

# Lawrence Berkeley National Laboratory

## Recent Work

**Title**

ISOTOPES OF RUBIDIUM, POLONIUM, AND BISMUTH

**Permalink**

<https://escholarship.org/uc/item/9nj3d6hh>

**Author**

Karraker, D.G.

**Publication Date**

1950-04-04

UNIVERSITY OF  
CALIFORNIA

*Radiation  
Laboratory*

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy  
which may be borrowed for two weeks.  
For a personal retention copy, call  
Tech. Info. Division, Ext. 5545*

BERKELEY, CALIFORNIA

## **DISCLAIMER**

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

UCRL-640  
No Distribution

THESIS

Cy. 2

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

~~MR. 230~~

Contract No. W-7405-eng-48

**UNCLASSIFIED**

Isotopes of Rubidium, Polonium, and Bismuth

D. G. Karraker

April 4, 1950

Berkeley, California

<u>Declassification Distribution</u>	<u>No. of Copies</u>
Patent Department	1
Technical Library, US AEC, Washington	2
Area Manager, Berkeley	1
Chemistry Division	8
Information Division	<u>10</u>
Total	22

Radiation Laboratory  
University of California  
Berkeley, California

ISOTOPES OF RUBIDIUM, POLONIUM, AND BISMUTH

D. G. Karraker  
Radiation Laboratory and Department of Chemistry  
University of California, Berkeley, California

ABSTRACT

Neutron deficient radioactive isotopes of rubidium, polonium, bismuth, and lead have been prepared by bombardments utilizing protons, deuterons, and helium ions from the 184-inch and 60-inch Berkeley cyclotrons. The results of this research are summarized in the table below.

Isotope	Half-life	Type of Radiation	Energy in Mev		Produced By
			Particles	Gamma-Rays	
Rb <sup>84</sup>	34 days	$\beta^+$ , K, e <sup>-</sup>	1.53 ( $\beta^+$ ) 0.37 (e <sup>-</sup> )	0.85	Br-a-n
Rb <sup>83</sup>	107 days	K	---	---	Br-a-2n
Rb <sup>82</sup>	6.3 hr.	$\beta^+$ , K	0.670	1.2, ~0.7	Br-a-n
Rb <sup>81</sup>	4.7 hr.	$\beta^+$ , K	0.990	0.95	Br-a-2n
Pb <sup>198</sup>	~25 min.	K	---	---	Tl-p-6n
Po <sup>205</sup>	1.5 hr.	K, a (0.5%)	5.2		Bi-p-5n
Po <sup>204</sup>	3.8 hr.	K, a (0.1%)	5.37		Bi-p-6n
Po <sup>203</sup>	48 min.	K, a (1%)	5.62		Bi-p-7n
Po <sup>202</sup>	52 min.	K, a (0.01%)			Bi-p-8n
Bi <sup>205</sup>	14.5 days	K		0.431, 0.550, 0.527, 0.746, 1.84	Po <sup>205</sup> K-decay
Bi <sup>202</sup>	95 min.	K			Po <sup>202</sup> K-decay

## PREFACE

This thesis involves isotope research on two different problems - the neutron deficient isotopes of rubidium and the neutron deficient isotopes of polonium. The objects of the two problems are quite similar, but the methods used are substantially different. The isotopes of rubidium were investigated with the aid of a mass spectrograph and a beta-ray spectrometer, while the polonium isotopes were investigated using the more traditional method of successive separations of daughter activities. The bismuth and lead isotopes investigated in this work have been identified through experiments designed to establish decay products and mass assignments of the polonium isotopes.

The author is greatly indebted to Dr. D. H. Templeton for the suggestion of the problems and direction of his research. The work on the isotopes of rubidium was done with the able collaboration of Dr. F. L. Reynolds, and Mr. G. D. O'Kelley was of great assistance in the operation of the beta-ray spectrometer. The author is grateful to Mr. J. T. Vale and Mr. B. Rossi and the crews of the 184-inch and 60-inch cyclotrons for their cooperation in the bombardments. This work was done under the auspices of the U. S. Atomic Energy Commission.

