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Neutron Deficient Radioactive Isotopes of Rhenium

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NEUTRON DEFICIENT RADIOACTIVE ISOTOPES OF RHENIUM

G. Wilkinson and H. G. Hicks

July 15, 1949

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Neutron Deficient Radioactive Isotopes of Rhenium

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University of California, Berkeley, California

ABSTRACT

A study has been made of neutron deficient radioactive isotopes of rhenium. Using the 60-inch Crocker Laboratory cyclotron, bombardments have been made of tantalum with 38, 30 and 19 Mev α-particles, of tungsten with 10 Mev protons, and of rhenium with fast neutrons.

Four new radioactive isotopes were observed; the characteristics of these and of the previously known 40 day Re^{184} which has been restudied are given in Table I.

Table I

Isotope	Type of Radiation	Half-Life	Energy of radiation in Mev Particles	Energy of radiation in Mev γ-rays	Produced by
Re^{182}	K or I.T. e^-,γ	64.0±0.5 hours	0.11,0.27(abs) 0.040,0.098 0.059,0.117 0.152,0.210 0.180,0.238 0.276,0.334 (spec)	L,K x-rays 0.22,1.5(abs) 0.110,0.129, 0.250(spec conv) 0.222,0.346 (spec and spec conv)	Ta-α-3n W-p-n
Re^{182}	K or I.T. e^-,γ	12.7 ± 0.2 hours	0.16,~1(abs) 0.040,0.098 0.059,0.117 0.152,0.210 0.180,0.238 0.276,0.334 (spec)	L,K x-rays 0.4,1.6(abs) 0.110,0.129, 0.250(spec conv); 0.222,0.346 (spec and spec conv)	Ta-α-3n W-p-n
Re^{183}	K,e^-,γ	~120 days	0.16(abs) 0.069,0.078 0.182,0.240 (spec)	L,K x-rays 1.0 (abs); 0.081,0.252 (spec conv)	Ta-α-2n W-p-n

Table I (cont'd)

Isotope	Type of Radiation	Half-Life	Energy of radiation in Mev Particles	Energy of radiation in Mev γ-rays	Produced by
Re^{184}	K,e^-,γ	40 days	0.2,0.7(abs) 0.031,0.040; 0.089,0.147; 0.135,0.193; 0.215,0.273; (spec)	L,K x-rays 0.043 0.205,0.285 (spec conv); 0.159(spec and spec conv)	Ta-α-n W-p-n Re-n-2n
Re^{184}	K or I.T., e^-,γ	2.2 days	0.2,1.1(abs) 0.052; 0.031,0.040; 0.089,0.147 (spec)	L,K x-rays 0.043(spec conv) 0.159(spec and spec conv)	Ta-α-n W-p-n

I. Experimental

Tantalum was bombarded on the 60-inch Crocker Laboratory cyclotron as the spectroscopically pure metal foil. After bombardment, the foil was dissolved in hydrofluoric acid, with the addition of the minimum of strong nitric acid. Rhenium carrier was then added, and hydrogen sulphide passed into the boiling solution for fifteen minutes. The rhenium sulphide was dissolved in a solution of hydrogen peroxide and sodium hydroxide, which was then "scavenged" by ferric hydroxide precipitations. The rhenium was then reprecipitated from strong hydrochloric acid solution as the sulphide. While this procedure has been found to give radiochemically pure rhenium, the following additional chemistry was performed in early bombardments: rhenium was distilled as the volatile chloride or oxide and recovered from the distillate by precipitation as sulphide. The latter was dissolved in a solution of hydrogen peroxide and sodium hydroxide and the rhenium then precipitated from the solution as nitron perrhenate by the well known procedure. Similar chemical methods were used for neutron bombarded rhenium metal and for proton bombarded tungsten.

The techniques of measurement of radiations by aluminum, beryllium and lead absorption, and the various assumptions made in the interpretation of results, have been described previously[1,2].

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(1) Geoffrey Wilkinson, Phys. Rev. 75, 1019 (1949).

(2) G. Wilkinson and H. G. Hicks, Phys. Rev. 75, 1370 (1949).

II. Rhenium Isotopes

Four new rhenium activities of half-lives 12.7 hours, 64.0 hours, $\sim$120 days and 2.2 days have been produced by α-particle bombardment of tantalum and have been allocated respectively to masses 182, 182, 183 and 184. The allocation of a previously reported 40 day rhenium activity to mass 184 has

also been confirmed.

A. 64.0 hour Re^{182}; 12.7 hour Re^{182}

In the bombardment of tantalum with 38 Mev α-particles, two activities of half-lives 12.7 hours and 64.0 hours were observed, in addition to the longer-lived isotopes. 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 α,4n reaction is shown by the constancy of the 12.7 hour to 64 hour intensity ratio (Table II), and particularly by the production of the 12.7 hour activity by 9 Mev proton bombardment of tungsten where the p,2n reaction does not occur. Further, the yield of the 140 day W^{181} (3) which would be produced as the

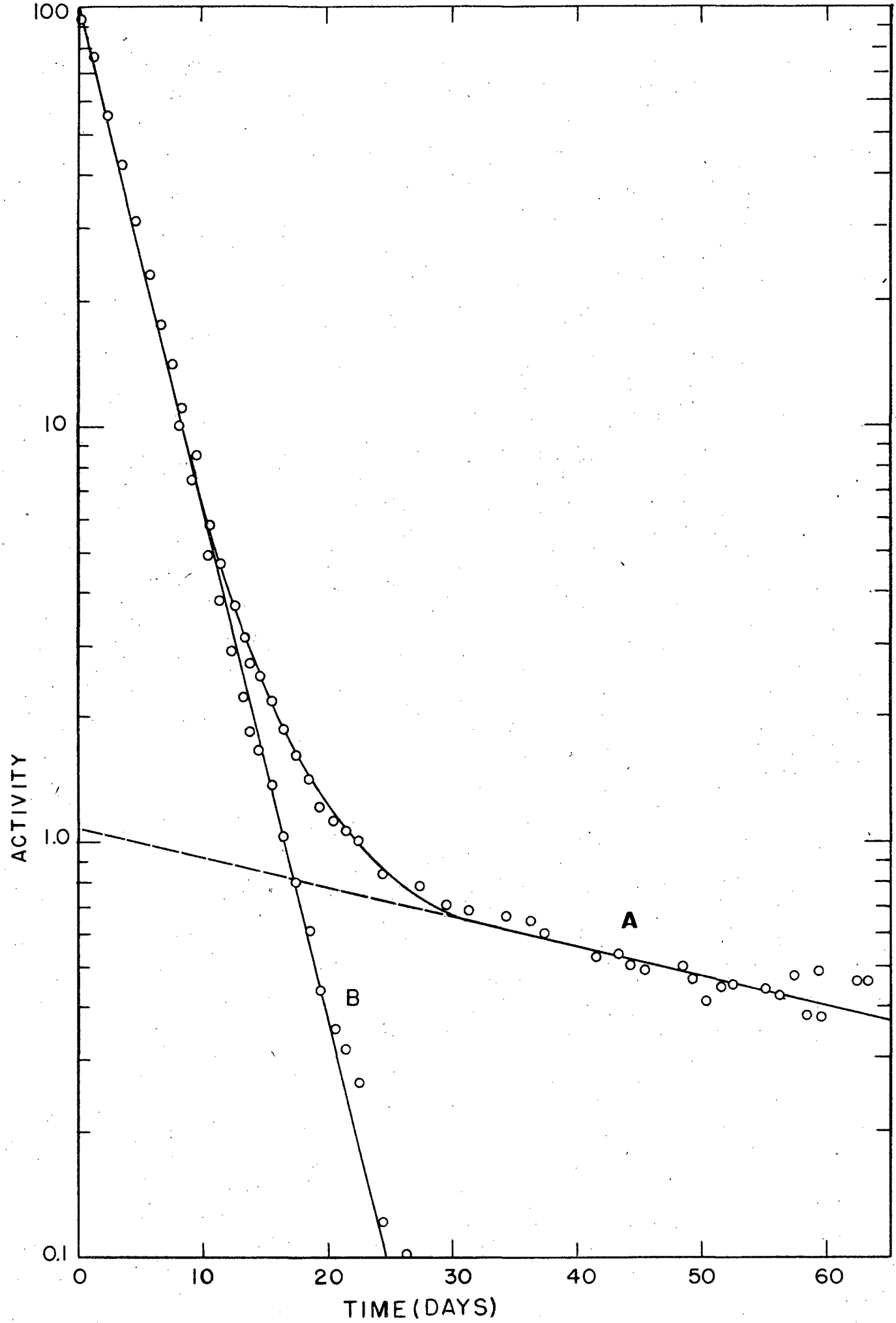
(3) Geoffrey Wilkinson, Nature 160, 864 (1947).

