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SOME NEW NEUTRON DEFICIENT RADIOACTIVE ISOTOPES OF THE RARE EARTH REGION

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Some New Neutron Deficient Radioactive Isotopes of the Rare Earth Region

Harry G. Hicks

ABSTRACT

Neutron deficient radioactive isotopes of neodymium, promethium, terbium, holmium, thulium, hafnium, tantalum, and rhenium have been prepared by cyclotron bombardment employing helium ions, deuterons, protons and fast neutrons from the 60-inch cyclotron of the Crocker Radiation Laboratory of the University of California.

Their radiation characteristics and half-lives have been measured. Mass allocations have been determined by chemical identification, and by cross-bombardment and yield techniques.

A limited number of these have been observed as products from 200 Mev deuteron bombardments of tantalum with the 184-inch cyclotron.

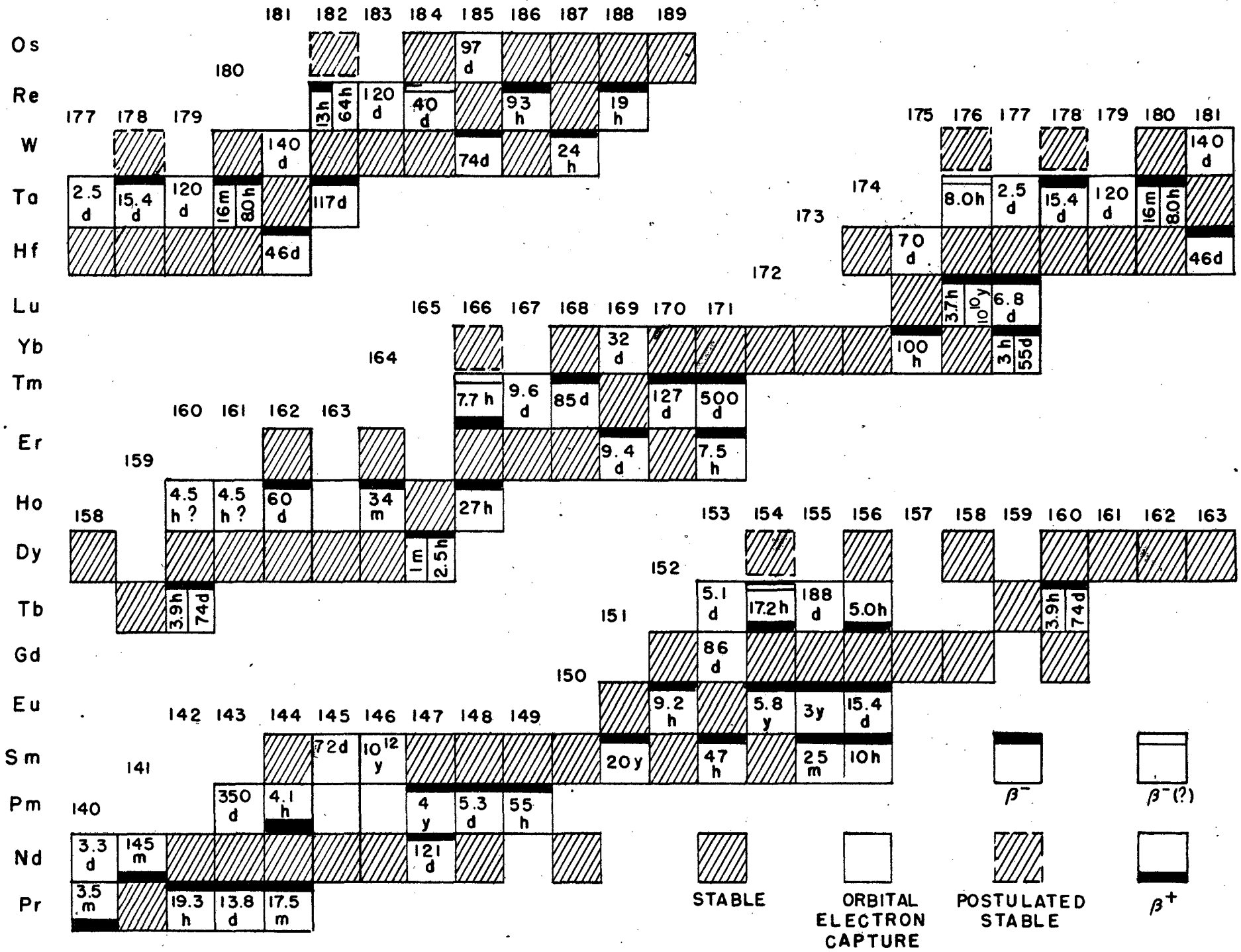
The production and properties of these new isotopes are summarized in Table I.

Table I

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	γ-rays	Produced by
Nd^{140}	K	3.3±0.1 days			Pr-d-3n
Pm^{144}	β^+	4.1±0.1 hours	1.3(β^+)		Pr-α-n
Tb^{153}	K,e^-,γ	5.1±0.1 days	0.15,0.2-0.4(e^-)	L,K x-rays 0.23,1.2	Eu^{151}-α-2n
Tb^{154}	K,e^-,β^+, γ(β^-?)	17.2±0.3 hours	0.13,0.5(e^-) 2.6(β^+)	L,K x-rays 1.3	Eu^{151}-α-3n Eu^{153}-α-n
Tb^{155}	K,e^-,γ	188 days	0.1(e^-)	L,K x-rays 1.4	Eu^{153}-α-2n
Tb^{156}	β^+	5.00±0.05 hours	1.3	L,K x-rays	Eu^{153}-α-n
Ho^{164}	β^-	34±1 minutes	0.95		Dy-p-n
Ho^{162}	K,e^-,β^-	60±2 days	0.12(e^-) 0.7(β^-)		Tb-α-n
$Ho^{160,161}$	K,e^-,β^+,γ	4.5±0.1 hours	0.09,>0.6(e^-) ~2(β^+)	L,K x-rays 1.1	Dy-p-n Tb-α-n or 2n
Tm^{166}	K,β^+,e^-,γ β^-	7.7±0.1 hours	0.24(e^-) ~1 (β^-)2.1(β^+)	L,K x-rays 1.7	Ho-α-3n Er-p-n
Tm^{167}	K,e^-,γ	9.6±0.1 days	0.21(e^-)	L,K x-rays 0.22,0.95	Ho-α-2n Er-p-n Ta-d-5z,16a

Table I(Cont'd)

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	γ-rays	Produced by
Tm^{168}	K, e^-, γ, β^-	85±2 days	0.16(e^-) 0.5(β^-)	L, K x-rays 0.21, 0.85	Ho-α-n Er-p-n
Hf^{175}	K, e^-, γ	70±2 days	0.3(e^-)	L, K x-rays 0.35	Lu-p-n Lu-d-2n
Ta^{176}	K, e^-, γ, β^-(?)	8.0±0.1 hours	0.12, 0.18(e^-) >1(β^-?)	L, K x-rays ~2	Lu-α-3n Ta-d-p, 6n
Ta^{177}	K, e^-, γ	2.50±0.05 days	0.11(e^-)	L, K x-rays ~1.4 (weak)	Lu-α-2n, 3n Hf-p-n Ta-d-p, 5n
Ta^{178}	β^-, γ	15.4±0.2 days	1.4	Very weak	Lu-α-n, 2n Hf-p-n
Ta^{179}	K, e^-, γ	>100 days			Lu-α-n Hf-p-n
Re^{182}	K or IT, e^-, γ, β^-(?)	12.7±0.2 hours	0.16(e^-) ~1(β^-?)	L, K x-rays 0.4, 1.8	Ta-α-3n W-p-n
Re^{182}	K or IT, e^-, γ	64.0±0.5 hours	0.11, 0.27(e^-)	L, K x-rays 0.22, 1.5	Ta-α-3n W-p-n
Re^{183}	K, e^-, γ	120 days	0.16(e^-)	L, K x-rays 1.0	Ta-α-2n W-p-n


Os
Re
W
Ta
Hf
Lu
Yb
Tm
Er
Ho
Dy
Tb
Gd
Eu
Sm
Pm
Nd
Pr
181 182 183 184 185 186 187 188 189
180
97 d
13h 64h 120 d 40 d 93 h 19 h
140 d 74d 24 h
2.5 d 15.4 d 120 d 16m 8.0h 117d
46d
177 178 179
175 176 177 178 179 180 181
174
173
172
8.0h 2.5 d 15.4 d 120 d 16m 8.0h 140 d
70 d
46d
3.7h 10y 6.8 d
100 h 3 h 55d
165 166 167 168 169 170 171
164
32 d
7.7 h 9.6 d 85d 127 d 500 d
9.4 d 7.5 h
160 161 162 163
159
4.5 h ? 4.5 h ? 60 d 34 m 27h
158
1m 2.5h
3.9h 74d
153 154 155 156 157 158 159 160 161 162 163
152
151
5.1 d 17.2h 188 d 5.0h
3.9h 74d
86 d
150
9.2 h 5.8 y 3y 15.4 d
142 143 144 145 146 147 148 149
72d 10^12 y 20y 47 h 25 m 10h
141
350 d 4.1 h 4 y 5.3 d 55 h
140
3.3 d 145 m 121 d
3.5 m 19.3 h 13.8 d 17.5 m
β⁻
β⁻(?)
STABLE
ORBITAL ELECTRON CAPTURE
POSTULATED STABLE
β⁺

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I. Introduction

Good radiochemical separations of the majority of the rare earth elements had been impossible before the application of ion exchange resins to this problem[1]. Cyclotron bombardments could now be undertaken to characterize their hitherto unknown neutron deficient radioactive isotopes with complete confidence in the speed and certainty of chemical identification. A systematic series of bombardments was undertaken to identify these isotopes in order to further the knowledge of neutron deficient isotopes and to allow interpretation of reactions with high energy particles on elements of higher atomic number.

The best general method of approach appeared to be the bombardment of the elements of odd atomic number; for with the exception of europium and promethium, they exist as single stable isotopes. Helium ions on these elements (Z) at 38 Mev and below, produce the α,3n, α,2n and α,n reactions only, for all the products of the α,p....n type of reaction are stable isotopes. When these product isotopes (Z + 2) had been allocated by cross bombardment and yield considerations, proton reactions on the adjacent element (Z + 1) would permit the identification and allocation of the remaining nuclides.

When sufficient knowledge of the region had been obtained in this way, it would be possible to bombard the elements of even atomic number with helium ions and allocate product isotopes from daughter activities.

Varying bombardment times enabled the detection of nuclides ranging in half-life from 20 minutes to greater than one year.

II. Experimental Methods

A. Bombardments

The most desirable target material in a bombardment is the metal because of its ease of manipulation. This was only possible with tantalum, tungsten and rhenium. The rare earths and hafnium necessarily must be bombarded as oxides.

When comparatively large amounts of the target element oxides were available, approximately one hundred milligrams of the oxide was spread on a grooved copper plate. The oxide was wetted with sodium silicate solution as an adhesive and dried under an infra red heat lamp. Additional protection was afforded by a 0.0002 inch tantalum foil over the target. Targets so prepared have withstood over one hundred microampere-hours of 20 Mev deuterons without loss.

In all cases where relatively small amounts of the target materials were available, ten to twenty milligrams of the finely powdered oxide were similarly mounted on a small grooved platinum boat which was silver soldered to a water cooled copper holder. Tantalum foil was wrapped around the boat and holder as additional protection.

Bombardments of many target materials have been made with 38 Mev helium ions, 19 Mev deuterons, 10 Mev protons and fast neutrons from the 60-inch Crocker Laboratory cyclotron. To reduce the energy of the beam incident on the target, suitable thicknesses of tantalum foil were placed over the target. The mass thickness in mg/cm^2 of the tantalum foil was obtained from curves by Rossi and Jones from the theoretical data of Aron, Hoffman and Williams.

Tantalum foil was bombarded with 200 Mev deuterons from the 184-inch cyclotron.

B. Chemical Separations[2]

The problem of chemical separations is divided into two general categories; the separation of hafnium, tantalum, tungsten and rhenium from them-

selves and from the rare earth elements, and the separation of the individual rare earths. Assume a bombardment of tantalum with 400 Mev helium ions to illustrate chemical separations.

1. Isolation of Rare Earth Fraction

Tantalum was dissolved in hydrofluoric acid with nitric acid being added dropwise. The fluoride ion was then complexed with boric acid and cerium III solution added to carry the rare earth activities. The rare earth fluoride was precipitated with additional hydrofluoric acid. The separation of the individual rare earths is discussed on page 10 et. seq.

2. Separation of Rhenium

The excess nitric acid was destroyed with hydrazine sulfate after the addition of carriers for hafnium, tungsten and rhenium. Rhenium was completely precipitated from the boiling hydrofluoric acid solution as the sulfide with hydrogen sulfide which was then dissolved in sodium hydroxide and hydrogen peroxide, and scavenged with ferric hydroxide. The solution was made at least 6N in hydrochloric acid, and the sulfide precipitated from the boiling solution. In cases where contamination might follow through this procedure, the sulfide was heated in a still with sulfuric acid to metathesize the sulfide to the volatile oxide. The distillate was collected in water, acidified, and the sulfide reprecipitated.

3. Separation of Tungsten

After the filtrate was boiled to expel hydrogen sulfide, hydrochloric acid, solid ammonium thiocyanate, metallic mercury and solid boric acid were added in that order to the boiling solution. This procedure produced an apple green thiocyanate complex of tungsten which extracted quantitatively into ethyl acetate. The organic layer was washed with 6N hydrochloric acid twice and evaporated. Just before dryness was reached, one cubic centimeter of 6N hydro-

chloric acid containing two or three drops of 30% hydrogen peroxide was added. This destroyed the tungsten blue and precipitated the trioxide. The washed trioxide was dissolved in concentrated sodium hydroxide and centrifuged to separate from any mercury carrying through the procedure. Tungsten oxide was reprecipitated from a boiling solution by the addition of concentrated nitric acid and a drop of wetting agent. The trioxide was washed, dried and weighed for chemical yield measurements.

4. Separation of Hafnium[3]

The tantalum and hafnium in the aqueous layer were precipitated as the hydrated oxides by the addition of an excess of boric acid, ammonium hydroxide and subsequent boiling. This ensured the solution of any mercury present and afforded an additional separation from tungsten and rhenium. This precipitate, after washing with concentrated ammonium hydroxide, was dissolved in hydrofluoric acid. The solution was adjusted to 3N in nitric and hydrofluoric acids and barium nitrate added. The radiochemically clean barium hafnium fluoride was precipitated, washed, and dissolved in hot 8N nitric acid which had been saturated with boric acid. The hydrated oxide was precipitated with ammonium hydroxide, ignited to the dioxide and weighed for chemical yield measurements. This procedure does not separate hafnium and zirconium, however, there was no evidence of zirconium activities noted.

5. Separation of Tantalum

Tantalum was precipitated from the fluoride solution as radiochemically pure potassium tantalum fluoride by the addition of an excess of saturated potassium fluoride solution. This precipitate was dissolved in 8N nitric acid saturated with boric acid, and the hydrated oxide precipitated with ammonium hydroxide. The hydrated oxide was ignited and weighed as Ta_2O_5 for chemical yield measurements.

6. Rare Earth Separation

The cerium III fluoride was washed and dissolved in 8N nitric acid saturated with boric acid. The hydroxide was precipitated with ammonium hydroxide, washed, dissolved in nitric acid, zirconium holdback agent added and the cycle repeated. Three such cycles were sufficient to ensure complete purification in nearly 100% yield. A small amount of lanthanum (1.0 mg) was added to the final solution in strong nitric acid, and the cerium oxidized to the IV oxidation state with sodium bismuthate. The cerium was then removed by precipitation of the cerium IV phosphate from 3N nitric acid solution. The lanthanum, with the rare earth activities, was recovered as the fluoride which was then converted to the chloride for subsequent separation of the individual rare earth activities on ion exchange resin columns.

7. Ion Exchange Resin Columns[1]

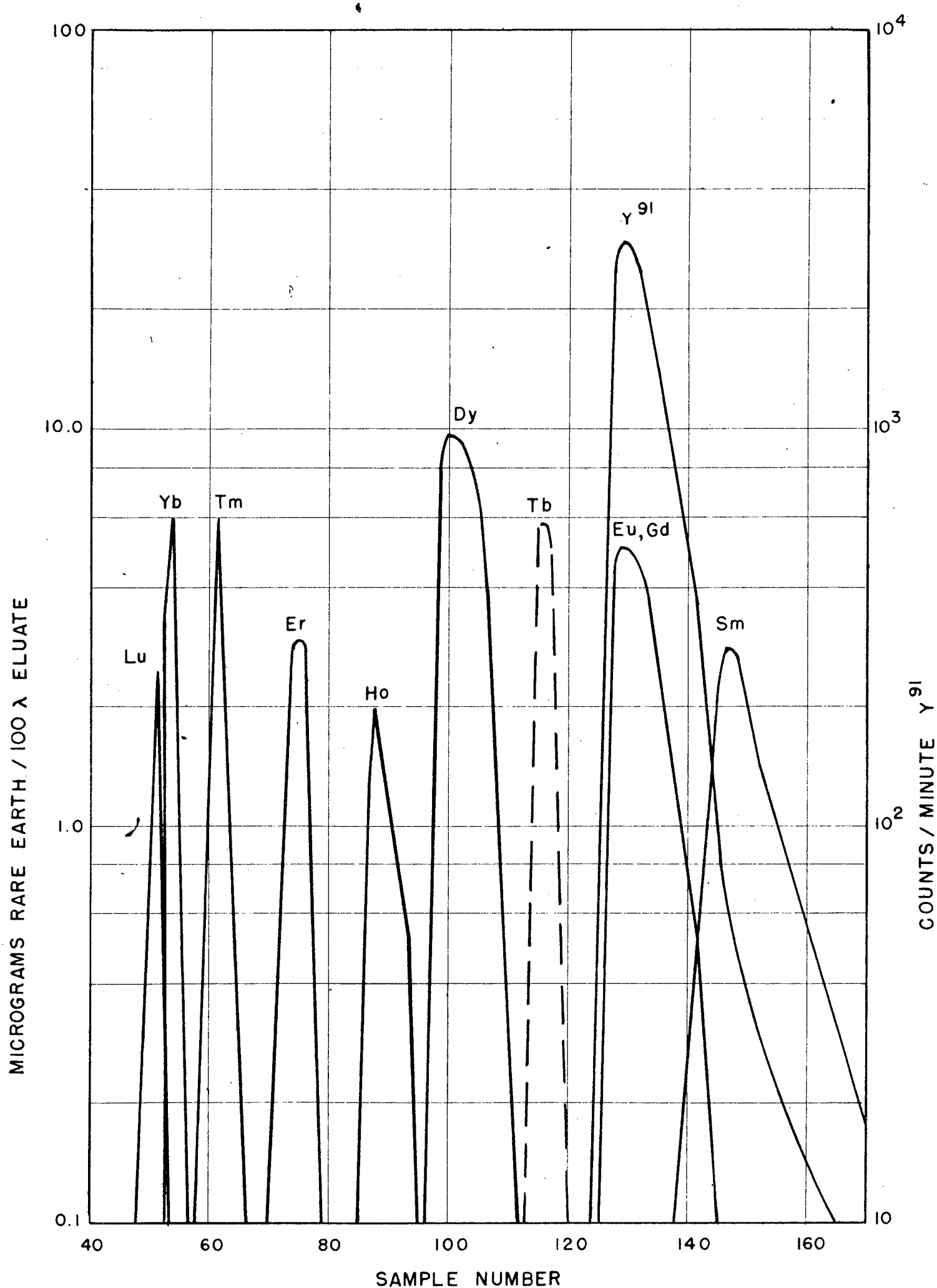
The first step in column separation was the careful adjustment of the rare earth chloride solution to about 0.1N in hydrochloric acid. The rare earth ions were then adsorbed on a small quantity of the ion exchange resin and transferred to the top of a resin column.

A complexing agent for the rare earths, ammonium citrate, was then passed through the column under strictly controlled conditions. The individual rare earths pass down the column with different rates; lutecium being the first to appear in the eluate, followed by ytterbium, thulium, erbium, etc., in succession.

To determine the characteristics of column separations and optimum separation conditions, macro amounts of inactive rare earths and radioactive tracer Y^{91} were used. The former were determined spectroscopically[4] and Y^{91} by its radioactivity (Fig. 1).

Fig. I <u>Standard rare earth elution</u>

50 x 0.4 cm Dowex-50 column. Flow rate, 0.08 $ml/cm^2/min$. Sampling time 15 minutes; eluate volume in milliliters is 0.282 x sample number. Y^{91} was measured by its radioactivity, other rare earths spectroscopically. Terbium is estimated since the cyanogen bands obscure the terbium lines. Eluting agent is citric acid adjusted to pH 3.05 with armonium hydroxide.


100
10.0
1.0
0.1
10^4
10^3
10^2
10
40
60
80
100
120
140
160
SAMPLE NUMBER
MICROGRAMS RARE EARTH / 100 λ ELUATE
COUNTS / MINUTE Y^{91}
Lu
Yb
Tm
Er
Ho
Dy
Tb
Eu, Gd
Y^{91}
Sm

8. Preparation of Resin

All column separations were made using Dowex 50[5], an aromatic hydrocarbon polymer containing many nuclear sulfonic acid groups. The resin was supplied as the sodium form in colloidal agglomerates, Batch 19891 M. The ammonium form of the resin was prepared by stirring with saturated ammonium chloride solution; excess ammonium chloride was removed by water washing. The resin was graded so that the settling time of the selected particles was less than five minutes for five centimeters in water at room temperature. The length of the preliminary resin column was 50 cm., with a diameter of 0.4 cm. In subsequent runs, it was found that 20 cm. x 0.4 cm. and 20 cm. x 1.0 cm. columns gave excellent separation.

9. Separation of Rare Earths[6]

The rare earths were precipitated as the hydroxides, washed and dissolved in a minimum of dilute hydrochloric acid. The pH was adjusted between the limits of 0.5 and 1.5 with hydrochloric acid only thus keeping the ionic strength of the solution at a minimum. This solution was then agitated with about 0.2 ml. of resin while standing in a hot water bath. This treatment continued for about 15 minutes. Longer contact would permit excessive diffusion of the ions into the interior of the agglomerates with subsequent slow elution and poor separation.

This resin was then transferred to the top of the resin column, and the eluting agent passed through.

The eluting agent employed was 0.25 M citric acid solution accurately adjusted to pH 3.05 with ammonium hydroxide. Care was taken to approach this value from the acid side so that readjustment with acid is unnecessary. It is very important for good separation that the ionic strength be held to a minimum, hence these precautions.

The flow rate was approximately 0.03 milliliter per square centimeter of column area per minute. With a column of diameter 0.4 cm. and rop size of 0.03 ml., this flow rate corresponded to one drop in three and one-half minutes, and approximated equilibrium conditions very closely.

The eluate was collected in 10 x 75 mm. rimless glass tubes placed in a perforated turntable, which could be rotated automatically at various time intervals, to place succeeding tubes under the column. Samples were normally collected for 15 minutes. Aliquots were taken with micropipettes and evaporated on stainless steel disks for counting. Macro amounts of rare earth elements were determined by spectrographic analysis of aliquot portions. To estimate the degree of separation achieved, the amount of rare earth thus determined was plotted against sample number. In all column runs, samples collected before the elution of one "free column volume" were discarded.

After the completion of a column run, any residual activity or rare earth was stripped from the column with 0.25 M ammonium citrate of pH 7 as eluting agent. After a thorough washing with water, the column was again ready for use.

10. Recovery of Rare Earths[6]

After separation on the column, the rare earths were recovered by evaporation of the citrate solution with nitric and perchloric acids and subsequent ignition. The rare earth residue was then extracted with hot nitric acid and the hydroxide precipitated with ammonium hydroxide. If the solution contained only trace amounts, a small amount of lanthanum was added as a carrier.

11. Further General Column Discussion[6]

In addition to strict control of the conditions mentioned above, it was also important for successful separations that the capacity of the resin for adsorption of rare earth ions not be exceeded. For the 0.4 and 1.0 cm. diameter columns, the maximum capacity corresponded to approximately ten milligrams

and one hundred milligrams of rare earth oxide respectively. When the starting materials contained significant amounts of other rare earth impurities such that purification prior to bombardment was necessary, a larger column which had a capacity for about one gram of rare earth oxide was used.[7] The drop rate was scaled up from the values used on the 0.4 cm. and 1.0 cm. diameter column runs, the amount of citrate flowing through unit cross-sectional area being kept constant.

The progress of any particular inactive rare earth through the column could be checked by using radioactive tracer for a neighboring heavier rare earth. After elution of the activity, the sample numbers of the desired inactive rare earth were estimated from the standard elution curve, Fig. 1. Aliquots of the samples were then examined spectroscopically to allow the pure rare earths to be isolated.

The standard elution curve shown in Fig. 1 was obtained with inactive materials and Y^{91} tracer. It should be noted that while the lutecium and ytterbium peaks were very close together, aliquots from peak samples have no detectable lines of each other or any other rare earth in their spectra. Thus samples of these neighboring elements can be obtained where the separation factor is at least one thousand to one.

A typical elution curve of a bombarded sample is shown in Fig. 2, where the dotted line shows the distribution of radioactivity produced by 38 Mev helium ion bombardment of holmium oxide. The solid lines show the distribution of inactive carriers of lutecium, ytterbium, erbium, and dysprosium added as references and determined spectroscopically.

12. Separation of Three Tracer Components

A further example of a column separation is shown in Fig. 3, when rare earths were present in tracer quantities only. In such cases, separation is generally cleaner and with less "tailing" of the peaks. The activities

Fig. 2 Column separation of thulium activities from Ho + α bombardment.

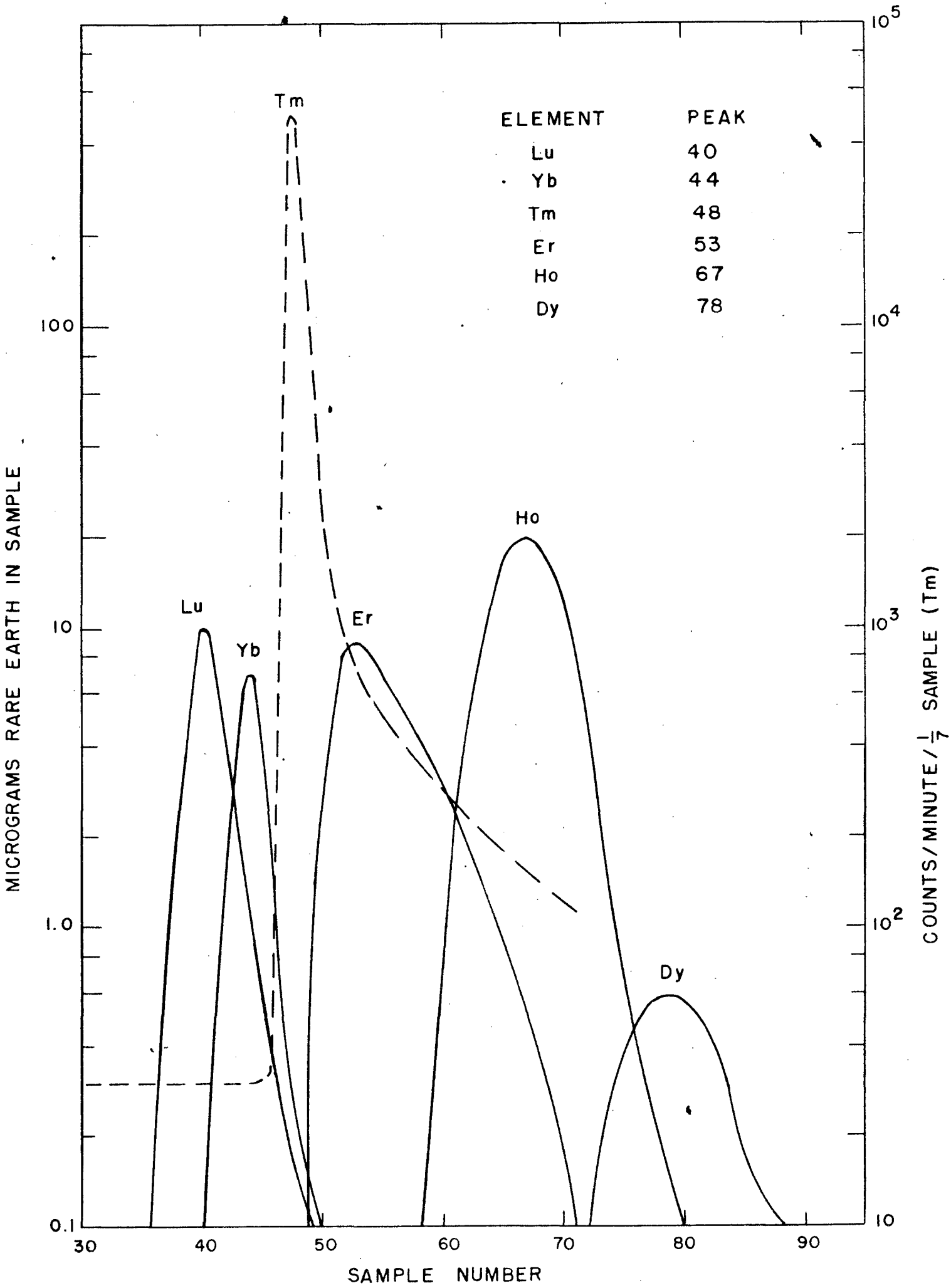
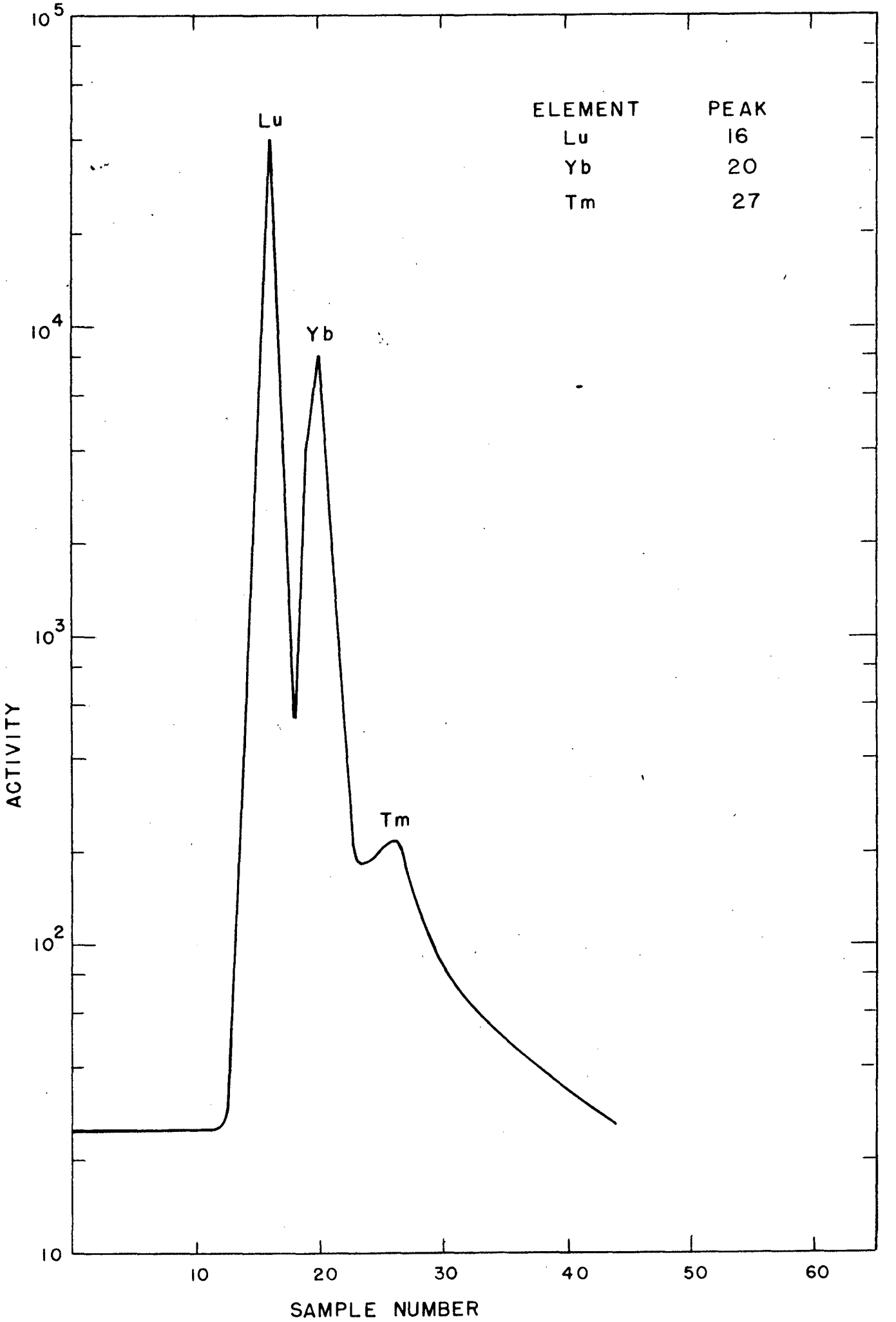

ELEMENT PEAK
Lu 40
Yb 44
Tm 48
Er 53
Ho 67
Dy 78
Tm
Lu
Yb
Er
Ho
Dy
MICROGRAMS RARE EARTH IN SAMPLE
COUNTS/MINUTE/1/7 SAMPLE (Tm)
SAMPLE NUMBER
0.1
1.0
10
100
10
10^2
10^3
10^4
10^5
30
40
50
60
70
80
90

FIG II

Fig. 3 Column separation of rare earth spallation products from 200 Mev deuteron bombardment of tantalum.


ELEMENT PEAK
Lu 16
Yb 20
Tm 27
Lu
Yb
Tm
ACTIVITY
SAMPLE NUMBER
10
10^2
10^3
10^4
10^5
10
20
30
40
50
60

shown in Fig. 3 were spallation products, obtained by the bombardment of tantalum with 200 Mev deuterons from the 184-inch cyclotron. Lanthanum was used as a carrier for these radioisotopes. A study of the radiation characteristics and decays of peak samples revealed no evidence of contamination.

13. Separation of Europium

Europium has been purified prior to, and recovered after bombardment by a modification of McCoy's method[8]. The europium in 6N hydrochloric acid was reduced to the divalent state by boiling with zinc amalgam. Other rare earths were precipitated by carbonate-free ammonia using cerium as a carrier. After centrifugying, the supernatant was withdrawn, the europium oxidized with ozone, and then precipitated with ammonia. This procedure was repeated with both fractions to insure complete separation.

14. Rare Earth Oxide Bombardments

When the rare earth oxides were bombarded, the rare earth chemistry formerly employed was modified. Since sodium silicate was used as a binder, and platinum, gold, mercury, copper, zinc and gallium activities could conceivably have been contaminants from the mounting, it was necessary to separate the rare earth activities from these elements.

The rare earth oxide was dissolved in hot concentrated nitric acid and the insoluble silica residues removed. Then the hydroxide was precipitated with ammonium hydroxide which reduced the volume of the material so that chemistry could subsequently be carried out in 15 cc. lusteroid and centrifuge test tubes. The hydroxide was then dissolved in 3N hydrochloric acid and silver nitrate added so that the silver chloride would mechanically carry and hold down any refuse coming from the target.

Holdback agents for all possible contaminants were added and the fluoride precipitated in the hot by hydrofluoric acid. The washed fluoride was dissolved

with the nitric-boric acid mixture, and the hydroxide precipitated with ammonium hydroxide. Two more such cycles were repeated and the final hydroxide dissolved in a minimum of concentrated hydrochloric acid, and transferred to a one cc. volumetric flask. Aliquots were then taken with micropipettes for counting, column separation and weighing to determine chemical yields. Yields were determined by precipitation of the oxalate, followed by ignition to the rare earth oxides and weighing.

C. Radioactive Measurement

Measurements were made with end-on Geiger-Müller counters which had 3 mg/cm^2 mica windows and were filled with 10 cm. argon and 1.0 cm. alcohol. Variations in counter efficiency were taken into account by the use of a UX_2 standard. The scaling circuit used was of conventional type, scale of 64 with 1.2% per thousand counts per minute coincidence corrections made. A crude beta-ray spectrometer of low resolution enabled the distinction of positive and negative electrons and served as a check on absorption measurements.

1. Absorption measurements[9]

Radiation characteristics of radioactive nuclides were determined by means of lead and aluminum absorptions of their radiations. Aluminum absorptions were taken in the conventional manner, and the range of electrons has been taken as the minimum absorber thickness required for total absorption, i. e., aluminum plus air gap plus window. Feather analyses were used to determine the range of beta particles. After an aluminum absorption, beryllium just sufficient to absorb all electrons present was interposed between the sample and counter. Aluminum absorbers were placed between the beryllium absorber and the sample until the electromagnetic absorption curve was completely determined. The amount of L x-rays was interpreted as the difference between the measured electromagnetic radiation and the extrapolation to zero absorber of the harder background.

Lead absorptions were taken in low geometry enabling the utilization of absorbers up to 45 g/cm^2 between the two beryllium absorbers. The absorber immediately above the sample was thick enough to remove all particles coming from the sample, and the second, just below the counter window, was thick enough to remove all secondary electrons, so that only x-rays and γ-rays were counted. A thick beryllium absorber was placed just below the sample to aid in minimizing scattering effects. It was found that when there was a hard γ-ray, the lead half-thickness as measured in the conventional lead cage, exceeded the mass half-thickness for very hard γ-rays ($\sim$17 g/cm^2). For this reason, all lead absorptions were carried out with a counter surrounded by as little matter as possible to minimize scattering effects. This "unshielded" arrangement gave reproducible results upon changing counters whereas those absorptions taken in the conventional lead shielding gave half-thicknesses for a 1.1 Mev γ-ray varying from 17 g/cm^2 to 29 g/cm^2 using different counter tubes.

2. Samples

Samples for counting were mounted on glass microscope cover slides. Where a minimum of backscattering was required, as in the measurement of the ratios of electrons to electromagnetic radiation, samples were mounted on mica of mass thickness $\sim$1 mg/cm^2.

3. Counting Efficiencies

Counting efficiencies of L x-radiation in the counters used were calculated from the absorption in the gas; for γ-radiation from 20 Kev to 0.5 Mev, a counting efficiency of 0.5% was assumed, with one percent per Mev thereafter.

4. Calibration

For quantitative counting, the counter geometry was calibrated by the use of a weighed UX_2 standard.

D. Calculations and Corrections

1. Calibration

In order that a cross-section could be calculated, the counter geometry had to be calibrated in terms of disintegrations per minute. This was done by counting a weighed sample of UO_2 through 30 mg/cm^2 of aluminum absorber. This allowed only the UX_2 beta to be counted. Knowing the number of UX_2 disintegrations in the UO_2, the geometry could be calculated after appropriate corrections for absorption in the sample, counter window and air gap, and added absorber.

All decay points were corrected for variation of counter efficiency by counting against an unweighed UX_2 standard and corrected to an arbitrary value of 1000 counts per minute of this UX_2 standard. This entailed merely the dividing of the decay count by the average value of this UX_2 standard before and after counting, and multiplying the quotient by 1000. This unweighed standard was counted against the weighed UX_2 standard and the geometry corrected to the arbitrary standard of 1000 counts per minute of the unweighed standard. Counts so corrected corresponded to 8.7 percent geometry in disintegrations per minute in the second shelf.

