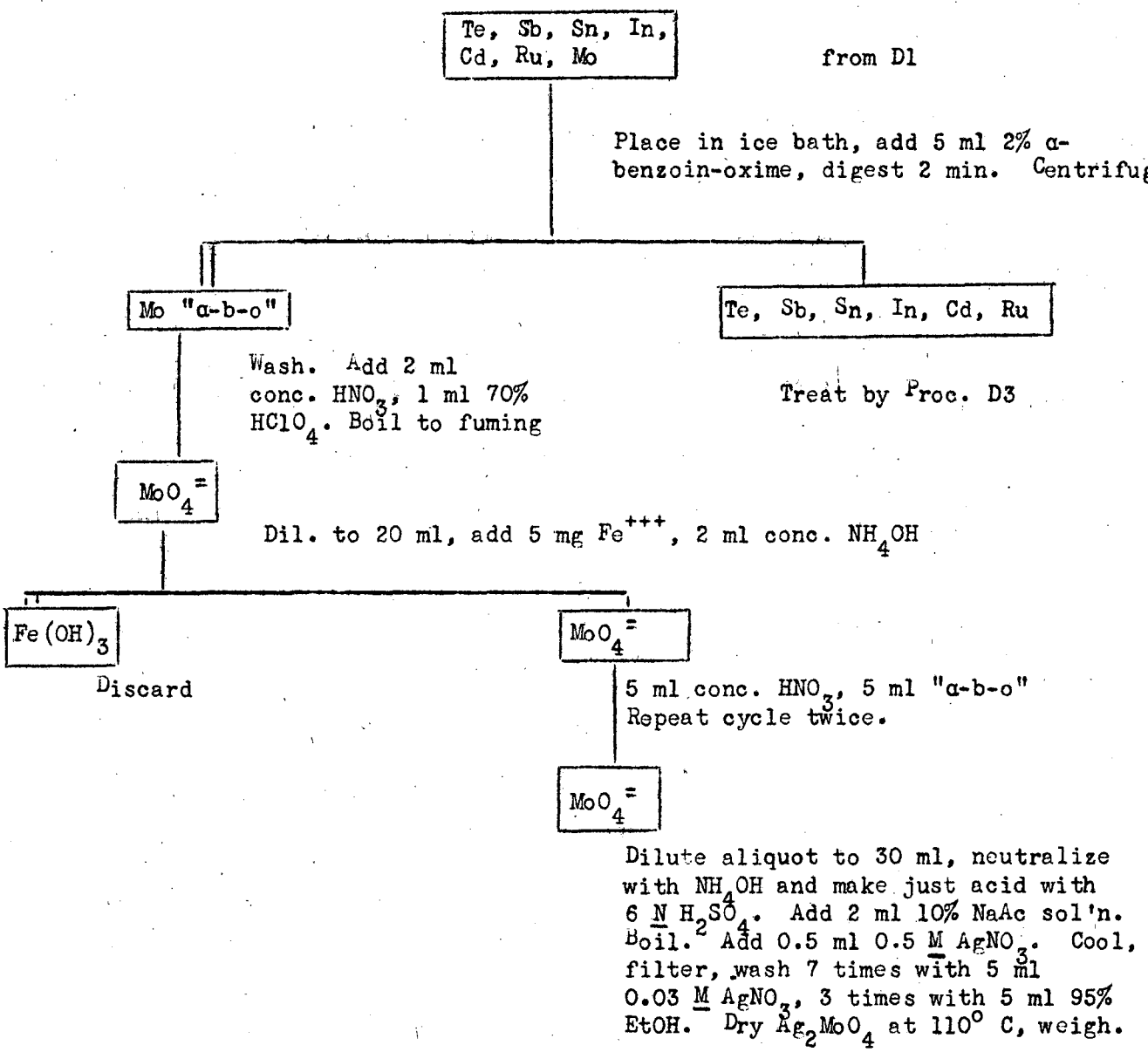
Procedure C - Isolation and Purification of Silver.



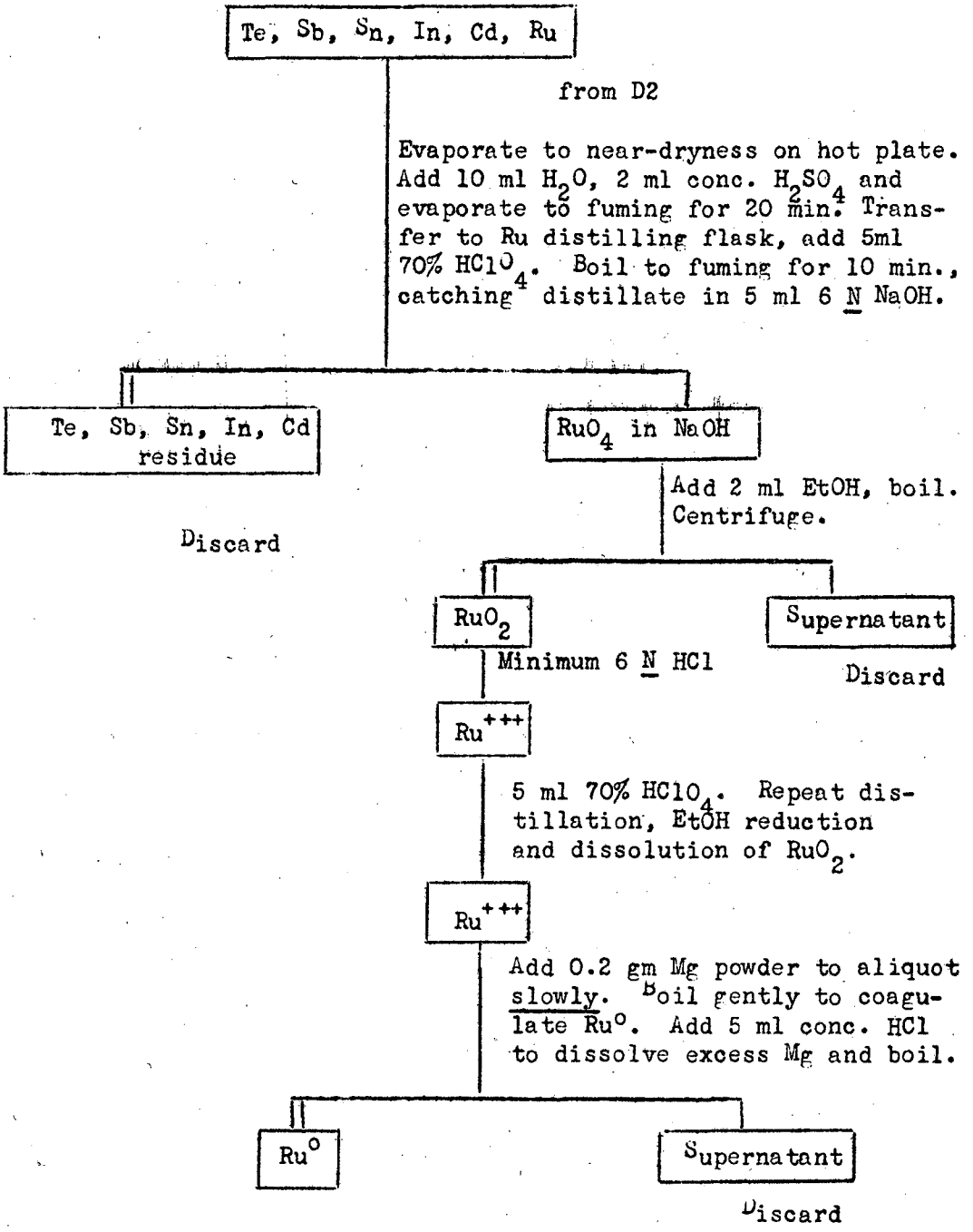
Procedure D1 - Isolation and Purification of Palladium.