daughter of any Re^{181} formed in an α,4n reaction of tantalum, agrees with that to be expected from the known deuteron contamination of the α-particle beam (<1%) and the measured cross section (5×10^{-2} barns) for the Ta-d-2n reaction.

64.0 ± 0.5 hour Re^{182}.

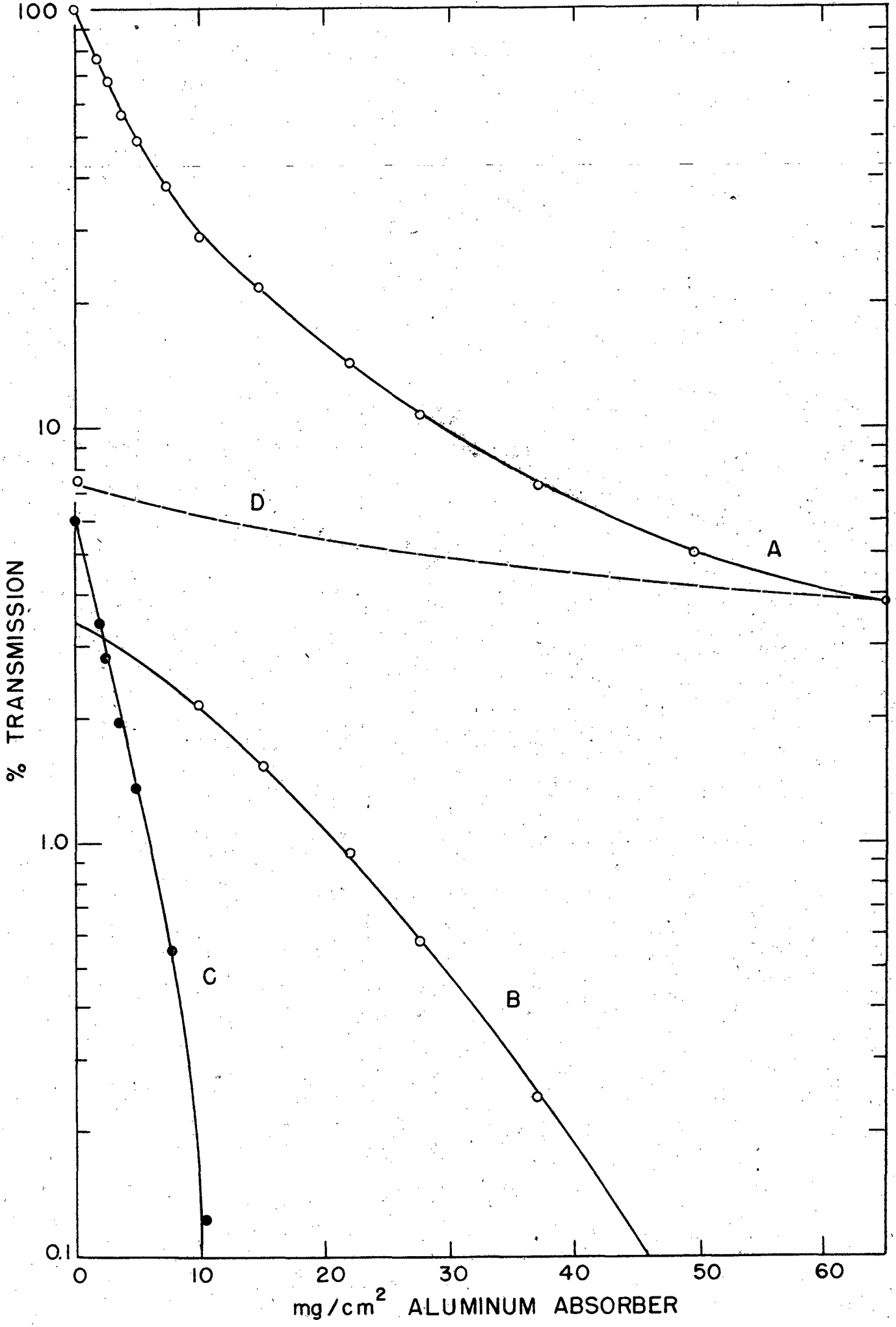
The gross decay of this activity, which comprises the bulk of the radioactivity in the 38 Mev α-particle bombardments of tantalum, has been followed through twelve half-lives to give a value of 64.0 ± 0.5 hours for the half-life. (Fig. I) The radiations consist of electrons, x-ray and hard γ-radiation, all of which have been separately followed and the same half-life obtained. The aluminum absorption of the radiations is shown in Fig. II. The electromagnetic radiation contribution was obtained from aluminum absorption measurements after removal of the electrons by beryllium absorbers. Two electron components have been resolved, of ranges 16 mg/cm^2 (~110 Kev), and 70 mg/cm^2

Figure I: Gross decay of 64.0 hour Re^{182}(B) and $\sim$120 day Re^{183}(A) activities from bombardment of tantalum with 38 Mev α-particles.



13200-1

Figure II: Aluminum absorption of 64.0 hour Re^{182} activity; K x-rays and γ-rays (A), 0.27 Mev electron (B), 0.11 Mev electron (C), and 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

13198 — 1

(0.27 Mev). The lead absorption curve (Fig. III) shows complex hard electromagnetic radiation of half-thicknesses 137 ± 5 mg/cm^2 (62 Kev), 550 mg/cm^2 (220 Kev) and 13.5 g/cm^2 lead (1.5 Mev) in addition to the soft radiation half-thickness 21 ± 1 mg/cm^2 aluminum measured in the aluminum absorption; the two soft components agree well with the expected L and K x-radiation of tungsten or rhenium. No positrons were detected in a very active sample studied on a crude beta ray spectrometer.