2. Decay Schemes

For simplicity, it was assumed that the orbital electron capturing isotopes studied here decayed by K electron capture only. Therefore, the amount of K x-rays counted should be a measure of the number of disintegrations in the sample.

In order to determine what fraction of the measured K x-radiation arose from the capture process, a rough decay scheme for each isotope must be postulated. It is realized that the ratios leading to these schemes may be in error by factors of two or more because of the crude techniques employed; however, it was felt that because many of these isotopes must be assigned by relative yield measurements, the use of such rough schemes was justified.

3. Corrections

In an attempt to make the ratios of the various radiations as nearly representative as possible, many corrections were employed. When ratios between electrons and electromagnetic radiation were desired, a very small mass of high specific activity was mounted on mica of surface density of 1-2 mg/cm^2. An aluminum absorption was taken in 8.7 percent geometry. Backscattering corrections were assumed to be small and hence neglected. The electrons were corrected for absorption in the air gap and counter window by extrapolation of the resolved absorption curve of the electrons. The energies of these electrons have been given with these corrections added. Their counting efficiency has been taken as one.

L x-rays were corrected for absorption in beryllium employed in conjunction with aluminum absorptions, air gap, and counter window by extrapolation, and for the Auger effect. The fluorescence yields were obtained from Compton and Allison[10], and were assumed to be equal to those of K x-rays of comparable energies. The counts as corrected were then divided by the Auger coefficient.

This left only the K x-rays and γ-radiation uncorrected. These appeared in aluminum absorption measurements as a straight line background after the L x-rays and electrons were completely absorbed. The ratio of the K x-ray to γ-rays was obtained from lead absorptions.

After resolution of the lead absorption, the various components were extrapolated to zero lead absorber. They were subsequently corrected individually for absorption in the beryllium employed and for the Auger effect as above. The total mass thickness of beryllium used varied from two to three grams per square centimeter. The ratio of K x-rays to γ-rays, as measured, was used to determine the number of counts per minute of each present in the electromagnetic background of the aluminum absorption.

4. Final Ratios

These corrected values were the counts per minute of each type of electron and electromagnetic quantum that would have been measured if the sample were inside the counter tube. The values were then divided by their respective counting efficiencies. Dividing all values by the number of K x-ray quanta thus calculated gave the final ratios upon which cross sections were calculated. If there existed means of producing a K x-ray quantum other than capture, i.e., a conversion electron line, this amount was subtracted from the K x-rays in the final ratio and only that remaining fraction of K x-ray counts was used to compute a cross section.

5. Cross Section Calculation

The calculation of an approximate cross section is now possible. An example is shown below for the reaction $Ho^{165}(\alpha,3n)Tm^{166}$.

The ratios for Tm^{166} are:

$e_1 : e_2 : \beta^+ :$ K x-rays $: \gamma$

0.05 :0.003:0.004: 1 : 0.3

counts per minute of K + γ in 8.7 percent geometry = 9200

$K/\gamma = 0.92$ as measured

$$K = \frac{0.92}{1.92} \times 9200 = 4400$$

$$\text{dis/sec at saturation} = \underset{a}{4400} \times \underset{b}{\frac{100}{0.5}} \times \underset{c}{\frac{100}{8.7}} \times \underset{d}{\frac{1}{60}} \times \underset{e}{\frac{100}{84}} \times \underset{f}{\frac{100}{65}} \times \underset{g}{\frac{1000}{5}} \times \underset{h}{\frac{100}{90}} \times \underset{i}{\frac{100}{80}} \times \underset{j}{\frac{100}{95}}$$

a = measured counts/minute

b = counting efficiency

c = geometry

d = to convert to counts/second

e = saturation of bombardment

f = decay since end of bombardment

g and h = chemical yield factor

i = Auger coefficient

j = contribution from capture

$$He^{++}/\text{second} = \frac{\overset{k}{5.3} \times \overset{l}{1.3} \times 10^{16}}{\underset{m}{11.7} \times \underset{n}{3600}}$$

k = microampere-hours of bombardment

l = number of He^{++} per microampere-hour

m = number of hours bombarded

n = seconds per hour

$$\text{Atoms/cm}^2 = \frac{\overset{o}{20.5} \times 10^{-3}}{\underset{q}{189}} \times \frac{\overset{p}{6.02} \times 10^{23}}{\underset{r}{0.7}}$$

o = grams of Ho_2O_3

p = Avogadro's number

q = 1/2 molecular weight of Ho_2O_3

r = area of target in cm^2

$$\sigma = \frac{\text{disintegrations/sec at saturation}}{\text{atoms/cm}^2 \times He^{++}/\text{second}}$$

σ = 1.1 barns.

III. Discussion of Results

A. Neodymium Isotopes

The isotope Nd^{141} has been reported[11] to have a 2.5-hour half-life and to emit 0.78 Mev positrons. The activity was produced by proton bombardment of praseodymium[11], by n,2n[11,12,13] and γ,n[11,13] reactions on neodymium, and possibly a d,H^3 reaction on neodymium[11,12]. Chemical separations were not made.

In the present work, chemical separations were made by ion exchange columns. The 2.5-hour activity has been characterized in more detail and allocated to

Nd^{141}, while a new isotope, Nd^{140} has been observed by Pr-d,3n reaction (Table II). This isotope decays by orbital electron capture to form the 3.5 minute positron-emitting Pr^{140}. In bombardments, spectroscopically pure Pr_6O_{11}[7] was used.

Table II

Isotope	Type of Radiation	Half-Life	Energy of Radiation Particles	Energy of Radiation γ-ray	Produced by
Nd^{140}	K	3.3 ± 0.1 days			Pr-d-3n
Nd^{141}	K, β^+ (2%), γ	145 ± 3 minutes	0.7(β^+)	K x-rays 1.05	Pr-d-2n Pr-p-n

1. 145-Minute Nd^{141}

Measurements of the radiation characteristics were made on unseparated praseodymium which had been bombarded with 10-Mev protons. The proof of the chemical identity by the standard ion-exchange resin column procedure generally used was almost impossible, since the neodymium fraction leaves the column only after about two days. A 4 cm x 0.4 cm column was therefore used, with conditions of flow, eluting agent, etc. the same as before. Although the separation of neodymium, and praseodymium is unsatisfactory from the chemical standpoint, these fractions leave the small column in about 12 hours, a time short enough to allow detection of the 145-minute activity. Such an experiment was made for a Pr + d bombardment. The ratio of the 145-minute activity to the 19.3-hour Pr^{142} activity formed by the d,p reaction was estimated, corrections for decay from a reference time being made for both activities. The ratio decreased rapidly in samples following the "break through" of active material showing that the 145-minute activity elutes before praseodymium. Further, spectroscopic analysis

showed that the 19.3-hour activity followed the praseodymium, while in the first active sample where the 145-minute decay was observed, praseodymium was below the limits of detection. The chemical identity of the 145-minute neodymium is, therefore, fairly certain.

On other bombardments, the gross decay was followed through eight half-lives, the hard γ-radiation, through six half-lives, and the positron decay, (Fig. 4), followed on a crude beta-ray spectrometer, also through six half-lives. The half-life is 145 $\pm$ 3 minutes.

The radiations consist of positrons, x-rays, and γ-rays. No negative electrons were observed. The aluminum absorption curve of the 145-minute activity from Pr + p bombardment, corrected for decay during the time of measurement, is shown in Fig. 5. The range of the electron, 245 mg/cm^2 (0.7 Mev) agrees with the value of the positron energy measured on the crude beta-ray spectrometer. The lead absorption (Fig. 6) shows electromagnetic radiations of half-thicknesses 39 mg/cm^2 (38 Kev), $\sim$4.5 g/cm^2 (0.5 Mev), and $\sim$11.5 g/cm^2 (1.2 Mev). The soft component agrees well with the K x-radiation of praseodymium, while the low abundant 0.5 Mev γ-ray is almost certainly annihilation radiation.

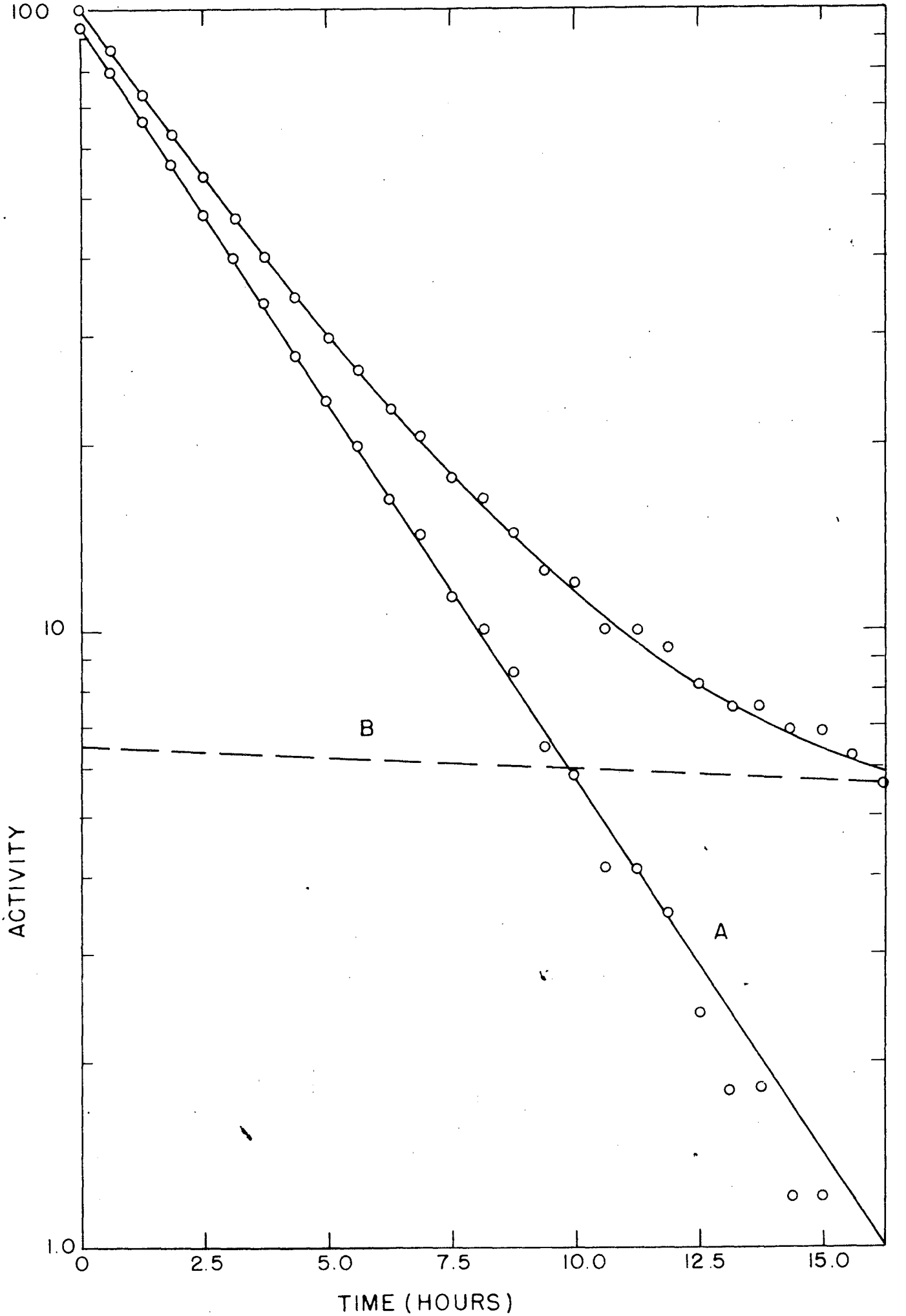
From the measurements, the following ratios were calculated:

0.7 Mev β^+	:	K x-rays	:	0.5 Mev γ	:	1.2 Mev γ =
0.02	:	1	:	0.02	:	0.02

It is clear that this isotope decays by orbital-electron capture with approximately 2% positron branching. The hard γ-radiation probably arises from an excited or metastable state of the daughter nucleus following electron capture.

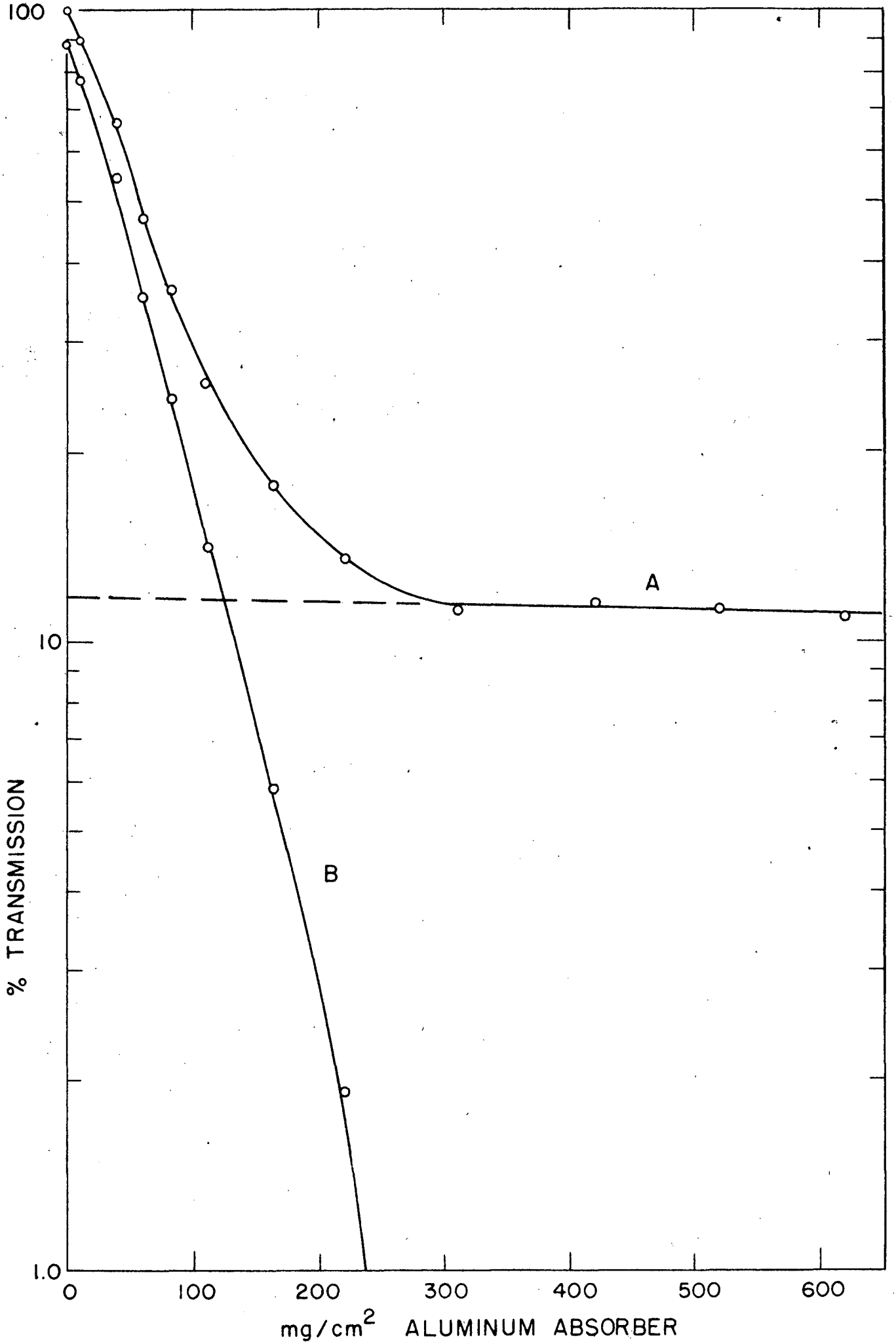
From the measured K x-ray intensities, together with data on chemical yields and bombardment, a cross section of 2.5 x 10^{-2} barns was calculated for the reaction $Pr^{141}(p,n)Nd^{141}$ with 10-Mev protons, assuming one K x-ray quantum represents one disintegration by orbital electron capture.

Fig. 4 Positron decay of Nd^{140}(A) and Nd^{141}(B) from Pr + d bombardment taken on crude beta-ray spectrometer.



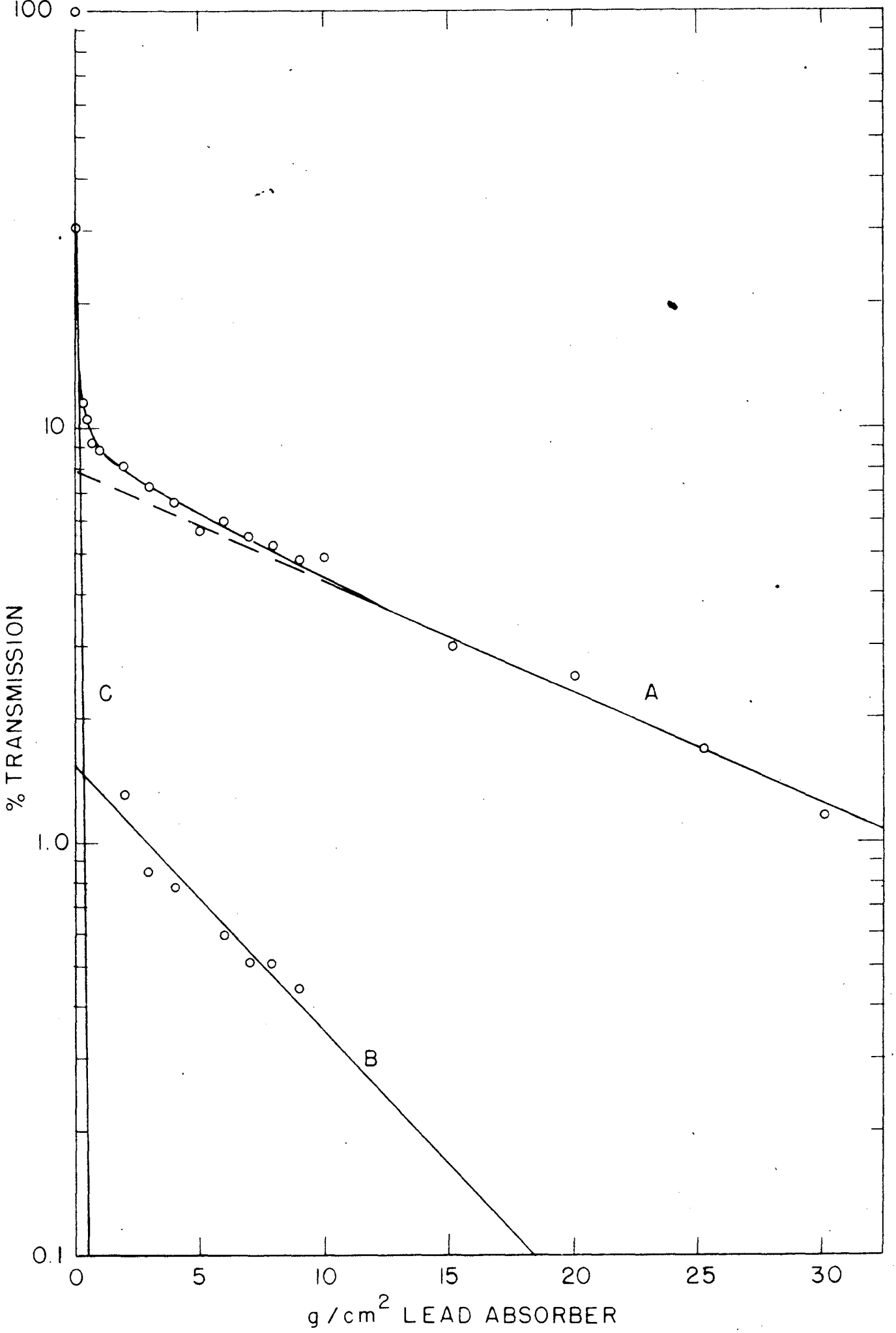
13265-1

Fig. 5 Aluminum absorption of 145-minute Nd^{141} from Pr + p bombardment. K x-ray and γ-ray background (A), 0.7 Mev positron (B).


100
10
1.0
% TRANSMISSION
A
B
0
100
200
300
400
500
600
mg/cm2 ALUMINUM ABSORBER

13262-1

Fig. 6 Lead absorption of 145-minute Nd^{141} from Pr + p bombardment. 1.2 Mev γ-ray (A), 0.5 Mev γ-ray (B), K x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
0
5
10
15
20
25
30
g / cm^2 LEAD ABSORBER
A
B
C
13261-1

2. 3.3 Day Nd^{140}

In the column separated neodymium fraction from Pr + d bombardments, a new long-lived activity was observed. The half-life, measured through seven periods, is 3.3 ± 0.1 days. The aluminum absorption (Fig. 7) shows a hard electron, range 1150 mg/cm^2 (2.4 Mev) with K x-ray and γ-ray background. The lead absorption (Fig. 8) shows components of half-thicknesses; 40 mg/cm^2 (38 Kev), 4.6 g/cm^2 (0.5 Mev), if one assumes that there is a small amount of a hard γ-ray of half-thickness 11.6 g/cm^2 (1.2 Mev).

The following ratios of electrons to electromagnetic radiation were obtained from the measurements:

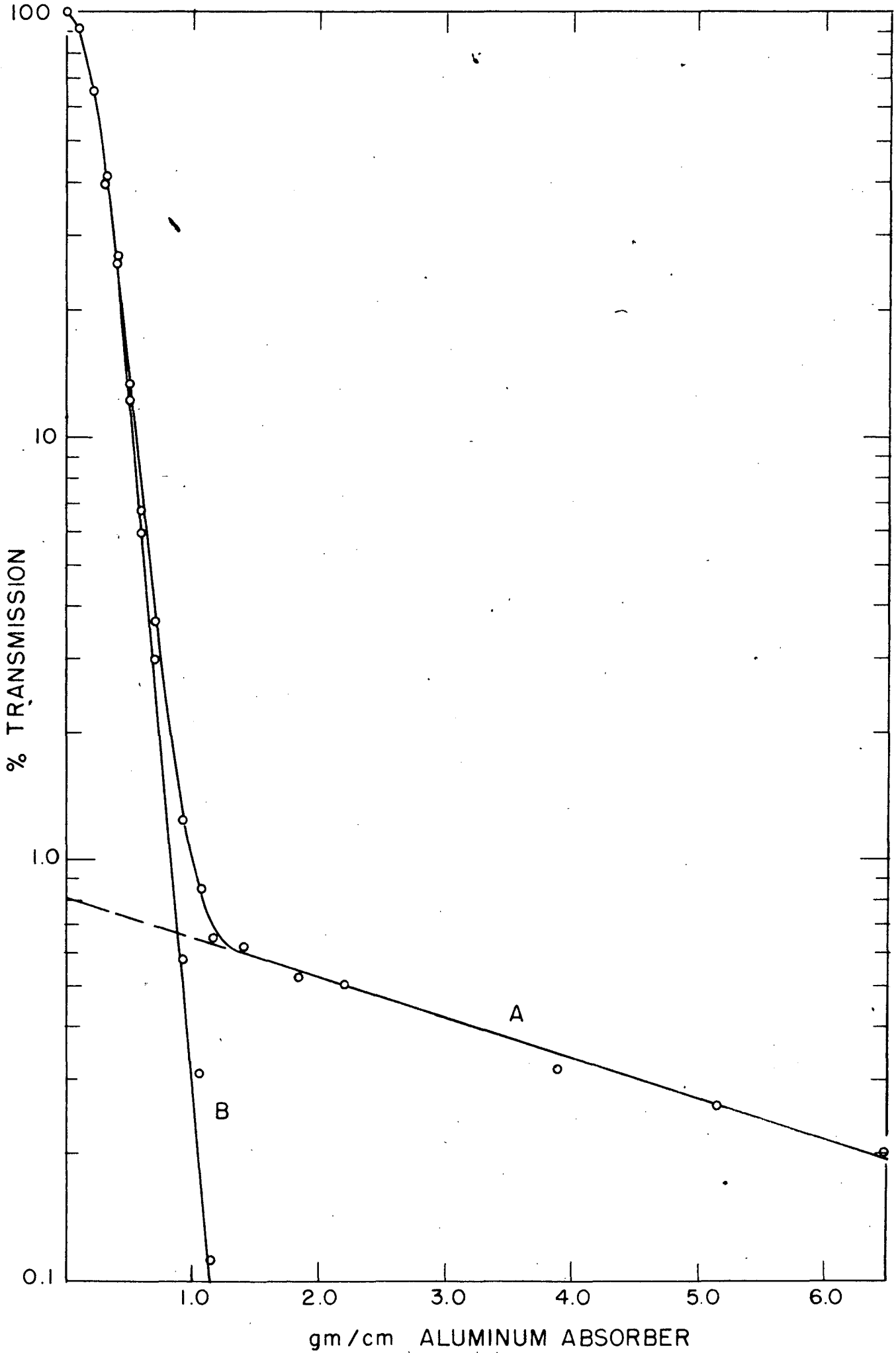
2.4 Mev β^+	: K x-ray	: 0.5 Mev γ	: 1.2 Mev γ =
0.2	: 1	: 0.2	: 0.01

Examination of the electron radiations on the crude beta-ray spectrometer showed that the hard electrons observed in the aluminum absorption were positrons of energy 2.4 Mev. The positron energy agrees well with that reported for the 3.5-minute Pr^{140}.[14]

The observed radiations of the 3.3-day activity are consistent with the isotope, Nd^{140}, decaying by orbital electron capture, in equilibrium with its 3.5-minute positron-emitting Pr^{140} daughter. The observed 0.5 Mev γ-radiation is undoubtedly annihilation radiation; the harder γ-ray may be associated with either Pr^{140} or Nd^{140}. From the ratio of positrons to K x-radiation, it follows that the daughter Pr^{140} activity is not a pure positron-emitter, but decays also by orbital electron capture, about two-thirds of the Pr^{140} disintegrations proceeding by this process.

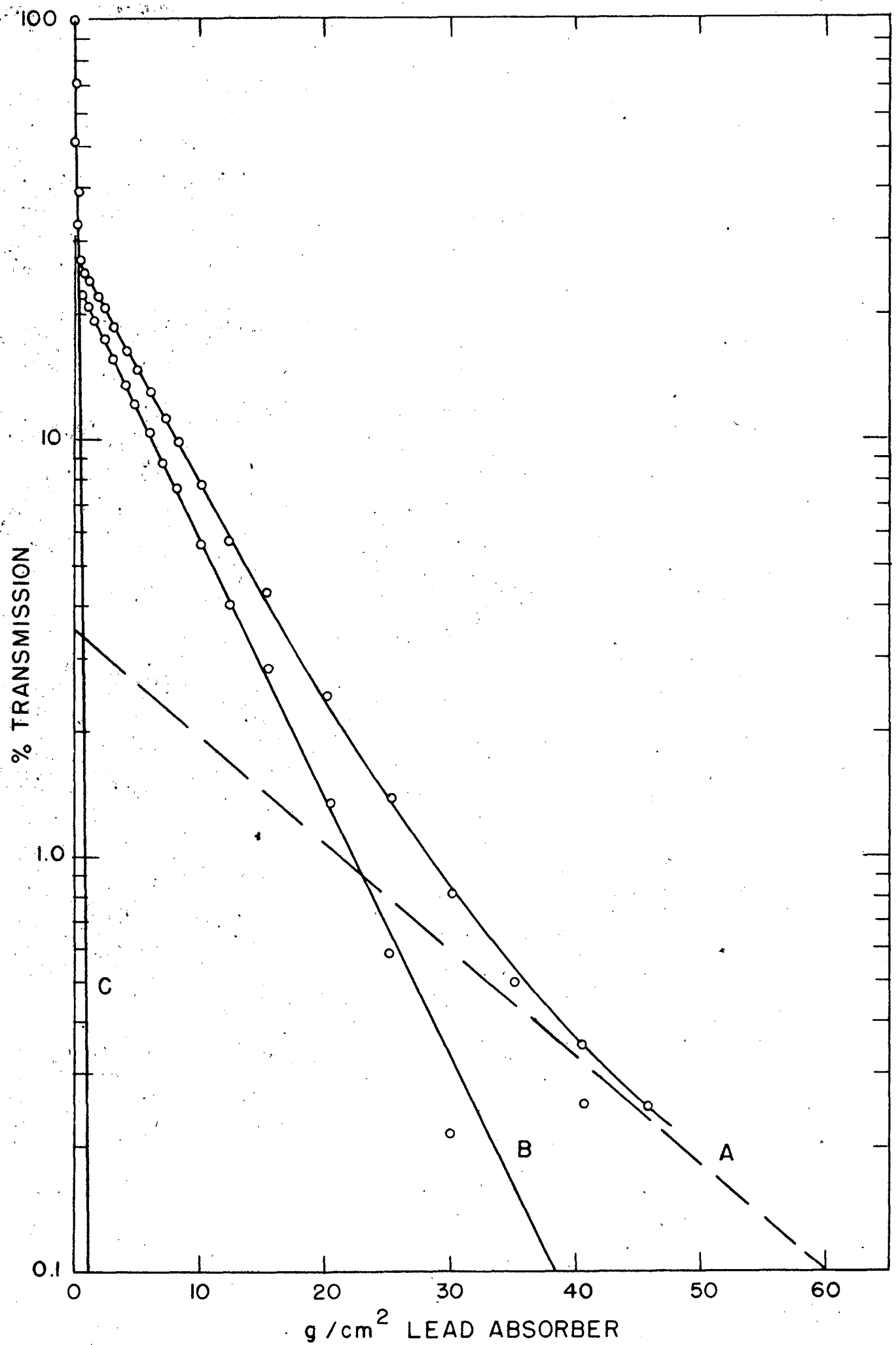
A rapid chemical separation of praseodymium was attempted.[15] The mixed rare earth chlorides were fused with potassium hydroxide at 450° C for periods of 5 to 15 minutes. After extracting the melt with water, the mixed oxides were leached with 1N acetic acid. The neodymium oxide which dissolves preferentially

Fig. 7 Aluminum absorption of 3.3 day Nd^{140} from column separated neodymium fraction of Pr + d bombardment. K x-ray and γ-ray background (A), 2.4 Mev positron (B).


100
10
1.0
0.1
% TRANSMISSION
1.0
2.0
3.0
4.0
5.0
6.0
gm/cm ALUMINUM ABSORBER
A
B

13264-1

Fig. 8 Lead absorption of 3.3 day Nd^{140} from column separated neodymium fraction of Pr + d bombardment. Postulated 1.2 Mev γ-ray (A), 0.5 Mev γ-ray (B), K x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
0
10
20
30
40
50
60
g /cm^2 LEAD ABSORBER
A
B
C

was recovered as hydroxide for counting. The praseodymium remains largely in the residue insoluble in acetic acid. Although the chemical separation achieved is unsatisfactory, the activity of the neodymium fraction has been observed to grow, showing the formation of a daughter activity. The half-life of the daughter appears to be about 1.5-minutes, somewhat shorter than the value reported[12] for Pr^{140}.

3. Discussion

Additional evidence for the allocation of the 3.3-day activity to Nd^{140} has been obtained from yields in the deuteron bombardment of praseodymium. It was assumed that 0.6 of the measured K x-radiation represented one disintegration of Nd^{140}. The cross section for production of the isotopes Pr^{142}, Nd^{141}, and Nd^{140} are given in Table III; the yields of Pr^{142} were calculated from the measured beta activity.

Table III

Cross Sections in Barns for Deuteron Reactions on Praseodymium

Half-Life	Deuteron Energy 19 Mev	9 Mev	Reaction	Isotope
19.3 hours	0.06	0.1	d,p	Pr^{142}
145 minutes	0.3	0.9	d,2n	Nd^{141}
3.3 days	0.08	---	d,3n	Nd^{140}

B. Promethium Isotopes

The two promethium isotopes summarized by Table IV were formed in the bombardment of praseodymium with helium ions of 38, 31, and 19 Mev energy.

Table IV

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	γ-rays	Produced by
Pm^{143}	K, γ, (β^-?)	350 days	0.7(β^-?)	L, K x-rays 0.95	Pr-α-2n
Pm^{144}	β^+	4.1±0.1 hours	1.3 β^+		Pr-α-n

1. 350 day Pm^{143}

A long-lived promethium activity has been previously reported by Wu and Segrè[16] in the helium ion bombardment of praseodymium, and by Kurbatov et. al.[11,17] in the deuteron bombardment of neodymium.

This activity has been confirmed and has been shown to be a promethium isotope by ion exchange resin column separation. Bombardments of column separated[5] praseodymium were made with various energies of helium ions. The decay has been followed through one half-life, and a value of 350 days obtained. A study of the radiations has shown hard negative electrons of range 220 mg/cm^2 aluminum (0.7 Mev) and soft electromagnetic radiation of half-thickness of 5.5 mg/cm^2 aluminum (5.8 Kev) (Fig. 9); electromagnetic radiation of half-thicknesses 50 mg/cm^2 lead (42 Kev) and 9.5 g/cm^2 lead (0.95 Mev) (Fig. 10.). The following ratios were obtained from these measurements:

0.7 Mev e^-	:	L x-rays	:	K x-rays	:	0.95 Mev γ
0.005	:	0.2	:	1	:	0.3

The isotope thus appears to decay by orbital electron capture with part of the disintegrations leading to an excited or metastable state of the daughter nucleus. The hard electron could be a beta particle[18] arising from a long-lived isomer of the 4-hour positron-emitter which decays by beta and orbital electron capture branching.

Fig. 9 Aluminum absorption of 350 day Pm^{143} from Pr + α bombardments. K x-ray and γ-ray background (A), 0.7 Mev electron (B), L x-rays (C).

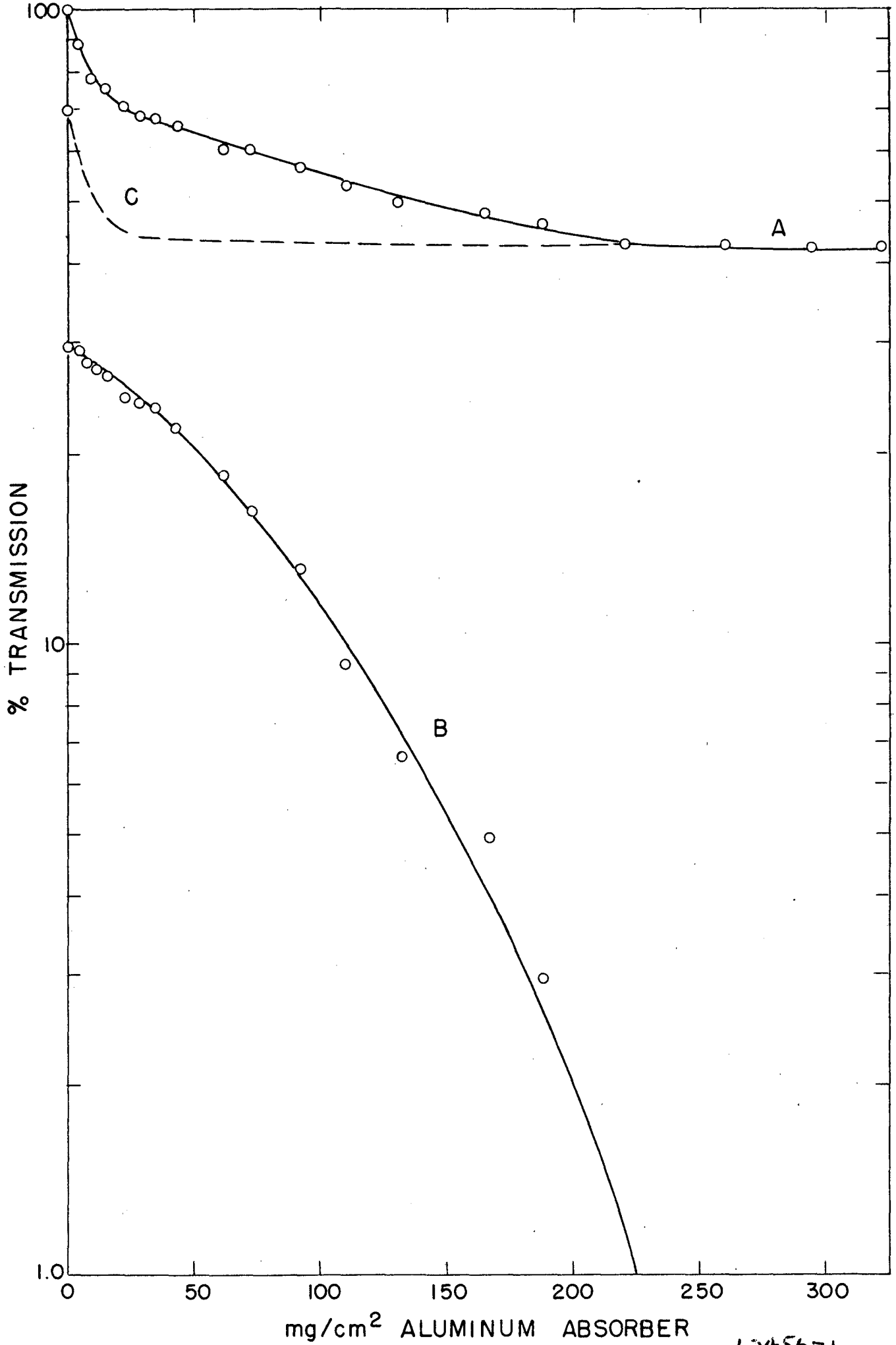
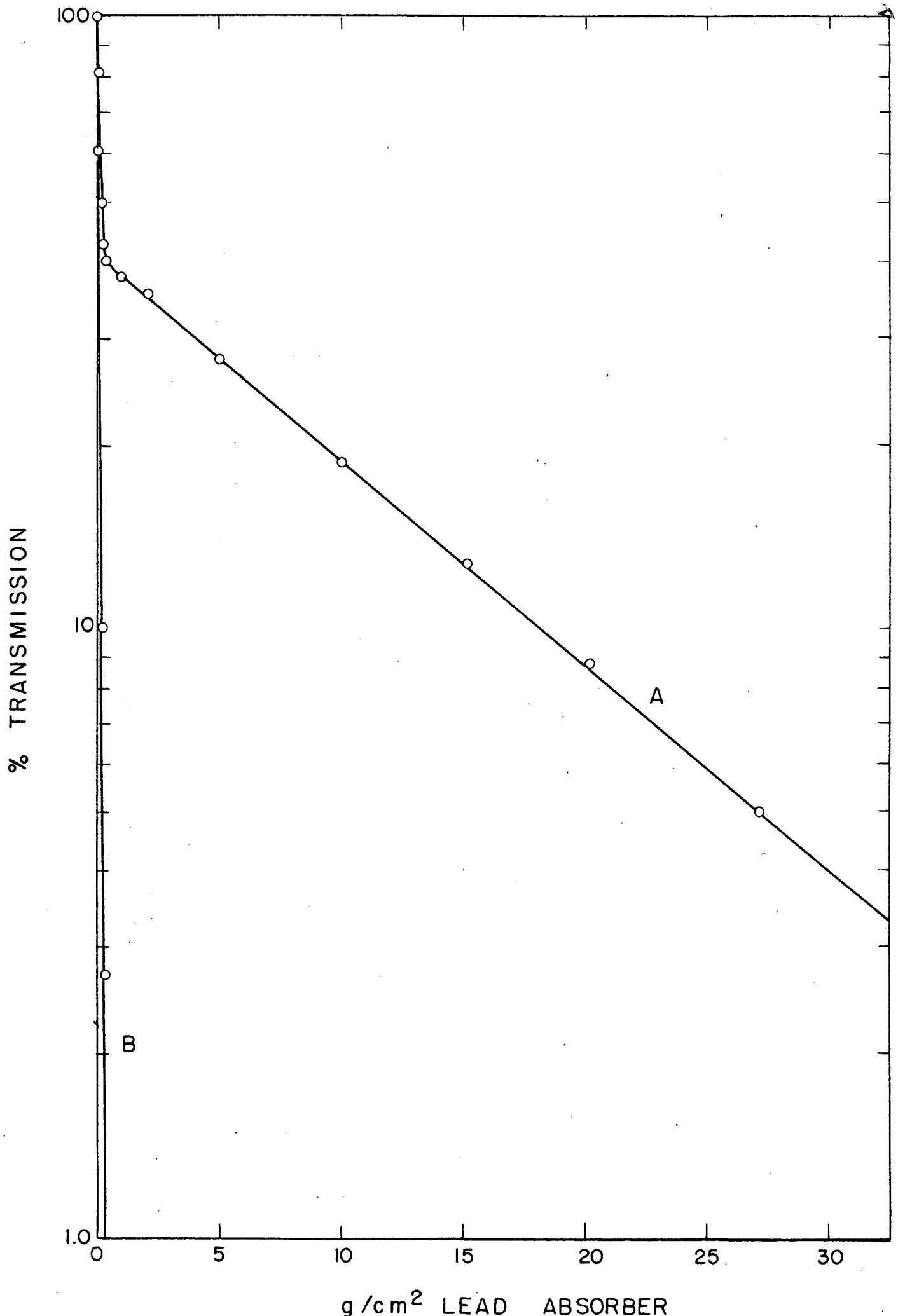

100
10
1.0
% TRANSMISSION
0
50
100
150
200
250
300
mg/cm2 ALUMINUM ABSORBER
A
B
C

Fig. 10 Lead absorption of 350 day Pm^{143} from Pr + α bombardment. 0.95 Mev γ-ray (A), K x-rays (B).