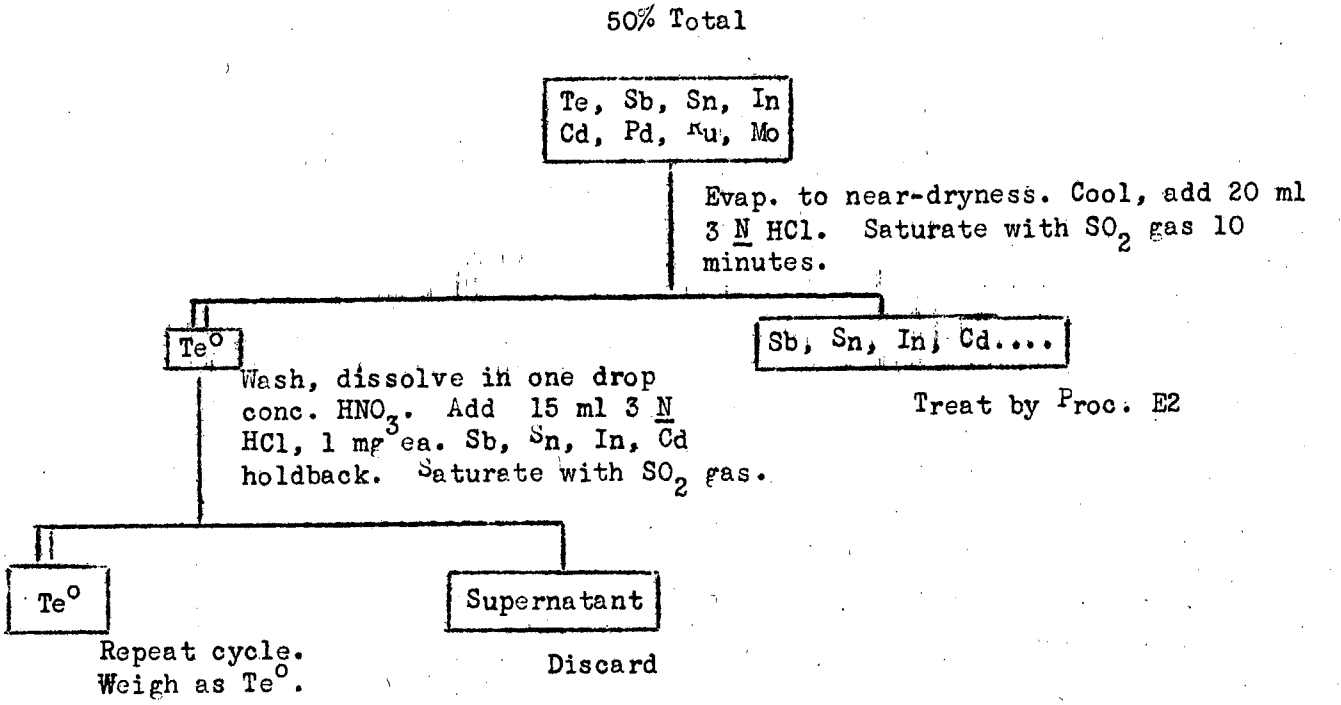
Procedure D2 - Isolation and Purification of Molybdenum.



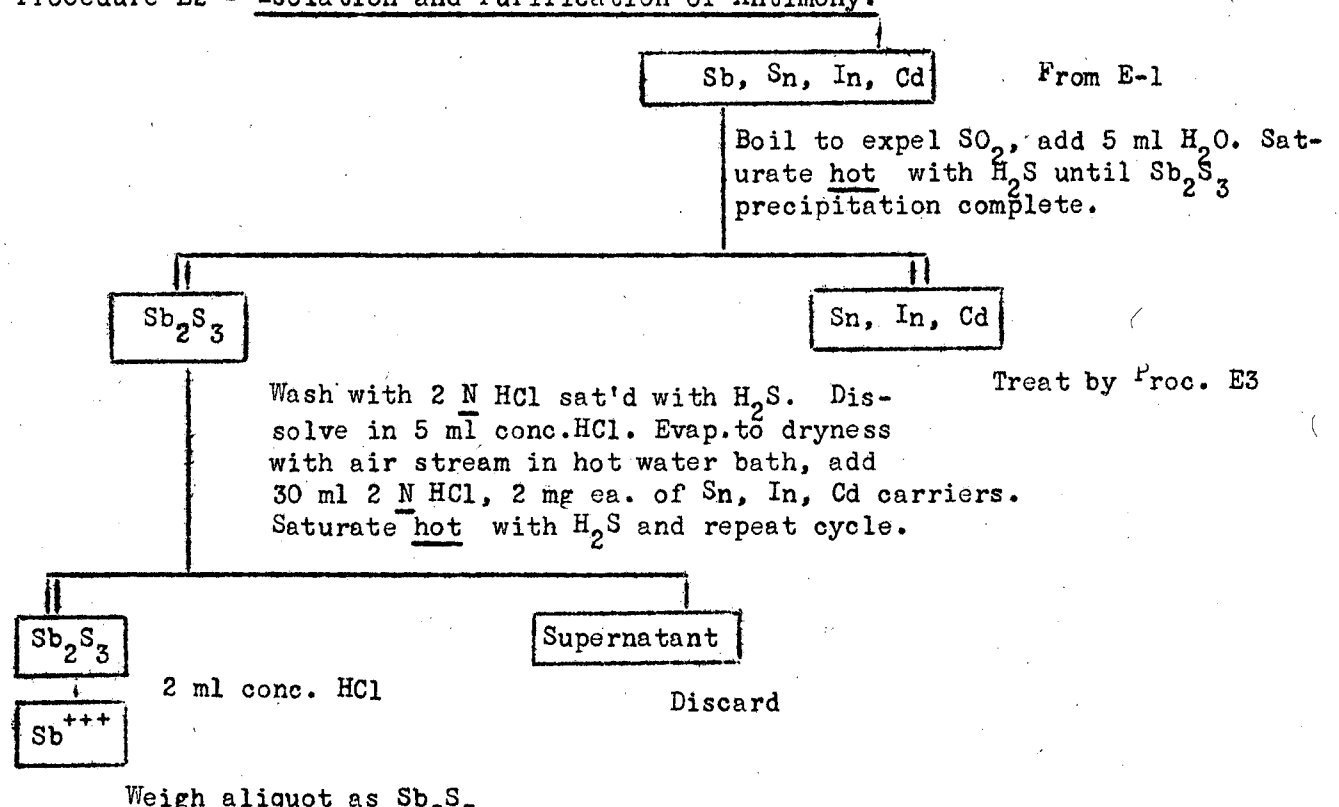
Procedure D3 - Isolation and Purification of Ruthenium.



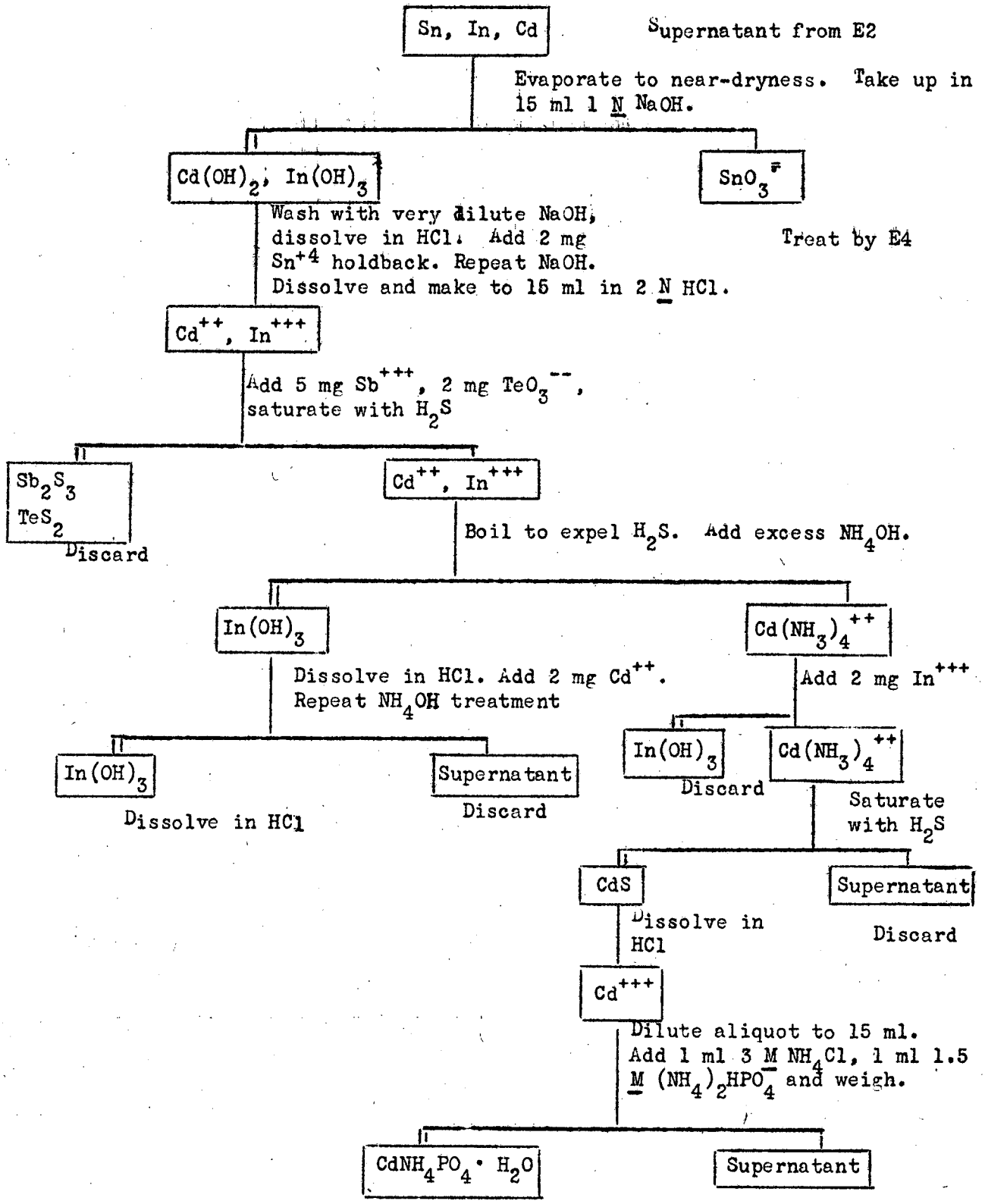
Procedure E1 - Isolation and Purification of Tellurium.