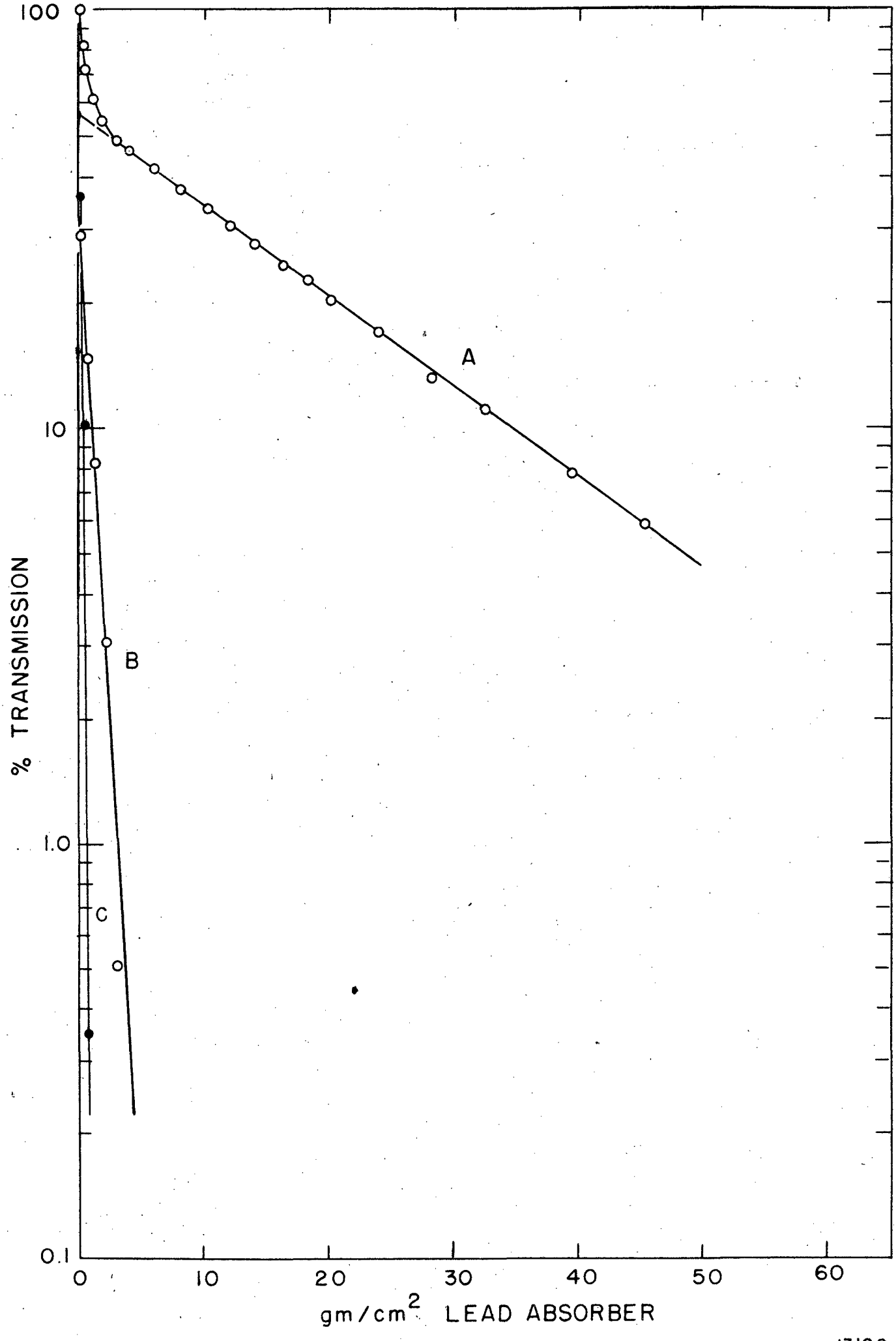
From the aluminum absorption of the radiations from a "carrierless" rhenium sample mounted on very thin mica backing, the ratios of the two electrons and L x-radiations to the harder electromagnetic radiations were determined; corrections were made for absorption of the soft radiations in the air gap and counter window were made. The ratios of the hard electromagnetic radiations were obtained from the lead absorption. Counting efficiencies assumed were 2.5% for L x-radiation, 0.5% for K x-radiation and 220 Kev γ-rays, and 1.5% for the 1.5 Mev γ-ray. Fluorescence yields of 0.8 and 0.5 were assumed for K and L x-rays respectively. The following ratios were obtained:

110 Kev e^- : 270 Kev e^- : L x-rays : K x-rays : 220 Kev γ : 1.5 Mev γ

= 1.4 : 0.2 : $\sim$2 : 1 : 0.7 : 0.45.

In view of the 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 activity observed in the bombardments along with the 64 hour rhenium. An attempt was made to separate the nuclear isomers. A carrierless solution of rhenium was prepared by removal of tantalum from a bombarded target after solution in nitric and hydrofluoric acids, by addition of boric acid followed by ammonia. The filtrate was evaporated with strong nitric acid and the solution, which should contain the radioactive rhenium as perrhenate, was diluted and neutralized with ammonia. If the 64 hour activity produced

Figure III: Lead absorption of 64.0 hour Re^{182} activity; 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
C
0
10
20
30
40
50
60
gm/cm² LEAD ABSORBER

13199-1

a lower isomeric daughter, part of the activity of the latter would be expected to remain in the III or IV oxidation states after decay of the parent. Any activity in the III or IV oxidation state could then be removed by scavenging the solution with ferric hydroxide. No evidence for such an isomer was observed, and it may be presumed that the very soft electrons of the 64 hour activity arise from conversion in 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 arise from conversion as well as from L or K orbital electron capture.

The relative yields for production of the 64-hour activity at the various energies of bombarding α-particles (Table II), can, however, be compared by taking the K x-rays as a reference.

12.7 hour Re^{182}

In bombardments of tantalum with α-particles, 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 bombardments (Fig. IV) were followed through four and eight half-lives respectively to give a value of 12.7 $\pm$ 0.2 hours for the half-life. The radiation characteristics were obtained by resolution of aluminum, beryllium and lead absorption curves, after the contribution of the longer-lived activities at the time of measurement had been subtracted. The aluminum and lead absorption curves for the 12.7 hour activity are shown in Fig. V and Fig. VI 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), 30 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 fol-

Figure IV: Decay of electromagnetic radiations of 12.7 hour Re^{182}(B) and 64.0 hour Re^{182} (A) activities from 10 Mev proton bombardment of tungsten.