100
10
1.0
% TRANSMISSION
A
B
0
5
10
15
20
25
30
g/cm² LEAD ABSORBER

Assuming one K quantum represents one disintegration by orbital electron capture, the cross sections for production of the isotope by bombardment with 38, 31, and 19 Mev helium ions were calculated, the values being given in Table V. From these values, which are compatible with an α,2n reaction, allocation of the isotope to mass 143 seems reasonable. While the isotopes of odd Z, even A are usually longer-lived than neighboring isotopes, an additional reason for the very long life of this isotope may be the fact that Pm^{143} has eighty-two neutrons, presumably forming a closed shell[19] and increasing stability.

2. 4.1 hour Pm^{144}

In all helium ion bombardments of praseodymium, a short-lived activity was observed in addition to Pm^{143}. The cross sections for 38, 31, and 19 Mev helium ions (Table V) agree with production by α,n reaction, and the isotope has been allocated to mass 144. The half-life, followed through eight periods, is 4.1 ± 0.1 hours. The radiations, all of which decay with the same half-life, consist of positrons, range 600 mg/cm^2 aluminum (1.3 Mev; Fig. 11), with K x-ray or γ-ray background. The radiations have been studied on a crude beta-ray spectrometer; no negative electrons were observed, but only a positron spectrum of endpoint corresponding to 1.3 Mev.

If a counting efficiency of 0.5% is assumed for K x-ray or γ-ray background, the ratio of positrons to electrons is 1:1 suggesting pure positron emission.

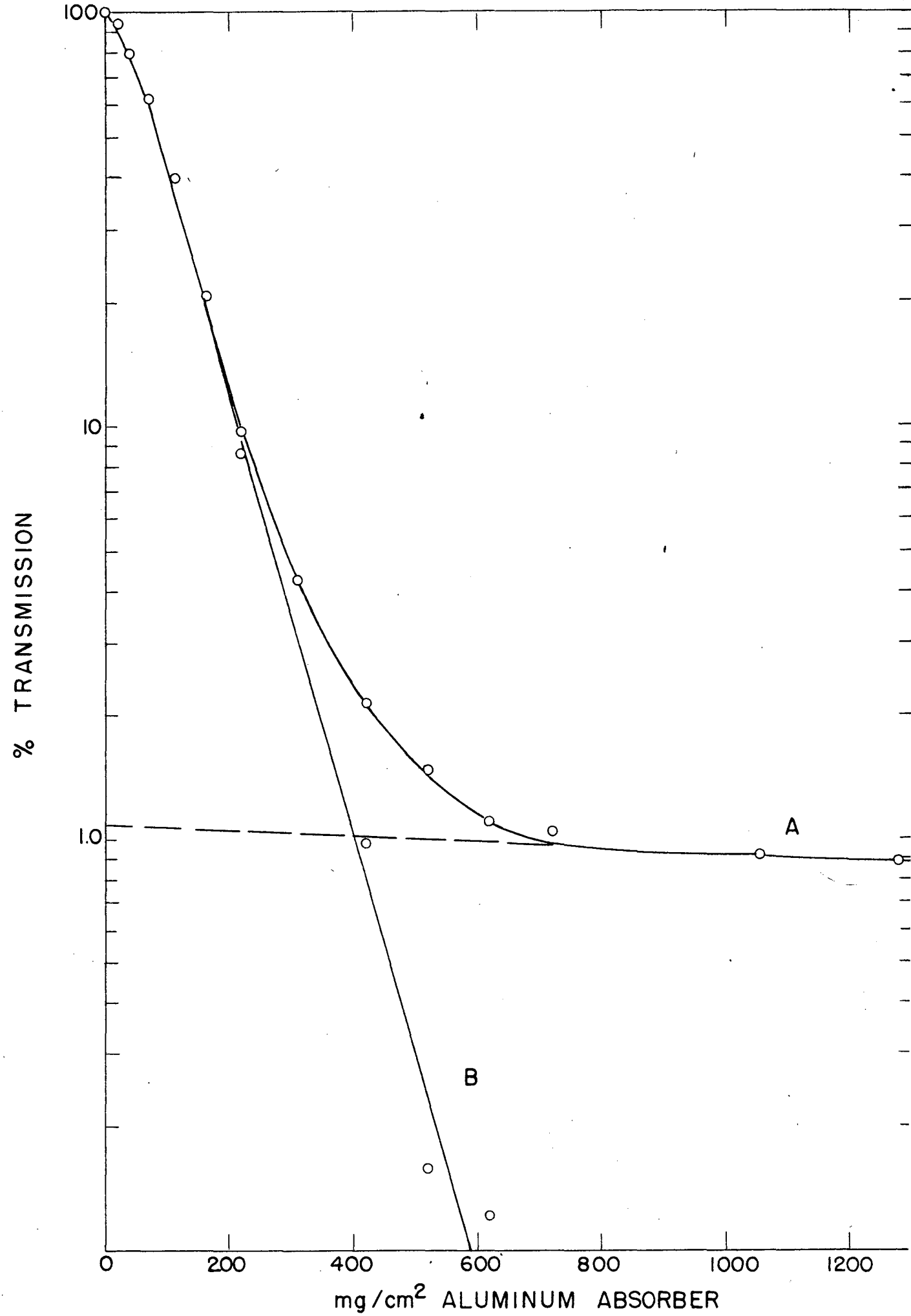
Table V

Cross Sections in Barns for Helium Ions on Praseodymium

Half-Life	38 Mev	31 Mev	19 Mev	Probable Reaction
350 day	0.05	0.1	1	α,2n
4.1 hours	10^{-5}	5×10^{-5}	0.02	α,n

Pm^{142}, the product of the α,3n reaction was not detected. It has a half-life either less than 20 minutes or very long.

Fig. 11 Aluminum absorption of 4 hour Pm^{144} from Pr + α bombardments. K x-ray and γ-ray background (A), 1.3 Mev positron (B).


100
10
1.0
% TRANSMISSION
A
B
0
200
400
600
800
1000
1200
mg/cm² ALUMINUM ABSORBER

C. Terbium Isotopes[20,21]

Europium prepared by amalgam reduction method[8] and by ion exchange resin column[1] was bombarded with 38, 31, and 19 Mev helium ions to produce the four new terbium isotopes summarized in Table VI

Table VI

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	Energy of Radiation in Mev γ-rays	Produced by
Tb^{153}	K, e^-, γ	5.1±0.1 days	0.15(e^-) 0.2-0.4	L, K x-rays 0.23,1.2	Eu^{151}-α,2n
Tb^{154}	K, e^-, β^+, γ (β^-?)	17.2±0.3hours	0.13,0.5(e^-) 2.6(β^+)	L,K x-rays 1.3	Eu^{151}-α,3n Eu^{153}-α,n
Tb^{155}	K, e^-, γ	188 days	0.1(e^-)	L,K x-rays 1.4	Eu^{153}-α,2n
Tb^{156}	β^+	5.00±0.05hours	1.3	L,K x-rays	Eu^{153}-α,n

1. 5.1 day Tb^{153}

A 5.1 day activity was found in high yields in 38 and 31 Mev helium ion bombardments of europium. It was followed through 9.6 half-lives (Fig. 12) to give a value of 5.1 ± 0.1 days. Aluminum absorption (Fig. 13) shows two electrons, range 32 mg/cm^2 (0.15 Mev) and 80-120 mg/cm^2 (0.2-0.4 Mev), soft electromagnetic radiation of half-thickness 7 mg/cm^2 (6.3 Kev), and hard electromagnetic radiation. Lead absorption (Fig. 14) shows three components of half-thicknesses 60 mg/cm^2 (46 Kev), 570 mg/cm^2 (0.23 Mev) and 11.6 g/cm^2 (1.2 Mev). The 6.3 Kev and 46 Kev radiations correspond to L and K x-radiation of gadolinium respectively. The following ratios were obtained from these measurements:

Fig. 12 Gross decay of 5.1 day Tb^{153}(B) and 188 day Tb^{155} from Eu + α bombardment.

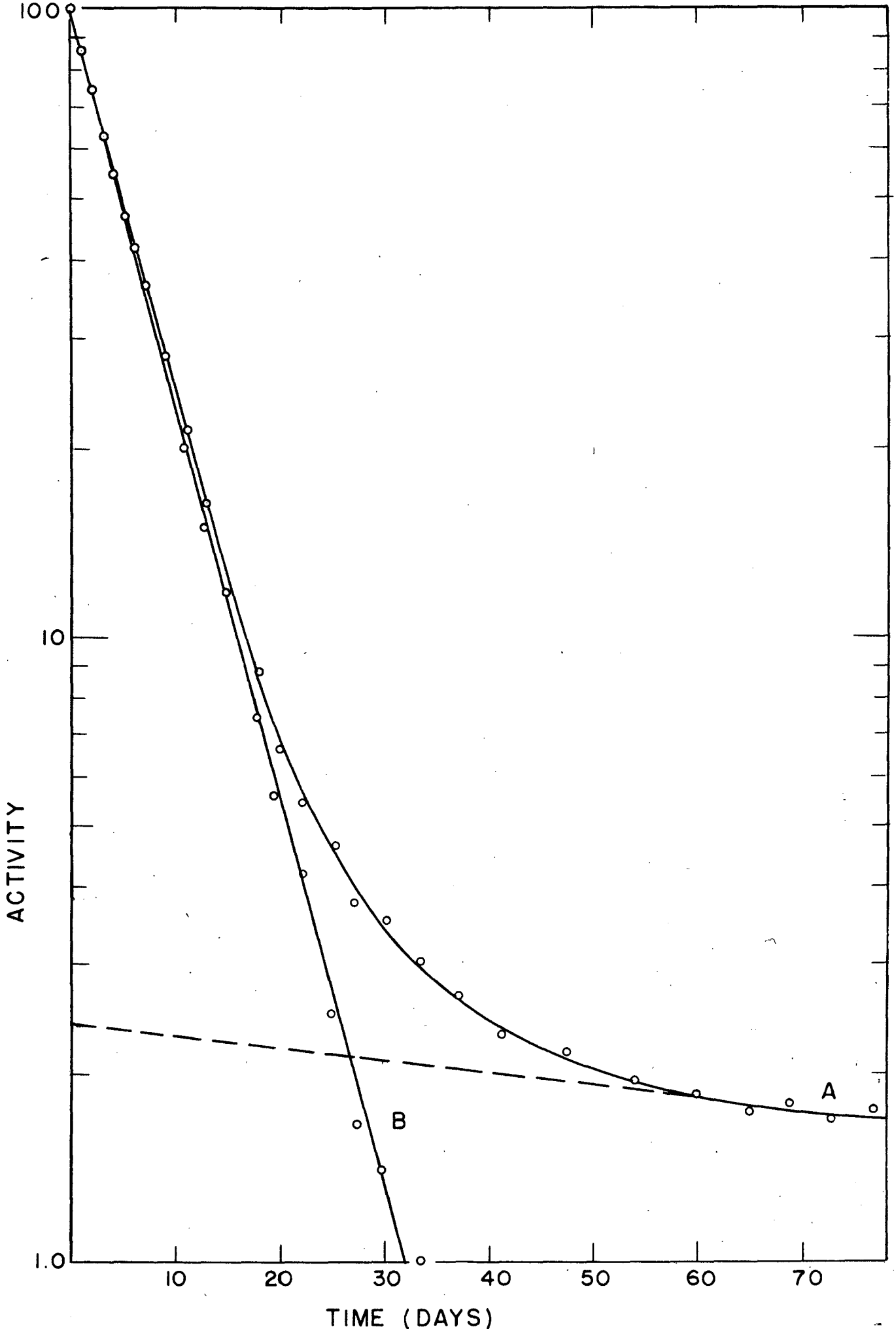

100
10
1.0
ACTIVITY
10
20
30
40
50
60
70
TIME (DAYS)
A
B

Fig. 13 Aluminum absorption of 5.1 day Tb^{153} from Eu + α bombardment. K x-ray and γ-ray background (A), 0.2-0.4 Mev electron (B), 0.15 Mev electron (C), L x-rays (D).

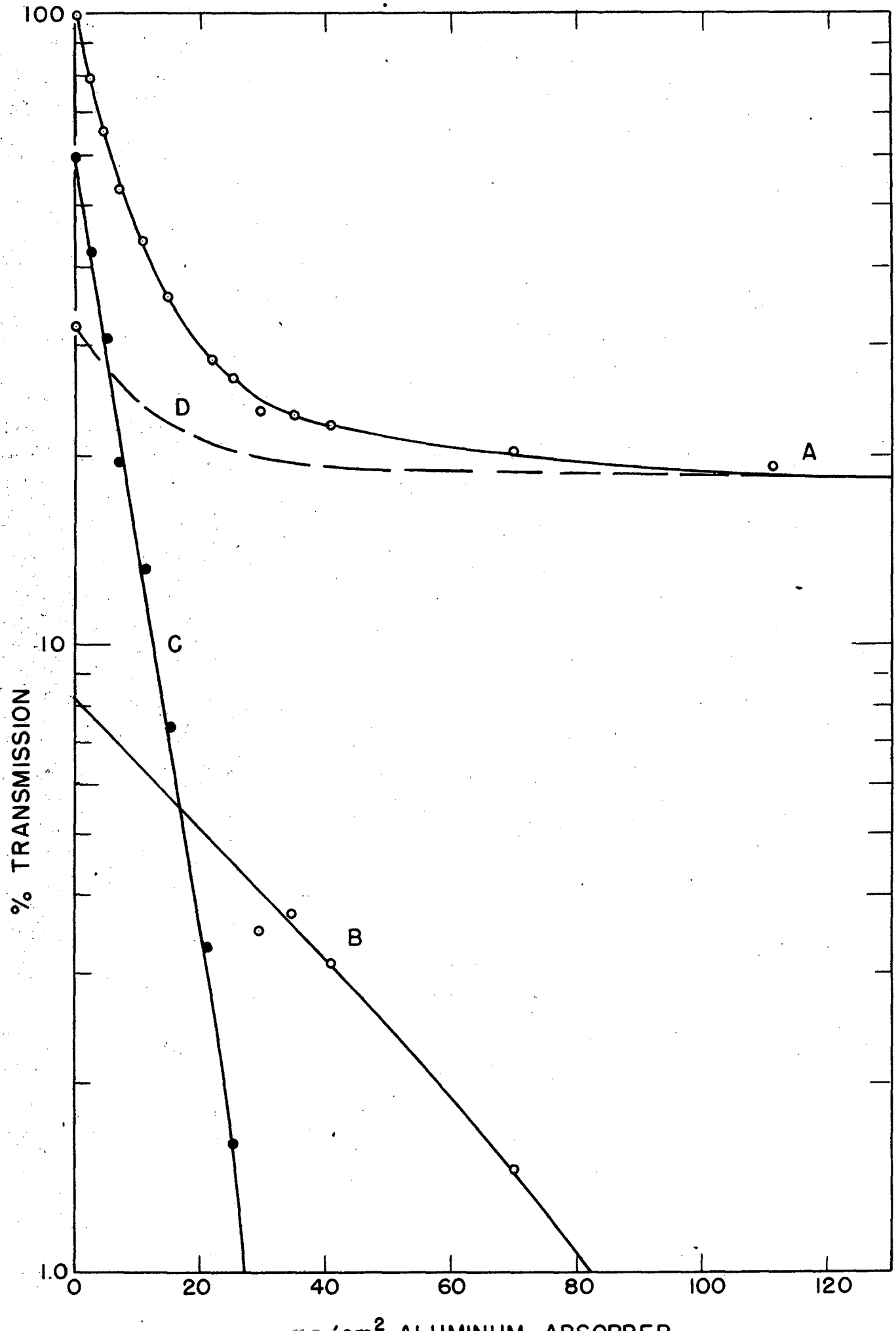
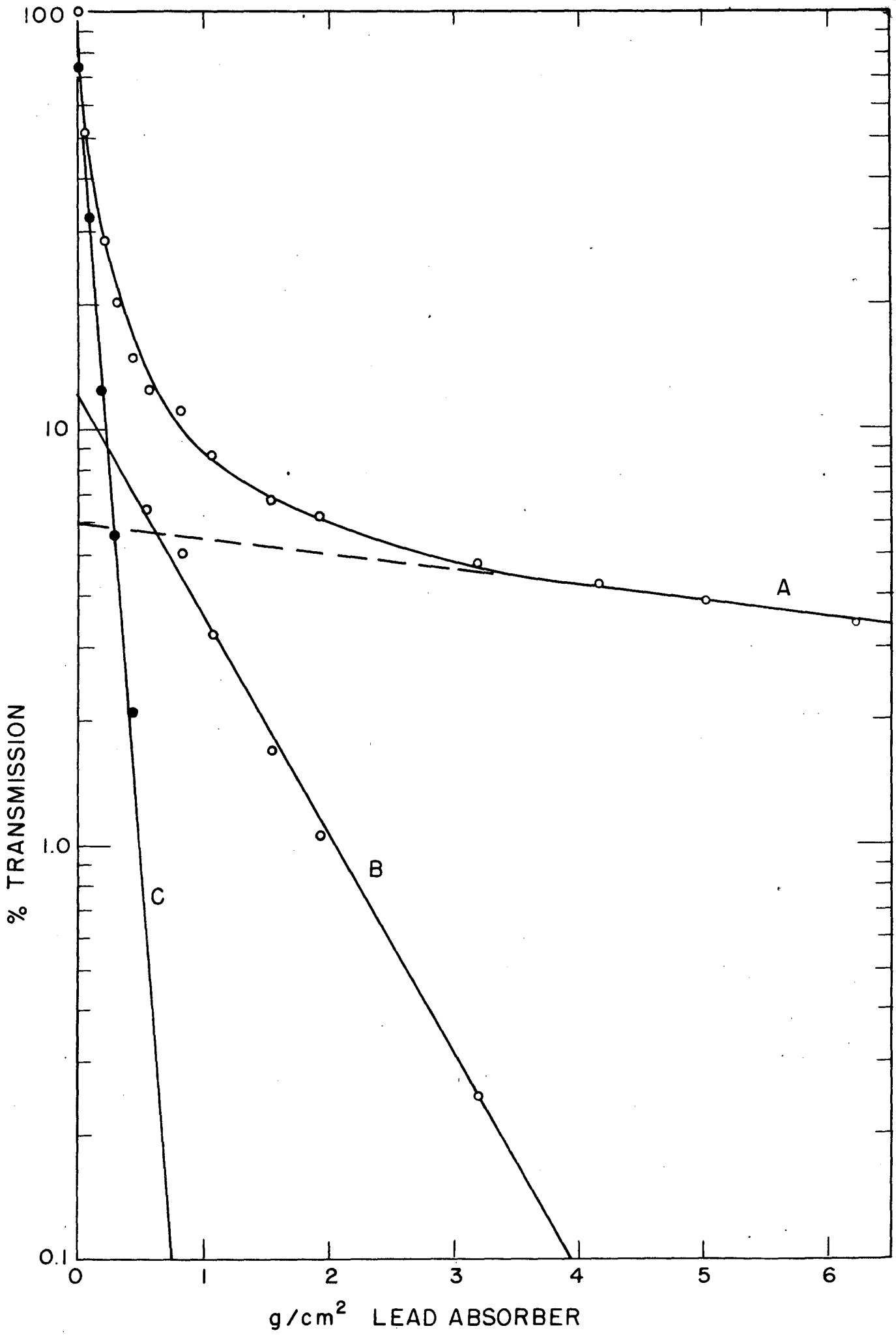

100
10
1.0
% TRANSMISSION
0
20
40
60
80
100
120
mg/cm² ALUMINUM ABSORBER
A
B
C
D

Fig. 14 Lead absorption of 5.1-day Tb^{153} from Eu + α bombardment. 0.23 Mev γ-ray (A), 1.2 Mev γ-ray (B), K x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
0
1
2
3
4
5
6
g/cm² LEAD ABSORBER
A
B
C

0.17 Mev e^-	0.2-0.4 Mev e^-	L x-rays	K x-rays	0.22 Mev γ	1.2 Mev γ
= 0.02	0.001	0.2	1	0.08	0.02

The soft electron corresponds well with the energy expected for conversion of the 0.22 Mev γ-ray in the K-shell. The isotope probably decays by orbital electron capture to two or more excited states of the daughter nucleus; one K x-ray quantum is assumed to represent one disintegration. The origin of the hard electron is not obvious.

2. 17.2 hour Tb^{154}

A 17-hour positron activity was observed in all bombardments of europium with helium ions. The activity was followed on the crude beta-ray spectrometer for 8.4 half-lives to give a value of 17.2 $\pm$ 0.3 hours. The gross and γ-decays (Figs. 15,16) also showed a 17 hour half-life.

Aluminum absorption (Fig. 17) shows electrons of range 28 mg/cm^2(0.13 Mev) and $\sim$300 mg/cm^2 (0.8 Mev), soft electromagnetic radiation of half-thickness 7 mg/cm^2 (6.3 Kev) and hard electromagnetic radiation. A positron of range corresponding to 2.6 Mev was found on the crude beta-ray spectrometer. The ratio between total negative electrons and positrons was determined by measuring the ratio of areas under each curve, (cf. Tm^{166} and Fig. 26). The lead absorption (Fig. 18) showed two components of half-thicknesses 60 mg/cm^2(46 Kev) and 12.2 g/cm^2 (1.3 Mev). The two soft quantum radiations correspond with L and K x-ray energies of gadolinium. Annihilation radiation is in too low abundance to be seen. The following ratios were obtained from these measurements:

0.13 Mev e^-	0.8 Mev e^-	2.6 Mev β^+	L x-rays	K x-rays	1.3 Mev γ-rays
0.1	0.02	0.004	0.3	1	0.03

It seems reasonable to conclude that the isotope decays at least 98% by orbital electron capture to two or more metastable levels of the daughter nucleus. According to Wilkinson's postulates concerning stable isotopes[18], the hard

Fig. 15 Gross decay of 5.0 hour Tb^{156} (C), 17.2 hour Tb^{154} (B), and 5.1 day Tb^{153} (A) from Eu + α bombardment.

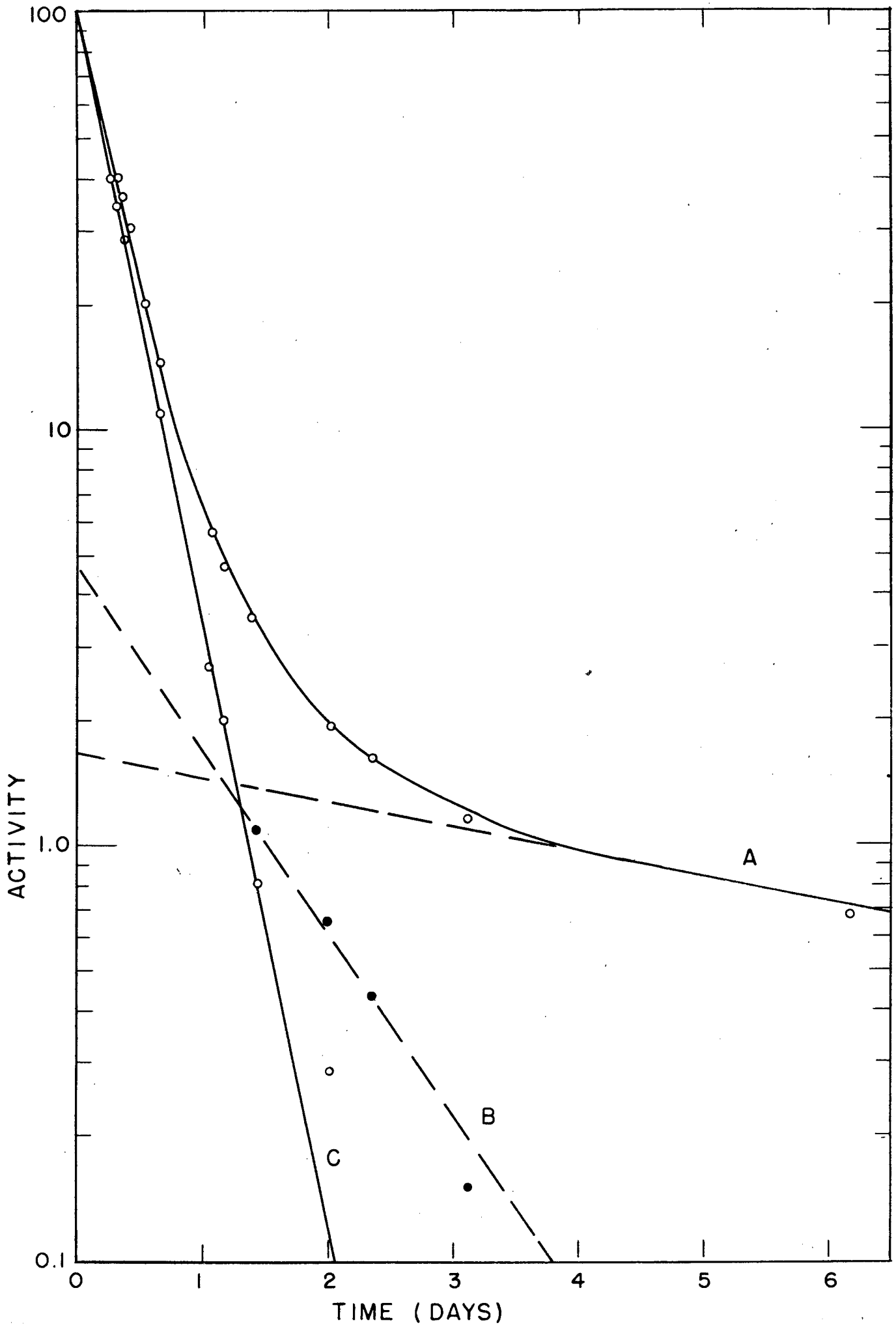

100
10
1.0
0.1
ACTIVITY
0
1
2
3
4
5
6
TIME (DAYS)
A
B
C

Fig. 16 Electromagnetic decay of 5.0 hour Tb^{156} (C), 17.2 hour Tb^{154} (B), and 5.1 day Tb^{153} (A) from Eu + α bombardment.

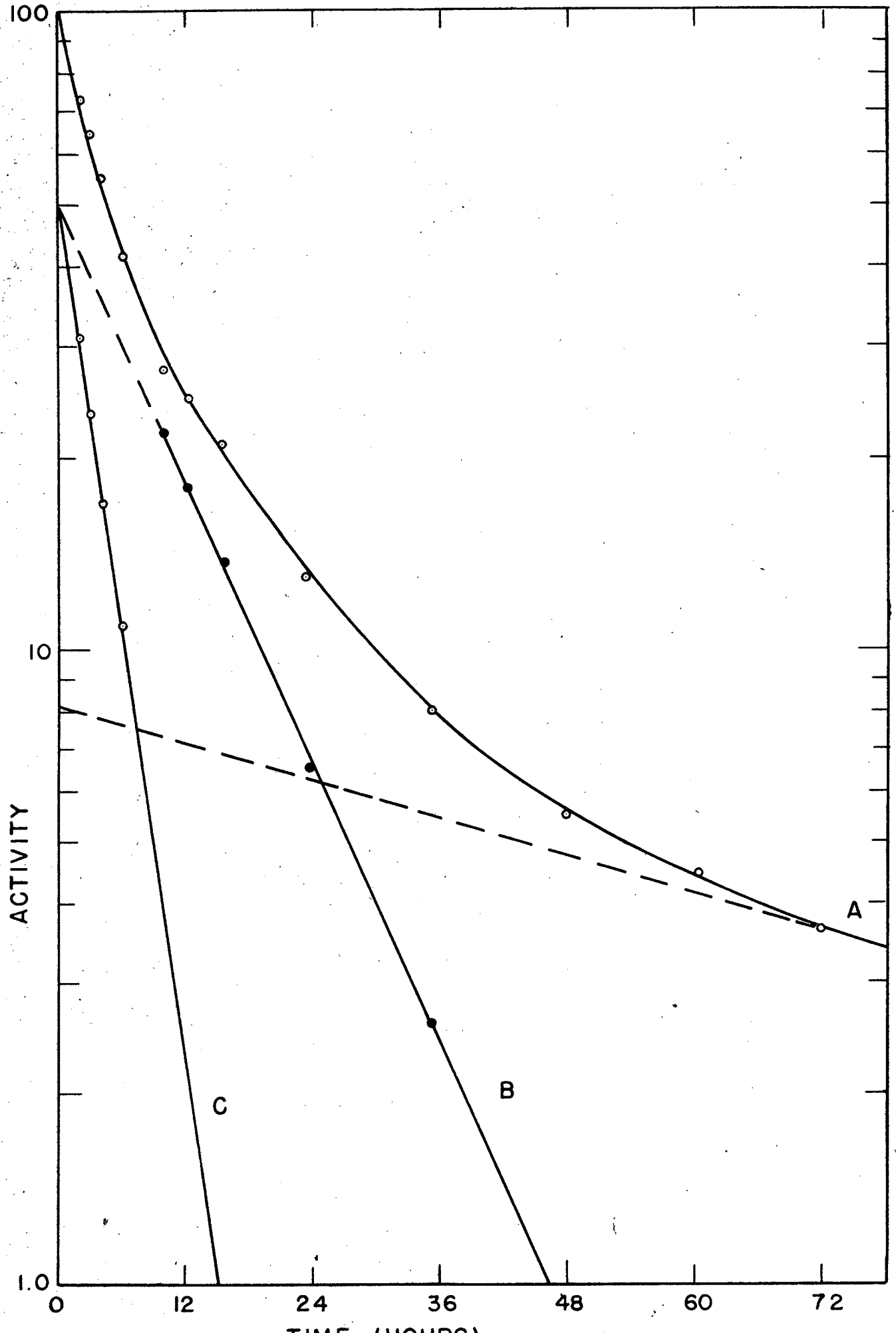

100
10
1.0
ACTIVITY
0
12
24
36
48
60
72
A
B
C

Fig. 17 Aluminum absorption of 17.2 hour Tb^{154} from Eu + α bombardment. K x-rays and γ-ray background (A), 0.8 Mev electron (B), 0.13 Mev electron (C) L x-rays (D).

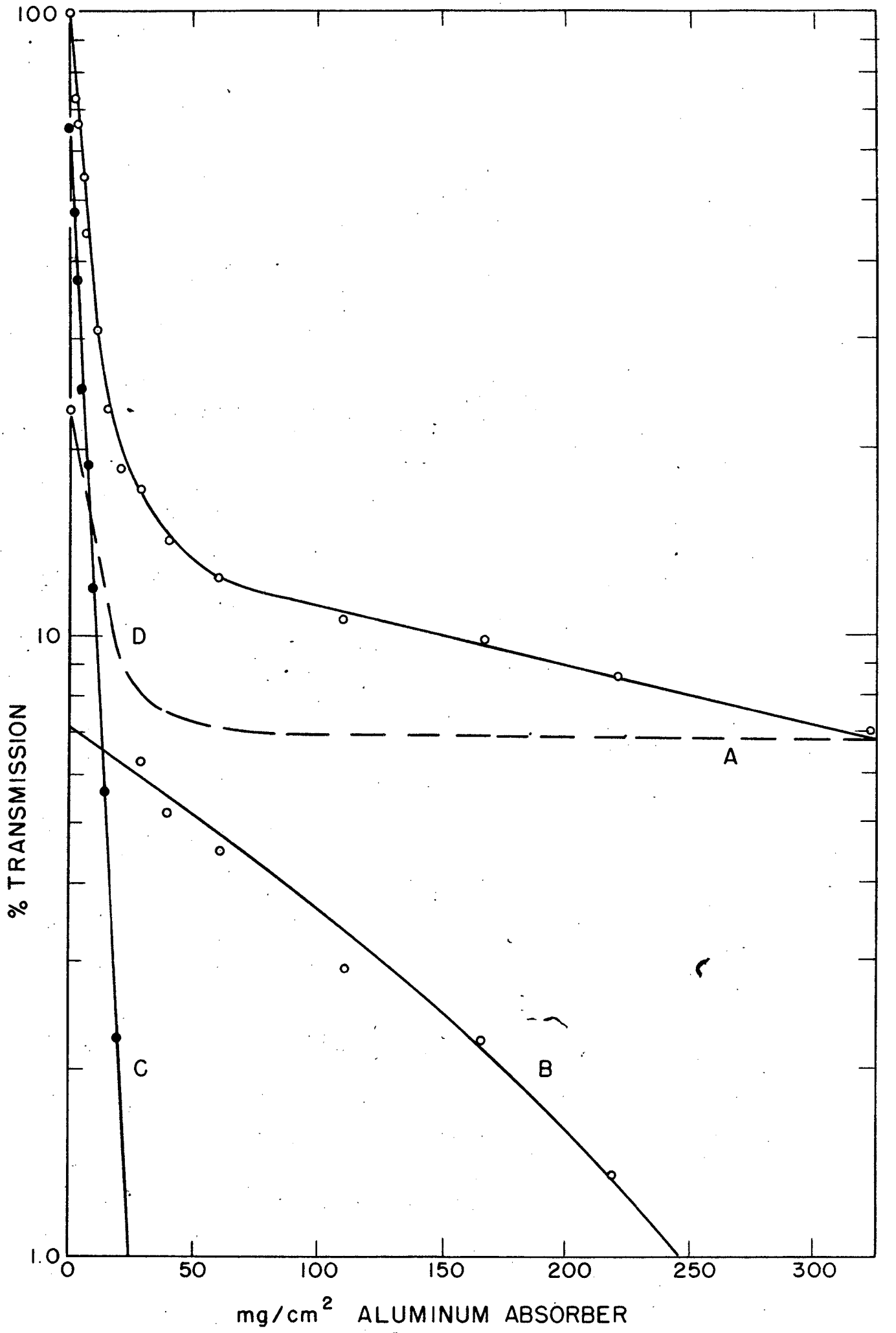
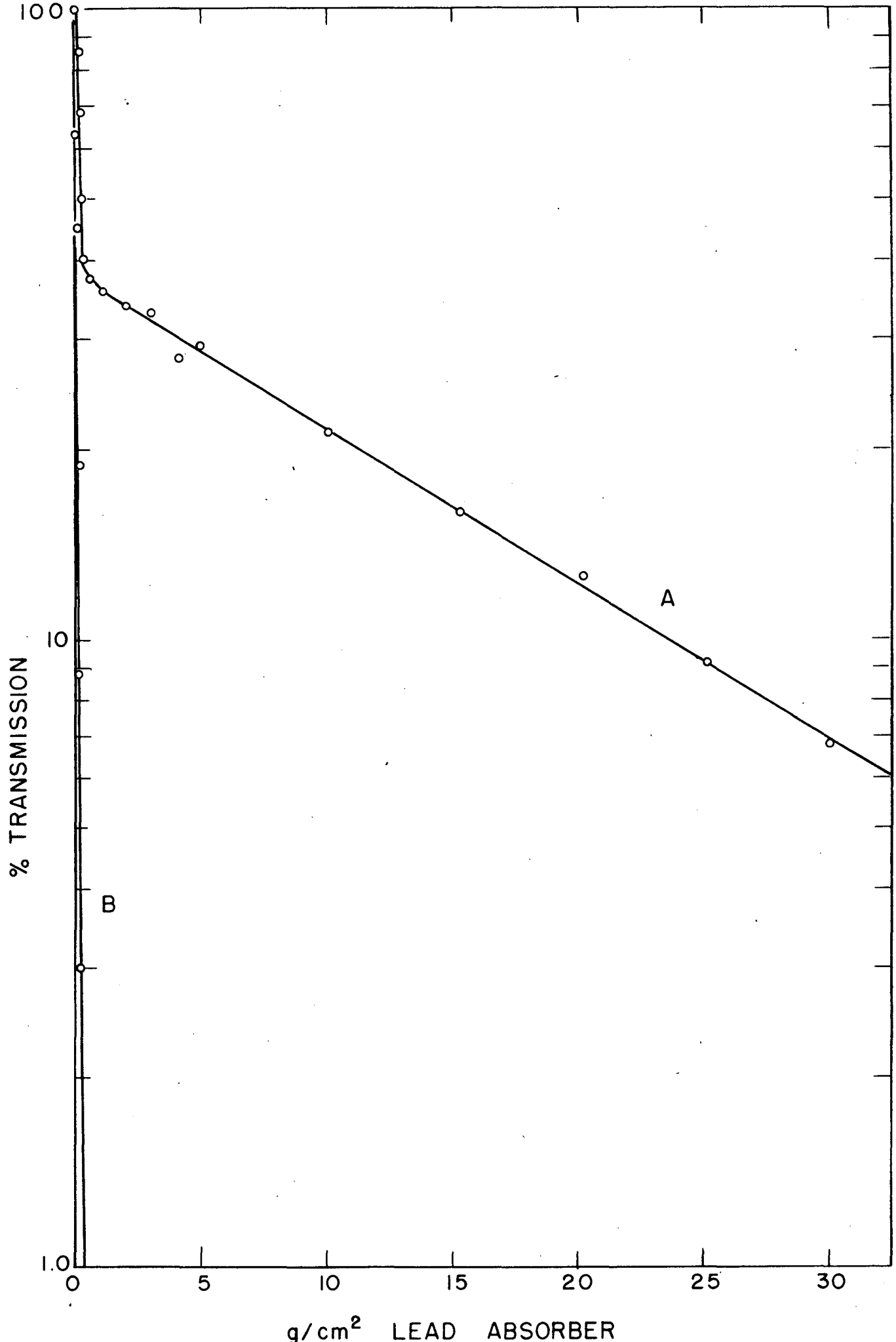

100
10
1.0
% TRANSMISSION
0
50
100
150
200
250
300
mg/cm^2 ALUMINUM ABSORBER
A
B
C
D

Fig. 18 Lead absorption of 17.2 hour Tb^{154} from Eu + α bombardment. 1.3 Mev γ-ray (A) K x-ray (B).