## CHAPTER I

### RADIOACTIVE ISOTOPES OF RUBIDIUM

#### I. INTRODUCTION

An investigation of the cyclotron-produced isotopes of rubidium has been completed, utilizing the mass spectrographic techniques of mass assignment introduced by Lewis, Hayden, and their co-workers.<sup>1</sup> Rubidium isotopes of masses 81 and 82 have been mentioned previously,<sup>2</sup> although their radiation characteristics were imperfectly reported. Barber<sup>3</sup> has reported a 40-day positron-emitter at mass 84 which coincides with a 34-day positron-emitter observed in this work and assigned to mass 84 on the basis of mass spectrographic evidence. A new 107-day electron-capture activity has been identified in this work and assigned to mass 83, also through the use of the mass spectrograph.

#### II. EXPERIMENTAL

##### A. Chemical Separations

Rubidium activities were produced by bombardment of bromine (as ammonium or cuprous bromide) with helium ions in the Berkeley 60-inch and 184-inch cyclotrons. Two principal procedures were used to separate the rubidium from the target material. In the case of ammonium bromide targets, 20-30 micrograms of inactive rubidium were added to the dissolved target material, the solution was evaporated to dryness and the ammonium bromide target material sublimed off by strong heating. The rubidium activities remained behind, and were dissolved in a small volume of water. This solution was divided into two portions. The major portion was used for mass spectrographic determination of the mass number without further purification. The remainder was purified further by scavenging with silver chloride, strontium carbonate, ferric hydroxide,



and lead sulfide. This repurified portion was used for decay and absorption measurements.

Cuprous bromide targets were used for the production of rubidium activity for beta-ray spectrometer measurements. In this case, the target was dissolved in hot aqua regia, and about 0.5 milligrams of inactive rubidium carrier was added. The copper was removed by precipitation as the sulfide, and the solution was scavenged with lanthanum hydroxide and strontium carbonate precipitates. The remaining solution was evaporated to dryness and heated strongly to sublime off ammonium salts, leaving solid rubidium chloride behind.

#### B. Mass Spectrograph Techniques

For use in the mass spectrograph, the active rubidium chloride, with few micrograms stable rubidium carrier, was evaporated on a tungsten filament, which was mounted in the source assembly of the mass spectrograph. The rubidium was ionized by heating the filament with an electric current. The  $\text{Rb}^+$  ions, after acceleration by a voltage drop and resolution by a 60 degrees magnetic field, were collected on an Eastman III-0 spectrographic plate, with the stable rubidium serving as a mass marker. To locate the radioactive masses, a second plate was placed emulsion to emulsion with the original plate before developing. The presence of a radioactive mass was detected by a blackening of the transfer plate. The length of time allowed for the transfer plate to remain in contact with the original necessarily varied with the half-life of the isotope expected, and for these experiments ranged from a few hours to six weeks.

The location of activity on the plates was also established by the use of a Geiger counter provided with a slit. It was possible to follow the decay of individual activities on the original plate. The shielding of the slit was not sufficient to prevent the count at each radioactive mass from being influenced by nearby radioactive masses.

### C. Spectrometer Techniques

The beta-ray spectrometer used was a  $255^\circ$  double-focusing type, similar to that invented by Svartholm and Seigbahn,<sup>4</sup> but with a 25 cm. radius instead of the 12 cm. radius of their first such machine. Samples were mounted on  $1-4 \text{ mg/cm}^2$  mica backing with between 0.2 and 1 milligram of material in the sample. The backscattering from these samples was quite marked at low energies, but was not serious in the region of interest (above 300 kev).

For samples of low activity, rough measurements were taken using a crude  $180^\circ$  spectrometer with about a 4 cm. radius. Its use was necessary to determine the range of the  $\text{Rb}^{84}$  positrons, and it also served to distinguish the sign of the particles in preliminary work.

### D. Radioactive Measurements

Decay and absorption measurements were made with end-on Geiger-Müller counters, with a window thickness of  $\approx 4 \text{ mg/cm}^2$  mica. Aluminum absorption measurements were taken in the conventional manner, the ranges of particles taken at the minimum absorber thickness required for total absorption. After an aluminum absorption measurement, sufficient beryllium to absorb all particles present was interposed between the sample and the counter. An aluminum absorption measurement of electromagnetic radiation was then taken. Subtraction of the electromagnetic radiation from the total then gives the shape of the particle absorption curve.