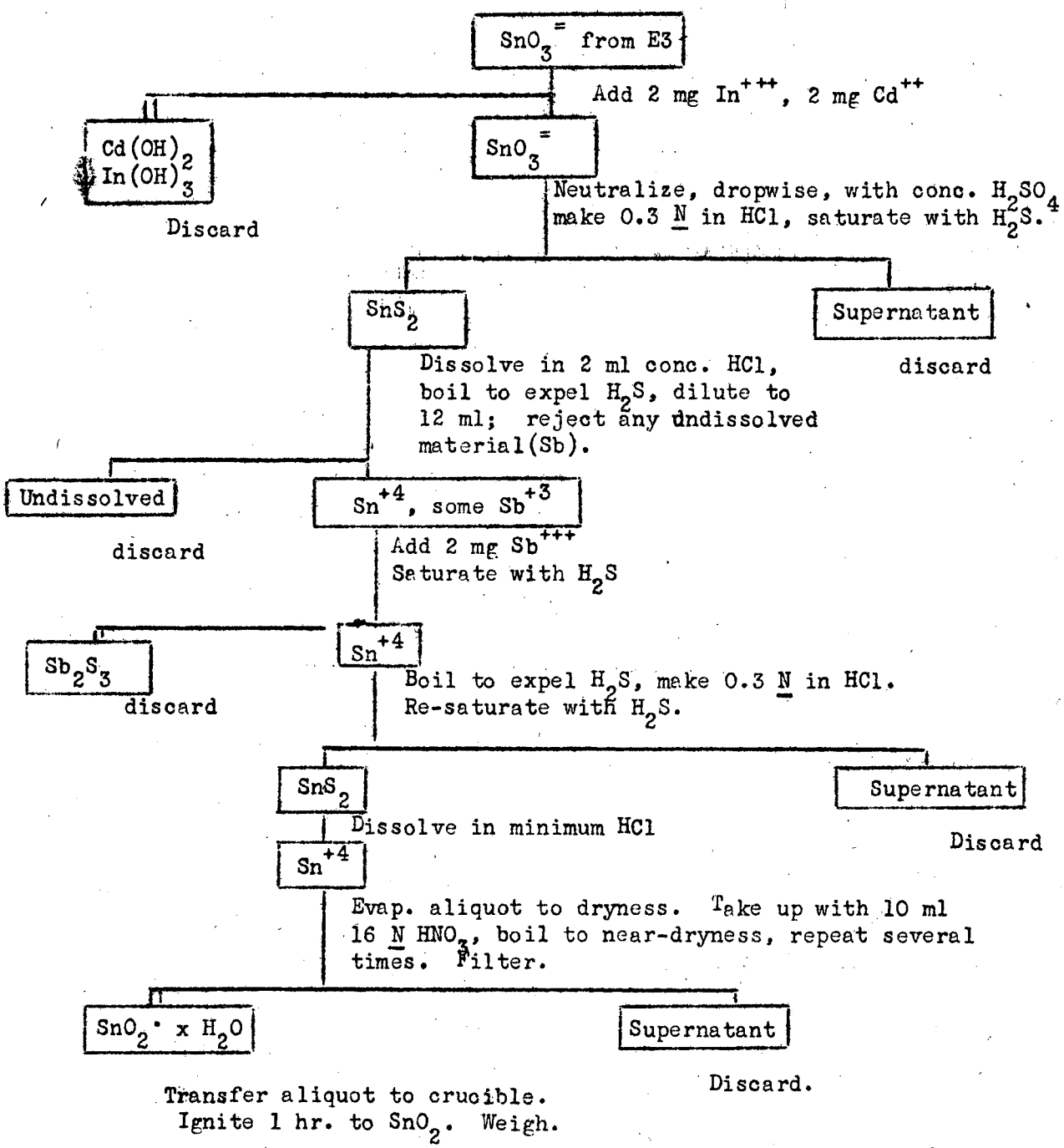
Procedure E2 - Isolation and Purification of Antimony.



Procedure E3



Procedure E-4 - Isolation and Purification of Tin



TELLURIUM FRACTION

140-day Te^{121}

This activity was ascertained through a complete absorption curve and also through a decay of at least one half-life. The "no-absorber" decay and the decay followed through beryllium both tailed off with this activity.

4.5-day Te^{119} and 6.0-day Te^{118}.

The characteristics of these 4.5-day Te^{119} and 6.0-day Te^{118} periods have been described in detail in Section II. Te^{119} was not detectable in the tellurium fraction decay followed without absorbers, being overshadowed by the 3.5-minute positron daughter of 6.0-day Te^{118}. However, when decay was followed through beryllium, approximately half of the 4-6 day activity was due to Te^{119}, the other half to Te^{118}. Lower energies (i.e.50 Mev) tend to favor the Te^{119} somewhat in relation to the Te^{118}. In all cases, the yield of the Te^{119} was determined through the presence of its 39-hour Sb^{119} daughter.

2.5-hour Te

This activity has not been extensively studied, other than to note that the decay mechanism appears to be one of positron emission. One would thus postulate a mass number of 117 or less. This appears to be borne out by the fact that, unlike the activities mentioned above, this period is not detectable at 50-Mev bombarding energy, and had undergone a sharper drop in yield at 75 Mev than the above.

ANTIMONY FRACTION

3-5-hour Sb Activities

Except at 50-Mev bombarding energy, a short-lived period with half-life ranging from three to five hours could be resolved from the decay curves without beryllium, with beryllium and with beryllium and lead. The period, or periods, thus contains a high abundance of x- and γ-rays, and electrons, indicating some conversion

This is probably a mixture of the 5.2-hour and 2.8-hour activity reported by Coleman and Pool[7]. These workers tentatively assigned the 5.2-hour period to Sb^{118} and the 2.8-hour period to Sb^{117}. Such an assignment might be compatible with the fact that these activities could only be formed, or at least detected, at energies of 75 Mev or higher.

39-hour Sb^{119}.

This 39-hour K-capture isotope was discussed in detail in Section II as the daughter of Te^{119}. It was found, however, that the activity was formed independently, at all bombardment energies, since the yield found in the Sb fraction was far too great to be accounted for on the basis of the amount grown from the tellurium in the irradiated metal between the time of bombardment and the time of separation of the tellurium fraction. The period could not be detected without absorber because of the large amount of the 2.8-day Sb^{122} present. With beryllium, however, the period was prominent, although it was naturally absent from the curve obtained by following decay through beryllium and lead.