100
10
1.0
% TRANSMISSION
A
B
0
5
10
15
20
25
30
g/cm² LEAD ABSORBER

negative electron might be a beta particle. If this is true, then the isotope decays 98% by the capture process, 2% by beta-branching, and 0.4% by positron emission. 0.9 K x-ray quanta have been assumed to represent one disintegration by orbital electron capture.

3. 188 day Tb^{155}

After the decay of the 5.1 day activity from Eu + α bombardments, a long-lived activity was observed (Fig. 12). It has been followed through one half-life to give a value of 188 days. Aluminum absorption (Fig. 19) showed a soft electron, range 14 mg/cm^2 (0.1 Mev), soft electromagnetic radiation of half-thickness 7 mg/cm^2 (6.3 Kev) and hard electromagnetic radiation. A lead absorption (Fig. 20) showed two components of half-thicknesses 60 mg/cm^2 (46 Kev) and 12.7 g/cm^2 (1.4 Mev). The two soft components agree with the energy of gadolinium L and K x-rays. The following ratios were obtained from these measurements:

0.1 Mev e^-	:	L x-rays	:	K x-rays	:	1.4 Mev γ
0.4	:	0.2	:	1	:	0.3

The isotope probably decays by orbital electron capture to two or more excited states of the daughter nucleus. 0.6 K x-ray quanta are assumed to represent one disintegration by orbital electron capture.

4. 5.0 hour Tb^{156}

A short-lived activity was observed in bombardments of europium with 19 Mev helium ions. Measured through nine half-lives (Figs. 15 and 16). The period was 5.0 ± 0.1 hours. Aluminum absorption (Fig. 21* showed only a hard electron, range 600 mg/cm^2 (1.3 Mev), soft electromagnetic radiation, half-thickness of 7 mg/cm^2 (6.3 Kev), and hard electromagnetic radiation. Study of this isotope on a crude beta-ray spectrometer showed only the presence of 1.4 Mev positrons which decayed with a 5.0 hour period; no negative electrons were observed for this isotope. Lead absorptions were not taken because of insufficient activity. The

Fig. 19 Aluminum absorption of 188 day Tb^{155} from Eu + α bombardment. K x-rays and γ-ray background (A) 0.1 Mev electron (B), L x-rays (C).

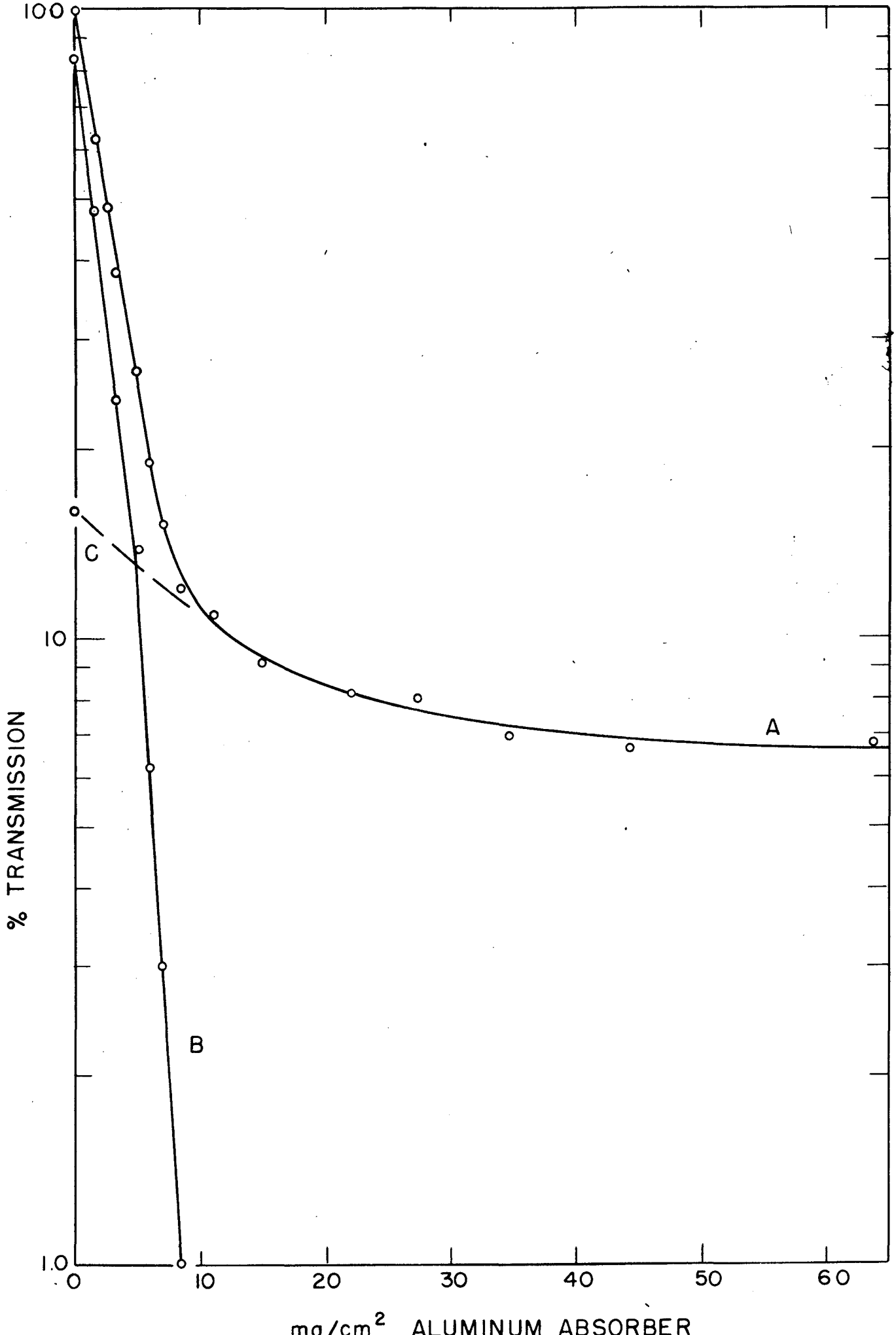

100
10
1.0
% TRANSMISSION
0
10
20
30
40
50
60
mg/cm² ALUMINUM ABSORBER
A
B
C

Fig. 20 Lead absorption of 188 day Tb^{155} from Eu + α bombardment. 1.4 Mev γ-ray (A), K x-ray (B).

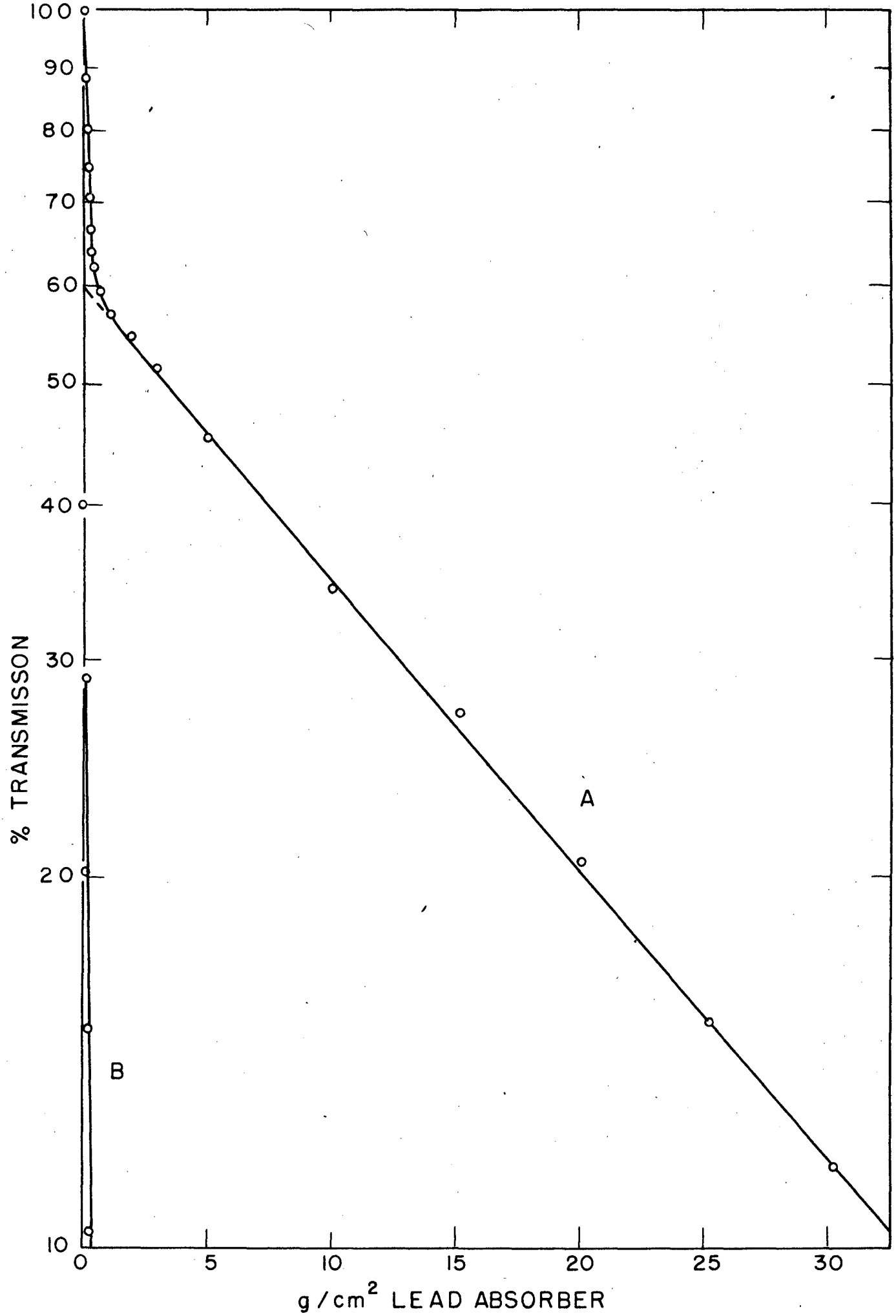
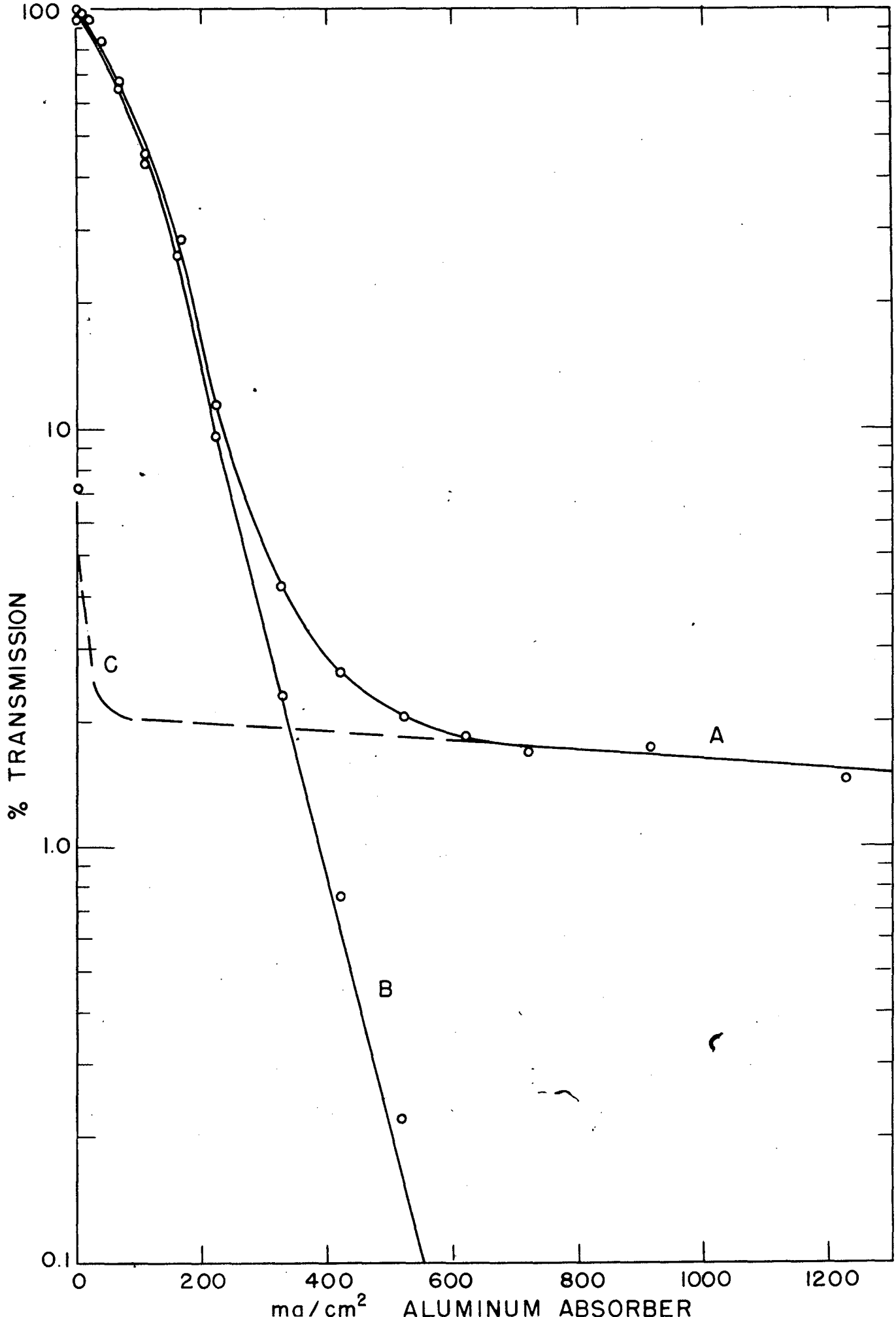

100
90
80
70
60
50
40
30
20
10
% TRANSMISSON
A
B
0
5
10
15
20
25
30
g / cm² LEAD ABSORBER

Fig. 21 Aluminum absorption of 5.0-hour Tb^{156} from Eu + α bombardment. K x-rays and γ-ray background (A), 1.3 Mev positron (B), L x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
0
200
400
600
800
1000
1200
mg/cm²
ALUMINUM ABSORBER
A
B
C

approximate ratios obtained from the aluminum absorption, assuming all x-radiation to be annihilation radiation, are as follows:

1.3 Mev β^+	L x-rays	K x-rays	γ-rays
0.25	0.13	1	0.25

The isotope thus appears to decay 20% by positron emission. Since Dy^{156} is known to be beta stable, Tb^{156} should show negative beta particle branching. Since this has not been observed, a long-lived negative beta particle emitting isomer must be postulated[18]. For cross-section calculations, it was assumed that one K x-ray quantum represented one disintegration by orbital electron capture.

5. Discussion

Europium has two stable isotopes, Eu^{151} and Eu^{153}, therefore, using 38, 31 and 19 Mev helium ions, one should see activities from five mass numbers, Tb^{152} to Tb^{156}. Only four activities were seen, leading to the conclusion that Tb^{152} is too short-lived (less than 20 minutes) to be observed. Table VII shows the measured cross sections in barns.

Table VII

Half-Life	Energy of Helium Ions 38 Mev	31 Mev	19 Mev	Probable Mass
5.1 days	0.4	6×10^{-3}	3×10^{-4}	153
17.2 hours	1.2	2×10^{-3}	6×10^{-5}	154
188 days	5×10^{-2}	2×10^{-3}	2×10^{-5}	155
5.0 hours	--	--	8×10^{-4}	156

At 31 and 19 Mev, the sum of the cross sections fall far short of one barn, so it appears that there are probbly undetected isomers of these masses. The assignments were made in order to be consistent with the variation of cross section with energy, and the generality that in this region, positron emitters of elements of

odd atomic number are of even mass number.

D. Holmium Isotopes[20,21]

Three new activities of holmium (Table VIII) were observed in a helium ion bombardment of terbium and proton bombardments of dysprosium.

Table VIII

Isotope	Type of Radiation	Half-Life	Energy of Radiations in Mev Particles	Energy of Radiations in Mev γ-rays	Produced by
Ho^{164}	β^-, γ	34 ± 1 minutes	0.95		Dy-p-n
Ho^{162}	K, e^-, β^-	60 ± 2 days	$0.12(e^-)$ $0.7(\beta^-)$		Tb-α-n
$Ho^{160,161}$	K, e^-, β^+, γ	4.5 ± 0.1 hours	$0.09, > 0.6(e^-)$ $\sim 2(\beta^+)$	L, K x-rays 1.1	Dy-p-n Tb-α-3n or 2n

1. 34-minute Ho^{164}

An activity of 47-minute half-life produced by fast neutron bombardment of holmium has been reported in an early paper by Pool and Quill[10].

The 34-minute beta active isotope here described was produced by short bombardments of very pure dysprosium oxide with 10 Mev protons. Identification of the activity with holmium has not been proved by chemical separation, but no activities due to known dysprosium isotopes have been observed, and longer-lived activities from the same bombardment have been shown to follow holmium chemistry. The decay has been followed for both electron and electromagnetic radiation. A half-life of 34 ± 1 minutes was obtained through eight half-lives of the gross decay (Fig. 22) and four half-lives of the γ-decay. The aluminum absorption curve of the 34-minute activity is shown in Fig. 23. Correction was made for decay of the sample during the time of measurement, and the contribution of the longer-

Fig. 22 Gross decay of 35 minute Ho^{164} (B) and 4.5-hour $Ho^{160,161}$ (A) from Dy + p bombardment.

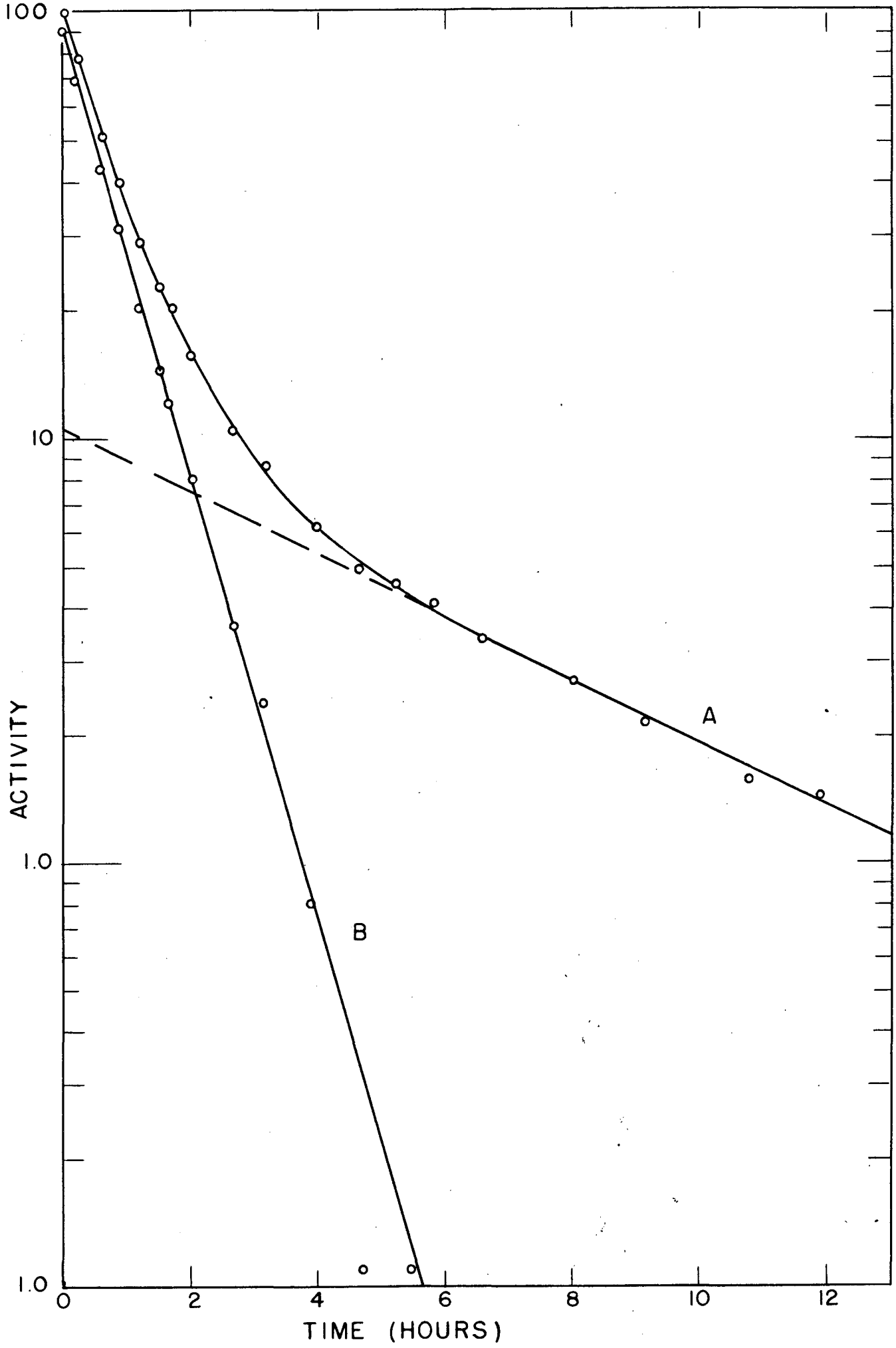
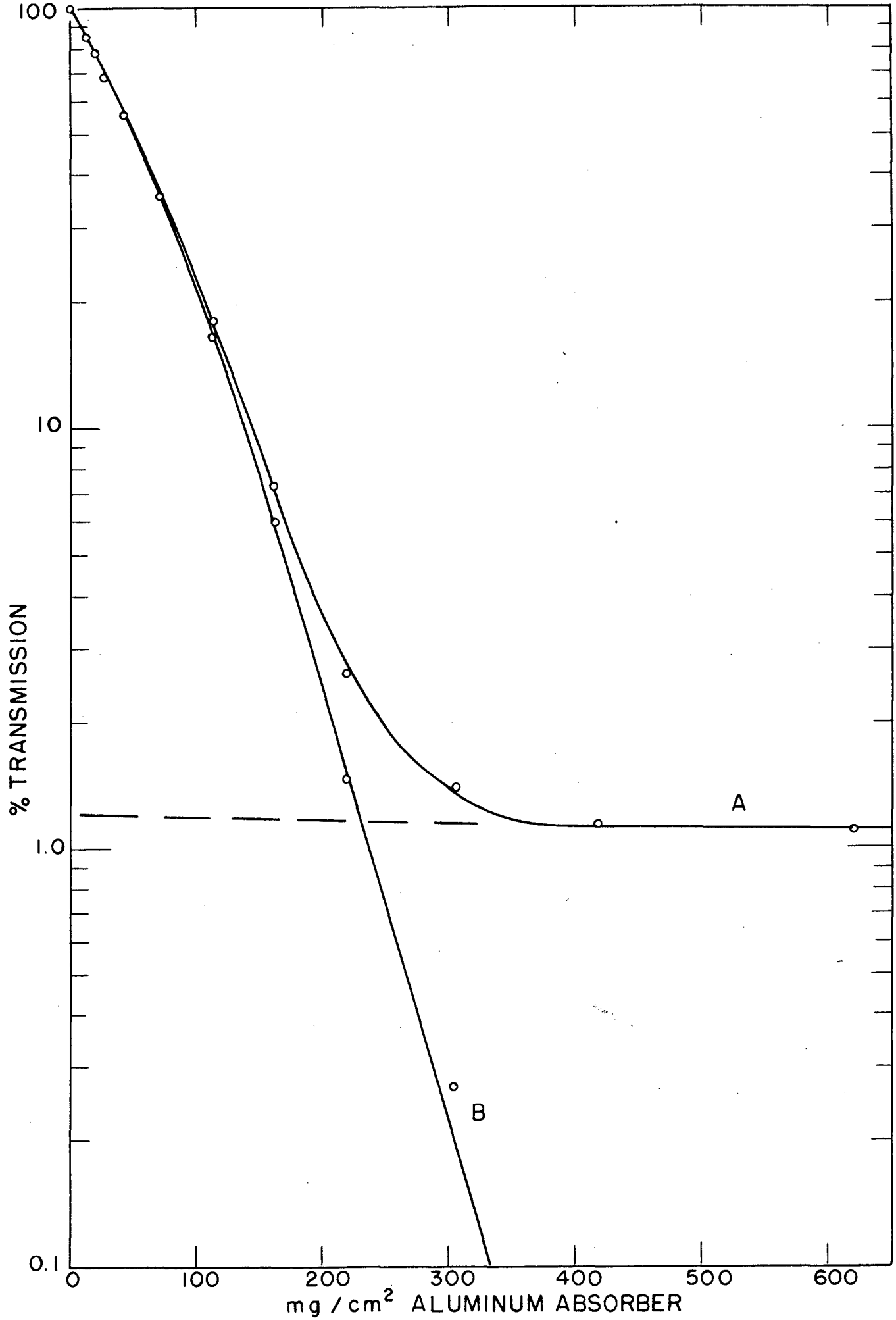

100
10
1.0
1.0
ACTIVITY
0
2
4
6
8
10
12
TIME (HOURS)
A
B

Fig.23 Aluminum absorption of 35 minute Ho^{164} from Dy + p bombardment. K x-ray and γ-ray background (A), 0.9 Mev beta (B).


100
10
1.0
0.1
% TRANSMISSION
0
100
200
300
400
500
600
mg / cm² ALUMINUM ABSORBER
A
B

lived holmium activities was subtracted. The lead absorption of the 34-minute electromagnetic radiation was not measured due to lack of intensity of the samples. From the aluminum absorption curves, the beta particle has a range of 340 mg/cm^2 aluminum (0.9 Mev). A Feather analysis of the curve gives an energy of 0.95 Mev. If a counting efficiency of one percent is assumed for the γ-radiation there is one γ-ray quantum per beta particle. No evidence has been found for any conversion electrons, either in aluminum absorptions, or on the magnetic counter.

2. 4.5-hour $Ho^{160,161}$

An activity of 4.5 hours half-life was observed in the column separated holmium fraction from both Tb + α and Dy + p bombardments. The radiation characteristics were measured on unseparated bombarded terbium samples; no activities other than those of holmium have been observed. The gross (Fig. 22) and electromagnetic radiations have been followed for decay to give a value of 4.5$\pm$0.1 hours through six half-lives. The aluminum absorption curve (Fig. 24) shows hard electromagnetic radiation, soft electromagnetic radiation of half-thickness 8.5 mg/cm^2 (6.8 Kev), hard electrons of range $>$200 mg/cm^2 ($>$0.6 Mev) and soft electrons of range 12 mg/cm^2 (0.09 Mev). Examination of the electrons on the crude beta-ray spectrometer demonstrated the presence of positrons of maximum energy about 2 Mev, but only in about one-hundredth the yield of hard electrons of energy about 1 Mev and mean energy about 0.3 Mev. The hard electron radiation comprises only a few percent of the total radiations measured in the aluminum absorption.

The lead absorption curve (Fig. 25) shows components of half-thicknesses 60 mg/cm^2 lead (45 Kev) and 11.0 g/cm^2 lead (1.1 Mev). The two soft electromagnetic radiations agree with the energies of dysprosium L and K x-radiation. From the measurements, the following corrected ratios were obtained:

0.09 Mev e^-	: total hard electron	: L x-rays	: K x-rays	: 1.1 Mev γ =
0.3	0.005	1	1	1

Fig. 24 Aluminum absorption of 4.5 minute $Ho^{160,161}$ from Dy + p bombardment. K x-ray and γ-ray background (A). >0.6 Mev electron (B), 0.09 Mev electron (C), L x-rays (D)

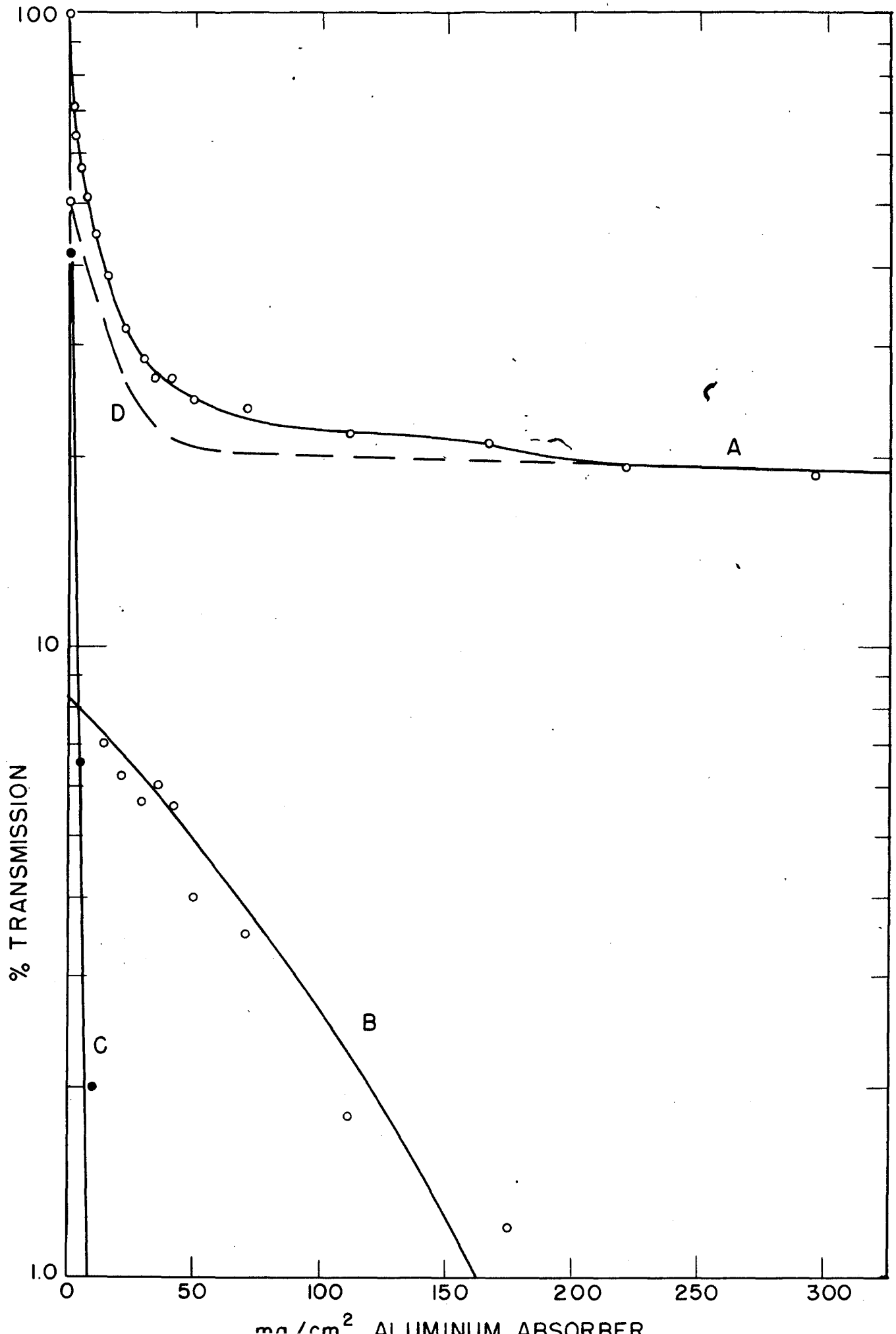
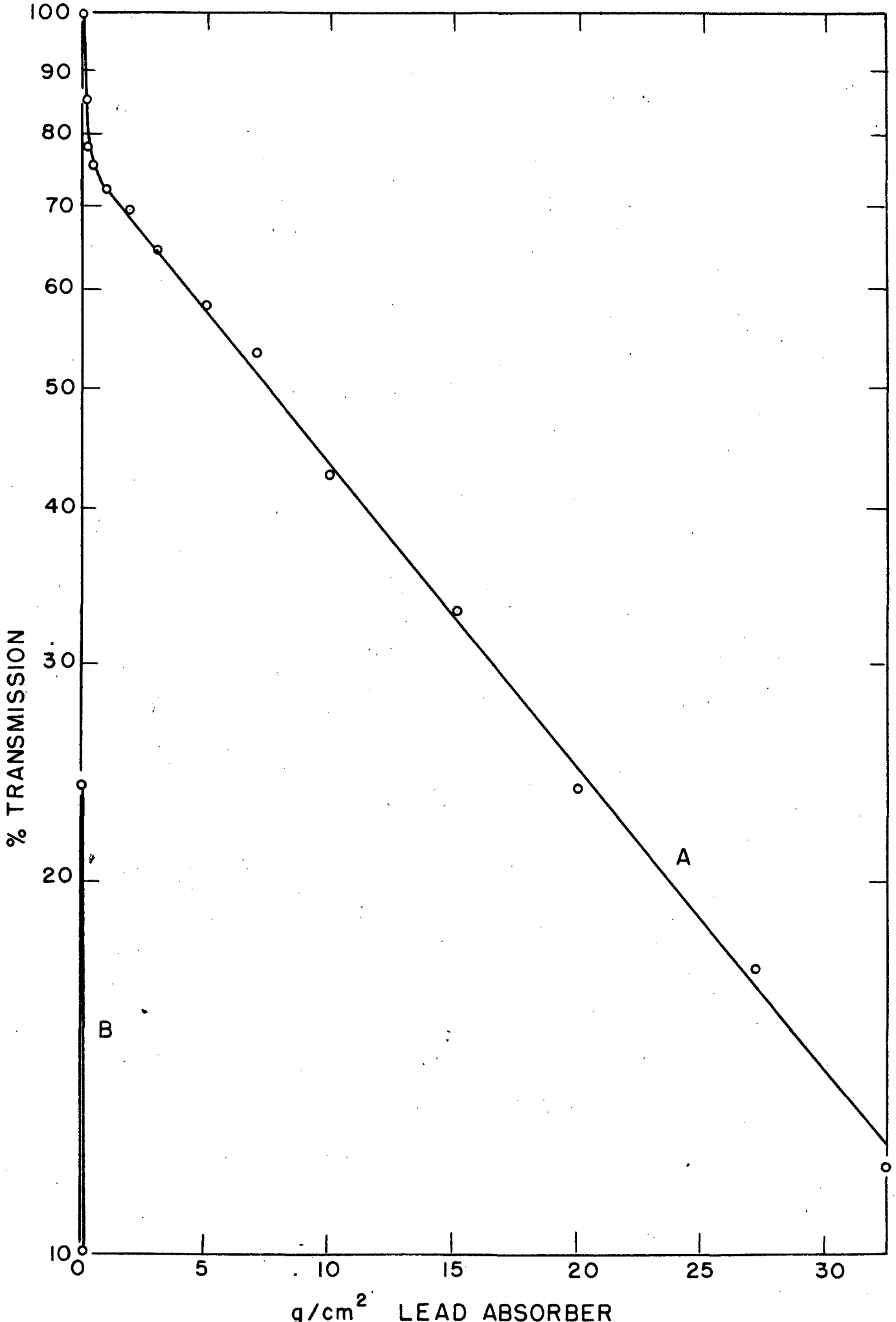

100
10
1.0
% TRANSMISSION
0
50
100
150
200
250
300
mg/cm² ALUMINUM ABSORBER
A
B
C
D

Fig. 25 Lead absorption of 4.5 hour $Ho^{160,161}$ from Dy + p bombardment 1.1 Mev γ-ray (A), K x-ray (B).


100
90
80
70
60
50
40
30
20
10
% TRANSMISSION
A
B
0
5
10
15
20
25
30
g/cm² LEAD ABSORBER

It seems reasonable to assume that the activity decays by orbital electron capture; since the positrons are only about one hundredth of the hard electrons, which may arise from beta particle branching if the isotope is Ho^{160} (18). The branching ratio for beta emission can be only of the order of 0.5% while that of positron emission can be only of the order of 0.005%.

3. 60-day Ho^{162}

After the decay of the 4.5-hour isotope a long-lived activity was seen in the Tb + α bombardment. The gross decay was followed through six half-lives to give a value of 60 ± 2 days. The aluminum absorption (Fig. 26) shows two electrons of ranges 15 mg/cm^2 (0.1 Mev) and 250 mg/cm^2 (0.7 Mev), Feather range 300 mg/cm^2 (0.8 Mev); and electromagnetic radiation of half-thicknesses 9 mg/cm^2 (6.5 Kev) and 1700-1800 mg/cm^2 (50 Kev). The soft radiation corresponds to dysprosium L x-radiation while the half-thickness value for the harder electromagnetic component indicates that it is probably nearly all dysprosium K x-rays. The following ratios were obtained from the absorption measurement:

0.1 Mev e^-	0.7 Mev β^-	L x-rays	K x-rays
0.12	0.13	1	1

Er^{162} is known to be a beta stable isotope, therefore it seems reasonable that the hard electron is a beta particle. The beta-branching ratio is then 11%, assuming one K x-ray represents one disintegration by orbital electron capture.

E. Thulium Isotopes[6,20,21]

The three new thulium isotopes summarized in Table IX have been prepared by cyclotron bombardment.

Fig. 26 Aluminum absorption of 60-day Ho^{162} from Tb + α bombardment. K x-ray and γ-ray background (A), 0.7 Mev beta (B), 0.1 Mev electron (C), L x-rays (D).