Soft electromagnetic components were interpreted as K x-rays, and determined by extrapolation of harder radiation back to zero aluminum absorber and subtraction of the harder components from the aluminum absorption of electromagnetic radiation.

Lead absorption measurements were taken at low geometry, using counters without the conventional lead shield. Lead absorbers were placed between two

beryllium absorbers of sufficient thickness to absorb all secondary particles.

### III. MASS ASSIGNMENTS

On all bombardments of bromine by helium ions ranging from 18 to 100 Mev in energy, two main activities appeared in decay measurements - one of 5-6 hours in half-life and one of the order of 1-3 months. Isotopes at masses 81 and 82 were observed by the mass spectrographic transfer technique at bombardment energies of 40 to 100 Mev. These activities gave radioactive transfers in a few hours (Fig. 1) and direct measurement of their decay with the slit counter showed that both were of the order of 5-6 hours in half-life, with mass 82 being slightly longer than 81. Transfer plates, taken over periods of the order of a month, resulted in radioactive transfers for masses 83 and 84, as well as 81 and 82. Since masses 83 and 84 did not transfer in a few hours, they were known to be the long-lived activities.

Bombardment of bromine with 18-Mev helium ions gave radioactive transfers as masses 82 and 84 only showing the major activity produced in these bombardments was the  $\alpha, n$  product. Decay measurements on the activities produced in this bombardment showed the half-life of the short component (mass 82) to be 6.3 hours, corresponding to the 6.5-hr. activity previously identified by Hancock and Butler<sup>5</sup> (erroneously assigned to mass 84) and also showed a 34-day positron activity (Fig. 2) now assigned to mass 84, corresponding to the 40-day rubidium reported by Barber.<sup>3</sup>

Absorption and crude spectrometer measurements on the activities produced at low energy showed that  $\text{Rb}^{82}$  had virtually no conversion electrons, whereas a strong conversion electron line had been observed at higher energy bombardments. Further work (described below) established that this line was due to a 13-second  $\text{Kr}^{81}$  daughter of  $\text{Rb}^{81}$ . Decay of this electron line, followed in the double-focusing spectrometer, gave a half-life of 4.7 hours for the

Fig. 1. Densitometer tracings of mass spectrographic  
plates (A) original plate (B) transfer plate.