5.7-day Sb^{120}

Described in full in Section III, this activity was very prominent in decay followed through beryllium and with beryllium and lead. It was also detectable in the decay followed with no absorber, but in the latter case the electrons were largely though not completely concealed by the large amount of Sb^{122} present.

2.8-day Sb^{122}

This beta-emitter was the predominating activity in the "no-absorber" decay but could not be detected at all in decay followed through absorbers owing to the predominance of the radiations associated with Sb^{119} and the 5.7-day Sb^{120}. As a consequence, the decay curves followed with no absorber were very different from those obtained with beryllium or with beryllium and lead. The identity of the 2.8-day activity was confirmed through a beta-ray absorption curve.

60-day Sb^{124}

All Sb decay curves "tailed" into this period which was followed over several half-lives and for which the very characteristic beta-ray absorption curve was determined in beryllium. The curve so obtained agrees closely with that reported by Meyerhoff and Scharff-Goldhaber[14].

In the bombardment of Sb with 400-Mev helium ions there is the possibility that the 2.7-year Sb^{125} be formed by $Sb^{123}(\alpha,2p)Sb^{125}$. However a beryllium absorption curve run on a sample which had stood for four months failed to reveal positive evidence of the presence of any period other than the 60-day Sb^{124}.

TIN FRACTION

4.5-hour Sn^{110}(?)

Already described in Section IV, this activity was detected as a spallation product only with 400-Mev helium ions and 200-Mev deuterons; at 100 Mev, there was no evidence of its presence in the tin fraction.

28-hour Sn^{121}

This activity appeared in bombardments at all energies from 50 to 400 Mev. Since there is no gamma ray associated with this period, it could be detected only in the decay followed without absorber.

14-day Sn (I.T.)

Over the range of bombarding energies from 50 to 400 Mev, this period, described in Section IV, is formed in yields somewhat greater than, but roughly proportional to, the 28-hour Sn^{121}. It is, therefore, likely that the activity has a mass number not far different from 121. Because Livingood and Seaborg[8] reported a 13-day tin activity which they obtained from the irradiation of cadmium with helium ions and reasoned that it could have a maximum mass number of 119, Perlman has tentatively assigned this the status of an excited state of the stable Sn^{119}.

110-day Sn^{113}

This period could be followed after decay of the 14-day isomeric activity was essentially complete. It could also be detected even in the presence of other activities by the removal of its 110-minute In^{113*} daughter.

INDIUM FRACTION

Periods of Several Hours

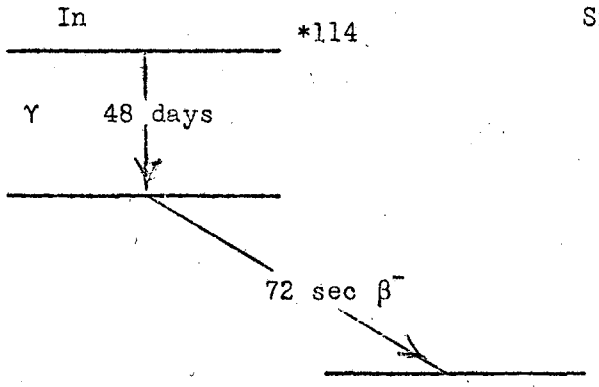
The indium fraction, like the antimony, always contained what appeared to be a complex mixture of activities ranging in half-life from less than an hour to about five hours. Consequently, resolution was not possible. It was known from following positron decay in the beta-ray spectrometer that the 20-minute positron[15] and the 65-70-minute In^{110} were certainly present, although no quantitative estimate could be made since the 105-minute In^{113*}, the 4.5-hour In^{115*}, the 54-minute In^{116} and the 117-minute In^{117} were also probably present in the complex mixture.

2.7-day In^{111}

This period which decays by electron capture was much more abundant at the higher bombarding energies than at low. It was also more prominent in decay followed through beryllium than in those decay curves taken with no absorber, due to the fact that this activity tailed into the 48-day In^{114*}, the latter being associated with less than 1% electromagnetic radiation. It was found that, on the average, there were about seven electron counts for each x-ray count associated with the 2.7-day activity. This would indicate somewhat less than 10% conversion of the gamma ray.

48-day In^{114}

This period, followed through five half-lives, was confirmed by its characteristic absorption curve through beryllium. The decay scheme is:



A beryllium absorption curve is given in Figure V-1, showing the presence of the conversion electron as well as the 2-Mev beta particle. The maximum energy of 2.0 Mev is in close agreement with that reported by Lawson and Cork[16]. The ratio of beta counts to counts due to the conversion line is seen to be roughly unity.

CADMIUM FRACTION

Short-Lived Periods

At high energies, at least one activity with a half-life of perhaps one to two hours was found in the cadmium fraction. However, as in the case of antimony and indium a graphical analysis could not be carried out successfully. Since this was not found at lower bombardment energies, it is likely that the period (or periods) is one of mass number less than the lowest stable cadmium, i.e., less than mass 106.

6.7-hour Cd^{107}

Decaying by K-electron capture, this activity could be detected by the separation of its 40-second silver daughter, Ag^{107*} as well as by its great prominence in the decay followed through beryllium. It could be only feebly detected without absorber, being masked by the shorter-lived (cf. above) and the longer-lived beta-activities (cf. below).

2.3-day Cd^{115*}

This beta-emitter was prominent in decay followed without absorber and almost undetectable in the decay followed through beryllium. The yield could also be verified by the separation, after equilibrium, of its 4.2-hour daughter, In^{115*}.

42-day Cd^{115}

No gamma being associated with this activity, it could only be observed in the decay followed without absorber. It decays directly to the ground state of In^{115}, hence has no detectable daughter.

330-day Cd^{109}

This long-lived activity was detectable in the decay followed through beryllium shortly after the 6.7-hour activity decay was essentially complete. Ultimately, of course, it was also detectable in the no-absorber decay after the 42-day activity was gone. Cd^{109} also has a 43-second Ag^{109*} daughter which could be separated from the parent. Figure V-2 shows the relative counting rate of this silver daughter with no absorber, and with nearly two grams of beryllium. The ratio of fifty, uncorrected for about 4-5 milligrams of air and mica window, indicates that the 0.094-Mev gamma ray is highly converted.

From the description of the cadmium activities, it is clear that the decay curves obtained with and without absorber are entirely different, the Cd^{107} and Cd^{109} being prominent in the absorber decay, while short-lived (probably positron) activities and the beta-emitters Cd^{115*} and Cd^{115} constituted most of the no-absorber decay.

SILVER FRACTION

Although the silver fraction contained several short-lived activities including at least the 24-minute Ag^{106}, a 70-minute period, and the 3.2-hour Ag^{112}, it was impossible to unambiguously analyze the decay curve followed without absorber.

24-minute Ag^{106} β^+

Although its presence could not be quantitatively determined, this period was found in the positron decay observed on the beta-ray spectrometer. It could not be satisfactorily analyzed in the decay curve followed on the Geiger counter.

70-minute β^+, K

This period, probably that reported by Enns[17] was detectable along with the 24-minute Ag^{106} described above, in the positron decay followed on the beta-ray spectrometer. It could not be satisfactorily resolved out of the decay curve of the silver fraction followed without absorber. However, through beryllium, this period is very prominent; in fact, it is the only short-lived activity found in the absorber decay. This fact leads one to conclude that the 70-minute period decays both by positron emission and by K-capture. The branching ratio could not be determined because of the presence of the other activities but it is likely that the greater percent decays by electron capture.

7.5-day Ag^{111} and 8.2-day Ag^{106}

These two activities, at first thought to be one and the same, both occur in spallation. The 7.5-day Ag^{111}, being a beta-emitter with no associated gamma radiation[18] would not show up in the decay followed through beryllium. Consequently when an activity of about this period did occur under the latter conditions, it was necessarily due to the x- and gamma-radiation associated with the 8.2-day Ag^{106} period. Furthermore, it was shown by Feather and Dunworth[19] that the ratio of beta to gamma radiation counts was 1.05 in Ag^{106}, so that only about 1% conversion accompanies the decay. As a result, the yields of the isotopes under consideration were such that nearly all of the 8-day activity followed without absorber was due to Ag^{111}, while all of the 8-day activity followed through beryllium was due to Ag^{106}.