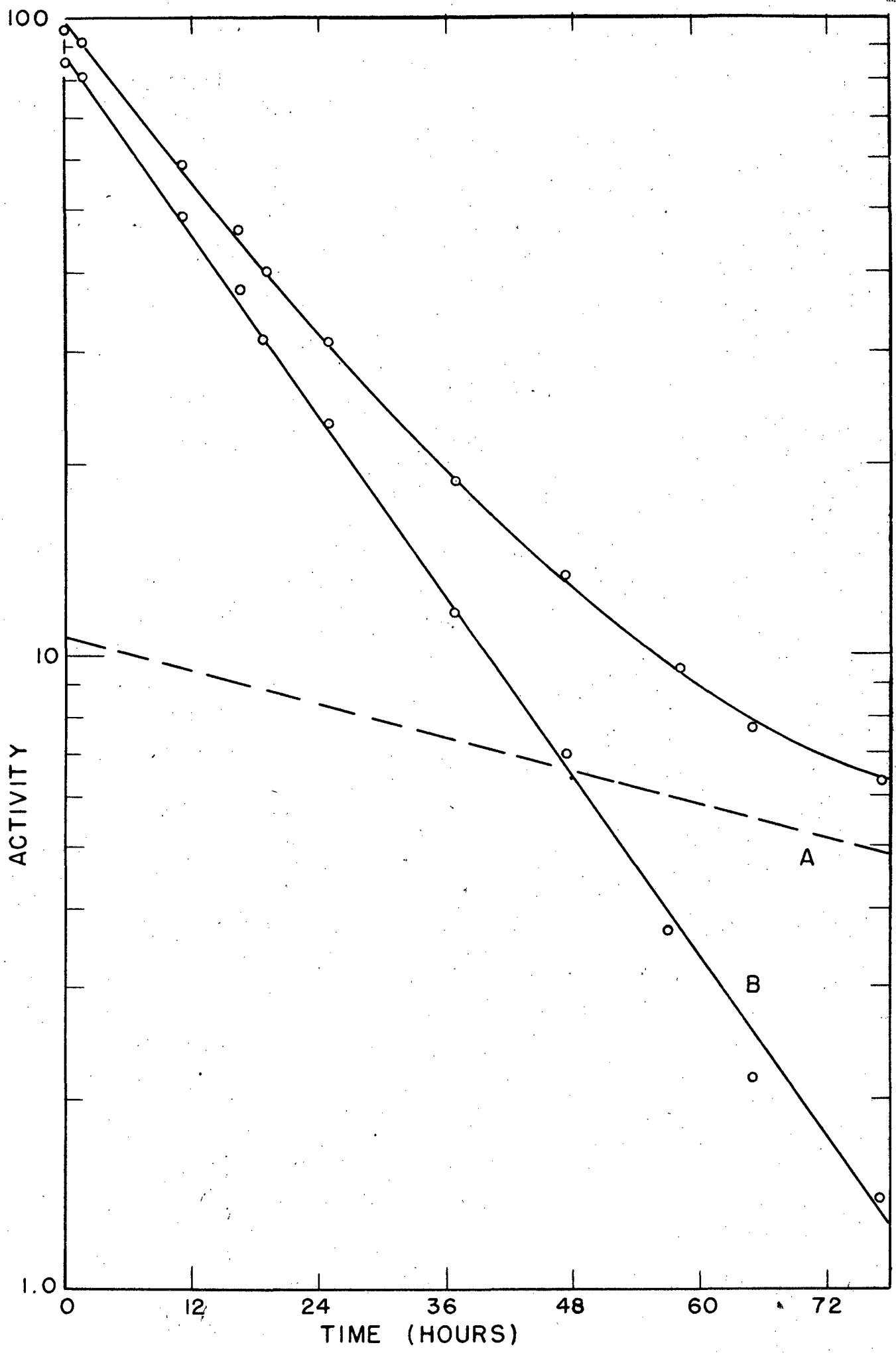
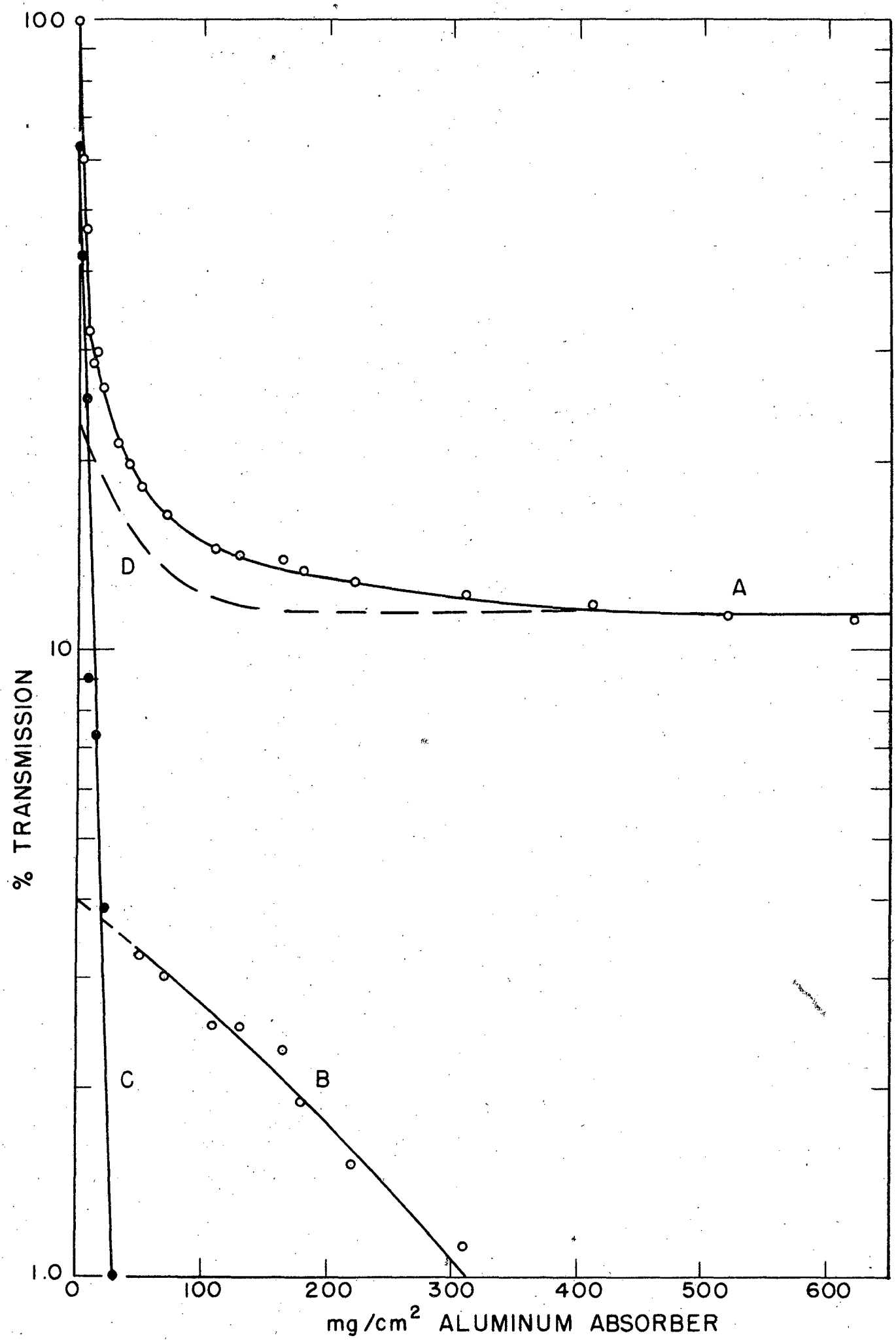
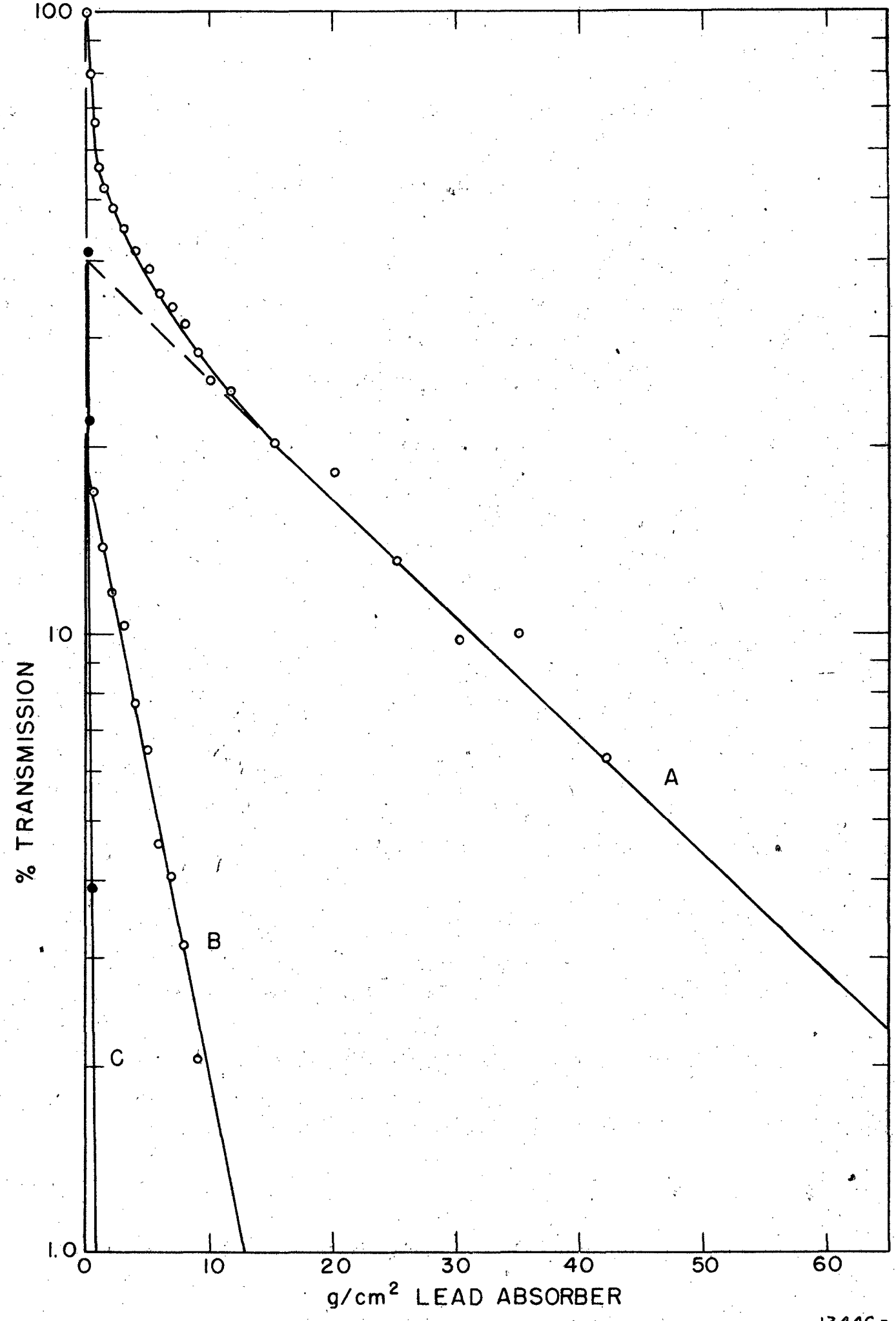