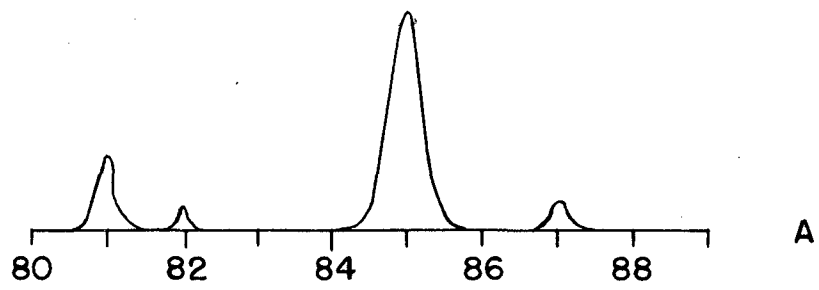
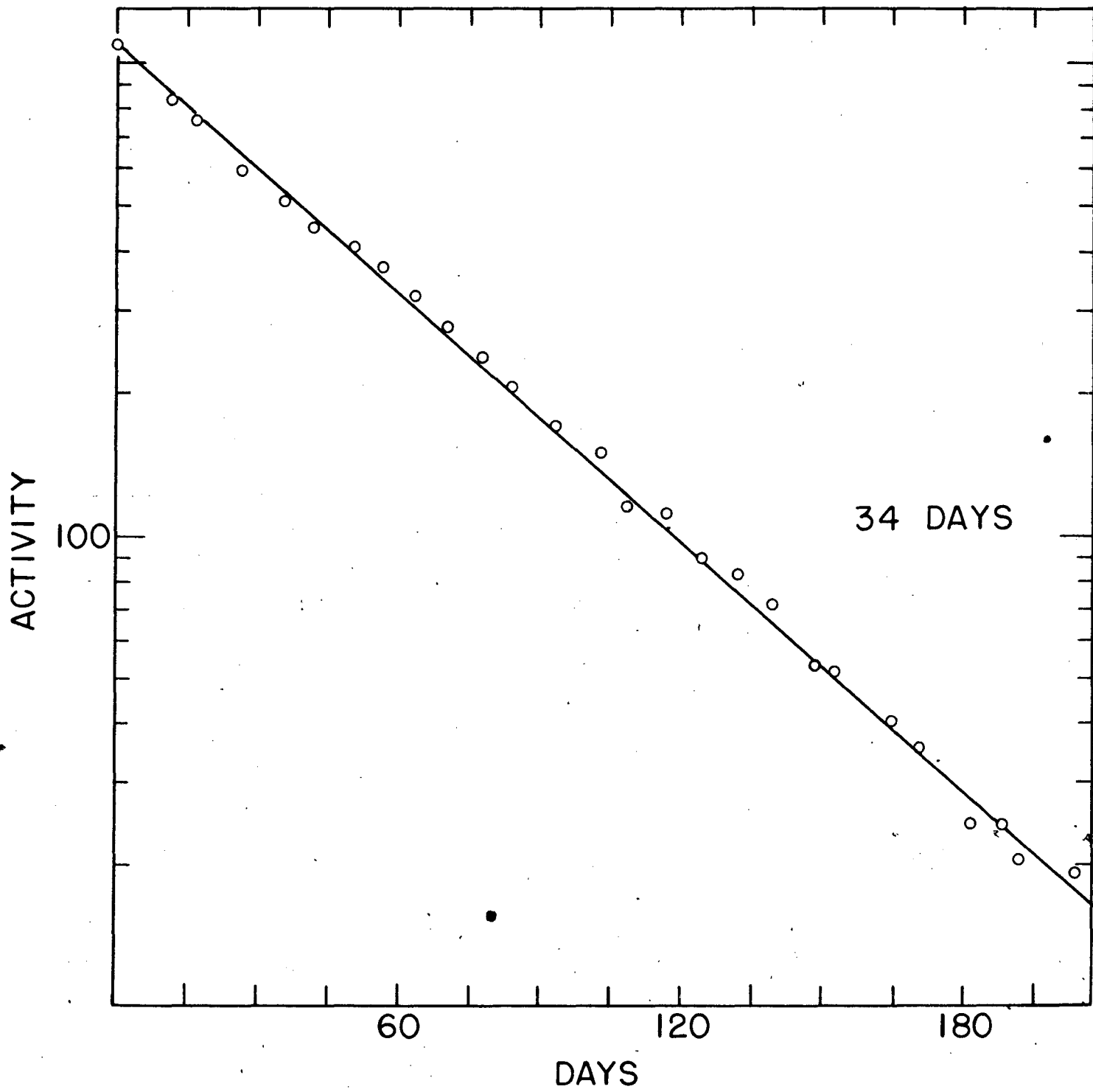


Fig. 2. Decay of 34-day Rb<sup>84</sup>.



half-life of  $\text{Rb}^{81}$  (Fig. 3). The half-life of  $\text{Rb}^{83}$  was determined by following the decay of a sample of rubidium activity produced by 60-Mev helium ion bombardment of bromine, very similar to the sample which gave a radioactive transfer at masses 83 and 84. Resolution of the decay curve showed a 107-day activity, assigned to mass 83, as well as the 34-day  $\text{Rb}^{84}$  (Fig. 4). A summary of the results is shown in Table I. Radiation characteristics of each isotope will be discussed individually.