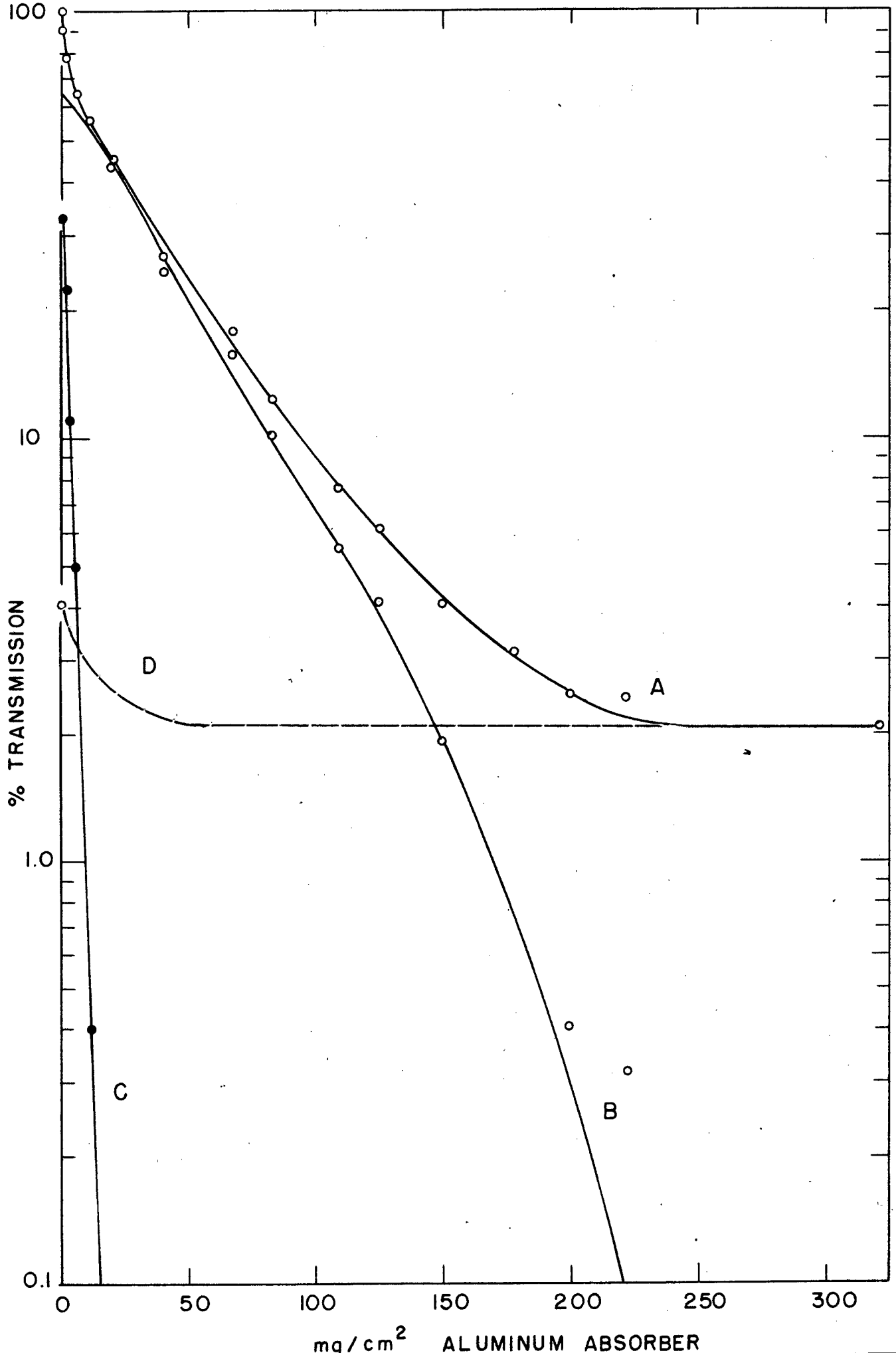

100
10
1.0
0.1
% TRANSMISSION
0
50
100
150
200
250
300
mg/cm2 ALUMINUM ABSORBER
A
B
C
D

Table IX

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	γ-ray	Produced by
Tm^{166}	$K, \beta^+, e^-, \gamma(\beta^-?)$	7.7 ± 0.1 hrs	$2.1(\beta^+) 0.24(e^-)$, $\sim 1(\beta^-?)$	L, Kx-rays 1.7	Ho-α-3n Er-p-n
Tm^{167}	K, e^-, γ	9.6 ± 0.1 days	$0.21(e^-)$	L, K x-rays 0.22, 0.95	Ho-α-2n Er-p-n Ta-d-5z, 16a
Tm^{168}	K, e^-, γ, β^-	85 ± 2 days	$0.16(e^-)$ $0.5(\beta^-)$	L, K x-rays 0.21, 0.85	Ho-α-n Er-p-n Tm-n-2n

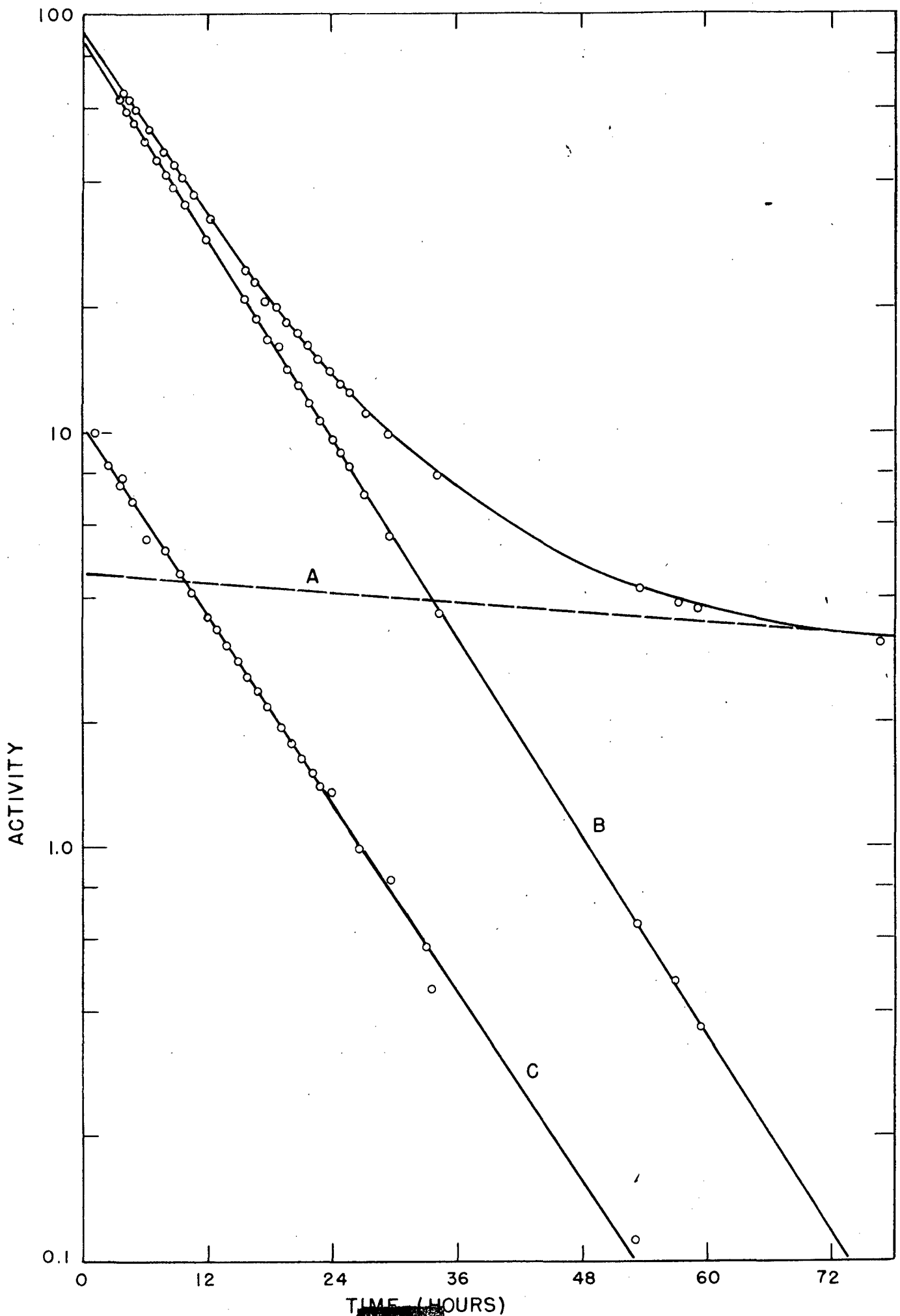
1. 7.7 hr Tm^{166}

The radiations of this isotope consist of positrons, several energies of negative electrons, L and K x-radiation, and very hard γ-radiation, all of which decay with the same half-life, 7.7 ± 0.1 hours. The gross decay of the isotope was followed through eight half-lives. (Fig. 27)

Examination of the isotope on the crude beta-ray spectrometer proved the presence of positrons. Fig. 28, Curve A, shows the measured distribution, with end point corresponding to 2.1 Mev maximum energy. The decay of the positrons, (Fig. 27, Curve C) was followed on the crude beta ray spectrometer through six half-lives in both Ho + α and Er + p bombardments.

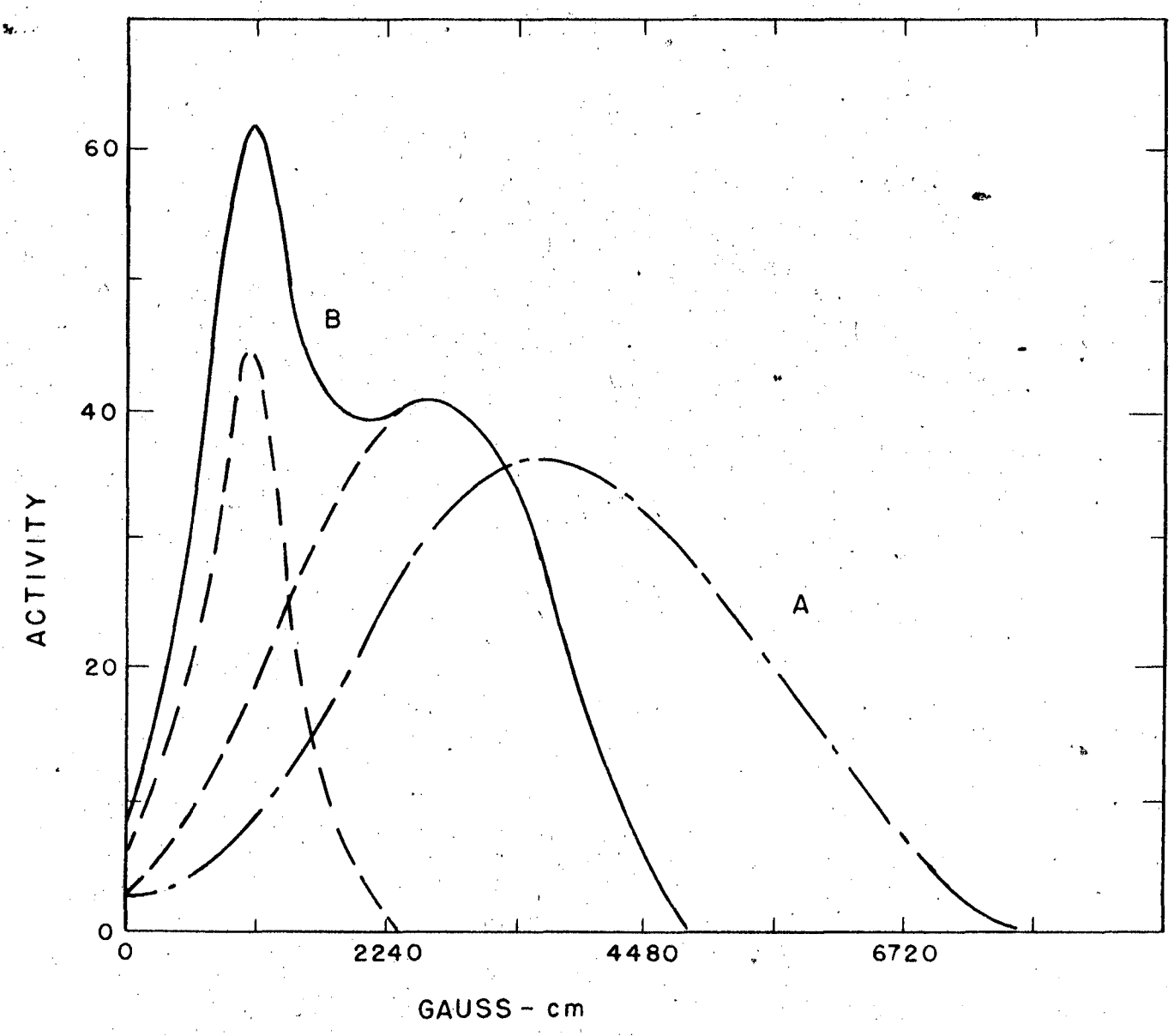
On the "negative sweep" of the crude beta ray spectrometer, two groups of negative electrons were observed (Fig. 28, Curve B) both of which decayed with 7.7-hours half-life. While the lower energy peak probably represents a single conversion electron, the second group of harder electrons has a much broader distribution of energies, and may be a mixture of several conversion electrons which the simple instrument is incapable of resolving. The maximum energy of this group

Fig. 27 Gross decay of 7.7-hour Tm^{166} (B) and 9.6-day Tm^{167} (A), and positron decay (C) measured on the crude beta-ray spectrometer from Ho + α bombardment.



13183-1

Fig. 28 Crude beta-ray spectrometer distribution of positrons (A) and electrons (B) of 7.7-hour Tm^{166} from Ho + α bombardment.


60
40
20
0
ACTIVITY
B
A
0
2240
4480
6720
GAUSS - cm

corresponds to 1 Mev, while the average energy is about 0.5 Mev.

The aluminum absorption of an infinitely thin sample is shown in Figure 29. The electromagnetic background was determined after the removal of all electrons by a 1400 mg/cm^2 beryllium absorber. No L x-radiation was observed here, although a direct beryllium absorption of the 7.7 hour activity suggests this to be present. The absorption curve shows electrons of ranges 70 mg/cm^2 and $\sim$960 mg/cm^2 aluminum, corresponding respectively to 0.24 and $\sim$2 Mev; the harder radiation is a complex mixture of positive and negative electrons. Lead absorption of the electromagnetic radiations (Fig. 30) shows two components of half-thicknesses 80 mg/cm^2 and 14.3 g/cm^2. The energy of the former, 50 Kev, agrees well with the value for erbium K x-radiation.

An approximate ratio of positive to negative electrons was obtained by measuring the areas under the curves (Fig. 28) from the crude beta-ray spectrometer. The ratio of both the soft and hard groups of electrons to the gross electromagnetic radiation was obtained from aluminum absorption measurements with appropriate corrections. The following approximate ratios were obtained from these measurements:

	0.24 Mev e^-	: 1 Mev e^-	: 2.1 Mev β^+	: K x-rays	: 1.7 Mev γ
=	0.05	: 0.003	: 0.004	: 1	: 0.3

The very complex radiations can be attributed to orbital electron capture in Tm^{166}, leading to excited or metastable levels of the daughter nucleus. Electrons arise from converted γ-ray transitions between the various levels or between these and the ground state. Positron emission may go to an excited level of the daughter nucleus, or directly to the ground state. Since the ratio of electrons to K x-radiation is only 0.05, approximately 95 percent of the K x-rays must arise from orbital electron capture, and it is assumed that 0.95 K quanta represent one disintegration. Thus, the branching ratio for disintegration by positron emission is about 0.5 percent.

Fig. 29 Aluminum absorption of 7.7-hour Tm^{166} from Ho + α bombardment K x-ray and γ-ray background (A), complex hard electrons (B) 0.24 Mev electrons (C).

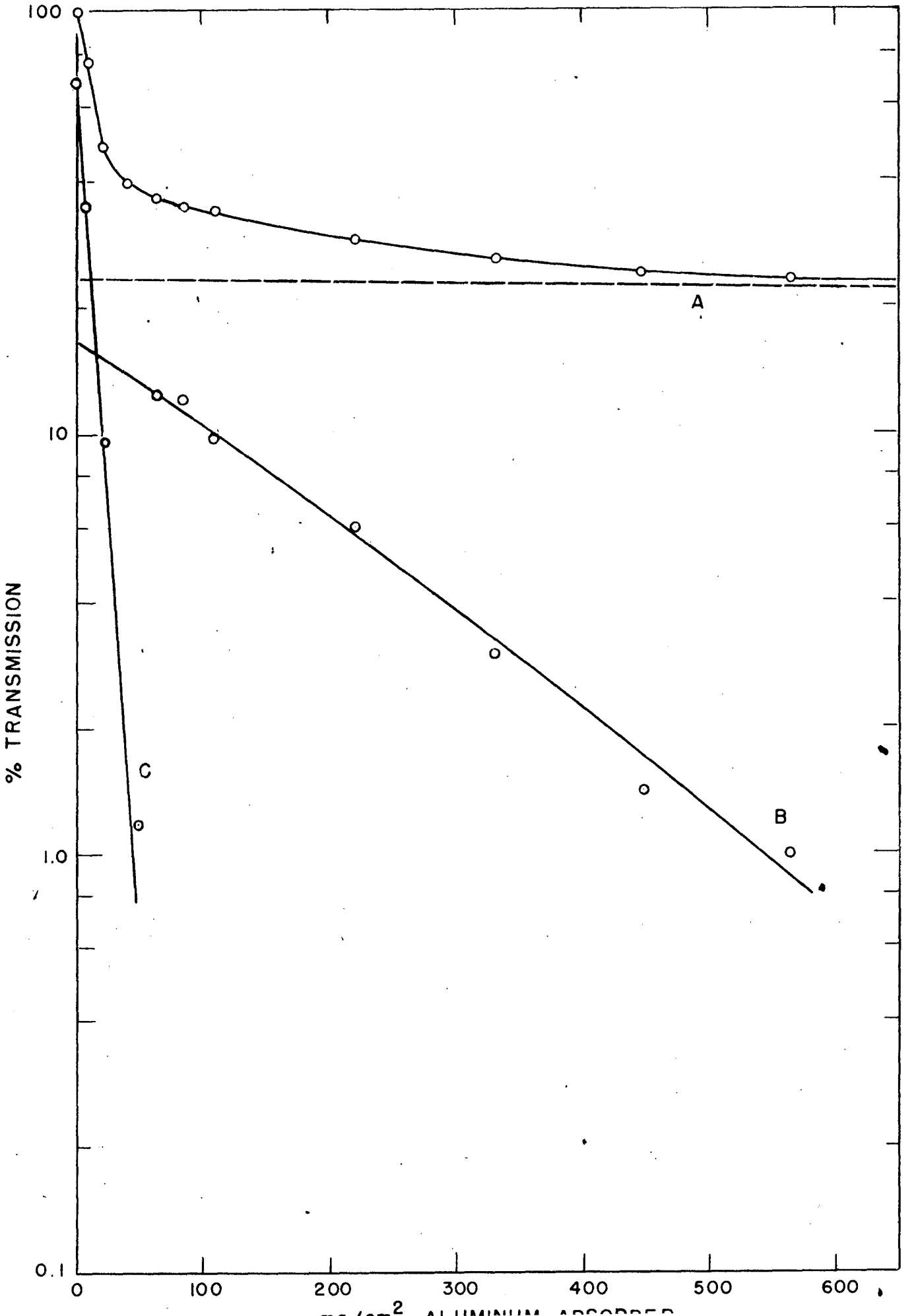
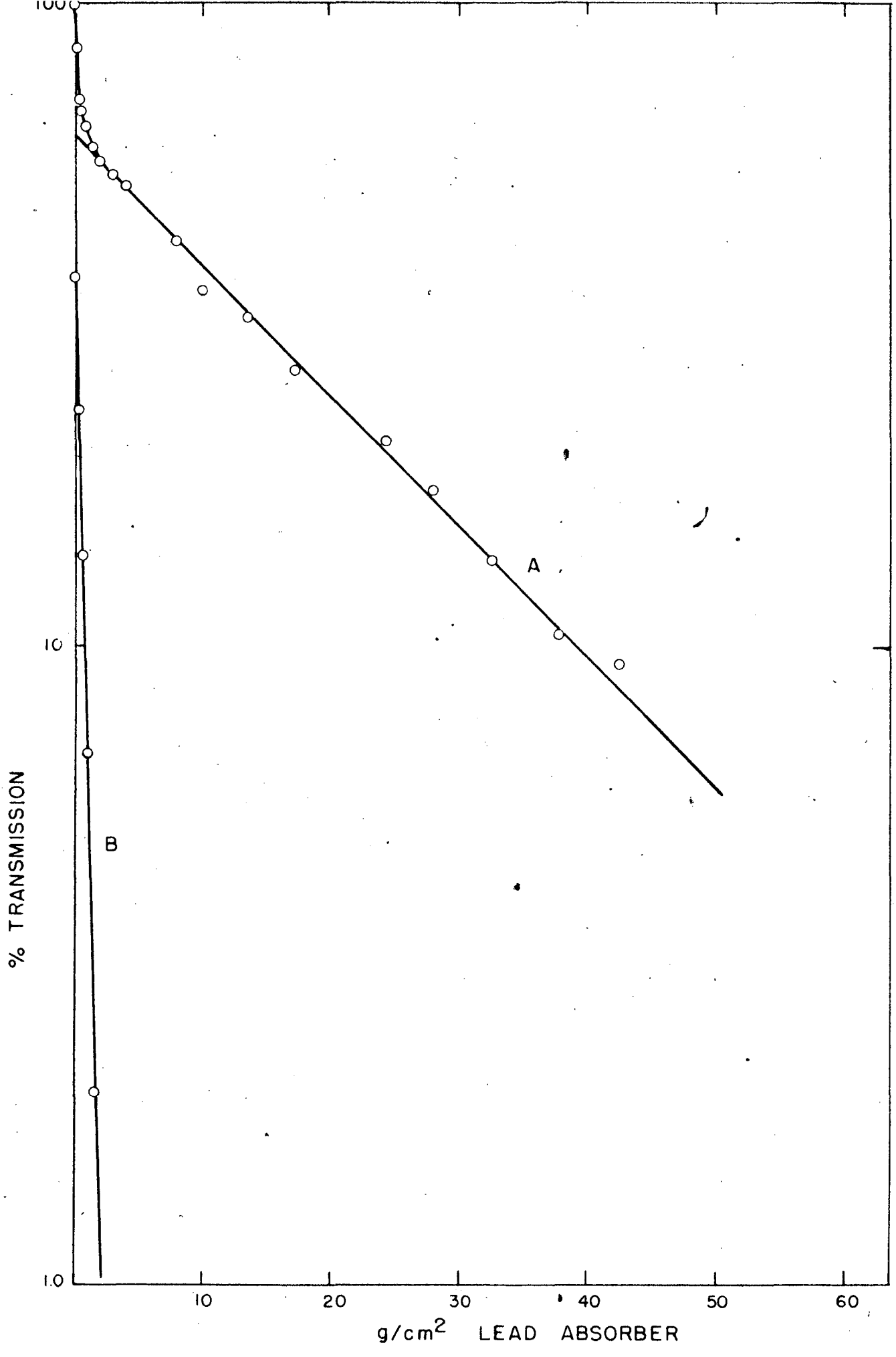

100
10
1.0
0.1
% TRANSMISSION
0
100
200
300
400
500
600
mg/cm2 ALUMINUM ABSORBER
A
B
C

Fig. 30 Lead absorption of 7.7-hour Tm^{166} from Ho + α bombardment. 1.7 Mev γ-ray (A) and K x-rays (B).



13189-1

It should be noted that the wide distribution of hard negative electrons observed (Fig. 28, Curve B) could be due to a continuous beta spectrum of maximum energy about 1 Mev arising from beta branching in Tm^{166}, since it is very possible that Yb^{166} is an unreported stable isotope of low abundance.[18]

If the hard negative electrons of Tm^{166} are a beta particle spectrum, the disintegration by beta emission is approximately 0.4% of that by orbital electron capture. The branching by beta and positron emission is thus almost the same.

2. 9.6 day Tm^{167}

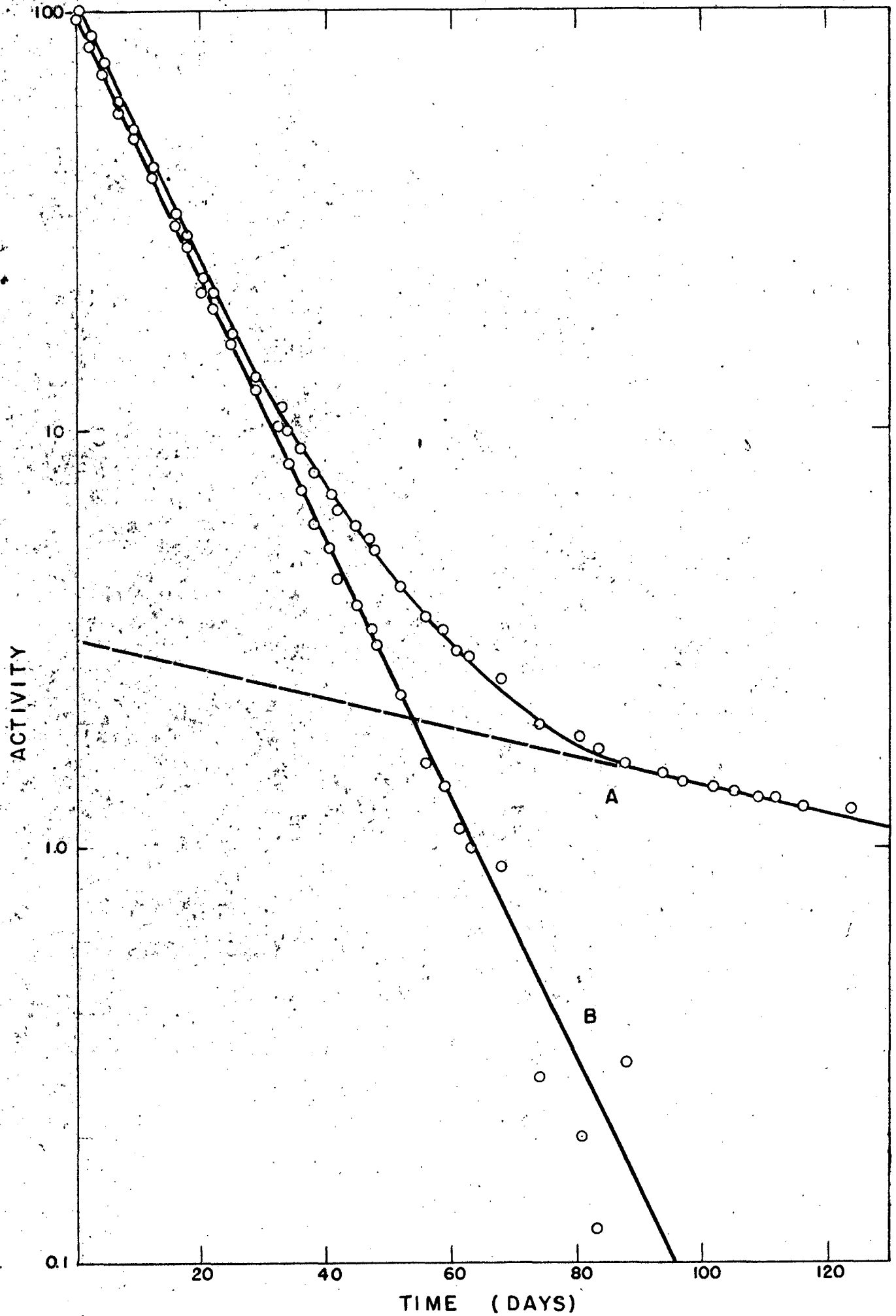
After decay of the 7.7 hour isotope from both Ho + α and Er + p bombardments, a residual activity consisting of 9.6 ± 0.1 day and 85 ± 2 day components was observed in the column separated thulium fraction. The gross decay of the 9.6 day activity was followed through ten, and γ-radiation, through nine half-lives (Fig. 31). The samples were studied on the crude beta-ray spectrometer, no positrons being observed. The aluminum absorption curve of an infinitely thin, column separated sample of the 9.6 day activity from Ho + α bombardment is shown in Fig. 32. The electrons have a range of 52 mg/cm^2 aluminum, corresponding to 0.21 Mev. The electromagnetic radiation consists of four components of half-thicknesses 7.5 mg/cm^2 aluminum, 80 mg/cm^2 lead, 770 mg/cm^2 lead, and 9.6 g/cm^2 lead (Fig. 33), corresponding respectively to 6 Kev, 50 Kev, 220 Kev, and 0.95 Mev. The two soft radiations agree well with the energies of erbium L and K x-radiation. The conversion line corresponds well with the expected energy of an electron arising from conversion of a 0.22 Mev γ-ray in the K shell. The conversion coefficient is about 2.

From the above measurements, the following corrected ratios were obtained:

	0.21 Mev e^-	:	L x-rays	:	K x-rays	:	0.22 Mev γ	:	0.95 Mev γ
=	0.3	:	0.2	:	1	:	0.1	:	0.01

Assuming that K x-radiation arises from the production of the conversion

Fig. 31 Gross decay of 9.6-day Tm^{167} (B) and 85 day (A) Tm^{168} from Ho + α bombardment.



[redacted]

13191-B

Fig. 32 Aluminum absorption of 9.6-day Tm^{167} from Ho + α bombardment. K x-ray and γ-ray background (A), 0.21 Mev electrons (B), and L x-rays (C).

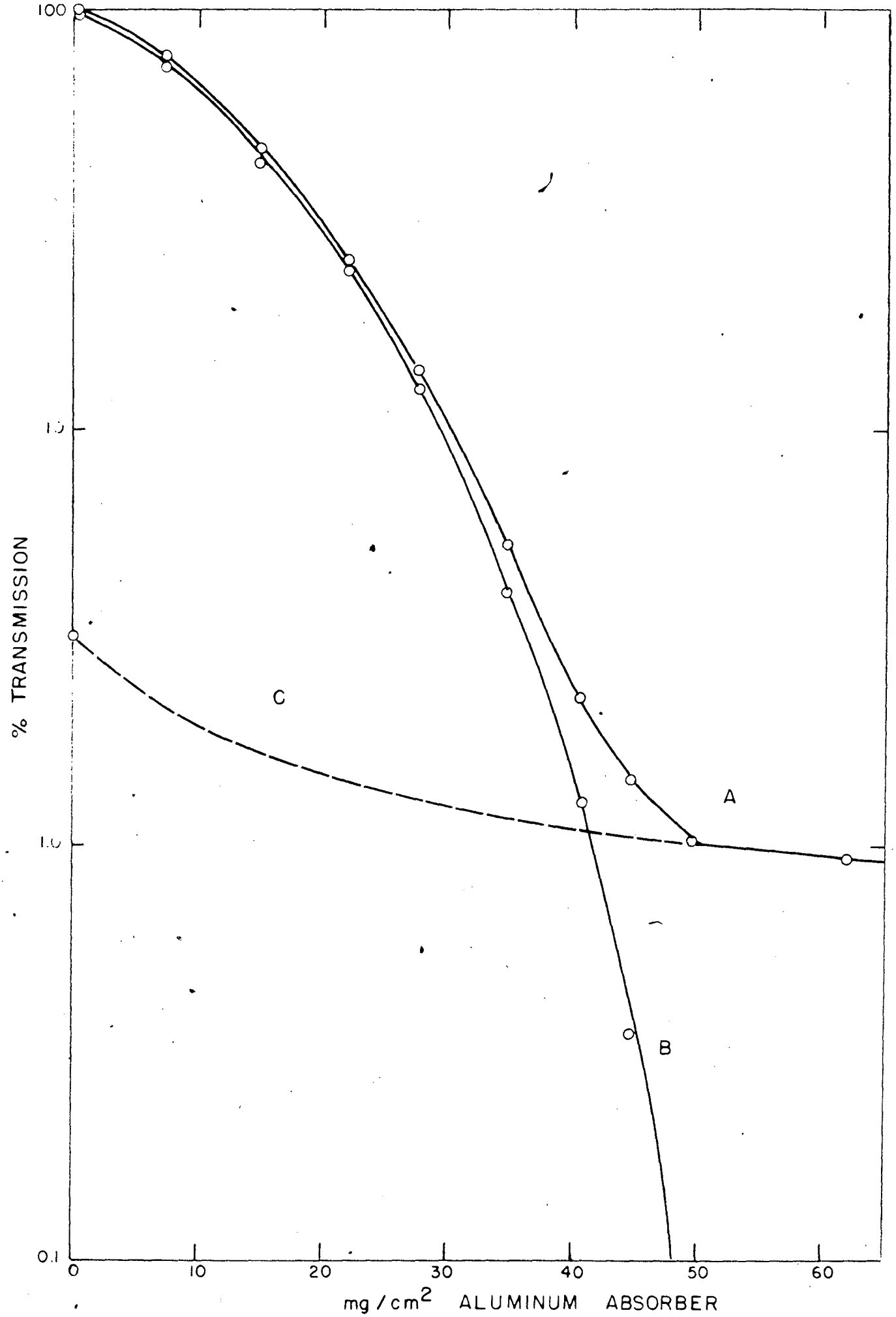
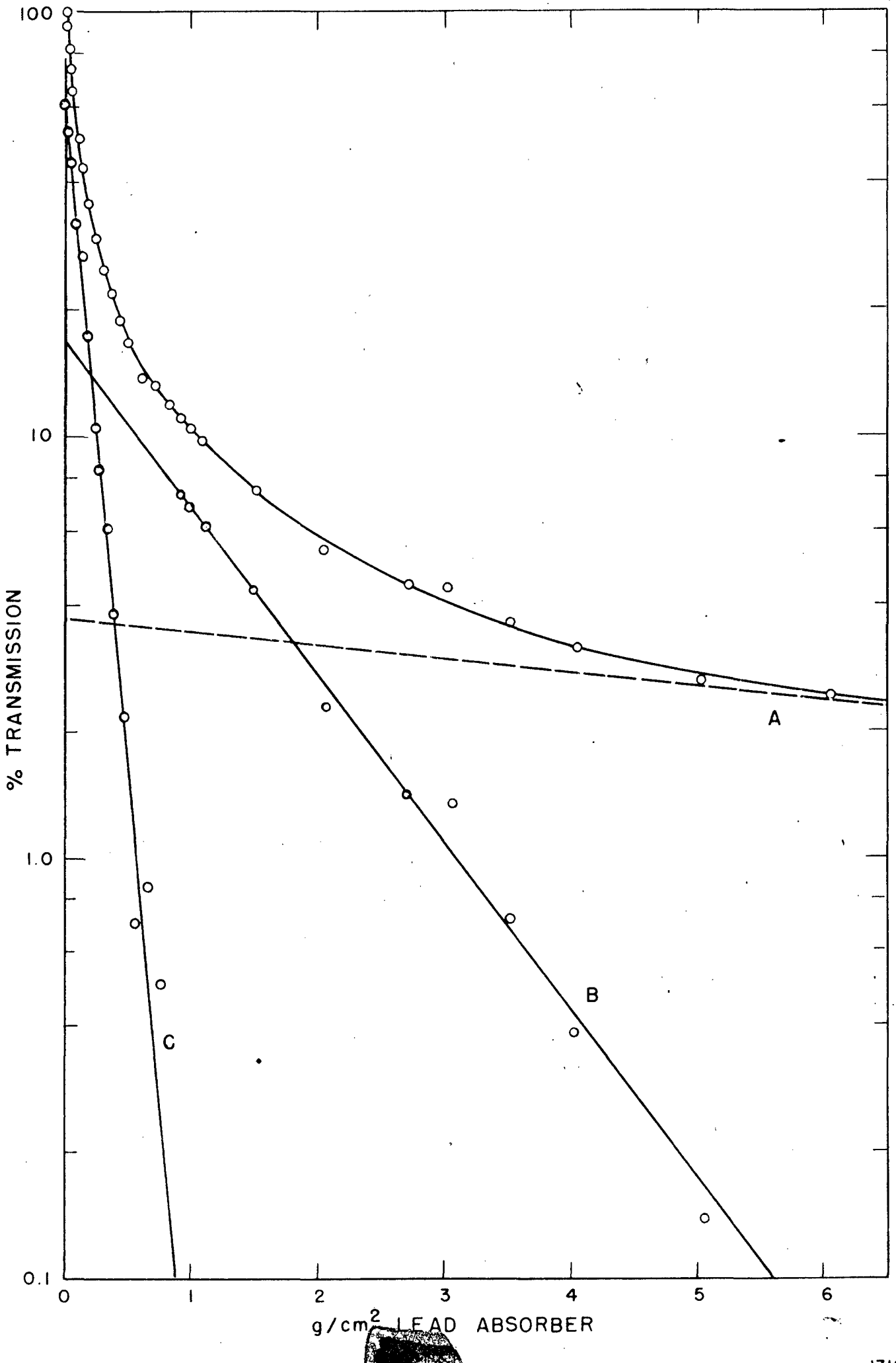

100
1.0
1.0
0.1
% TRANSMISSION
0
10
20
30
40
50
60
mg / cm2 ALUMINUM ABSORBER
A
B
C

Fig. 33 Lead absorption of 9.6-day Tm^{167} from Ho + α bombardment. 0.95 Mev γ-ray (A), 0.22 Mev γ-ray (B), K x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
0
1
2
3
4
5
6
g/cm² LEAD ABSORBER
A
B
C

electrons, then 0.7 K x-ray quanta probably represent one disintegration by orbital electron capture.

This isotope has also been produced by the bombardment of tantalum with 190 Mev deuterons from the 184-inch cyclotron. The reaction producing the isotope is designated Ta-d-5z,16a.

3. 85 day Tm^{168}

After the decay of the 9.6-day isotope, a small residual activity of long-life (Fig. 31) was found in the column separated thulium fractions from the Ho + α bombardments at 38 Mev. In order to study this long-lived activity, 80 mg of pure holmium oxide was bombarded with 19 Mev helium ions for 54 microampere-hours. The decay of electron and electromagnetic radiations of the isotope have been followed through three half-lives to give a value of 85 ± 2 days. The aluminum absorption curve (Fig. 34) shows electrons of ranges 29 mg/cm^2 (0.16 Mev) and 150 mg/cm^2 (0.5 Mev). The soft electromagnetic radiation corresponding to L x-ray energies was determined after removal of electrons by beryllium. The lead absorption (Fig. 35) of hard radiation shows components of half-thicknesses 77 mg/cm^2 (49 Kev), ~750 mg/cm^2 (0.21 Mev), and 8.8 g/cm^2 (0.85 Mev). The first of these corresponds well with the value for erbium K x-radiation. From these measurements, the following approximate ratios were obtained:

	0.16 Mev e^-	:	0.5 Mev e^-	:	L x-rays	:	K x-rays	:	0.21 Mev γ	:	0.85 Mev γ
=	0.15	:	0.02	:	0.2	:	1	:	0.2	:	0.8

The energy of the soft electrons agrees well with that to be expected for K shell conversion of the 0.21 Mev γ-ray. The conversion coefficient is about 0.8. Assuming that K x-rays arise from conversion, then about 0.75 of the observed K x-radiation may be considered to represent one disintegration by orbital electron capture.

Since the 85 day activity is allocated to Tm^{168}, the possibility of beta

Fig. 34 Aluminum absorption of 85-day Tm^{168} from Ho + α bombardment. K x-ray and γ-ray background (A), complex hard electron (B), 0.16 Mev electron (C), L x-rays (D).