It was found, furthermore, that the 8-day counting rate through beryllium decreased with bombarding energy in a manner different from the corresponding rate without absorber--further proof that two different 8-day periods are present.

45-day Ag^{105} (?)

There is doubt as to the mass assignment. However, this activity was found in bombardments at 200 and at 400 Mev. It was more prominent in decay followed through beryllium than in decay with no absorber. Resolution from the decay curve was difficult owing to the fact that the activity "tailed" into the 225-day Ag^{110}--a time long compared with the time of the experiment.

225-day Ag^{110}

For the reason mentioned above, it was difficult to obtain an entirely satisfactory resolution of the 225-day and 45-day periods.

3.2-hour Ag^{112}

This activity was observed in all bombardments from 50 to 400 Mev. It was less prominent at 400 and 200 Mev owing to the increased abundance of other silver activities at these higher energies. At the lower energies the period was easily confirmed by the absorption curve and beta-ray distribution curve of the very hard beta-particle.

PALLADIUM FRACTION

The palladium fraction contained so many activities whose half-lives were of the same order of magnitude that the resolution of the decay curves proved almost impossible. Instead, daughter separations were made wherever possible.

In bombardments below 200 Mev, the yields of all activities of or below palladium were so small that they were not measured.

21-hour Pd^{112}

This period could easily be identified by its 3.2-hour Ag^{112} daughter. Decrease in yield of the latter as a function of time served to determine the

amount of Pd^{112} present.

26-minute Pd^{111}

When the initial palladium separation was performed rapidly enough so that the final purified fraction still contained some of the 26-minute period, the first silver daughter fraction contained the 7.5-day Ag^{111} as well as the 3.2-hour period.

12-hour Pd^{109}

Also a beta-emitter, this period was determined by the separation of its 43-second daughter Ag^{109*}. It was assumed that all the 109 isotope decayed to this excited state of silver.

17-day Pd^{103}

When the palladium was allowed to age for about five weeks, the sole remaining period was this activity. It was characterized by almost pure x-rays. Actually, its Rh^{103*} daughter has a conversion line, but its range is less than that of the mica window and air present in the counting arrangement.

10-hour Pd^{101}

This period was detected by measurement of the decreased yield of 4.2-day Rh^{101} daughter separated at successive time intervals. A complete description of this isobaric pair is given in Section I.

4.0-day Pd^{100}

Also described in Section I, this activity was measured, as above, by the successive decrease in the amount of 21-hour Rh^{100} daughter separated from it.

RUTHENIUM FRACTION

4.5-hour Ru^{105}

Ru^{105} was the only short period found in the ruthenium fraction. Since the decay chain is

$$Ru^{105} \xrightarrow{4.5\text{-hr. }\beta^-} Rh^{105} \xrightarrow{1.5\text{ d. }\beta^-} Pd^{105}$$

the yield of the Ru^{105} was checked by allowing a sample of the ruthenium fraction

to decay for one day, distilling the ruthenium from perchloric acid and collecting the residual Rh^{105} daughter.

2.7-day Ru^{97}(?)

This activity could be detected in the decay both with and without absorber but was more prominent in the former.

42-day Ru^{103}

Detectable in the no-absorber decay, this period and the 2.7-day Ru^{97}(?) were also found in the ruthenium distilled from the Rh^{105} daughter after aging for one day.

One-year Ru^{106}

This period was detected through the tailing of the 42-day Ru^{103} and earlier through a very hard beta-ray component which was presumed to be due to the 30-second Rh^{106} daughter of Ru^{106}. The relative quantities by the two methods were in essential agreement.

MOLYBDENUM FRACTION

67-hour Mo^{99}

Mo^{99} was the only activity found in the molybdenum fraction, no activity shorter than one day being looked for. The beta-ray absorption curve confirmed the activity, which was followed through six half-lives.

YTTRIUM FRACTION

80-hour Y^{87}

No yttrium activities of half-life less than one day were sought. Initial growth in the decay curve suggested the pair

$$Y^{87} \xrightarrow{K} Sr^{87*} \xrightarrow{2.7\ hr.} Sr^{87}$$

This was confirmed by separating from an equilibrium mixture the 2.7-hour Sr^{87*} in the following manner. To the $Y(NO_3)_3$ solution was added 2 mg. $Sr(NO_3)_2$.

This was evaporated to dryness and taken up in fuming nitric acid. The insoluble $Sr(NO_3)_2$ was centrifuged, washed, dissolved in one drop of water, and repurified by addition of 2 mg. Y^{+++} and repetition of the above procedure.

The decay curve for the yttrium fraction did not give very close agreement with the value of 80 hours for the half-life of Y^{87}. This suggests that the 62-hour Y^{90} was also present, although in an indeterminable amount.

57-day Y^{91}

The 80-hour yttrium tailed into this period which was followed for nearly three half-lives.