Table I  
Rubidium Radioactivities

Mass	Half-life	Mode of Decay	Energy in Mev	
			Particles	Gamma-Rays
81	4.7 hr.	$\text{K}, \beta^+$	$\beta^+$ , 0.995	0.95
82	6.3 hr.	$\text{K}, \beta^+$	$\beta^+$ , 0.670	~0.7, 1.2
83	107 da	$\text{K} (?)$	---	---
84	34 da	$\text{K}, \beta^+$	$\beta^+$ , 1.5 $e^-$ , 0.37	0.85

#### IV. RADIATION CHARACTERISTICS

##### A. 34-day $\text{Rb}^{84}$

Measurement on a crude  $180^\circ$  spectrometer have shown that  $\text{Rb}^{84}$  has associated positron of 1.5-Mev energy, as determined by a Kurie plot of the spectrum (Fig. 5). A broad electron distribution centered about 0.37 Mev was also observed. From the width of the distribution, the electrons observed are certainly complex, and may contain contributions from the 107-day  $\text{Rb}^{83}$ . K x-rays have been identified by absorption in aluminum (Fig. 6) and a gamma-ray of 0.85 Mev (half-thickness, 8.5 grams of lead) has been identified by absorption in lead (Fig. 7). The small amount of  $\text{Rb}^{83}$  present has been



Fig. 3. Decay of electron line of  $\text{Rb}^{81}$ .

ACTIVITY

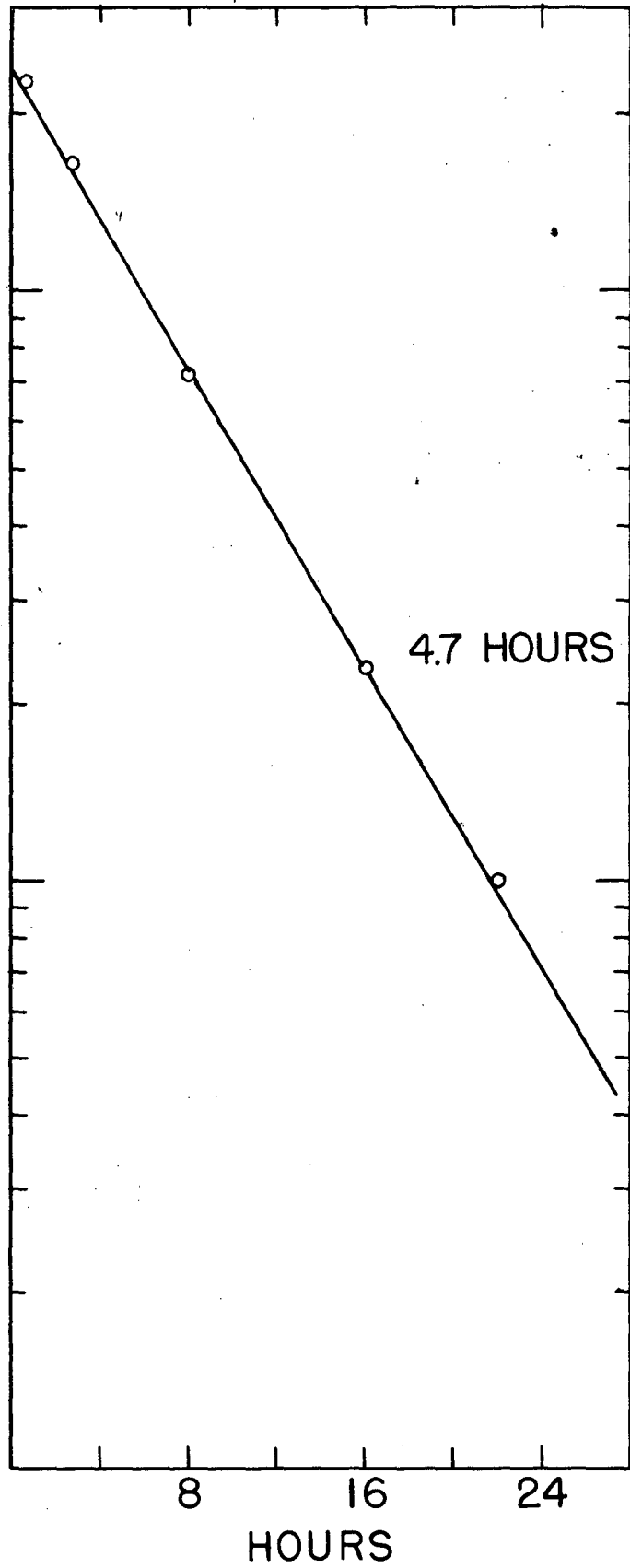


Fig. 4. Decay of long-lived rubidium activities  
(A) 34-day  $\text{Rb}^{84}$  (B) 107-day  $\text{Rb}^{83}$ .