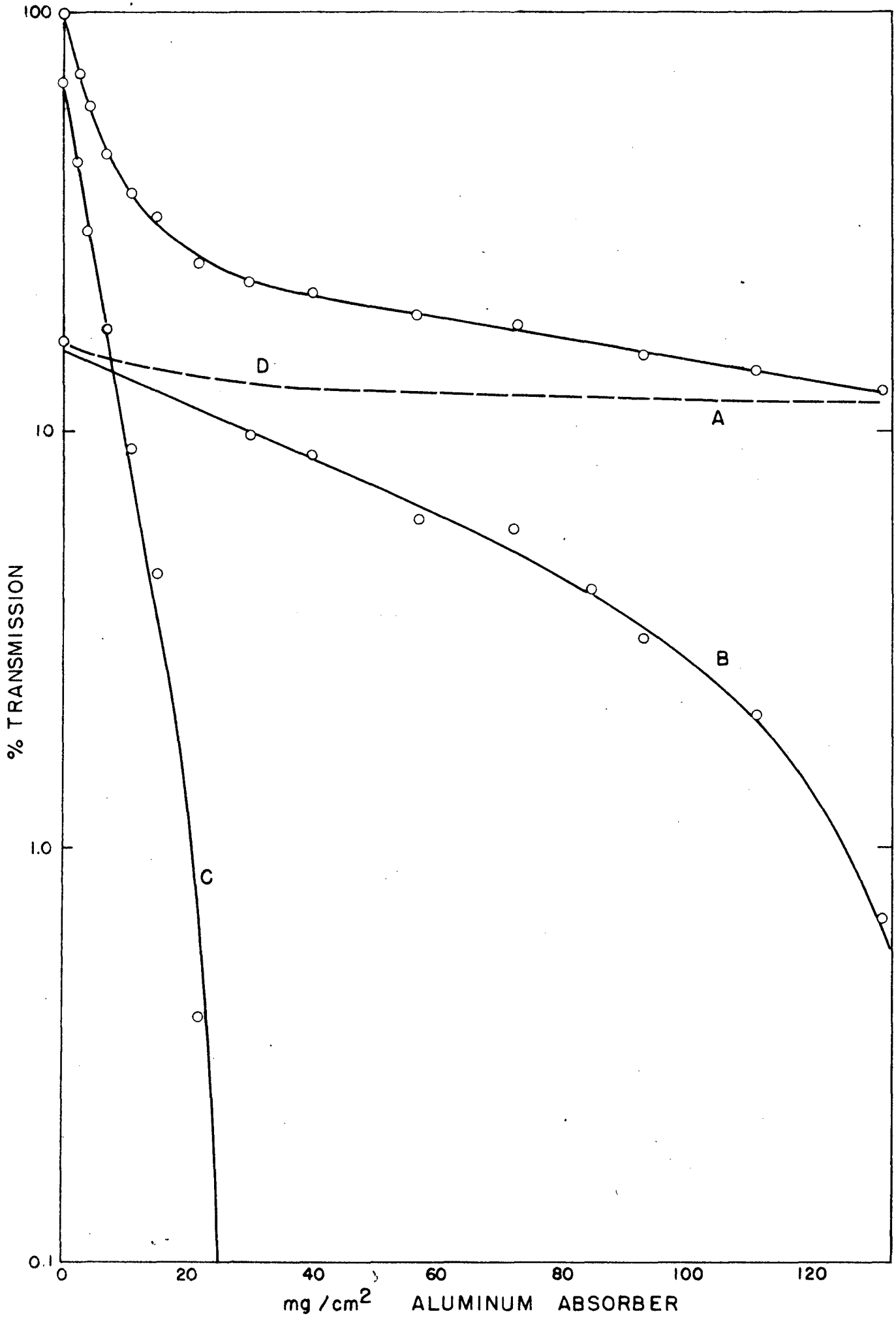
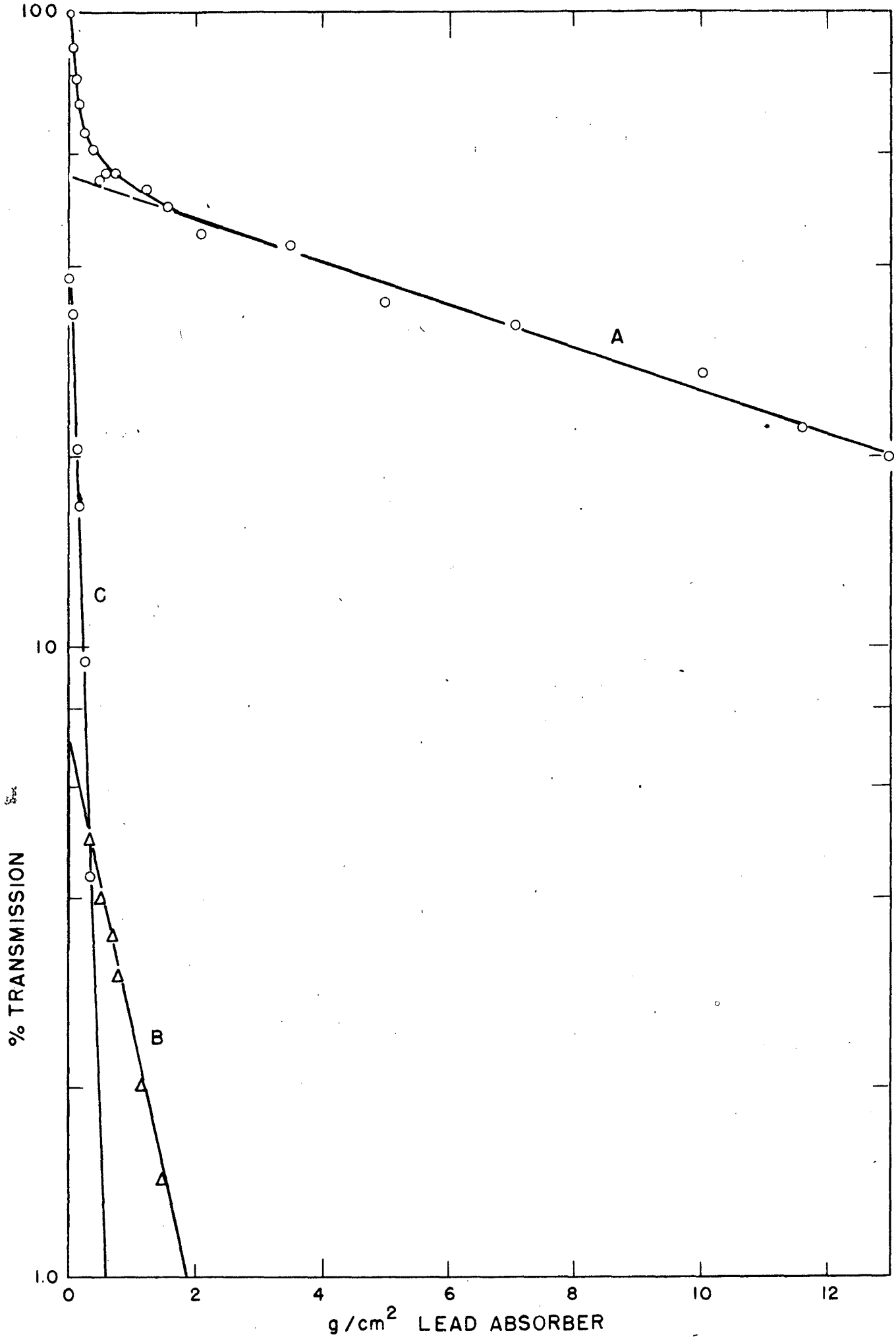

100
10
1.0
0.1
% TRANSMISSION
0
20
40
60
80
100
120
mg /cm2 ALUMINUM ABSORBER
A
B
C
D

Fig. 35 Lead absorption of 85 day Tm^{168} from Ho + α bombardment 0.85 Mev γ-ray (A), 0.21 Mev γ-ray (B), K x-rays (C).


100
10
1.0
% TRANSMISSION
A
B
C
0
2
4
6
8
10
12
g/cm^2 LEAD ABSORBER

branching exists, and the 0.5 Mev electrons observed are probably beta-radiation.[18] With the previous assumptions regarding K x-rays, then about two percent of the disintegrations would proceed by beta-emission.

The 85-day isotope has also been observed in bombardments of erbium with 10 Mev protons and thulium with fast neutrons. In both cases, the 127 day Tm^{170} is also formed. The latter is reported to have no γ-radiation[22], and the x and γ-radiation observed in the thulium fraction decays with a half-life of 85 days. The allocation to mass 168 on the basis of reaction yields is thus confirmed.

4. Discussion

The cross sections for the formation of the three thulium isotopes are given in Table X. The relative yields of the isotopes at various energies are significant, and allow allocation of the 7.7 hour, 9.6 day, and 85 day activities to masses 166, 167 and 168 respectively on the basis of α,3n, α,2n and α,n reactions.

Table X

Activity	Energy of α-particles in Mev 38	31	19	Probable Reaction	Probable Isotope
7.7 hours	1.1	5×10^{-4}	--	Ho-α-3n	166
9.6 days	7×10^{-3}	0.1	10^{-3}	Ho-α-2n	167
85 days	10^{-4}	3×10^{-3}	0.2	Ho-α-n	168

The yields of the three isotopes in Er + p bombardments are in agreement with the above allocations. Correcting the observed yields for isotopic abundances of the erbium isotopes, the isotopic cross sections for all three cases, assuming the p,n reaction, are close to 2×10^{-2} barns. This cross section value was obtained for the reaction $Lu^{175}(p,n)Hf^{175}$.[3]

No evidence of thulium activities of half-life shorter than those reported has been observed in Ho + α and Er + p bombardments.

F. 70-day Hf^{175} (3)

In the hafnium fraction from the bombardments of column separated lutecium with 19 Mev deuterons and 10 Mev protons, a single radioactivity with half-life of 70 ± 2 days has been observed. No evidence of zirconium contamination or shorter-lived activities has been seen.

An aluminum absorption curve of an "infinitely thin" sample mounted on mica is shown in Fig. 36. The lead absorption curve (Fig. 37) was measured in an unshielded counter. The radiations consist of electrons, range 82 mg/cm^2 (0.3 Mev); electromagnetic radiations of half-thickness 14.5 mg/cm^2 aluminum (8.2 Kev), 100 mg/cm^2 lead (55 Kev), 2.2 g/cm^2 lead (0.35 Mev), and 13.3 g/cm^2 lead (1.5 Mev). The energies of the two soft electromagnetic components correspond well with those to be expected for the L and K x-radiation of lutecium. The 0.3 Mev electron corresponds to the energy expected for conversion of the 0.35 Mev γ-ray in the K-shell.

The following ratios were obtained from the above measurements:

	0.3 Mev e^-	:	L x-rays	:	K x-rays	:	0.35 Mev γ	:	1.5 Mev γ
=	0.1	:	0.1	:	1	:	0.2	:	0.05

The isotope thus probably decays by orbital electron capture, mainly to the ground state, and also to two or more excited or metastable levels of the daughter nucleus. 0.9 K quanta are assumed to constitute one disintegration by orbital electron capture.

For 10 Mev protons and 19 Mev deuterons on natural lutecium, the cross sections are respectively 3×10^{-2} and 5×10^{-2} barns. These values are reasonable for the p,n and d,2n reactions. Thus the isotope has been allocated to mass 175.

Fig. 36 Aluminum absorption of 70-day Hf^{175} from Lu + p bombardment. K x-ray and γ-ray background (A), 0.3 Mev electron (B), L x-rays (C).

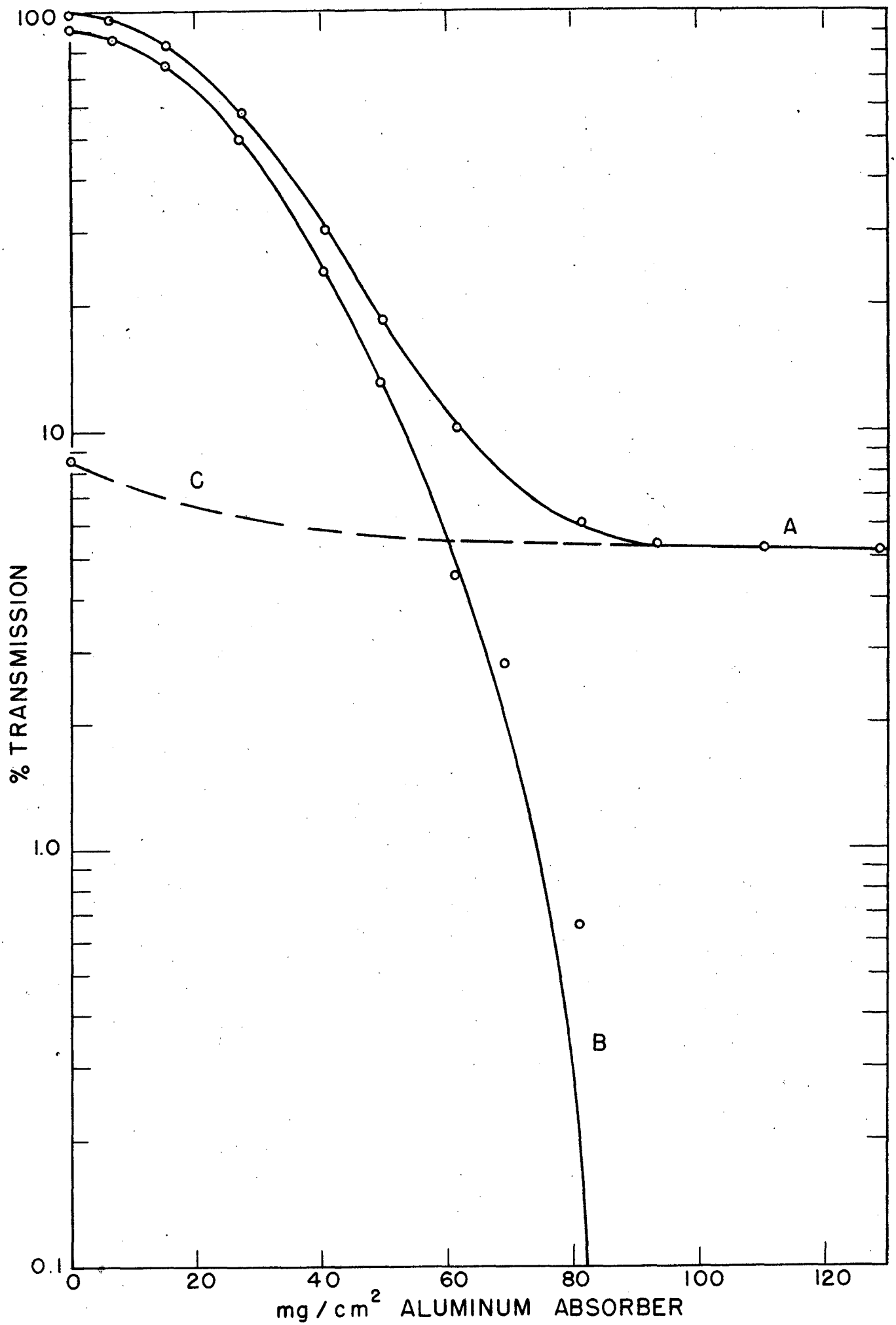
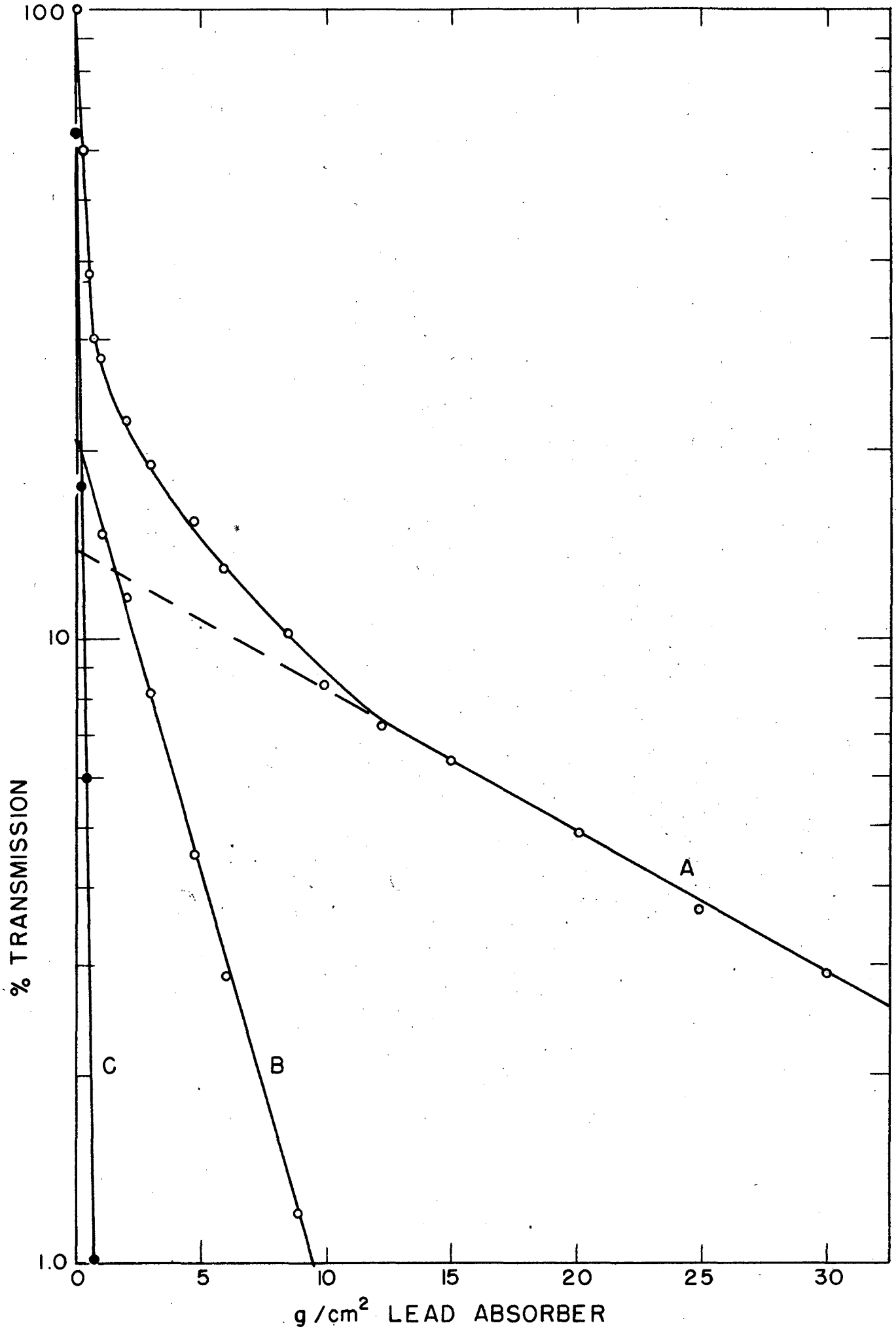

100
10
1.0
0.1
% TRANSMISSION
0
20
40
60
80
100
120
mg / cm² ALUMINUM ABSORBER
A
B
C

Fig. 37 Lead absorption of 70-day Hf^{175} from Lu + p bombardment. 1.5 Mev γ-ray (A), 0.35 Mev γ-ray (B), K x-rays (C).


100
10
1.0
% TRANSMISSION
0
5
10
15
20
25
30
g /cm² LEAD ABSORBER
A
B
C

G. Tantalum Isotopes[20, 21]

Four new tantalum activities summarized in Table XI have been produced by bombardment of lutecium with helium ions and hafnium with protons. The mass allocations have been made by yield determinations in Lu + α bombardments. The 8.0-hour and 2.5-day activities have also been identified in the tantalum fraction from bombardment of 200 Mev deuterons using the 184-inch cyclotron.

Table XI

Isotope	Type of Radiation	Half-Life	Energy of Radiation in Mev Particles	γ-rays	Produced by
Ta^{176}	K, e^-, γ, (β^-?)	8.0±0.1 hours	0.12, 0.18 (e^-) >1 (β^-?)	L, K x-rays ~2	Lu-α-3n Ta-d-p6n
Ta^{177}	K, e^-, γ	2.50±0.05 days	0.11 (e^-)	L, K x-rays ~1.4 (weak)	Lu-α-2n, 3n Hf-p-n Ta-d-p, 5n
Ta^{178}	β^-, γ	15.4±0.2 days	1.4	Very weak	Lu-α-n, 2n Hf-p-n
Ta^{179}	K, e^-, γ	>100 days			Lu-α-n Hf-p-n
Ta^{180}	K, γ, β^- (11%)	8.00±0.05 hours	0.7 (β^-)	K x-rays 1.3	Ta-n-2n
Ta^{180}	K or IT e^-, β^-, γ	16±2 min	~0.2 e^- ~0.6 β^-		Ta-n-2n

1. 8.0-hour Ta^{176}

The chemically separated tantalum from 38 Mev helium ion bombardment of lutecium shows the 8.0-hour activity in high yield.

The decay of the gross and electromagnetic radiations were followed separately

through about eight half-lives (Fig. 38); after correction for longer-lived backgrounds, a half-life of 8.0 ± 0.1 hours was obtained from several measurements.

In Figure 39 is shown the aluminum absorption of the radiations of the 8.0-hour activity. Electrons of ranges ~19 mg/cm^2 (0.12 Mev), ~40 mg/cm^2 (0.18 Mev), and >400 mg/cm^2 (> 1 Mev) and electromagnetic radiation of half-thickness 16 mg/cm^2 (8.5 Kev) were observed. The lead absorption (Fig. 40) shows electromagnetic radiations of half-thicknesses ~115 mg/cm^2 (58 Kev) and 15.5 g/cm^2 (~2 Mev). The following ratios were obtained from the above measurements:

	0.12 Mev e^-	:	0.18 Mev e^-	:	1 Mev e^-	:	L x-rays	:	K x-rays	:	2 Mev γ
=	~1	:	~0.04	:	~0.02	:	~0.5	:	1	:	0.6

The isotope thus probably decays by orbital electron capture followed by the converted γ-ray transitions from excited or metastable levels of the daughter nucleus. The 1 Mev electron may be a beta particle.[18] 0.5 K quanta were assumed to represent one disintegration by orbital electron capture.

2. 2.5 day Ta^{177}

After decay of the 8.0-hour activity, an activity of 2.5 days half-life has been observed; the resolved gross decay of the 2.5-day activity was followed through eight half-lives, and the decay of the electromagnetic radiation through nine half-lives to give a half-life of 2.50 ± 0.03 days (Fig. 38, 41).

In Fig. 42 is shown the aluminum absorption after contribution (10%) of the 15-day tantalum activity at the time of measurement was subtracted. Resolution shows an electron of total range 16 mg/cm^2 (0.11 Mev) together with soft electromagnetic radiation of half-thickness 16 mg/cm^2 (8.5 Kev) and hard electromagnetic background. The lead absorption of electromagnetic radiation (Fig. 43) shows components of half-thicknesses 120 mg/cm^2 (58 Kev) and ~13 g/cm^2 (1.4 Mev). The following ratios were obtained from the above measurements:

Fig. 38 Gross decay of 8.0-hour Ta^{176} (B) and 2.50 day Ta^{177} (A) with 15.4 day background (C) from Lu + α bombardment.

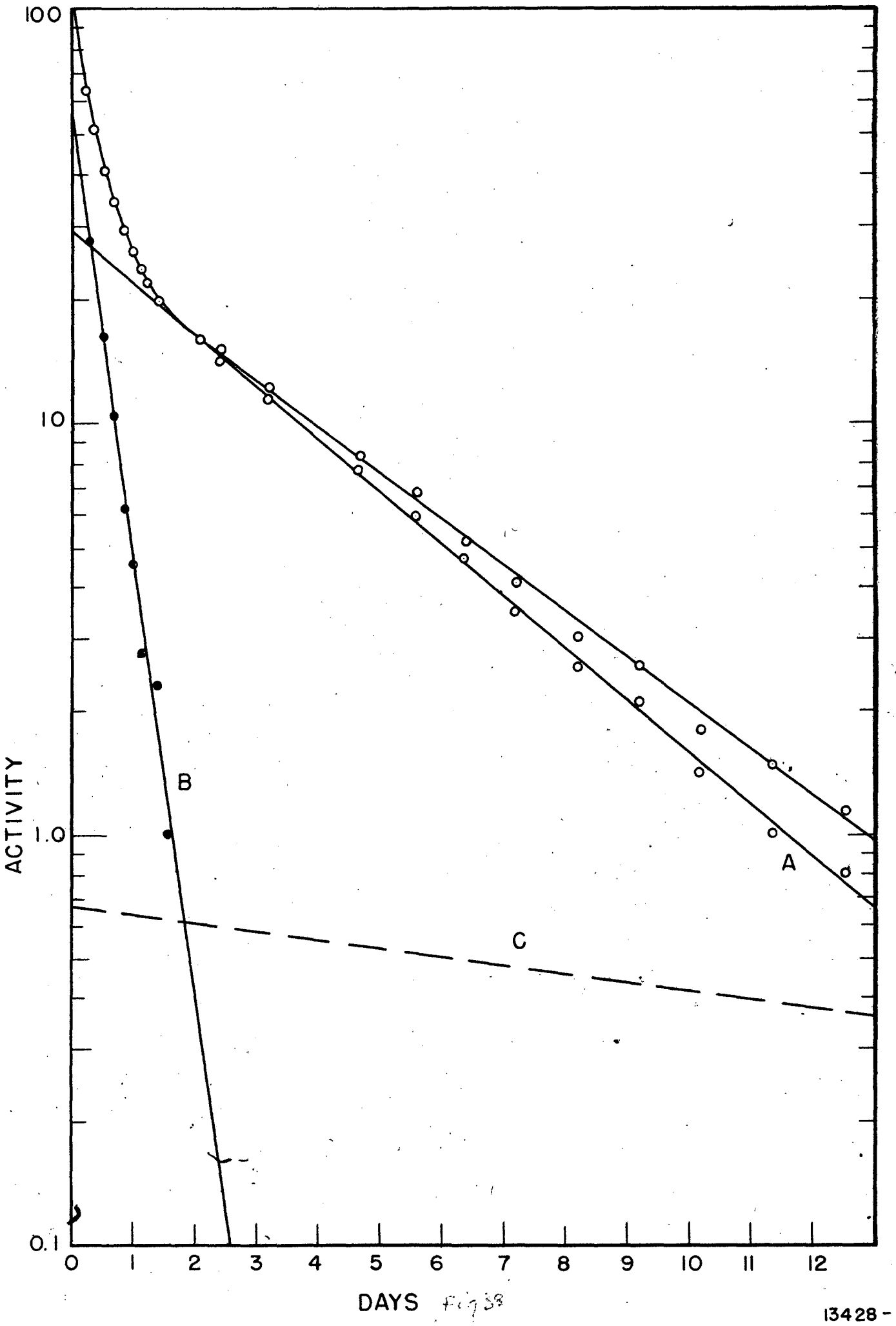


Fig 38

13428-1

Fig. 39 Aluminum absorption of 8.0-hour Ta^{176} from Lu + α bombardment. K x-ray and γ-ray background (A), >1 Mev electron (B), 0.18 Mev electron (C), 0.12 Mev electron (D), L x-rays (E).

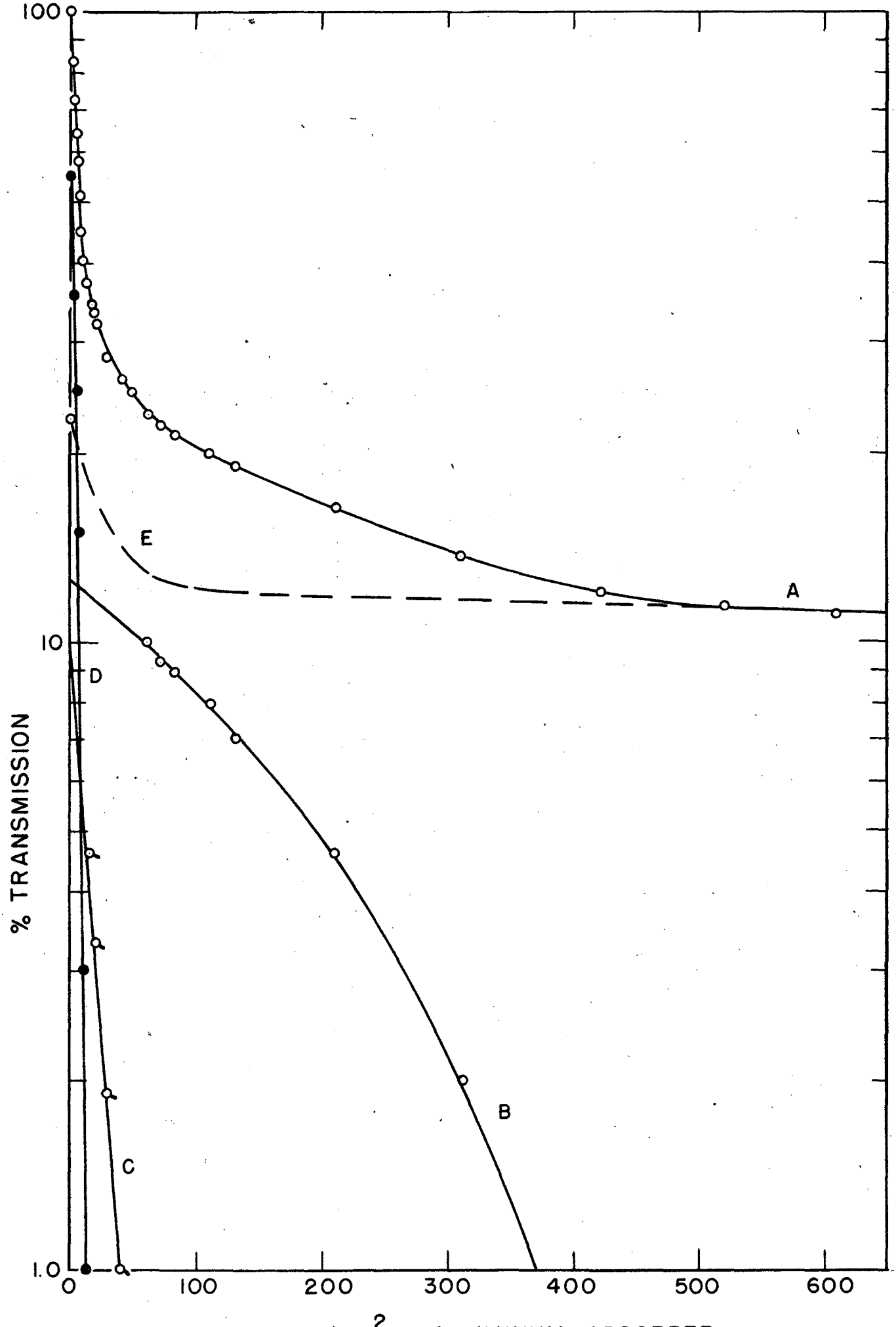

100
10
1.0
% TRANSMISSION
0
100
200
300
400
500
600
A
B
C
D
E

Fig. 40 Lead absorption of 8.0-hour Ta^{176} from Lu + α bombardment.

$\sim$2 Mev γ-ray (A), K x-rays (B).

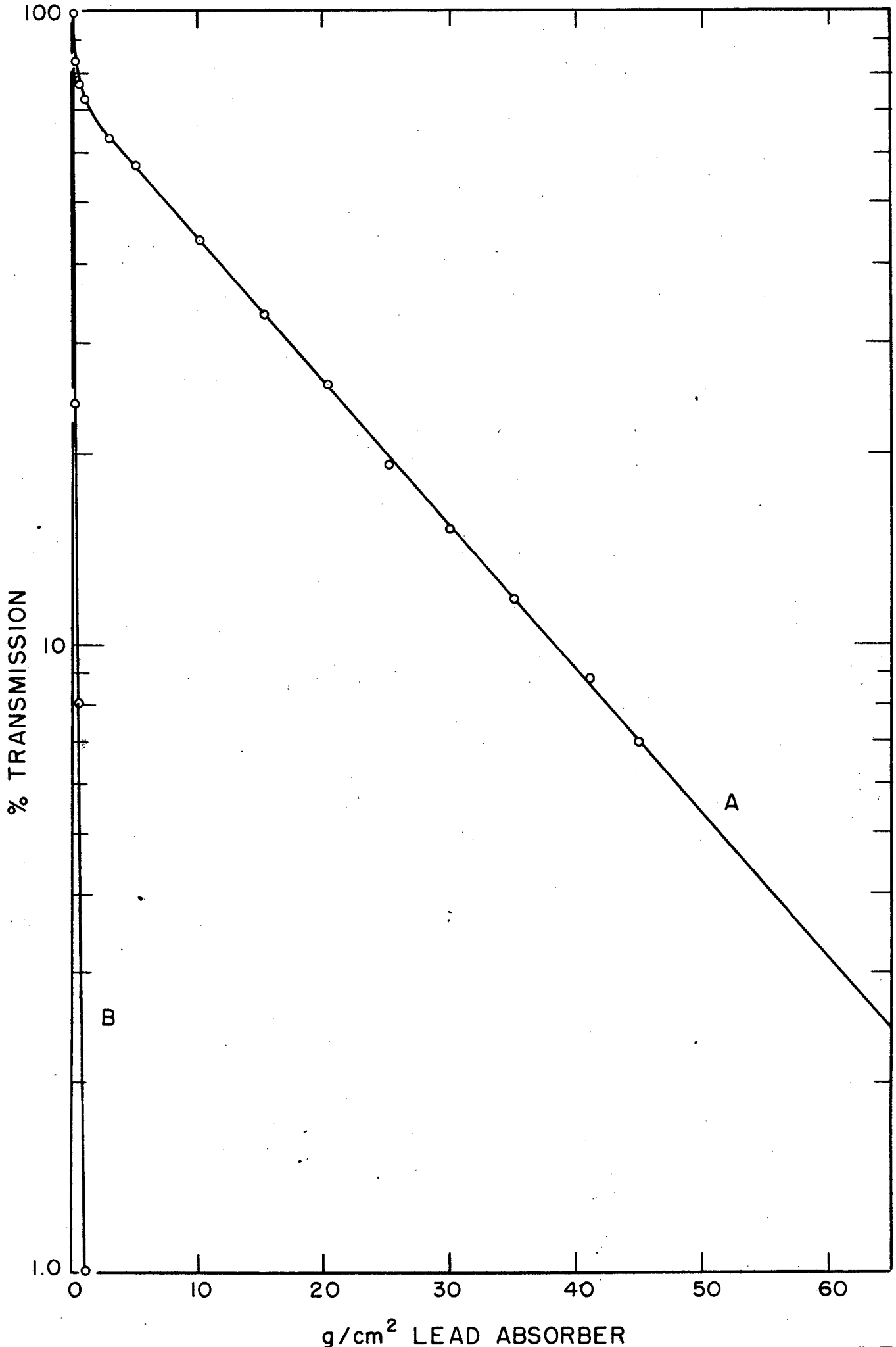

100
10
1.0
% TRANSMISSION
0
10
20
30
40
50
60
g/cm² LEAD ABSORBER
A
B

Fig. 41 Gross decay of 2.50-day Ta^{177} (C), 15.4 day Ta^{178} (B) and ~120 day Ta^{179} from Lu + α bombardment.

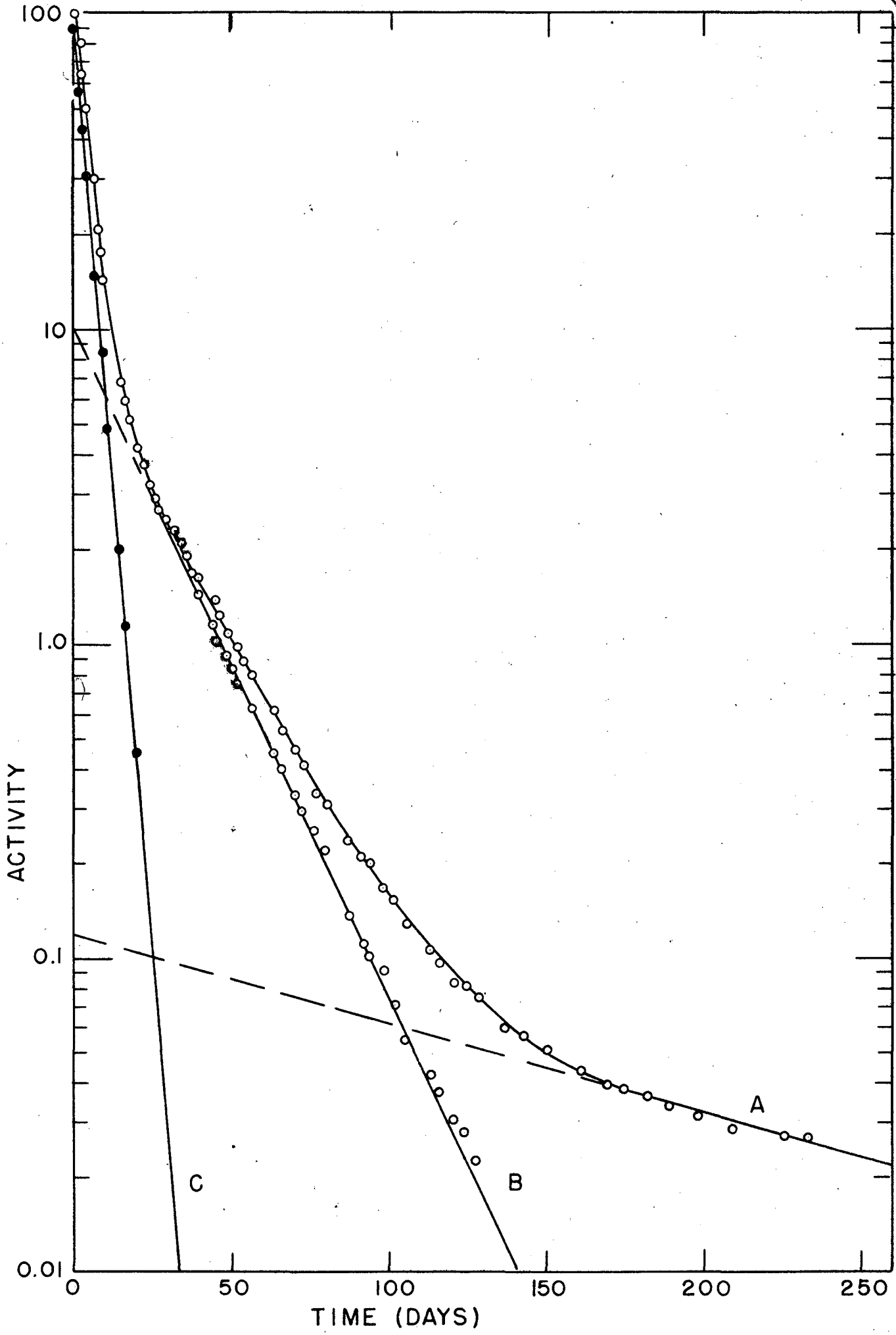

100
10
1.0
0.1
0.01
ACTIVITY
0
50
100
150
200
250
TIME (DAYS)
A
B
C

Fig. 42 Aluminum absorption of 2.50-day Ta^{177} from Lu + α bombardment. K x-ray and γ-ray background (A), 0.11 Mev electron (B) and L x-rays (C).