TABLE V-1

SPALLATION PRODUCTS OF ANTIMONY IN RELATION TO PERIODIC TABLE

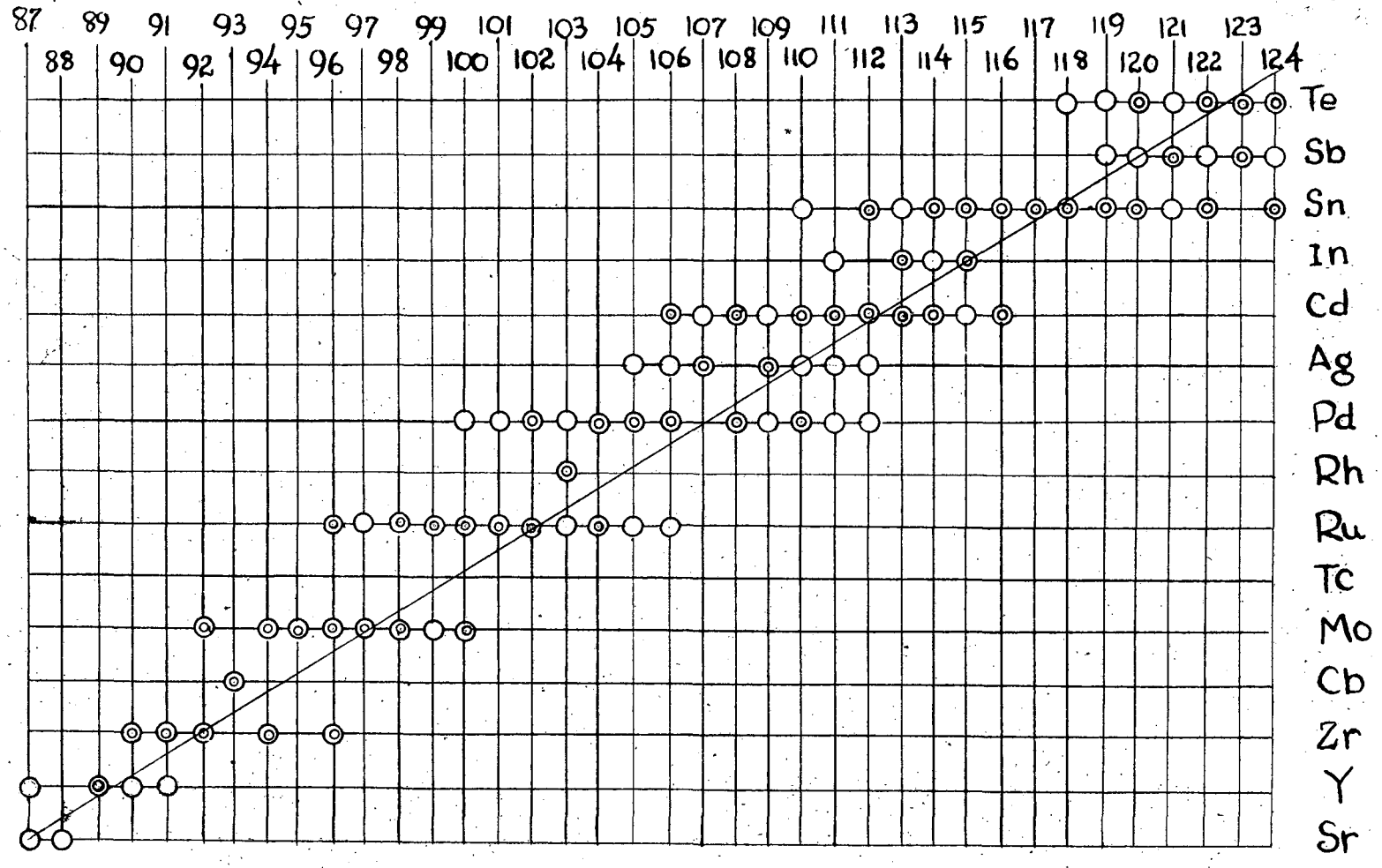


TABLE V-2

CROSS SECTION IN BARNS FOR FORMATION OF SPALLATION PRODUCTS OF ANTIMONY AT VARIOUS ENERGIES

ELEMENT	MASS	$T_{1/2}$ TYPE	400α	200D	200D	200D	100D	75D	50D
Te	121	140d IT			0.013	0.009			
	121	17 d K							
	119	4.5d K	0.067		0.021	0.023	0.066	0.065	0.077
	118	6.0d K	0.029		0.018	0.017	0.053	0.031	0.023
		2.6hr β^+	0.020	-	-	0.021	0.020	0.0044	$< 7 \times 10^{-5}$
Sb	124	60d β^-	0.015		0.0011	0.0016	0.0025	0.0015	
	122	2.8d β^-	0.072		0.042	0.045	0.065	0.028	0.031
	120	5.7d K	0.18		0.070	0.14	0.16	0.040	0.047
	119	39hr K	0.10		-	0.16	0.18	0.063	0.029
Sn	121	28 hr β^-	0.0022		-	-	8.1×10^{-4}	3.2×10^{-4}	1.4×10^{-4}
	?	14d IT	0.028		0.017	-	0.017	0.0056	0.0022
	113	110d K	0.030		0.017	-	0.0062	7×10^{-4}	$< 3 \times 10^{-5}$
	110?	4.5hr K	0.008				$\leq 10^{-5}$		
In	114	48d IT, β^-	0.023	0.008	0.015	0.016	0.0051	7.4×10^{-4}	$< 2 \times 10^{-6}$
	111	2.7d K	0.16		0.08	0.065	9.5×10^{-4}	$\leq 10^{-5}$	-
Cd	115*	2.3d β^-	3.5×10^{-4}	1.1×10^{-4}	1.5×10^{-4}	1.8×10^{-4}	2.6×10^{-5}	4.1×10^{-6}	$\leq 1.7 \times 10^{-6}$
	115	42d β^-	1.1×10^{-3}		7.0×10^{-4}	6.5×10^{-4}	1.1×10^{-4}	8.8×10^{-6}	$\leq 4.1 \times 10^{-6}$
	109	330d K	0.014	2.7×10^{-3}	3.2×10^{-3}	3.6×10^{-3}	$\leq 1.3 \times 10^{-4}$		
	107	6.7hr K	0.043	-	-	0.005	$< 5 \times 10^{-6}$	-	-
Ag	112	3.2hr β^-	5.7×10^{-4}	0.0014		2.8×10^{-4}	3.8×10^{-5}	2.9×10^{-6}	4.2×10^{-7}
	111	7.5d β^-	2.3×10^{-3}	2.2×10^{-3}	4.5×10^{-4}	8.0×10^{-4}	8.0×10^{-5}	3.5×10^{-6}	$< 1.7 \times 10^{-7}$
	110	225d K		7.6×10^{-3}					
	106	8.2d K	3.5×10^{-2}	1.5×10^{-2}	7.0×10^{-3}	1.2×10^{-2}	$< 10^{-5}$		
	105?	45d K		1.0×10^{-2}					

Table V-2(cont'd)

CROSS SECTION IN BARNS FOR FORMATION OF SPALLATION PRODUCTS OF ANTIMONY AT VARIOUS ENERGIES

ELEMENT	MASS	$T_{1/2}$ TYPE	400a	200D	200D	200D	100D	75D	50D
Pd	112	21hr β^-		9×10^{-7}	7×10^{-7}		$<10^{-7}$		
	111	26min β^-		3×10^{-5}					
	109	13hr β^-		8×10^{-6}	4.7×10^{-5}				
	103	17d K		1.2×10^{-4}	5.5×10^{-5}				
	101	8-10hr β^+,K							
	100	4d K		2.4×10^{-4}	2.3×10^{-6}				
Ru	106	1yr β^-		-	2.1×10^{-6}				
	105	4.2hr β^-		8×10^{-6}	-				
	103	40d β^-		4.1×10^{-6}	3.9×10^{-6}				
	97	2.7d K		6×10^{-6}	1.4×10^{-5}				
Mo	99	67hr β^-			2.3×10^{-6}				
Y	91	57d β^-			9×10^{-7}				
	87	80hr K			3×10^{-7}				

QUANTITATIVE RESULTS OF SPALLATION OF ANTIMONY AND DEPENDENCE ON BOMBARDMENT ENERGY

Table V-1 depicts a chart of the region studied. The hollow circles represent radioactive products positively identified in the spallation process, the concentric circles the naturally occurring stable species of the elements.

The data in Table V-2 comprise the results of yields for thirty six different activities for bombarding energies of 50, 75, 100-and 200-Mev deuterons, and for 400-Mev helium ions.

Serber's theory[20] of high-energy nuclear transformations obtained on the 184" cyclotron predicts the formation of products ranging from few particles ejected from the compound nucleus to many ejected. Even a brief inspection of Tables V-1 and V-2 shows this to be so. According to this theory many-particle ejection would result from the projectile particle "sticking" in the nucleus and distributing its energy over the entire nucleus, in which case the resulting reaction would be explained on the basis of the liquid drop model, with subsequent evaporation of many nucleons. Such a product is typified by Mo^{99}. On the other hand, the mean free path of a high-energy projectile in a nucleus being very large, a "grazing" collision might result in only a comparatively small fraction of the total energy of the projectile transferred to the nucleus, with a consequent loss of very few particles. Thus, products such as Sb^{124}, Sb^{122}, and Te^{121} are obtained, products which would be expected of deuterons of much lower energy, say, 20 Mev.

Perhaps the stripping process[21] should be mentioned at this point. By this mechanism, either of the two component particles of a deuteron may imbed itself in a target nucleus while the other component continues along its path. Thus the deuteron is "stripped" of either its neutron or proton. In effect then, because of stripping, three incident particles are possible: the deuteron, the neutron, and the proton. However, it has been estimated by Horning[22] that the stripping

process in the region of antimony occurs for only about 20% of the deuterons. Thus, although this value is appreciable, nevertheless unchanged deuterons are responsible for most of the transformations observed.

These general ideas having been noted, more specific details from Table V-2 will now be considered.

DECREASE IN YIELD WITH DEUTERON ENERGY AS DEPENDENT ON MASS NUMBER

It will be noted that the yields of most antimony and tellurium isotopes are relatively constant in the range of 50-Mev to 200-Mev deuterons. These isotopes all have mass numbers within about five mass units of the target nuclei. The one exception to this is the 2.6-hour tellurium activity, which, because of its short half-life probably has a low mass number.

The yield of Sn^{121}, as compared to that of Sn^{110} and Sn^{113} is also relatively constant with varying bombardment energy.

One is therefore led to assume that the picture is rather general: yields of products of high mass numbers are less affected by changes in deuteron energy than are products of relatively low mass numbers. Thus, the fourteen-day excited state of tin described in Section IV is thought to be of relatively high mass number, since its yield varies with energy in approximately the same manner as does that of Sn^{121}. It will be noted that nothing has thus far been said about the magnitude of the cross-sections observed; merely the change in cross section with energy.

The analogy is seen to hold roughly throughout the entire region in which data are available; e.g., the yields of Cd^{107} and Cd^{109} decrease much more markedly than do either of the isomers of Cd^{115}. Ag^{105} and Ag^{106} decrease very much more rapidly than do either Ag^{111} or Ag^{112}, as the energy available for disintegration is decreased.

The facts observed from such variation in yield with energy appear to have a rather simple explanation: as more energy becomes available for the formation of lower mass numbers, then such reactions will naturally occur.