100
10
1.0
ACTIVITY
0
12
24
36
48
60
72
TIME (HOURS)
A
B
13450-1

Figure V: Aluminum absorption of 12.7 hour Re^{182} activity; K x-rays and γ-rays (A) $\sim$1 Mev electron (B), 0.16 Mev electron (C), and L x-rays (D).


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

13445-1

Figure VI: Lead absorption of 12.7 hour Re^{182} activity; 1.6 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
13446-1

lowing ratios were obtained:

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.

B. 40 day Re^{184}; 120 day Re^{183}

After decay of the shorter lived periods in Ta + α and W + p bombardments, a complex long lived activity remains in the rhenium fractions. This activity has been resolved into two components of half-lives 40 ± 2 days and 120 days, the measurements being carried through several half-lives. The 40 day activity has been also produced by fast neutron bombardment of rhenium. The radiation characteristics obtained by resolution of absorption and decay curves (Fig. VII, VIII) are the same in all bombardments, and agree well with those reported by other workers for a 50 day Re^{184} isotope(4). The approximate

(4) See G. T. Seaborg and I. Perlman, Revs. Mod. Phys. 20, 585 (1948).

ratios of the various radiations of the 40 day activity are:

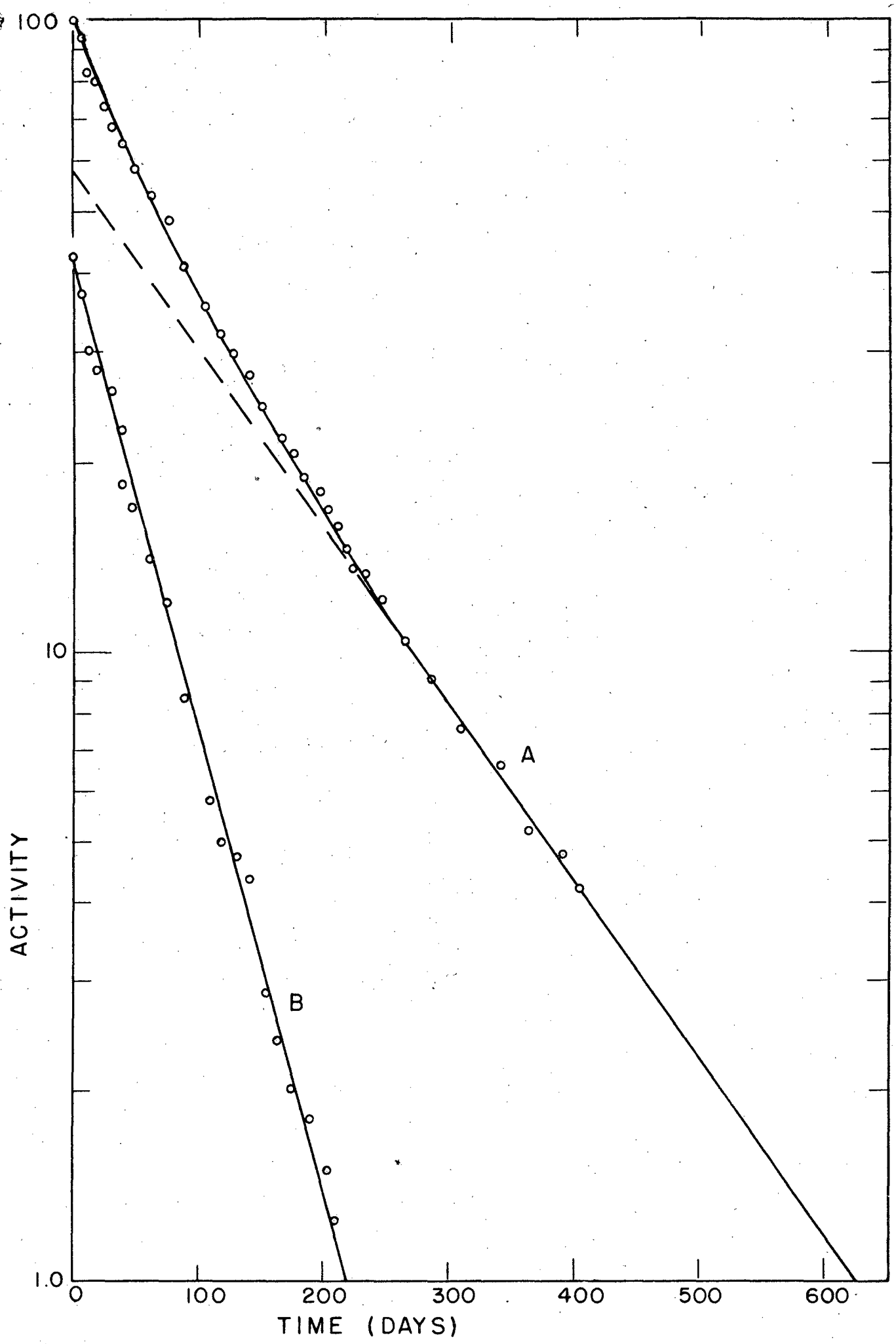
0.2 Mev e^-: 0.7 Mev e^-: L x-rays: K x-rays

0.35 : 0.004 : 0.35 : 1

In the rhenium fraction 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 α-particles from the 60 inch Crocker Laboratory cyclotron.