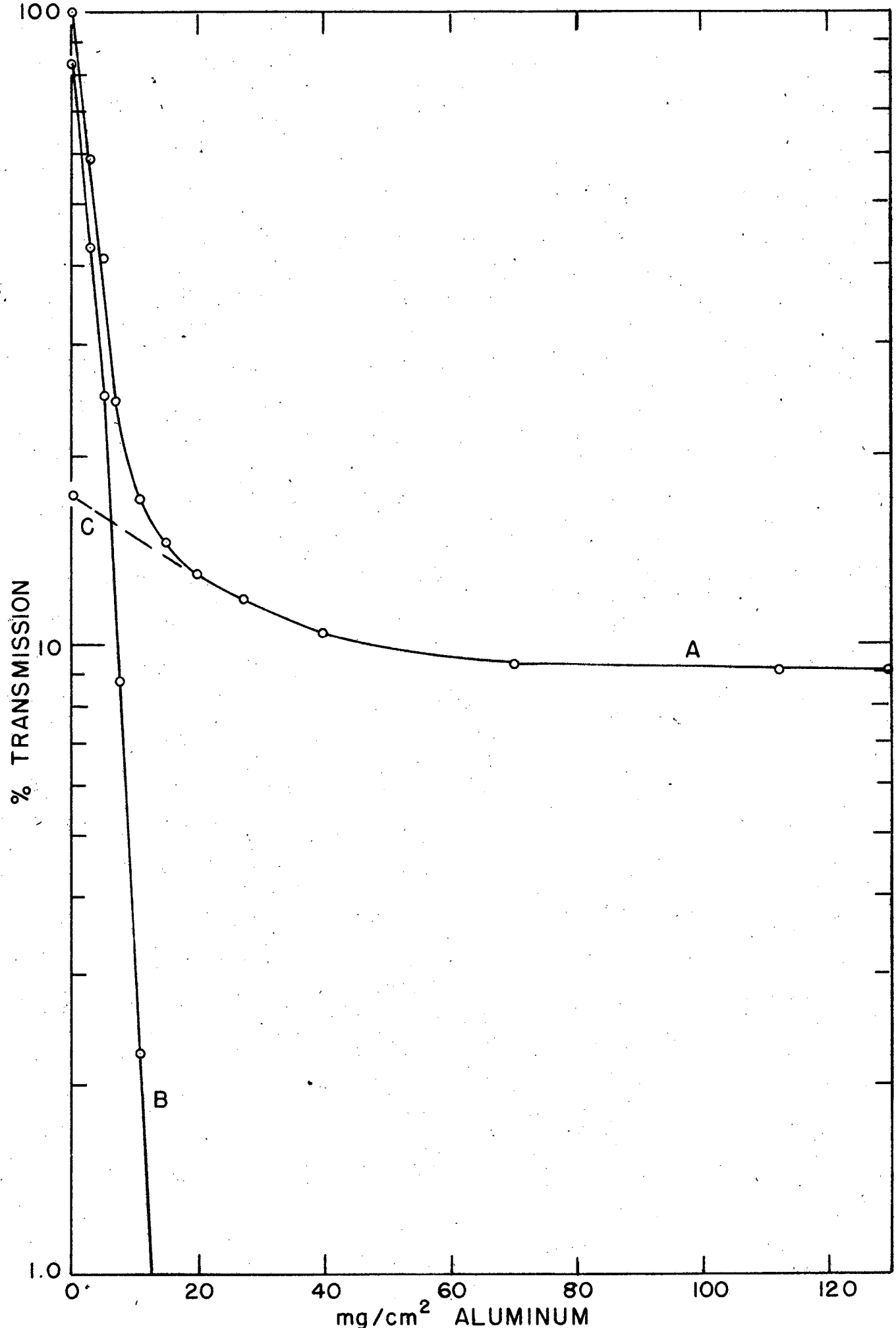
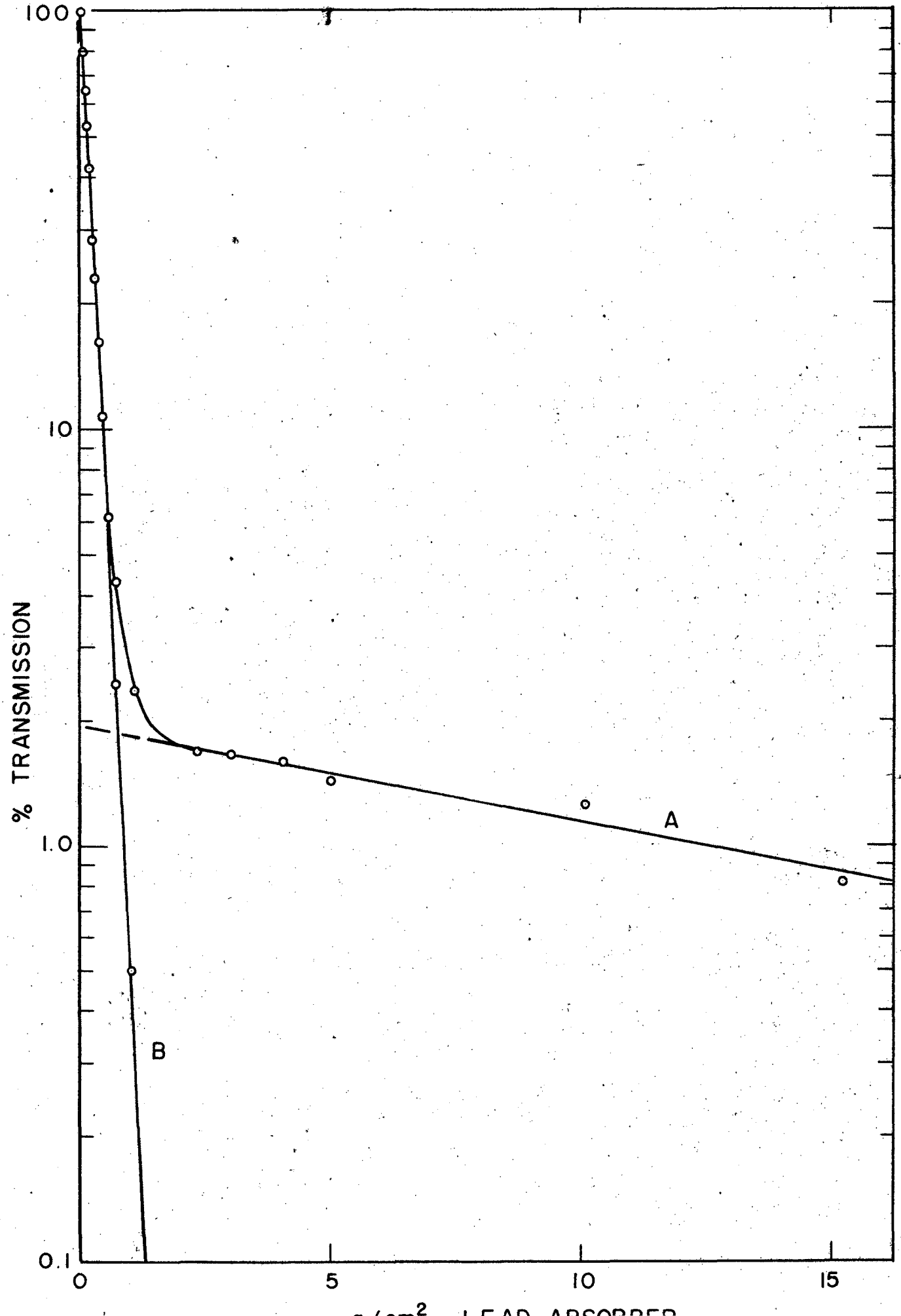

100
10
1.0
% TRANSMISSION
0
20
40
60
80
100
120
mg/cm² ALUMINUM
A
B
C

Fig. 43 Lead absorption of 2.50-day Ta^{177} from Lu + α bombardment. 1.4 Mev γ-ray (A), K x-rays (B).


100
10
1.0
0.1
% TRANSMISSION
0
5
10
15
g/cm² LEAD ABSORBER
A
B

	0.11 Mev e^-	: L x-rays	: K x-rays	: 1.4 Mev γ
=	$\sim$0.3	: 0.35	: 1	: 0.005

It appears that this isotope decays by orbital electron capture followed by converted γ-ray transitions between metastable levels of the daughter nucleus. 0.7 K quanta were taken to be one disintegration by orbital electron capture.

3. 15.4-day Ta^{178}

The half-life of this isotope resolved from the Lu + α bombardment (Fig. 41) is 15.4 $\pm$ 0.2 days through six half-lives. The aluminum absorption curve (Fig. 44) shows the radiation to consist almost entirely of hard electrons, range 610 mg/cm^2 (1.4 Mev). The shape of the absorption curve and the low percentage of electromagnetic radiation suggests beta-radiation and allocation to mass 178.[18] The beta-radiation has been confirmed by study of the isotope on the crude beta-ray spectrometer. The Feather range 770 mg/cm^2 corresponds to 1.65 Mev, maximum energy.

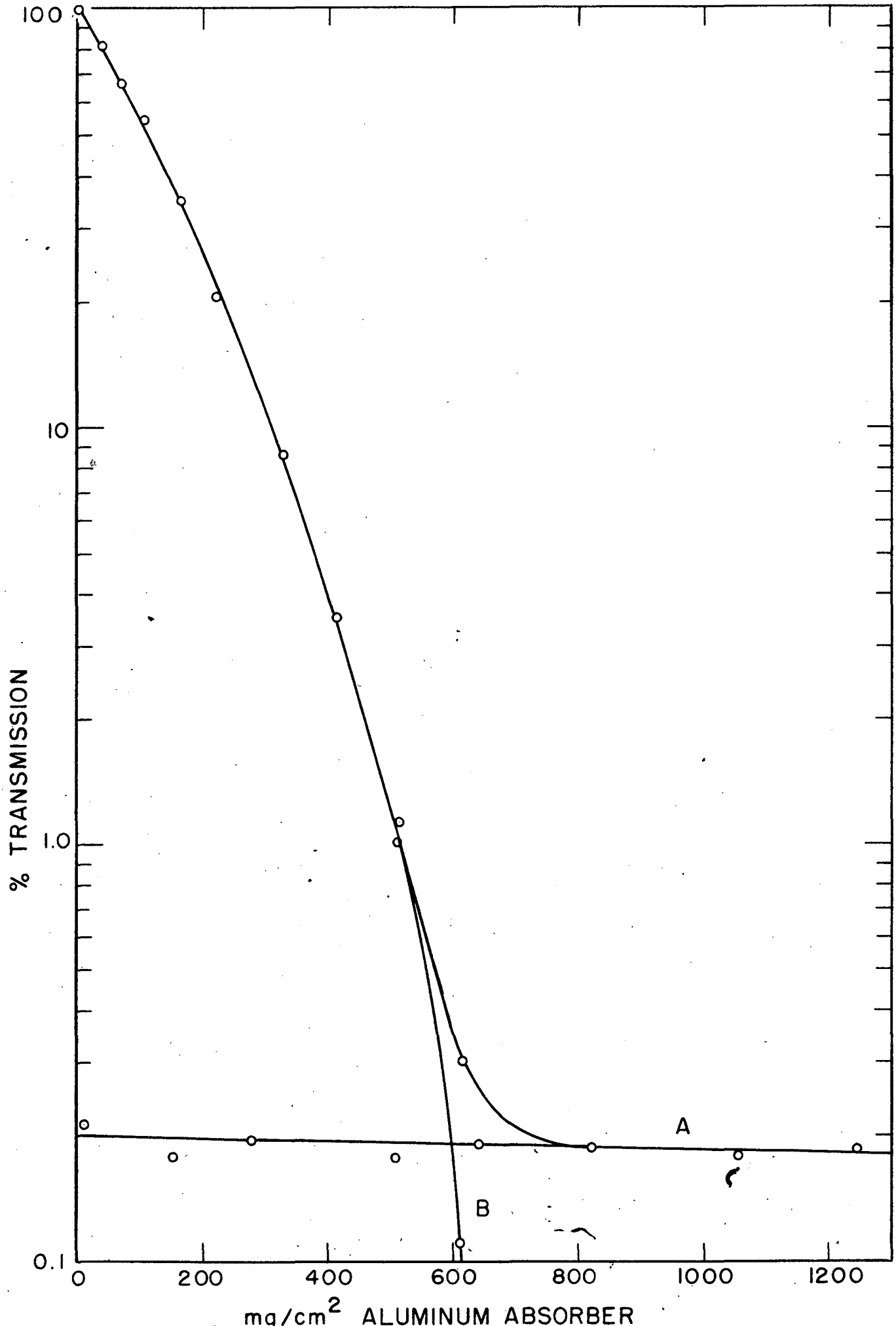
Sufficient intensity was not available to allow a lead absorption of γ-radiation, but from the decay of tantalum electromagnetic radiation and aluminum absorption measurements, an upper limit of 0.002 γ or K x-ray quanta per electron is obtained. Assuming 1% counting efficiency for such radiation, the γ/β ratio is less than 0.2, which further confirms the designation of the 15.4 day activity as a beta-emitter. The isotope has been observed also in the tantalum fraction from Hf + p bombardments; the resolved decay and radiation characteristics agree with those from Lu + α bombardments.

4. >100 day Ta^{179}

After decay of shorter-lived activities, tantalum fractions show, in both Lu + α and Hf + p bombardments, a long-lived activity. The half-life at present appears to be $\sim$120 days.

The aluminum absorption shows electrons, range $\sim$20 mg/cm^2 (0.12 Mev), soft

Fig. 44 Aluminum absorption of 15.4-day Ta^{178} from Lu + α bombardment. K x-ray and γ-ray background (A), 1.4 Mev beta (B).


100
10
1.0
0.1
% TRANSMISSION
0
200
400
600
800
1000
1200
mg/cm2 ALUMINUM ABSORBER
A
B

electromagnetic radiation, half-thickness $\sim$16 mg/cm^2 and K x-rays or γ-radiation. For estimation of yields, one K x-ray quantum was assumed to indicate one disintegration.

A more intense sample of this activity has been recently prepared by proton bombardment of hafnium, and a more detailed report will be published subsequently.

5. 8.0-hour and 16-minute Ta^{180} (21)

Two activities of half-lives 8.2-hours and 14-minutes have been produced by n,2n and γ,n reactions in tantalum.[23] These activities have now been re-examined. Very pure tantalum was bombarded with fast neutrons from Be + d reaction; the short-lived activity was studied without chemical separation, but separation was made for the 8-hour activity.

The decay of the gross and electromagnetic radiations of the longer-lived activity were followed through ten half-lives (Fig. 45) and a value of 8.00 $\pm$ 0.05 hours obtained. A small long-lived background was identified as the 117-day Ta^{182} produced by neutron capture. The aluminum absorption curve of the eight-hour activity (Fig. 46) shows beta-particles of range 210 mg/cm^2 (0.6 Mev), Feather range 238 mg/cm^2 (0.7 Mev) and hard electromagnetic radiation. The lead absorption (Fig. 47) shows components of half-thickness 115 mg/cm^2 (58 Kev) and 12.2 g/cm^2 (1.3 Mev); the energy of the former corresponds well with the energy of hafnium K x-radiation. From the measurements, the following ratios were obtained:

0.7 Mev β^-	:	K x-rays	:	1.3 Mev γ =
0.13	:	1	:	0.02

The isotope thus appears to decay predominantly by orbital electron capture. Taking one K x-ray quantum as representing one disintegration by orbital electron capture, the branching by beta-particle emission[18] is around 11%.

Fig. 45 Gross decay of 8.00-hour Ta^{180} (B) and 117 day Ta^{182} (A) from Ta + n bombardment.

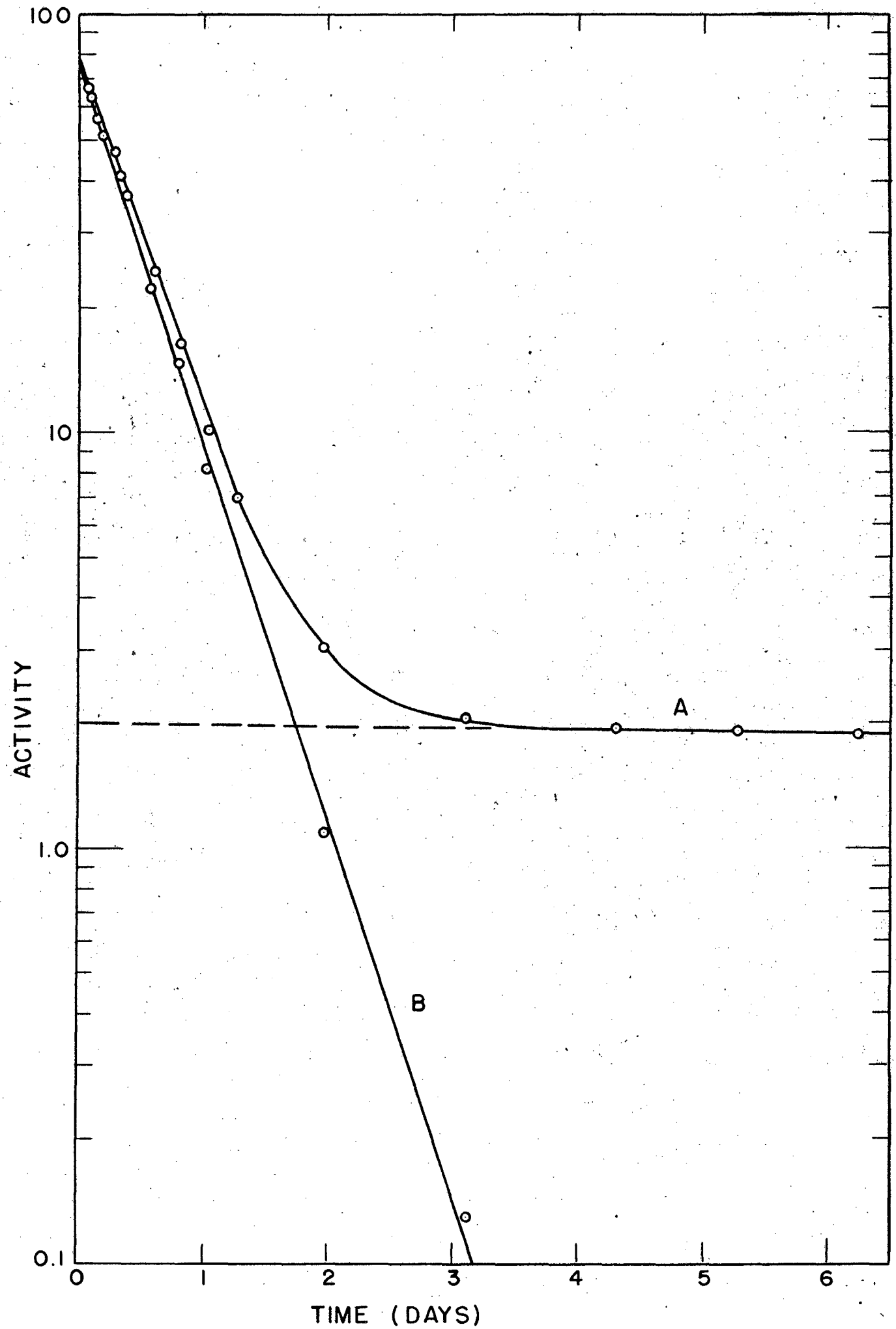

100
10
1.0
0.1
ACTIVITY
0
1
2
3
4
5
6
TIME (DAYS)
A
B

Fig. 46 Aluminum absorption of 8.00-hour Ta^{180} from Ta + n bombardment. K x-ray and γ-ray background (A), 0.7 Mev beta (B).

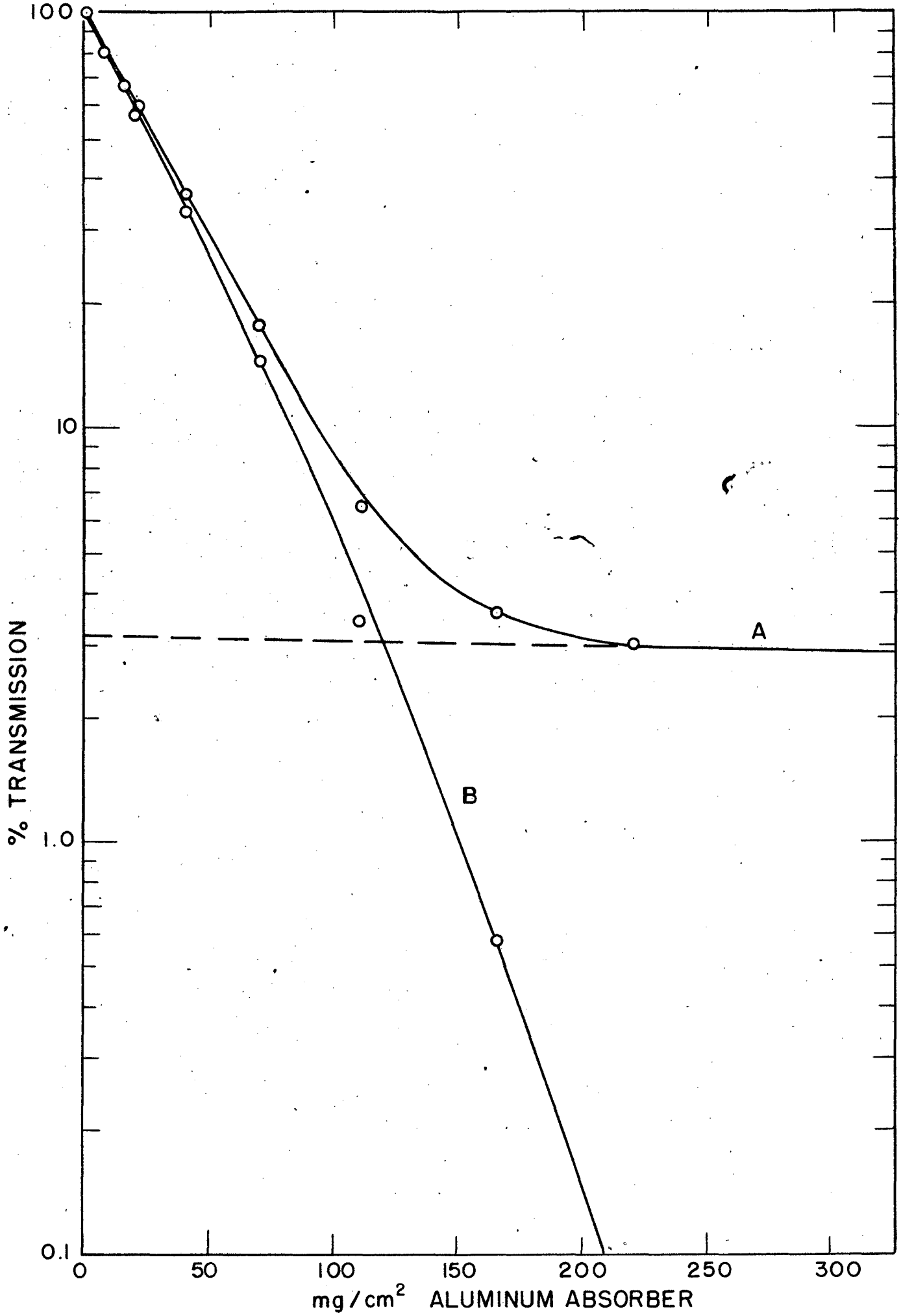
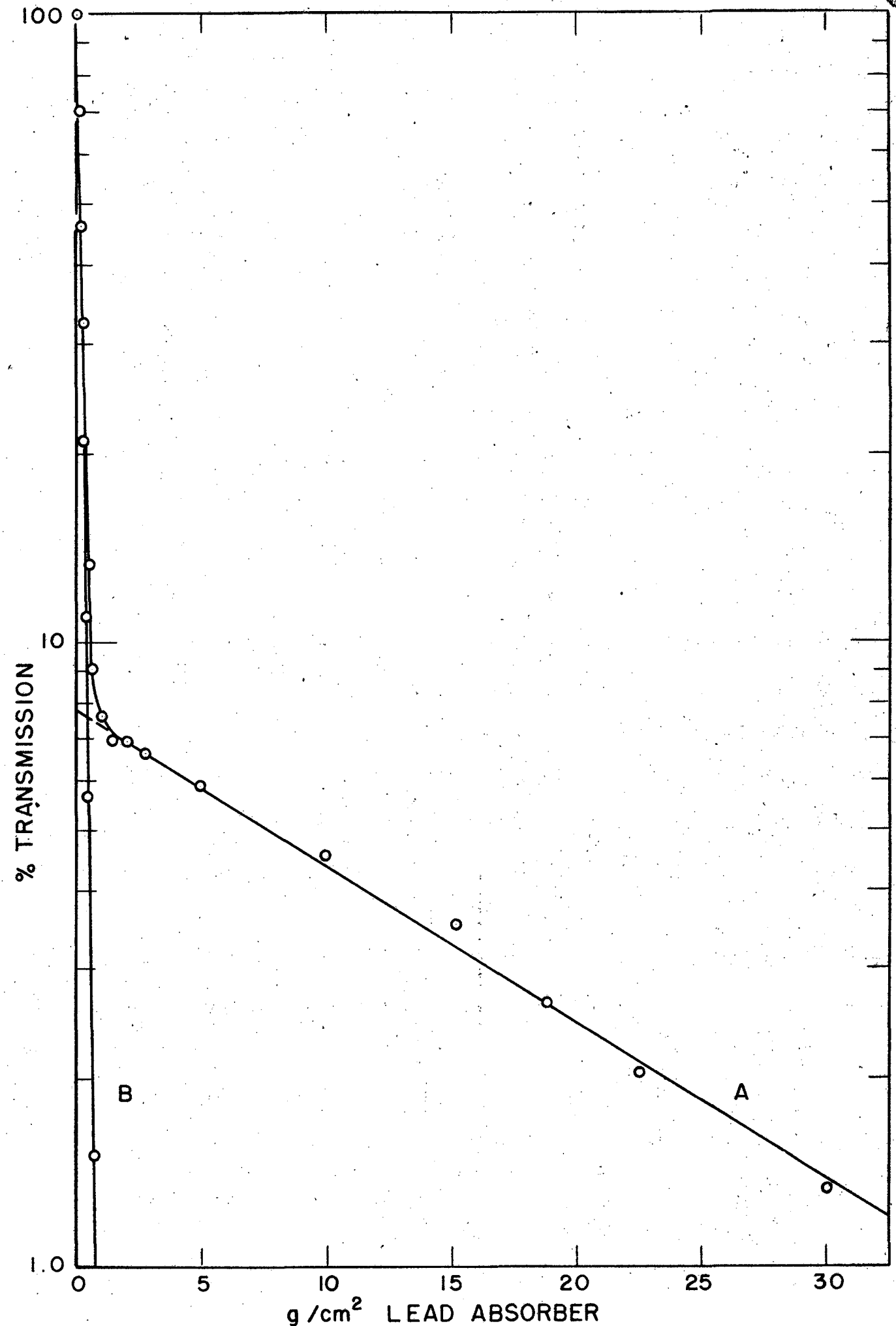

100
10
1.0
0.1
% TRANSMISSION
0
50
100
150
200
250
300
mg / cm² ALUMINUM ABSORBER
A
B

Fig. 47 Lead absorption of 8.00-hour Ta^{180} from Ta + n bombardment. 1.3 Mev γ-ray (A), K x-rays (B).


100
10
1.0
% TRANSMISSION
0
5
10
15
20
25
30
g /cm² LEAD ABSORBER
A
B

The short-lived isomer was studied in short bombardments and a half-life of 16 ± 2 minutes obtained. This activity is formed in very low yield, the saturation intensity being only about 1% that of the 8.0-hour activity. A rapid measurement of the aluminum absorption of the mixed activities was made and the contribution of the 8-hour activity subtracted. The resolved aluminum absorption curve (Fig. 48) of the 16-minute activity shows very soft electrons, range $\sim$20 mg/cm^2 (0.12 Mev), hard electrons or beta particles, range $\sim$180 mg/cm^2 ($\sim$0.6 Mev) together with K x-ray or γ-ray background. The approximate ratios of the radiations are:

0.12 Mev e^-	:	0.6 Mev β^-	:	K + γ-rays
$\sim$0.2	:	$\sim$0.05	:	1

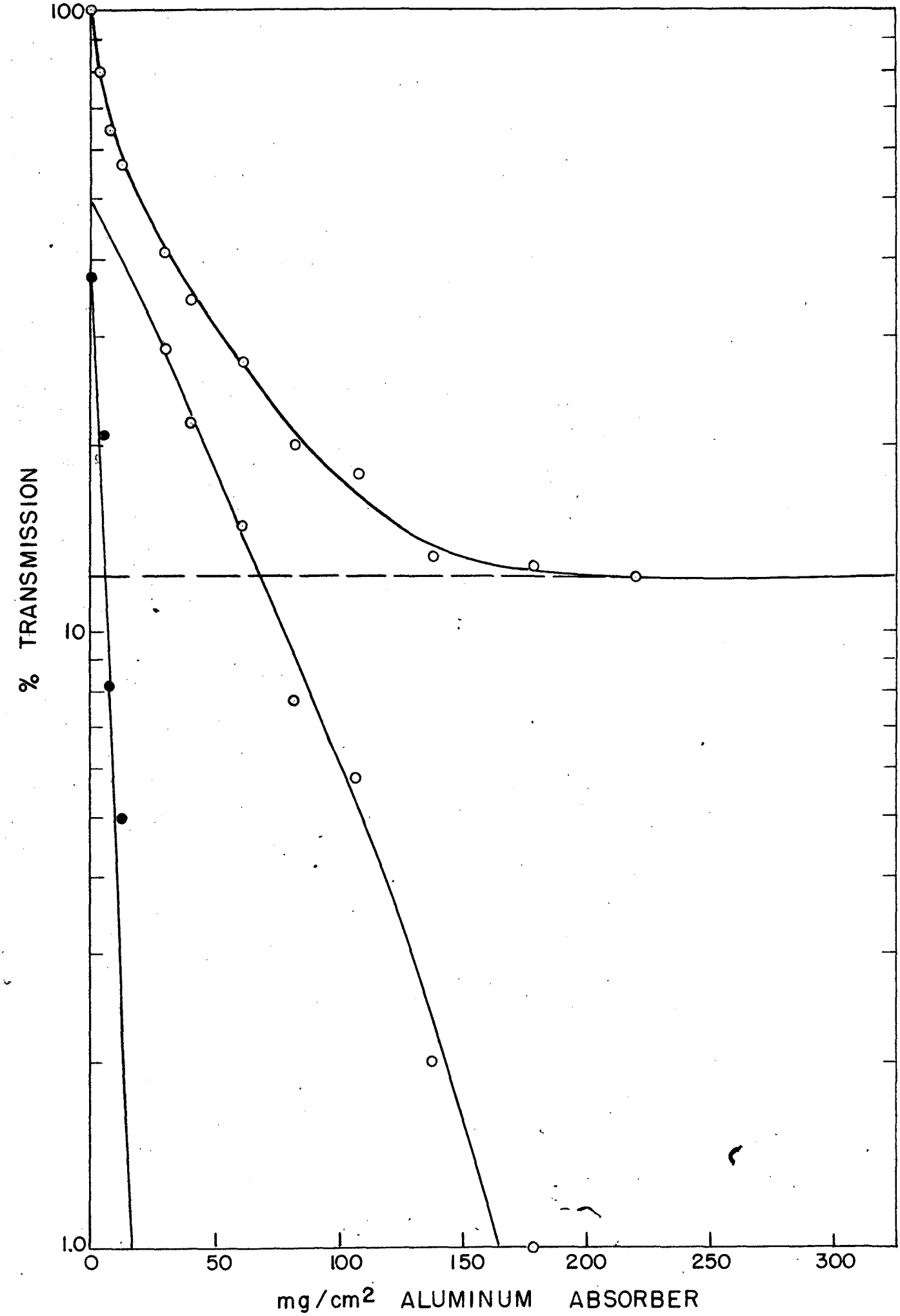
Since the soft electron was not observed in the decay of the 8.0-hour activity, it seems unlikely that these electrons arise from γ-ray transitions in hafnium following orbital electron capture, and the 16-minute activity may decay, partially at least, by isomeric transition. The hard particle present suggests a few percent branching by beta-particle emission[18].

6. Discussion

The relative yields of the tantalum isotopes have been estimated for 38, 31, and 19 Mev helium ion bombardments. In order to show most readily the yield variation with energy (Table XII), the yield of the most abundant activity in each bombardment is taken to be unity. In parentheses are given the approximate cross sections in barns. Since lutecium has two stable isotopes, tantalum isotopes of masses 177 and 178 can be formed by both α,3n and α,2n, and α,2n and α,n reactions respectively, while isotopes of masses 176 and 179 can only be formed by Lu^{175}-α-3n and Lu^{176}-α-n reactions respectively.

It should also be noted that such cross sections may not comprise the whole α,xn reaction since undetected short-lived activities may also be formed.

Fig. 48 Aluminum absorption of 16-minute Ta^{180} from Ta + n bombardment. K x-ray and γ-ray background (A), 0.6 Mev beta (B), 0.2 Mev electron (C).


100
10
1.0
% TRANSMISSION
0
50
100
150
200
250
300
mg/cm2 ALUMINUM ABSORBER

The variations of the observed yields (Table XII) shows that allocation of the 8.0-hour, 2.5-day, 15.4-day, and ~120-day activities to masses 176, 177, 178 and 179 respectively is reasonable; further, these variations and the order of magnitude of the yields themselves agree reasonably with those found for helium ion bombardments of holmium[6].

Table XII

Tantalum Activity	Energy of Helium Ions in Mev 38	31	19	Probable Reaction	Mass
8.0-hours	1	$0.09(10^{-2})$		$Lu^{175}\alpha,3n$	176
2.5 days	0.05	1(0.1)		$Lu^{175}\alpha,2n$ $Lu^{176}\alpha,3n$	177
15.4 days	0.006	$0.002(2 \times 10^{-4})$		$Lu^{175}\alpha,n$ $Lu^{176}\alpha,2n$	178
> 100 days				$Lu^{176}\alpha,n$	179

The yields of the 2.5-day, 15.4 day and >100-day activities in the 10 Mev proton bombardment of hafnium agree with the present allocations. The presence of the 8.0-hour Ta^{176} is masked by the simultaneous formation of the 8.0-hour Ta^{180} (21) in Hf + p bombardments.

H. Rhenium Isotopes[20,21]

Three new rhenium activities of half-lives 12.7 hours, 64-hours, and ~120 days (Table XIII) have been produced by helium ion bombardment of tantalum and proton bombardment of tungsten and have been allocated to masses 182, 182, and 183 respectively. The allocation of a long-lived rhenium activity[24] to mass 184 has also been confirmed.

Table XIII

Isotope	Type of Radiation	Half- Life	Energy of Radiation in Mev Particles	γ-rays	Produced by
Re^{182}	K or IT e^-,γ (β^-?)	12.7 hours	0.16(e^-) ~1 (β^-?)	L,K x-rays 0.4,1.8	Ta-α-3n W-p-n
Re^{182}	K or IT e^-,γ	64.0±0.5 hours	0.11,0.27(e^-)	L,K x-rays 0.22,1.5	Ta-α-3n W-p-n
Re^{183}	K,e^-,γ	~120 days	0.16(e^-)	L, K x-rays 1.0	Ta-α-2n W-p-n
Re^{184}	K,e^-,β^-,γ	40 days	0.1,0.22(e^-) 0.86(β^-)	0.17, 1.05	Ta-α-n W-p-n Re-n-2n

1. 64.0-hour Re^{182}, 12.7-hour Re^{182}

In the bombardment of tantalum with 38 Mev helium ions, two activities of half-lives of 12.7 hours and 64.0-hours were observed, in addition to the longer-lived isotopes of masses 183 and 184. The two short-lived isotopes were observed in the same ratio, but with greatly lowered intensities at 30 Mev, and not at all at 20 Mev bombarding energy. This observation is consistent with production by α,3n reaction. That the 12.7-hour activity cannot be Re^{181} produced by the α,4n reaction is shown by the constancy of the 12.7-hour to 64-hour intensity ratio, and particularly by the production of the 12.7-hour activity in high yield by proton bombardment of tungsten at energies (10 Mev) where the p,2n reaction does not occur. Further, the yield of the 140-day W^{181} (25) which would be produced as the daughter of any Re^{181} formed in the helium ion bombardment of tantalum, agrees well with that to be expected from the known deuteron contamination of the helium ion beam (< 1%) and the measured cross section (2×10^{-2} barns) for the Ta-d-2n reaction.

2. 64.0-hour Re^{182}

The gross decay of this activity, which comprises the bulk of the radioactivity in the 38 Mev helium ion bombardment of tantalum, has been followed through twelve half-lives to give a value of 64.0 ± 0.5 hours for the half-life (Fig. 49). The radiations consist of several electrons, x-rays, and hard γ-radiation, all of which have been followed separately to give this same half-life. The aluminum absorption of the radiations is shown in Fig. 50. Two electron components have been resolved of ranges 16 mg/cm^2 (0.11 Mev) and 70 mg/cm^2 (0.27 Mev). The lead absorption curve (Fig. 51) shows complex electromagnetic radiation of half-thicknesses 140 mg/cm^2 (62 Kev), 550 mg/cm^2 (0.22 Mev) and 13.5 mg/cm^2 (1.5 Mev) in addition to the soft radiation, half-thickness 21 mg/cm^2 aluminum (9.3 Kev) from the aluminum absorption. The two soft components agree well with the expected L and K x-radiation of tungsten. No positrons were detected in a very active sample studied on a crude beta-ray spectrometer. From these measurements, the following ratios were obtained:

	0.11 Mev e^-	:	0.27 Mev e^-	:	L x-rays	:	K x-rays	:	0.22 Mev γ	:	1.5 Mev γ
=	1.4	:	0.2	:	$\sim$2	:	1	:	0.7		0.45

In view of the very complex nature of the radiations, it was thought that a shorter-lived lower isomer of the 64-hour activity might be present - possibly the 12.7-hour rhenium activity. The attempt outlined below was made to separate such a lower isomer.

A carrierless solution of rhenium was prepared from the tantalum target after solution in nitric and hydrofluoric acids. Then the tantalum was precipitated as the hydrated oxide by addition of boric acid, followed by ammonium hydroxide. The filtrate was evaporated with strong nitric acid and the solution which should contain the radioactive rhenium as the perrhenate, was diluted and made alkaline with ammonia. If the 64-hour activity produced a lower isomeric daughter, part

Fig. 49 Gross decay of 64.0 hour Re^{182} (B) and 120 day Re^{183} (A) from Ta + α bombardment.