POSSIBLE MECHANISM FOR SPALLATION AT 200 MEV

There is, however, a fact which is not obvious at all. It will be noted that, at 200 Mev, the yields of Cd^{107} and Cd^{109} greatly exceed that of both isomers of Cd^{115}. The same is true for Ag^{105} and Ag^{106} as compared to Ag^{111} and Ag^{112}, as well as In^{111} compared to In^{114}, and Sn^{110} and Sn^{113} as compared to Sn^{121}. For 400-Mev helium ions, the situation is emphasized to an even greater extent. Why then do the yields of the neutron-deficient isotopes attain values so much greater than those ever attained by the "heavier" isotopes, or beta emitters? Although no simple explanation is immediately obvious, one argument may be advanced as follows:

A highly-excited nucleus might emit either neutrons, protons, or alpha particles. Table V-3 gives a rough measure of the average binding and coulomb energies for these particles in the antimony region[22].

Table V-3

Particle	B.E. (Mev)	C.E. (Mev)	Total
n	8	0	8
p	8	8	16
α	3	16	19

Only about 8 Mev are therefore required for neutron ejection as compared to twice this quantity for either proton or alpha particle emission. Although sufficient energy is certainly available for emission of any of these, multiple neutron emission would seem to be the most likely especially since there are more neutrons than protons. Of course, the above value of neutron binding energy

applies only to the first neutron emitted, or perhaps the first several. As the nucleus becomes further degraded of neutrons, the binding energy of the neutrons is increased, with the result that the excited nucleus will then emit protons. Consequently, it might be expected that the most probable reaction would be multiple neutron emission followed by a number of protons sufficient to produce a nucleus relatively stable with respect to its total remaining neutrons and protons. Nuclei so formed will be neutron-deficient, hence lie to the left of the line of maximum stability shown in Table V-1. This line represents a certain relationship between the neutrons and protons in a nucleus, given by

$$Z = \frac{A}{2}\left(\frac{1}{1 + \frac{\gamma A^{2/3}}{4\alpha\epsilon}}\right) \qquad \text{Eq. (1)}$$

Eq. (1) results from obtaining the minimum (i.e., the derivative) of the expression[23,24] for the binding energy of a nucleus Z^A

$$E = A\epsilon - \alpha\epsilon\frac{(A-2Z)^2}{A} - \frac{\gamma Z^2}{A^{1/3}} \qquad \text{Eq. (2)}$$

where α, γ and ϵ are constants.

In eq. (1), Z is not necessarily an integer. Several values of Z have been calculated for various values of A, the line connecting these thus being the line of maximum stability - i.e., for a given Z, the binding energy is a maximum.

The binding energy per particle is only slightly higher on either side of this line so that one must not expect only those isotopes to form which lie alongside - or very close to - this line. Furthermore, since neutron emission seemed a likely means for the excited nucleus to lose its excess energy, it might be expected that the greater yield of the product nuclei would lie on the neutron- deficient side of this line. This is certainly the case. All of those isotopes which lie to the right of the stability line, from Sn to Ag, are formed in lower yield at 200 Mev than those which lie to the left of this line (This statement is readily

verified by inspection of Tables V-1 and V-2. Ag^{110} is formed at 200 Mev in yield much larger than for either Ag^{111} or Ag^{112} - in fact, in yield comparable to that of Ag^{105} and Ag^{106}. This was at first puzzling; but if we admit of the above explanation, it is seen that Ag^{110}, unlike the low-yield group of beta emitters, actually falls on the line of maximum stability and could thus be formed by the neutron-emission process. Another result of this idea is that In^{114}, a beta emitter, is formed in very much higher yield than any other beta emitter. This also was difficult to understand until it was seen that In^{114} actually lies on the left side of the line of maximum stability, hence can be reached via multiple neutron emission.

The fact that isotopes are formed at all to the right side of this line is easily explained on the basis of alpha particle emission. This process may be less likely than neutron emission but it certainly occurs. Since alpha emission increases the ratio of neutrons to protons in a nucleus, beta-emitters should be expected to result from alpha particle ejection, but in lower yield than the neutron-deficient isotopes resulting from multiple neutron ejection. This phenomenon is precisely what is observed.

To carry the case further, as the energy available for disintegration is decreased, alpha particle emission may become favored over the emission of the equivalent in neutrons and protons; hence the beta emitters would become more prominent as the energy becomes decreased.

DISCONTINUITY OF YIELDS AT PALLADIUM

While the above ideas seem satisfactory for those elements from tin to silver, yields of isotopes below silver do not appear to fit into this scheme. It will be noticed, for example, that the yields of Pd^{100} and Pd^{103} are not appreciably different from those of Pd^{111} and Pd^{112}.

But even more noteworthy is the fact that the yields themselves of palladium, ruthenium, etc., are suddenly several orders of magnitude lower than those of silver, cadmium, etc. A possible explanation of this phenomenon might be that Pd^{111} and Pd^{112} would be formed in lower yield than the corresponding silver isobars since the former involves emission of a charged particle not required for the formation of Ag isotopes. It is to be noted that the yield for both Ag and Pd is in the direction 111>112. Thus Pd^{112} is farther from the line of maximum stability, as inspection of Figure V-1 shows. As for the low-mass palladium isotopes, the small yield may simply be due to the fact that the energy available for the production of them is near the threshold, for a total of about twenty mass units must be ejected to arrive at these palladium activities. One might therefore expect that neutron-deficient palladium isotopes would be formed in much greater yield with 400-Mev helium ions. The heavier palladium isotopes might, on the other hand, show a relatively small increase in yield.

ISOBAR FORMATION

The example cited above of the independent formation of Pd^{112} and Ag^{112} is one of several cases of isobar formation found in antimony spallation. Others are: Te^{121} and Sn^{121}, Te^{119} and Sb^{119}, Ag^{112} and Pd^{112}, In^{111}, Ag^{111} and Pd^{111}, Sn^{110} and Ag^{110}, Ag^{106} and Ru^{106}, Ag^{105} and Ru^{105}, and Pd^{103} and Ru^{103}. The isobars of mass 119 are formed in similar yield because of the possibility of forming Sb^{119} by a neutron loss reaction, but in every other case except the 110 isobaric pair, the isobar of higher atomic number is formed in greater yield. It will be noted from Table V-1 that in all cases except that of Ag^{110}, the isobar of lower Z lies to the right of the line of maximum stability, the isobar of higher Z to the left of this line. Hence one should expect greater yields of the latter since they can be formed by emission of fewer charged particles. In the case of the 110 isobaric pair, however, Ag^{110} itself lies at the most stable configuration.

This might be expected to somewhat counter the effect of neutral particle emission to the extent that the yield of the two periods might be comparable. This is observed.

It is interesting to note that, in that instance in which data are available, the yield of the isobar with higher Z approached that of the isobar with lower Z as the bombarding energy is decreased. The case of In^{111} and Ag^{111} demonstrates this. At 75 Mev bombarding energy, the yields of these isobars are probably similar.

RELATIVE YIELDS OF (d,p) AND (d,p2n) REACTIONS (OR THEIR EQUIVALENTS)

The cross-section for formation of Sb^{122} with 200-Mev deuterons is approximately an order of magnitude greater than that of Sb^{124}. The latter can be formed only by

$$Sb^{123}(d,p)Sb^{124}$$

or by the equivalent neutron capture reaction. However, Sb^{122} can be formed by two reactions:

$$Sb^{121}(d,p)Sb^{122} \qquad \text{and by}$$

$$Sb^{123}(d,p2n)Sb^{122}$$

It therefore appears that the relatively greater yield of Sb^{122} in comparison to Sb^{124} is due to the (d,p2n) reaction. The d,p2n reaction is therefore about thirty times as probable as the (d,p) reaction for 200-Mev deuterons. This ratio has decreased to about 20 for 75-Mev deuterons, and is in the direction which would be anticipated since lowering of the bombarding energy has increased the cross section for fewer particles ejected.

An interesting corollary of the above reaction is the cross-section for formation of Sb^{120} by the reactions:

$$Sb^{121}(d,p2n)Sb^{120}$$

$$Sb^{123}(d,p4n)Sb^{120}$$

If one assumes the cross-section for formation by the former process to be the same as that of Sb^{122} from Sb^{123}, then the difference in the actual values and those for the (d,p2n) reaction should give an indication of the cross-section for the (d,p4n) reaction. These differences are given as for the various bombarding energies in Table V-4:

Table V-4

energy	for d,p4n
400	0.11 barns
200	0.03
200	0.03
100	0.10
75	0.012
50	0.016

Although these values are not very well suited to quantitative expression, the trend is probably significant and would indicate that the cross-section for (d,p4n) decreases somewhat like that of the (d,p2n) reaction, as energy is decreased.