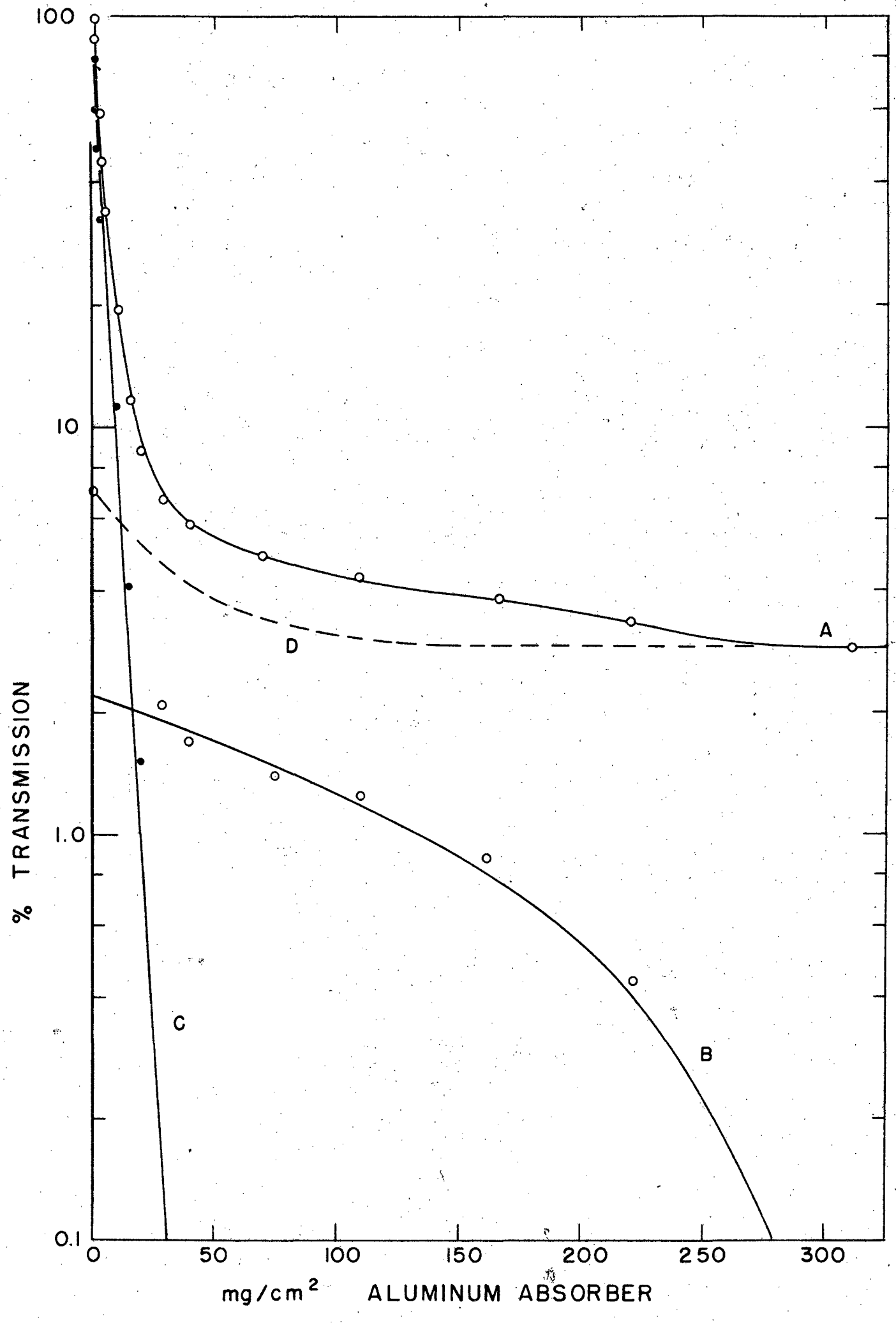
The aluminum absorption of an "infinitely thin" sample of the 120 day activity mounted on thin mica is shown in Fig. IX, and the lead absorption on a more active sample, in Fig. X. The radiations consist of electrons, total

Figure VII: Gross decay of 40 day Re^{184}(B) and ~120 day Re^{183}(A) activities from 10 Mev proton bombardment of tungsten.



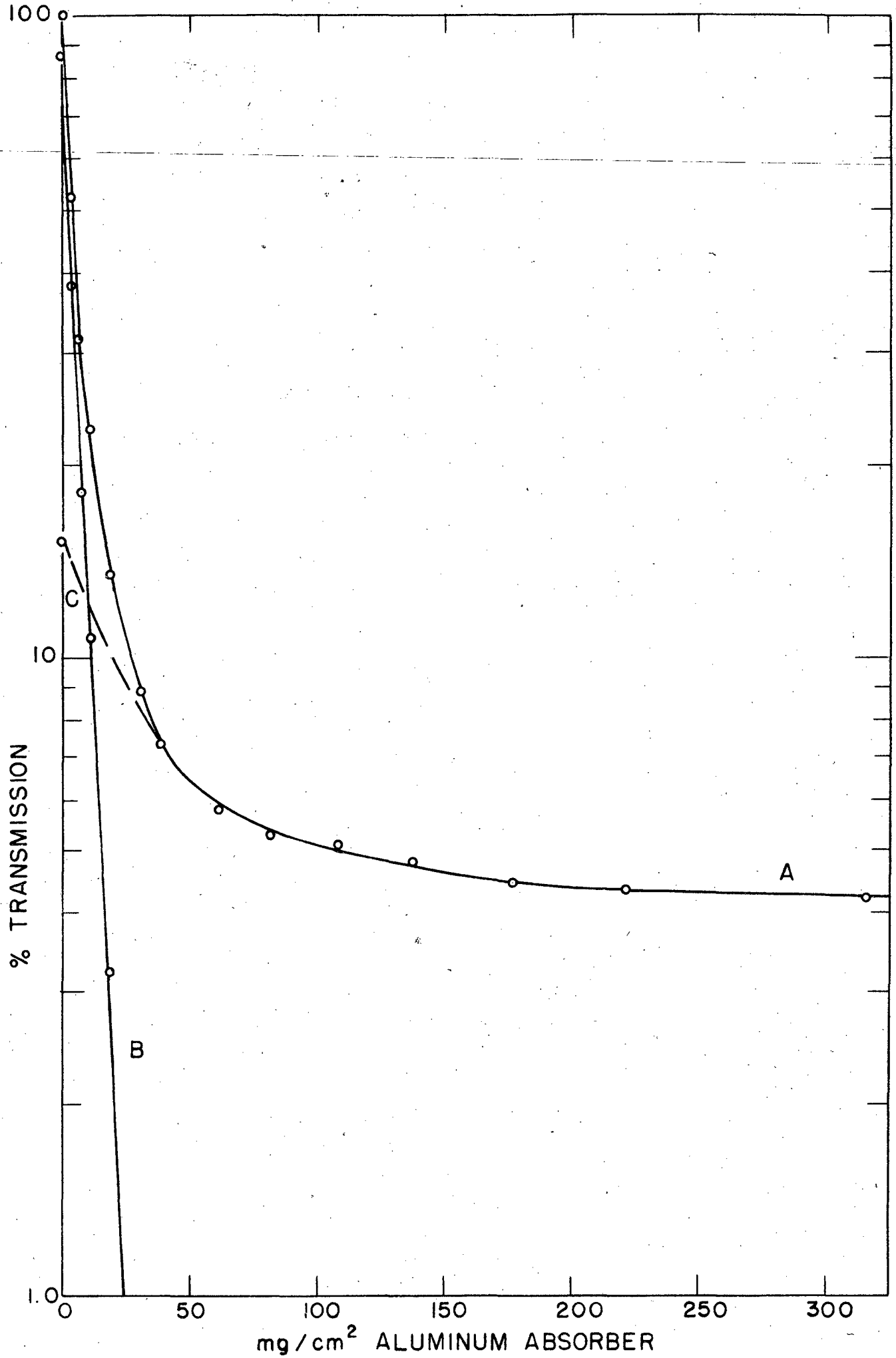
13462-1

Figure VIII: Aluminum absorption of 40 day Re^{184} activity; K x-rays and γ-rays (A), 0.7 Mev electron (B), 0.2 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