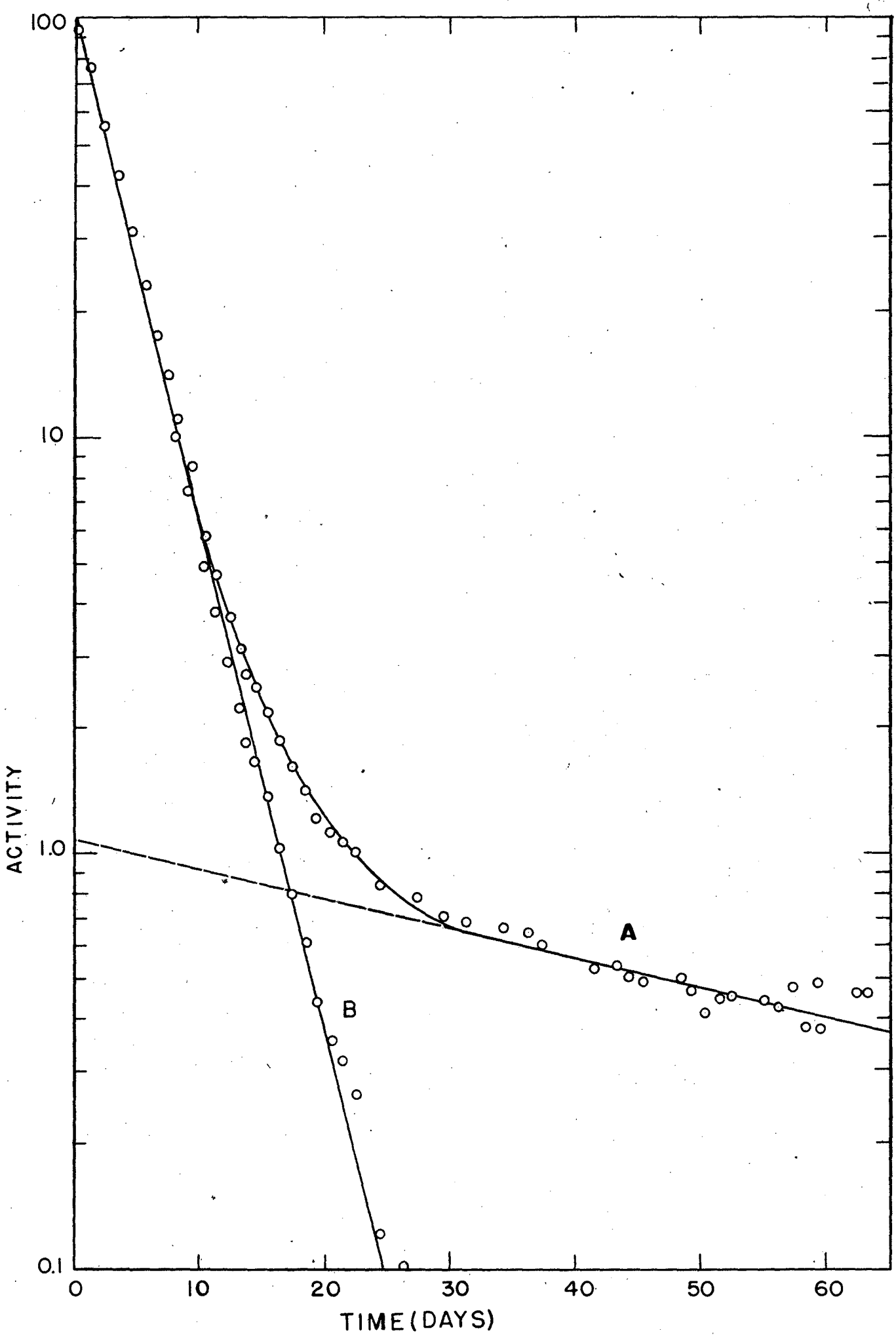

100
10
1.0
0.1
ACTIVITY
0
10
20
30
40
50
60
TIME (DAYS)
A
B

Fig. 50 Aluminum absorption of 64.0-hour Re^{182} from Ta + α bombardment. K x-ray and γ-ray background (A), 0.27 Mev electron (B), 0.11 Mev electron (C), L x-rays (D).

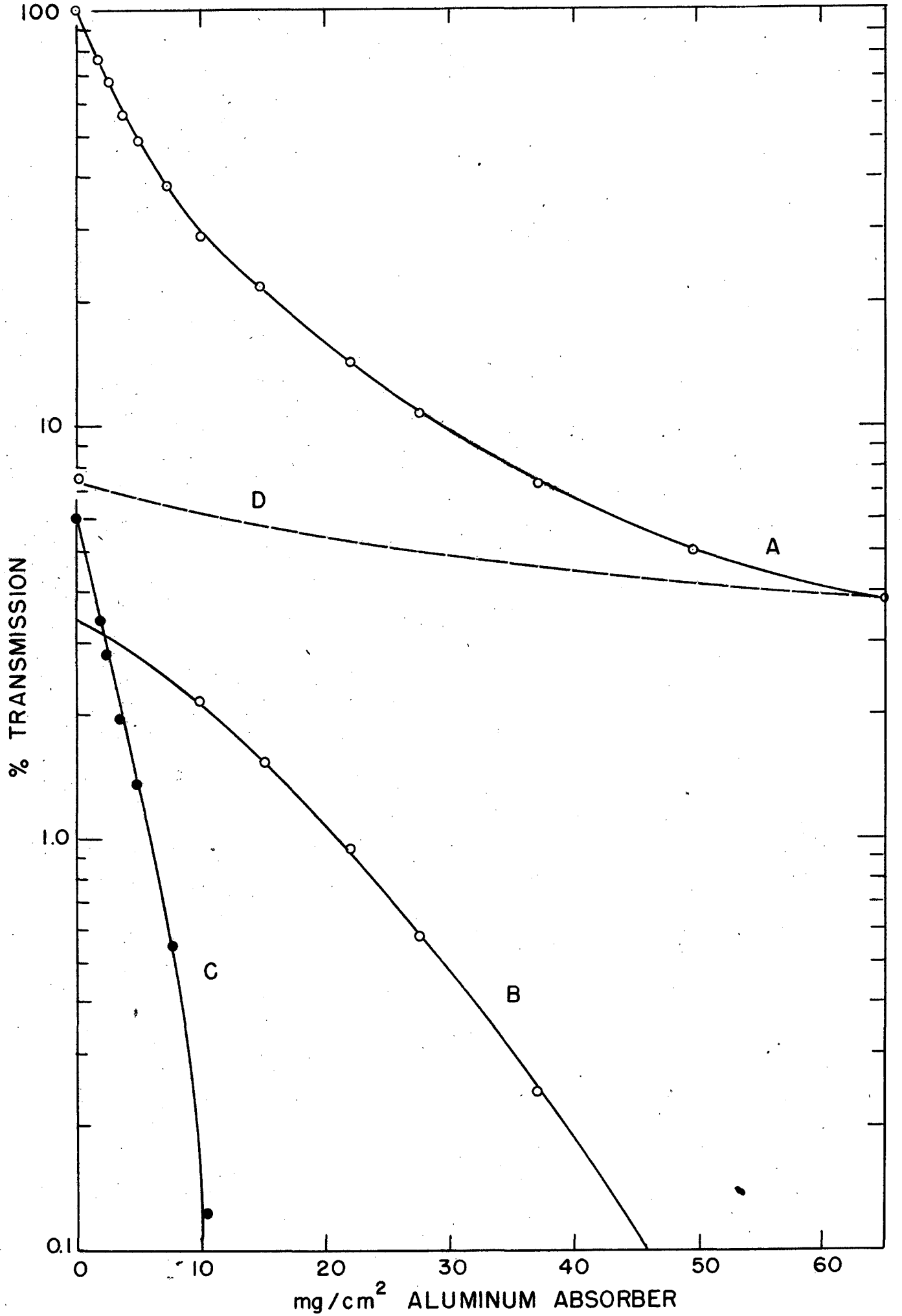
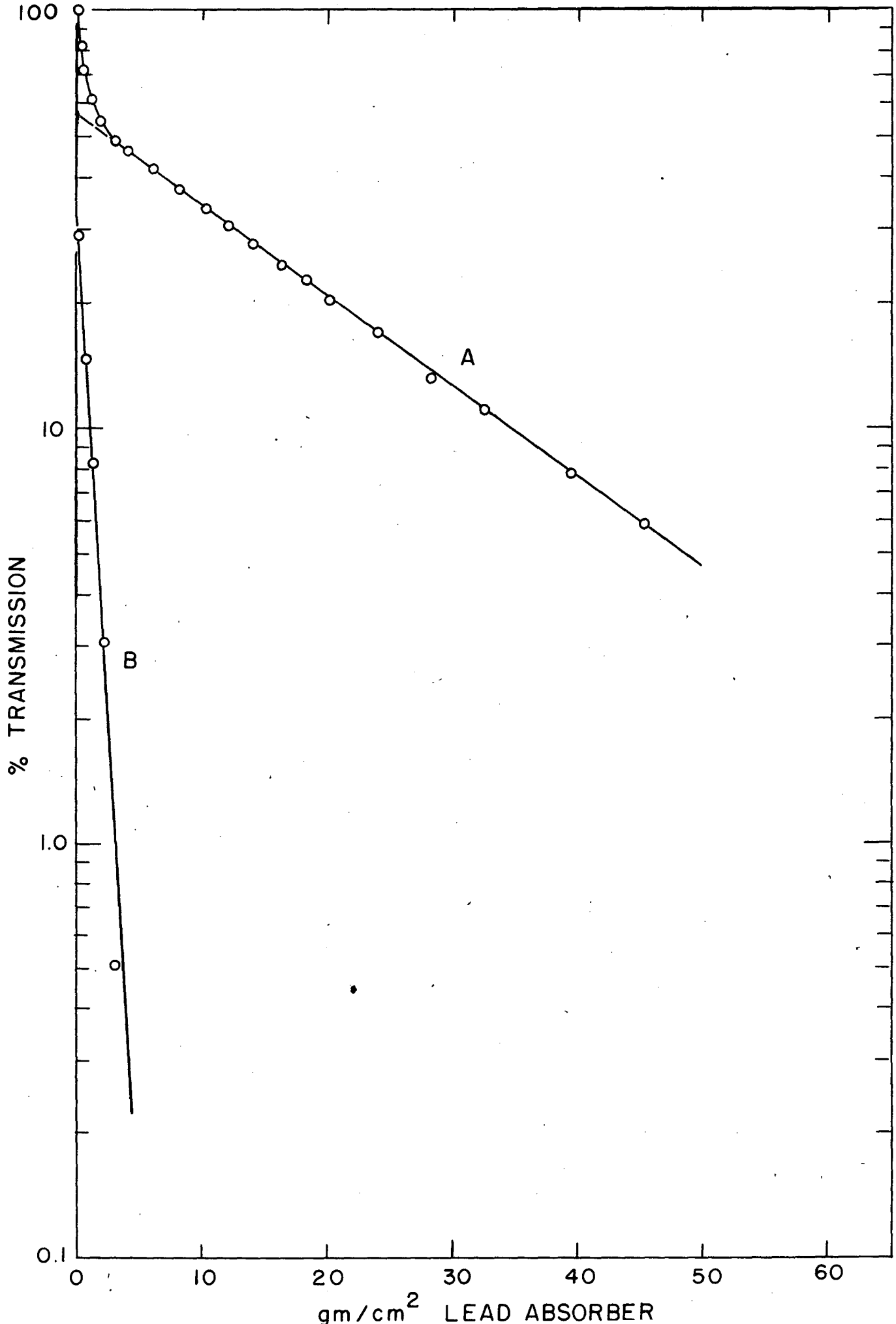

100
10
1.0
0.1
% TRANSMISSION
A
B
C
D
0
10
20
30
40
50
60
mg/cm² ALUMINUM ABSORBER

Fig. 51 Lead absorption of 64.0-hour Re^{182} from Ta + α bombardment. 1.5 Mev γ-ray (A), 0.22 Mev γ-ray (B), K x-rays (C).


100
10
1.0
0.1
% TRANSMISSION
A
B
0
10
20
30
40
50
60
gm/cm^2 LEAD ABSORBER

of the activity of the latter could be expected to remain in the III or IV oxidation states, which could be removed by scavenging the solution with ferric hydroxide. No evidence for such an isomer was observed, and it may be presumed that the various electrons of the 64-hour activity arise from conversion of γ-ray transitions from metastable levels in the daughter nucleus following orbital electron capture.

It is somewhat difficult to decide what radiations constitute one disintegration, since x-rays can come from conversion as well as L or K orbital electron capture. The relative yields at various bombarding energies in Table XIV were calculated on the assumption that one K x-ray quantum represents one disintegration.

3. 12.7 hour Re^{182}

In bombardments of tantalum with helium ions, an activity of 12.7 hours half-life was found to accompany the 64-hour Re^{182}. The activity was found also in the bombardment of tungsten with 10 Mev protons. The decay of the gross and electromagnetic radiations from both Ta + α and W + p (Fig. 52) were followed through four and eight half-lives respectively to give a value of 12.7 $\pm$ 0.2 hours. The radiation characteristics were obtained by resolution of aluminum and lead absorption curves after subtraction of the contribution of longer-lived activities at the time of measurement. The aluminum and lead absorption curves for the 12.7 hour activity are shown in Fig. 53 and Fig. 54 respectively. The radiations consist of electrons of ranges 35 mg/cm^2 (160 Kev) and $\sim$400 mg/cm^2 ($\sim$1 Mev); and electromagnetic radiations of half-thicknesses 20 mg/cm^2 aluminum (9.3 Kev), 140 mg/cm^2 lead (62 Kev), 3.0 g/cm^2 lead (400 Kev) and 15 g/cm^2 (1.8 Mev). The two soft electromagnetic radiations correspond well with tungsten or rhenium L and K x-radiation. From the measurements, the following ratios were obtained:

Fig. 52 Electromagnetic decay of 12.7 hour Re^{182} (B) and 64.0-hour Re^{182} background (A) from Ta + α bombardment.

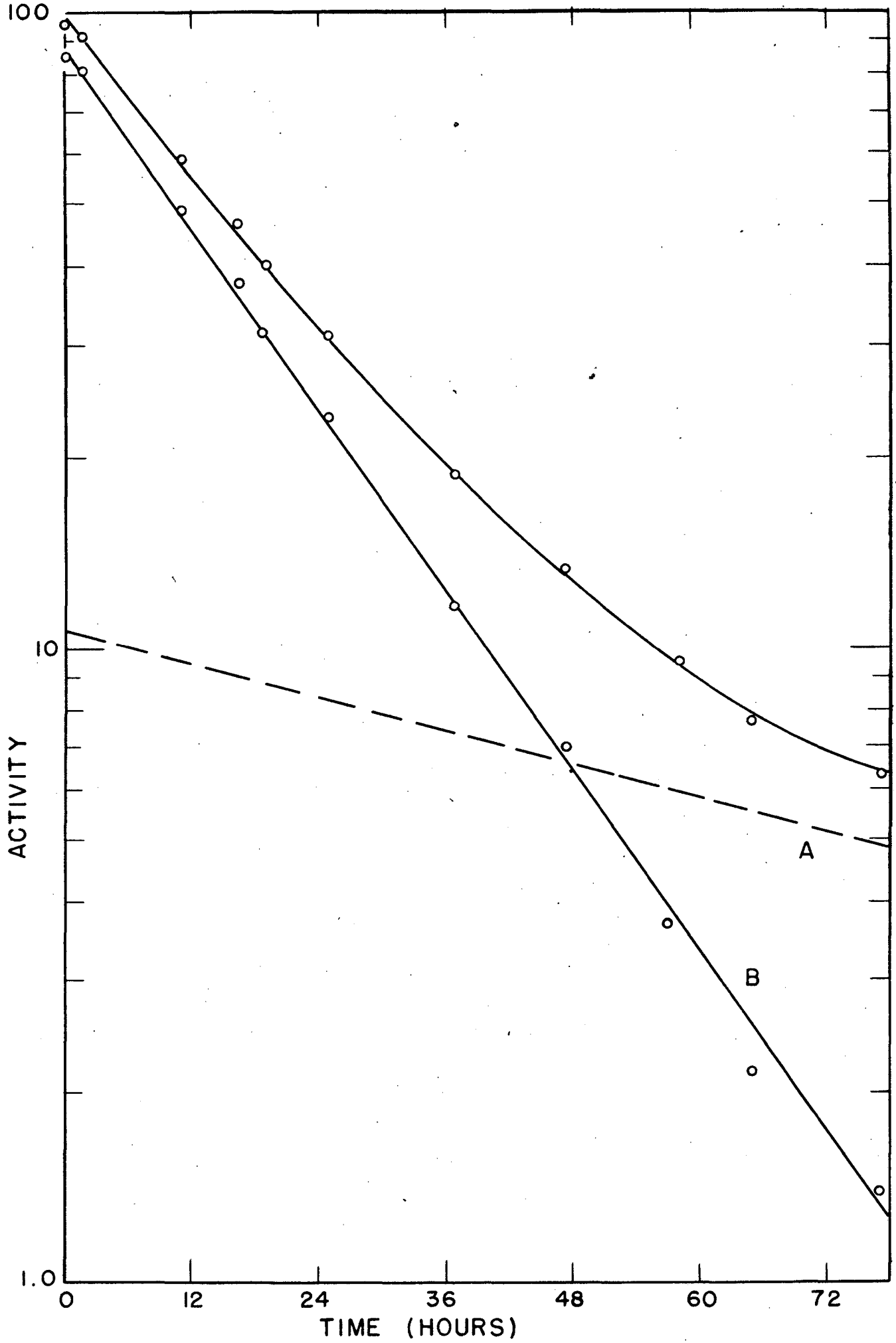

100
10
1.0
ACTIVITY
0
12
24
36
48
60
72
TIME (HOURS)
A
B

Fig. 53 Aluminum absorption of 12.7 hour Re^{182} from Ta + α bombardment. K x-ray and γ-ray background (A). 1 Mev electron (B), 0.16 Mev electron (C), L x-rays (D).

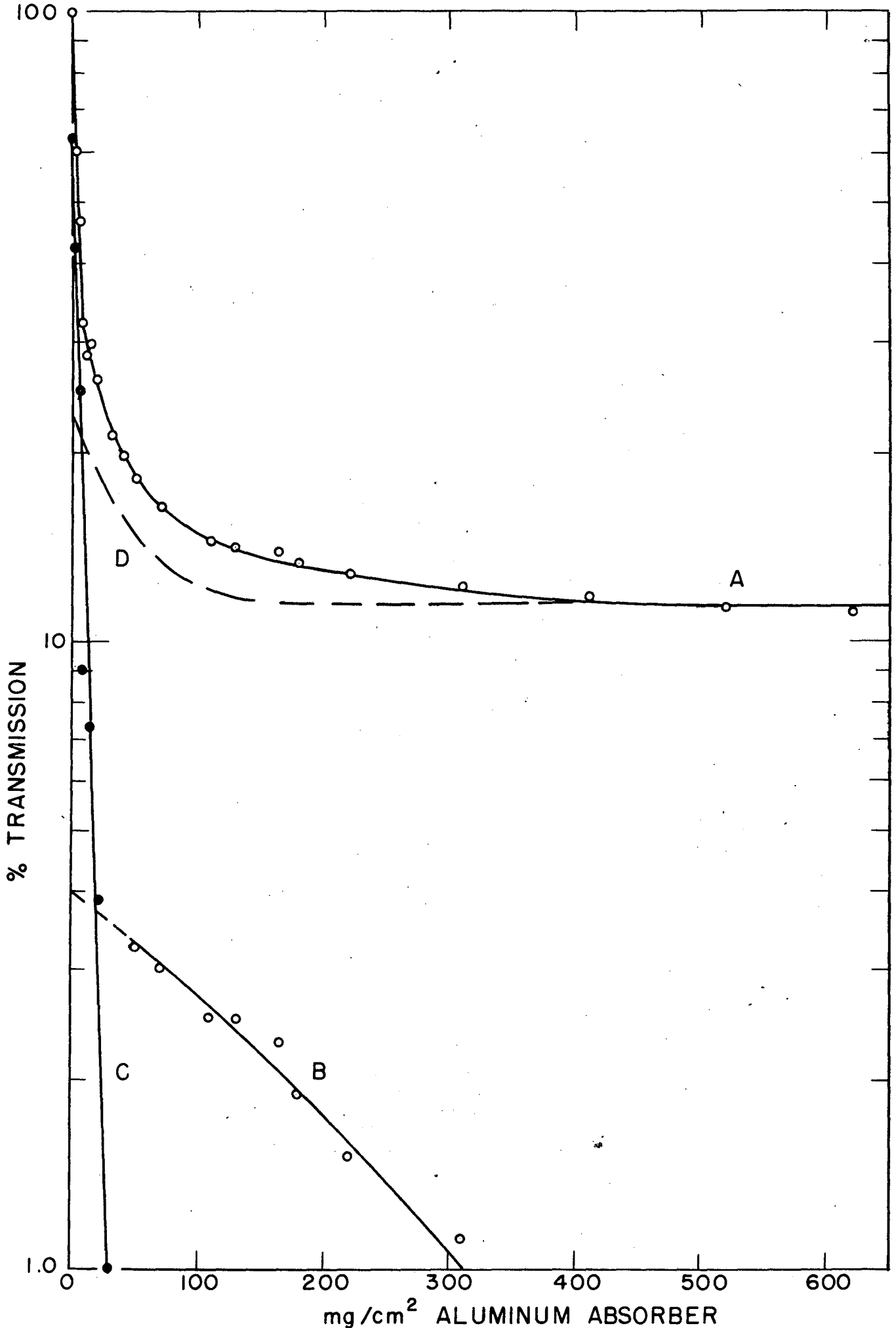
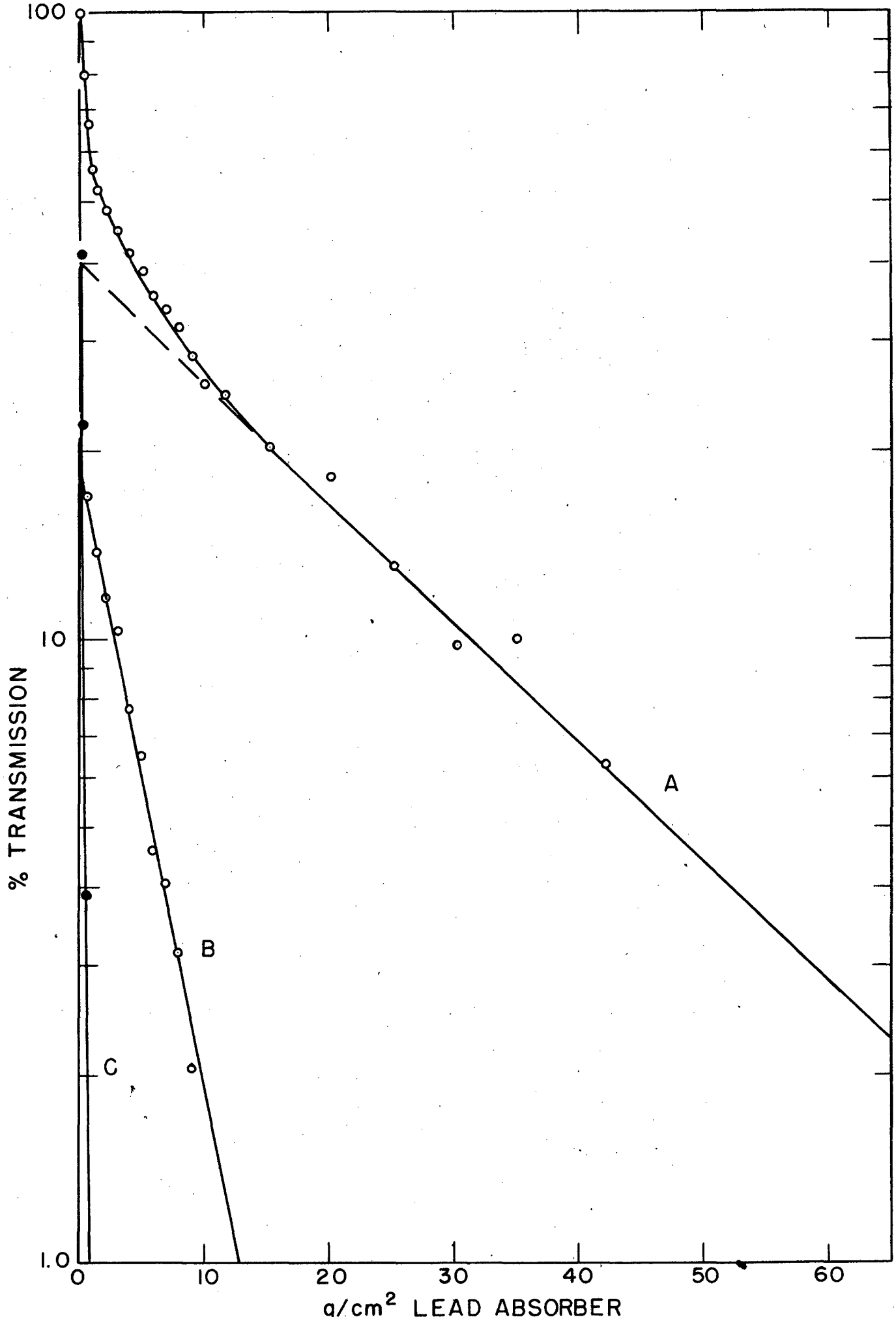

100
10
1.0
% TRANSMISSION
0
100
200
300
400
500
600
mg/cm^2 ALUMINUM ABSORBER
A
B
C
D

Fig. 54 Lead absorption of 12.7 hour Re^{182} from Ta + α bombardment. 1.8 Mev γ-ray (A), 0.4 Mev γ-ray (B), K x-rays (C).


100
10
1.0
% TRANSMISSION
0
10
20
30
40
50
60
g/cm² LEAD ABSORBER
A
B
C

160 Kev e^-	~1 Mev e^-	L x-rays	K x-rays	0.4 Mev γ	1.8 Mev γ
0.04	0.003	0.5	1	0.25	0.15

The isotope thus appears to decay by orbital electron capture, although partial decay by isomeric transition to the 64-hour activity is not excluded. The hard negative electron observed in the radiation of the 12.7-hour activity may be a beta-particle, in which case, the branching by beta disintegration to an unreported beta stable osmium isotope[18], Os^{182}, would be approximately 0.3%. Since no hard electrons were observed in the radiations of the 64-hour activity, this may be presumed to be a lower isomer of the 12.7-hour isotope.

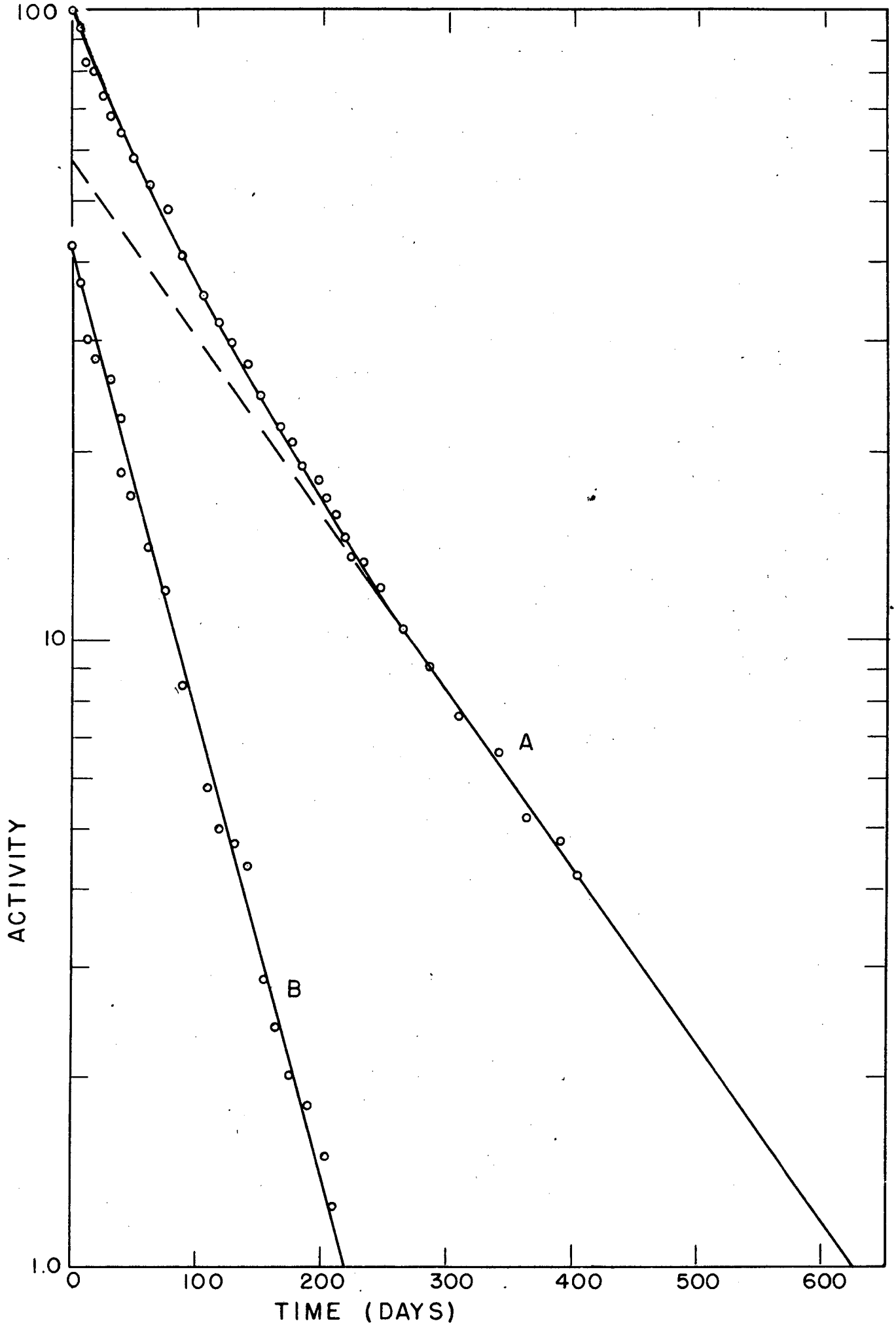
For calculation of relative yields one K x-ray is assumed to represent one disintegration by orbital electron capture or isomeric transition.

4. 120 day Re^{183} and 40 day Re^{184}

After decay of the short periods in Ta + α, W + p, and Re + n bombardments, a long-lived activity remains. A 50 day activity, allocated to Re^{184} and decaying by both orbital electron capture and beta emission is known, and has been well characterized[21, 24]. The long-lived activity now obtained from fast neutron bombardment of rhenium has a half-life of 40 $\pm$ 2 days measured through 2 periods. The resolution of the long period decays from Ta + α and W + p bombardments gives also the 40-day half-life (Fig. 55). This is somewhat shorter than that obtained by previous workers[24], but since the decay has been followed through several periods, 40 days is considered to be a better value. The measured radiation characteristics agree with those obtained by others.

In the rhenium fractions from Ta + α and W + p bombardments, an activity of ~120 days half-life has been observed after decay of the 40-day period. A pure intense source of this isotope was obtained from aged tungsten exit strips which had received deuterons, protons, and helium ions from the 60-inch Crocker Laboratory cyclotron.

Fig. 55 Gross decay of 40 day Re^{184} (B) and 120-day Re^{183} (A) from Ta + α bombardment.


100
10
1.0
ACTIVITY
0
100
200
300
400
500
600
TIME (DAYS)
A
B

The aluminum absorption of an infinitely thin sample is shown in Fig. 56, and the lead absorption of a more active sample, in Fig. 57. The radiations consist of electrons, range 35 mg/cm^2 (0.16 Mev) and electromagnetic radiations of half-thicknesses $\sim$21 mg/cm^2 aluminum (9.4 Kev), 140 mg/cm^2 lead (62 Kev), and 10 g/cm^2 lead (1.0 Mev). On the crude beta-ray spectrometer, only a single peak of very soft electrons of $\sim$0.15 Mev energy were observed; no positrons were detected in a very active sample. The following ratios were obtained from these measurements:

0.16 Mev e^-	: L x-rays	: K x-rays	: 1 Mev γ
$\sim$0.4	: $\sim$1.1	: 1	: 0.1

The isotope appears to decay by orbital electron capture, with electrons arising from subsequent γ-ray transitions. The isotope has been allocated to mass 183 on the basis of yields (Table XIV) in the helium ion bombardments of tantalum; the absence of hard electrons or beta particles is also in agreement with the present allocation[18].

5. Discussion

Because of the complexity of decay schemes, only the relative yields in helium ion bombardments at various energies can be considered reliable. These were calculated taking one K x-ray quantum as representative of one disintegration by orbital electron capture. The results agree in trend with those for helium ion reactions at the same energies on other elements.

Fig. 56 Aluminum absorption of 120-day Re^{183} from old tungsten-exit strip. K x-ray and γ-ray background (A), 0.16 Mev electron (B), L x-rays (C).

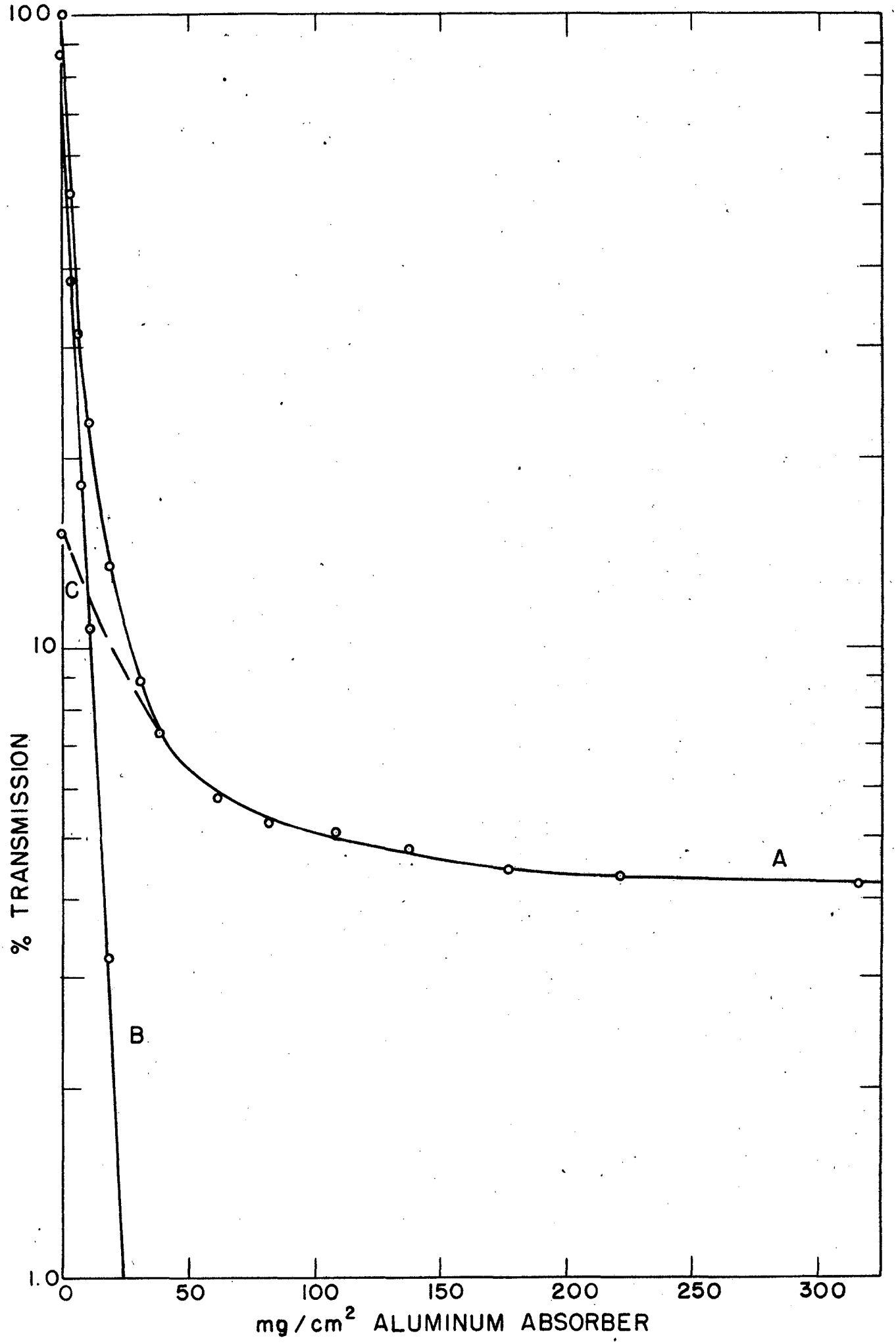

100
10
1.0
% TRANSMISSION
0
50
100
150
200
250
300
mg / cm² ALUMINUM ABSORBER
A
B
C

Fig. 57 Lead absorption of 120-day Re^{183} from old tungsten-exit strip. 1.0 Mev γ-ray (A), K x-rays (B).

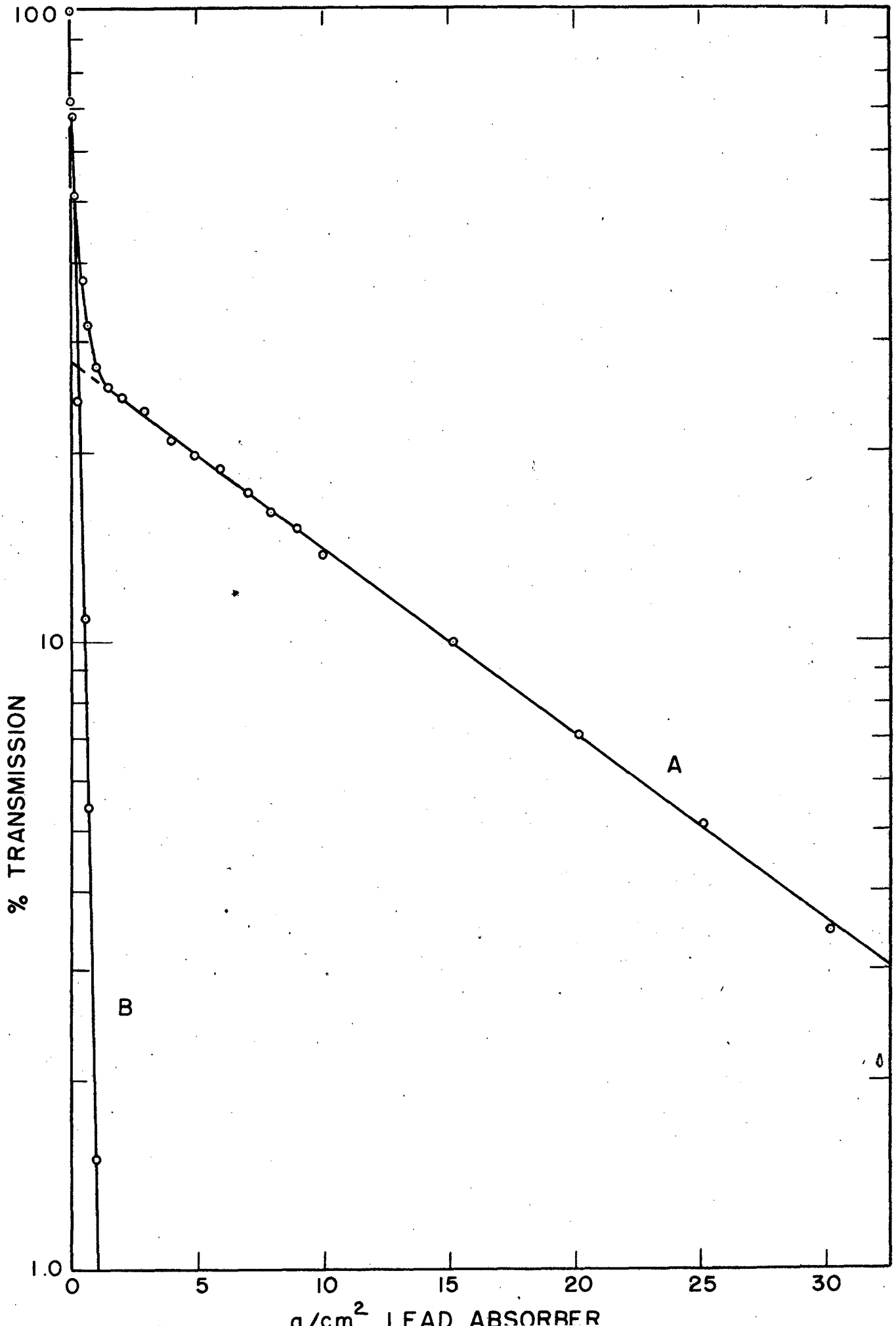

100
10
1.0
% TRANSMISSION
A
B
0
5
10
15
20
25
30
g/cm² LEAD ABSORBER

Table XIV

Half-Life	Helium Ion Energy 38	31	19	Probable Reaction
12.7 hours	0.5	0.5	--	α,3n
64.0 hours	1	1	--	α,3n
120 days	0.02	0.5	0.7	α,2n
40 days	0.01	0.07	1	α,n

IV Acknowledgements

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