ISOMER FORMATION

An interesting situation has presented itself in the cadmium fraction. Cd^{115} is known to consist of two independent isomers decaying as follows:

$$Cd^{115} \xrightarrow{42\ d\ \beta^-} In^{115}_{4.2\text{-hr I.T.}}$$

$$Cd^{115*} \xrightarrow{2.3\ d\ \beta^-} In^{115*} \xrightarrow{4.2hr\ I.T.} In^{115}$$

In slow-neutron fission of uranium[25] and in the fission of thorium with 40-Mev helium ions[26], the ratio of 2.3-day Cd^{115} to 43-day Cd^{115} is ten. This has been suggested as evidence by Newton[26] that the Ag^{115} precursor of these two isomers branch decays by this ratio.

Seren, Engelkemeier, et al.[27] have reported that the ratio of 2.3-day to 43-day isomer formed in slow-neutron capture of cadmium is about five. On the other hand, it is stated that the fast-neutron formation of Cd^{115} from indium via

$$In^{115}(n,p)Cd^{115}$$

gives a ratio for 2.3-day to 43-day isomer of a bout one.

It has been observed in antimony spallation (cf. Table V-2) that the ratio of 2.3-day to 43-day Cd^{115} is about one-third. Furthermore, this ratio remains sensibly constant over the range of bombarding energies from 400 Mev (helium ions) to 50 Mev (deuterons).

One may thus summarize in Table V-5 the relative yields of the Cd^{115} isomers as obtained in various processes.

Table V-5

COMPARISON OF ISOMERIC RATIO OF 2.3-DAY AND 43-DAY Cd^{115}

Method	Ratio, 2.3 d/43 d
fission	10
slow n capture	5
$In^{115}(n,p)$	1
Sb spallation	1/3

Although several other cases of nuclear isomerism occur in this region (e.g. Te^{121}, Sb^{120}, and Ag^{106}) none is suitable to determination with the certainty of the Cd^{115} pair.

ACKNOWLEDGEMENTS

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APPENDIX

Beryllium Absorption Characteristics

In the introduction it was pointed out that because of the frequent occurrence of K x-rays whose half-thickness could become confused with that of many beta particles in absorption curves run with aluminum absorbers, it was necessary to resort to the use of beryllium to distinguish between beta particles and x-rays of about 20-25 kilovolts energy. Since it was desirable - at least in the case of previously unreported isotopes - to run Feather analyses on many beta-ray absorption curves, it was essential to have beryllium absorption data for some standard beta ray whose absorption characteristics have been studied with aluminum. Feather[5] has determined the range of the β^- particle from radium E (Bi^{210}) to be 476 mg/cm^2 of aluminum. Since the electron density of most materials is essentially constant, and since beta particle absorption is due chiefly to the interaction of beta rays with these electrons, one can arbitrarily take the exact maximum range of the radium E beta particles as 476 mg/cm^2 of matter. Likewise Feather found that the maximum range for the beta particles of UX_2 (1.14-m Pa^{234}) was 1104 mg/cm^2 of Al, and so for any absorber. If these values be assumed for the exact range of RaE and UX_2 in beryllium, then it is possible, through the use of the beryllium absorption curves shown in Figs. A-I and A-II to construct "Feather Analyzers", devices by means of which it is possible to compare the range of an unknown beta ray over its entire range, with the curves for known beta rays, RaE and UX_2. A summary discussion of the method of the Feather Analysis has been treated by Glendenin[28].

The RaE curve was obtained from a sample of 20-year RaD-E-F in equilibrium. The soft RaD beta was initially stopped by 15 mg of aluminum.

The UX_2 absorption curve was run on a sample of UX_1-UX_2. The UX_1 (24-day Th^{234}) was co-precipitated on LaF_3 from a solution of uranyl nitrate. This

procedure carries the Th^{234} on the LaF_3 leaving other activities in solution. The 1.14-min. Pa^{234} soon grows in. Figs. A-I and A-II show that the ranges in beryllium are indeed visually similar to those in aluminum given by Feather. Of course, the arbitrary values mentioned above were used.

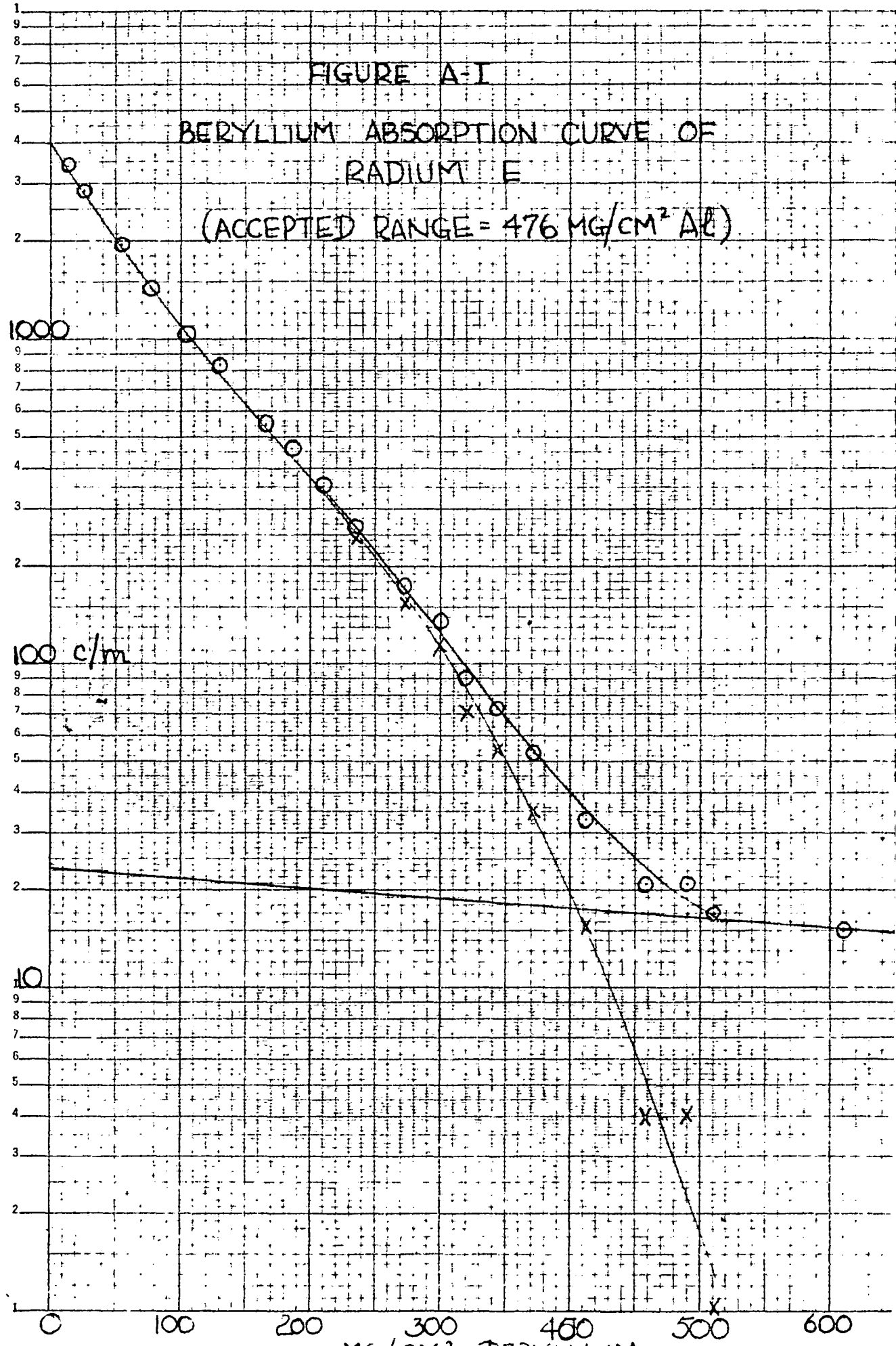
FIGURE A-I
BERYLLIUM ABSORPTION CURVE OF RADIUM E
(ACCEPTED RANGE = 476 MG/CM² Al)
1000
100 c/m
10
0
100
200
300
400
500
600
MG/CM² BERYLLIUM

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