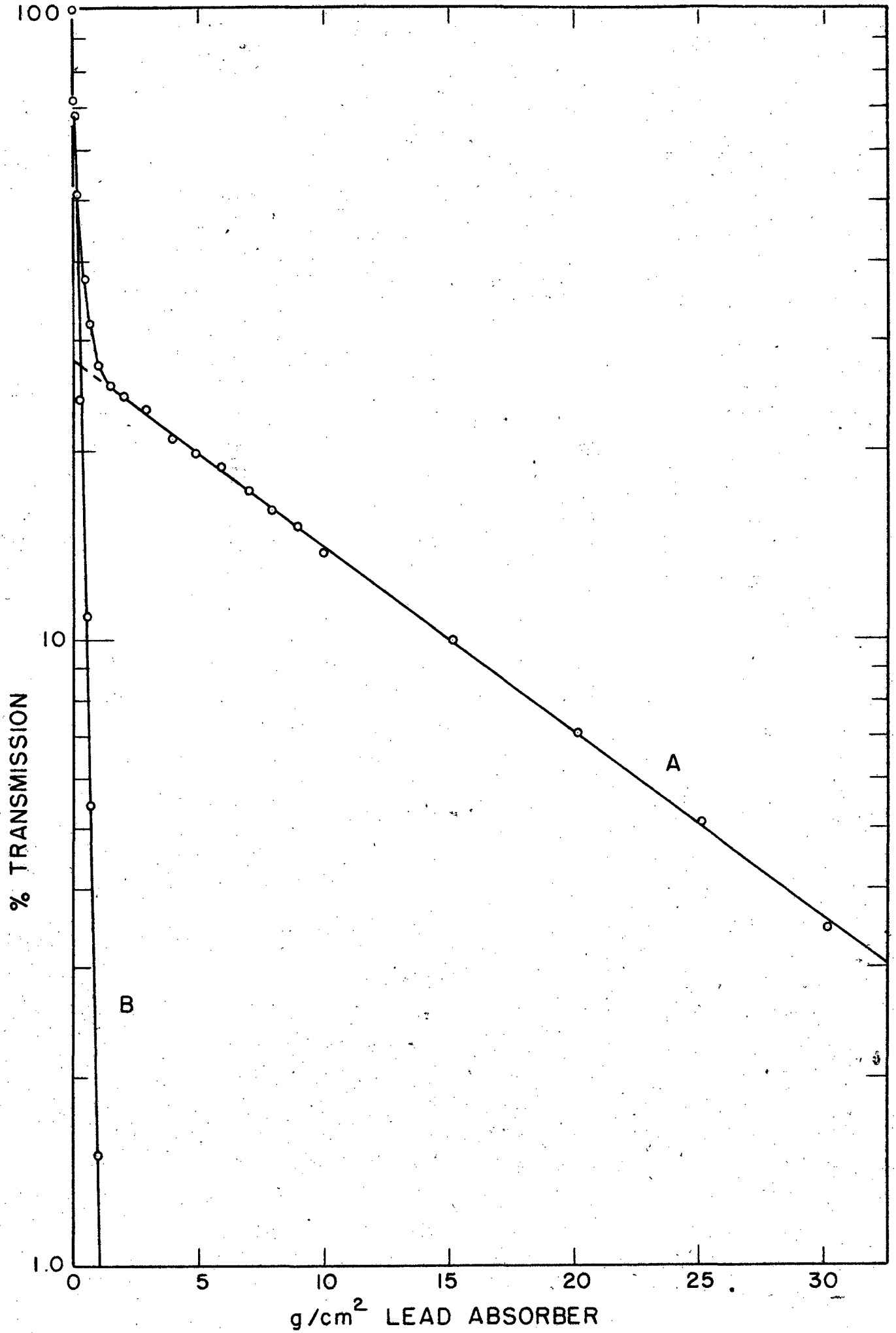
13878-1

Figure IX: Aluminum absorption of $\sim$ 120 day Re^{183} activity; K x-rays and γ-rays (A), 0.16 Mev electron (B), L x-rays (C).


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

13460

Figure X: Lead absorption of ~120 day Re^{183} activity; 1.0 Mev γ-ray (A), K x-rays (B).



13461-1

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 spectrograph 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 by the usual procedure.

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

The isotope thus 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 in the α-particle bombardments.

C. 2.2 day Re^{184}.

In the 19 Mev α-particle bombardment of tantalum, an activity of 2.2 days half-life has been observed in addition to the longer periods. The activity which was not observed in the higher energy bombardments because of the low yields and masking by other activities, is allocated to Re^{184} on the basis of its production by α,n reaction on tantalum. That the activity was not detected by other workers who studied the n,2n reactions in rhenium and p,n reactions in tungsten is not surprising in view of the similarity in half-life to the 98.2 hour Re^{186} which would effectively mask the shorter half-life.

The decay of the activity from Ta + α bombardment is shown in Fig. XI; the half-life is 2.2 $\pm$ 0.1 days measured through seven periods. The radiation characteristics were obtained by resolution of aluminum absorption (Fig. XII) and decay curves. The radiations consist of electrons of ranges 50 mg/cm^2 (0.2 Mev) and $\sim$450 mg/cm^2 aluminum (1.1 Mev), soft electromagnetic radiation of half-thickness 21 mg/cm^2 (9.4 Kev) and hard electromagnetic radiation. Lead absorptions were not taken because of insufficient activity. The following approximate ratios were calculated from the aluminum absorption:

Figure XI: Gross decay of 2.2 day Re^{184}(B) and 40 day Re^{184} activities from 19 Mev α-particle bombardment of tantalum.

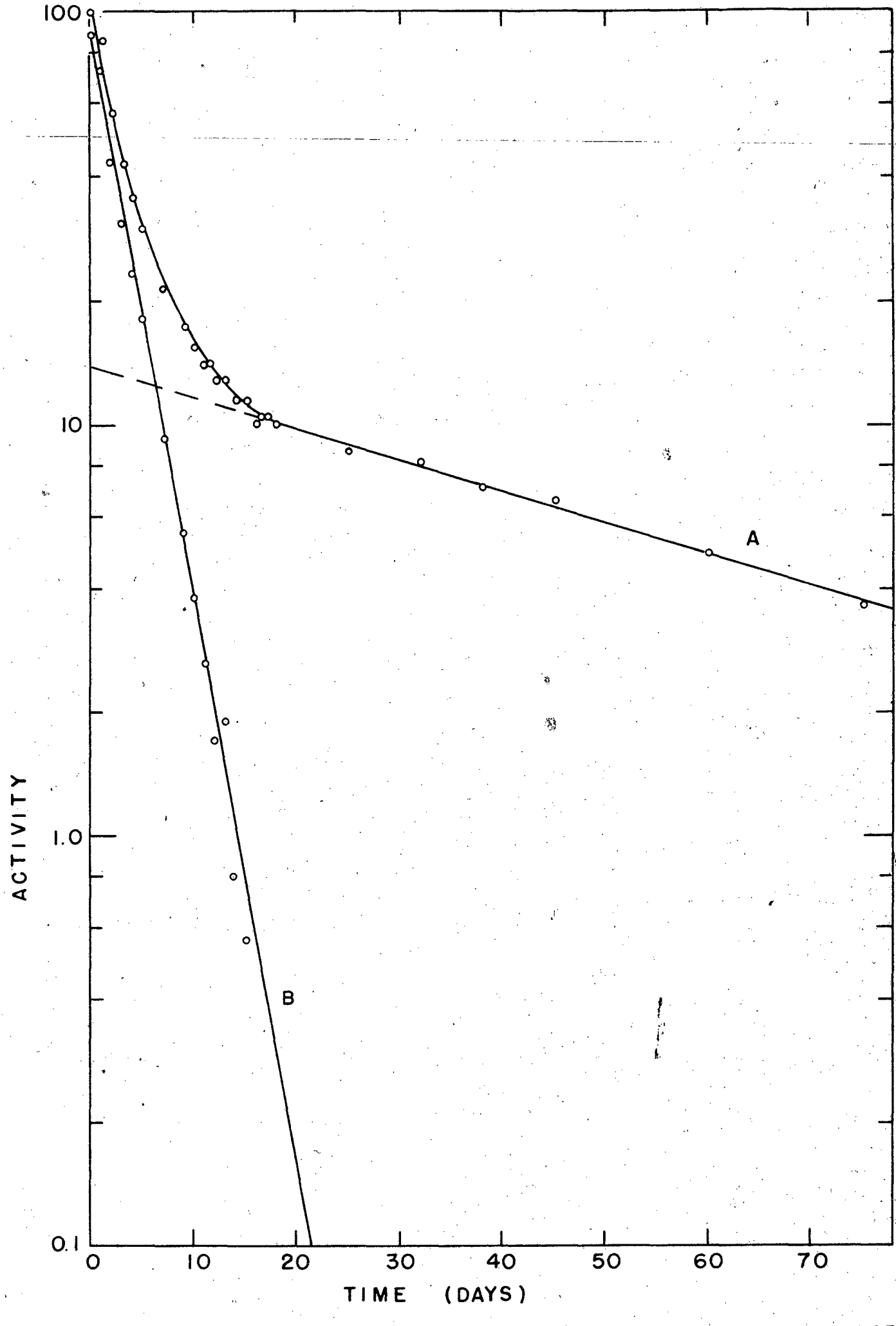
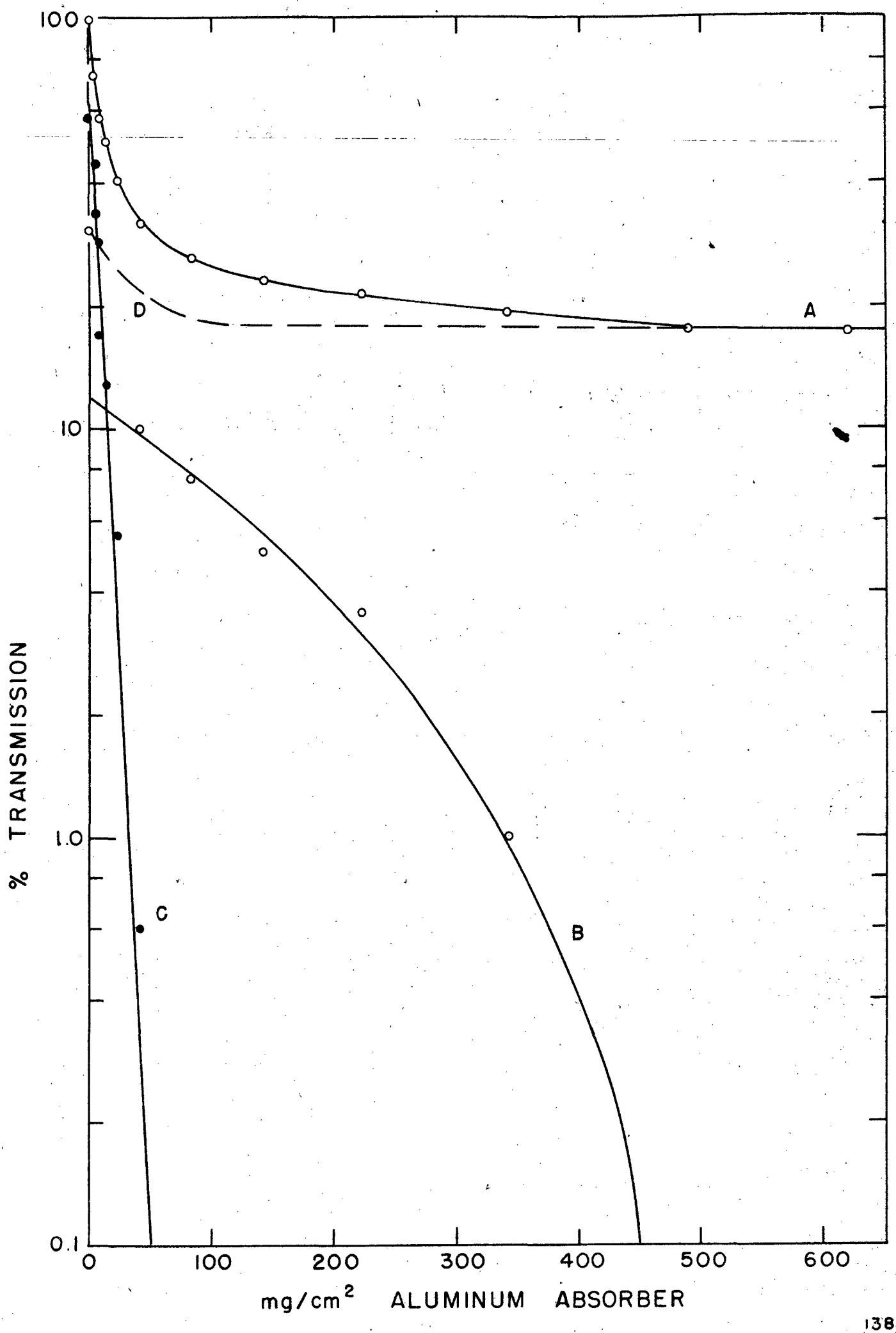

100
10
1.0
0.1
ACTIVITY
0
10
20
30
40
50
60
70
TIME (DAYS)
A
B
13879-1

Figure XII: Aluminum absorption of 2.2 day Re^{184} activity; K x-rays and γ-rays (A), 0.22 Mev electron (B), 1.1 Mev electron (C), L x-rays (D).


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

0.2 Mev e^-	1.1 Mev e^-	L x-rays	K + γ rays
0.02	0.003	0.09	1

III. Discussion.

In order to clarify the status of the five rhenium activities, very active samples were prepared for study on a magnetic deflection beta ray spectrometer, by Prof. A. C. Helmholz and Mr. R. W. Hayward. The 64.0 hour and 12.7 hour activities appear to have identical radiations and since the isotopes have been allocated to Re^{182}, it is very probable that they are independent isomers decaying by orbital-electron capture to excited or metastable states of W^{182}. No evidence of beta particle emission was found in any isotope and the suggestion of W. H Sullivan[4] that the 40 day Re^{184} decays at least partially, by beta emission is not confirmed.

The mass assignments have been made according to measurements on cross sections for production of the isotopes by Ta-α,xn reactions at various bombarding energies. In view of the complex radiations in each case, it is impossible to determine even a rough disintegration scheme by the techniques used, and consequently, the measured K x-radiation is taken as a measure of one disintegration by orbital electron capture. In Table II, the various yields are given relative to those of the 40 day isotope which is unequivocally allocated to mass 184 on the basis of its production by Ta-α,n and Re-n,2n reactions. The variations in yields of the isotopes at α-particle bombarding energies of 38, 30 and 19 Mev, then allow fairly certain mass allocations; the observed yield trends agree with those observed in bombardments of other elements[2].

The present allocations are in agreement with the formation of the rhenium activities by proton bombardment of tungsten.

Table II

Relative Yields of Rhenium Isotopes from
α-particle Bombardment of Tantalum

Activity	Energy of α-particles in Mev			Probable Reaction	Mass Assignment
	38	30	19		
12.7 hours	50	7	--	α,3n	182
64.0 hours	100	14	--	α,3n	182
~120 day	2	7	0.7	α,2n	183
40 day	1	1	1	α,n	184
2.2 day	masked	masked	24	α,n	184

V. Acknowledgments.

We wish to thank Professor J. G. Hamilton, Messrs. T. Putnam and B. Rossi, and the crew of the 60-inch Crocker Laboratory cyclotron for their cooperation and assistance in making bombardments. We are grateful to Professor A. C. Helmholz and Mr. R. W. Hayward for their helpful interest in making measurements on their beta-ray spectrometer. We are indebted to Professors G. T. Seaborg and I. Perlman for their continued encouragement and advice.

This work was performed under the auspices of the Atomic Energy Commission.