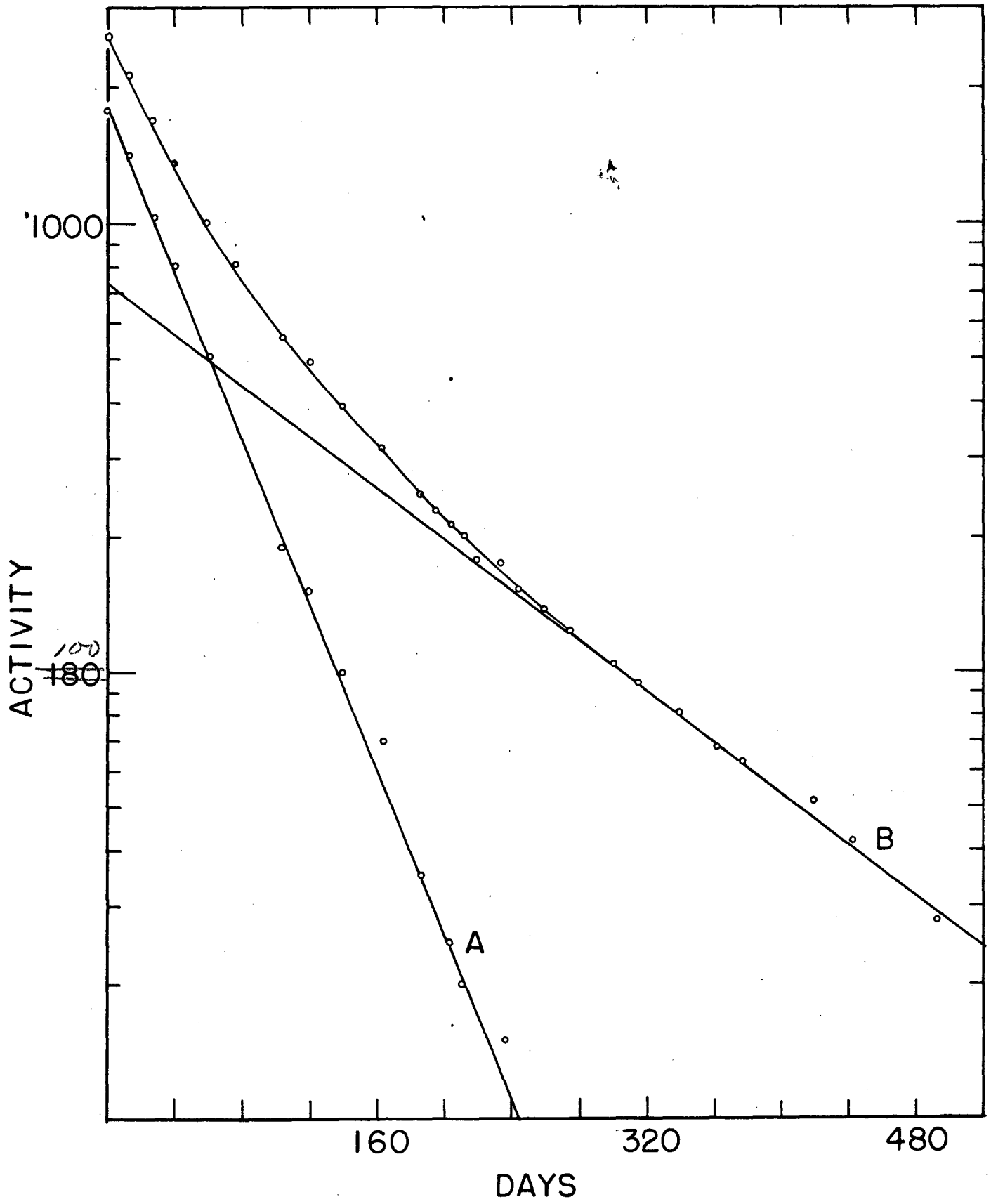


Fig. 5. Kurie plot of  $\text{Rb}^{84}$  positrons  
E max. = 1.53 Mev.

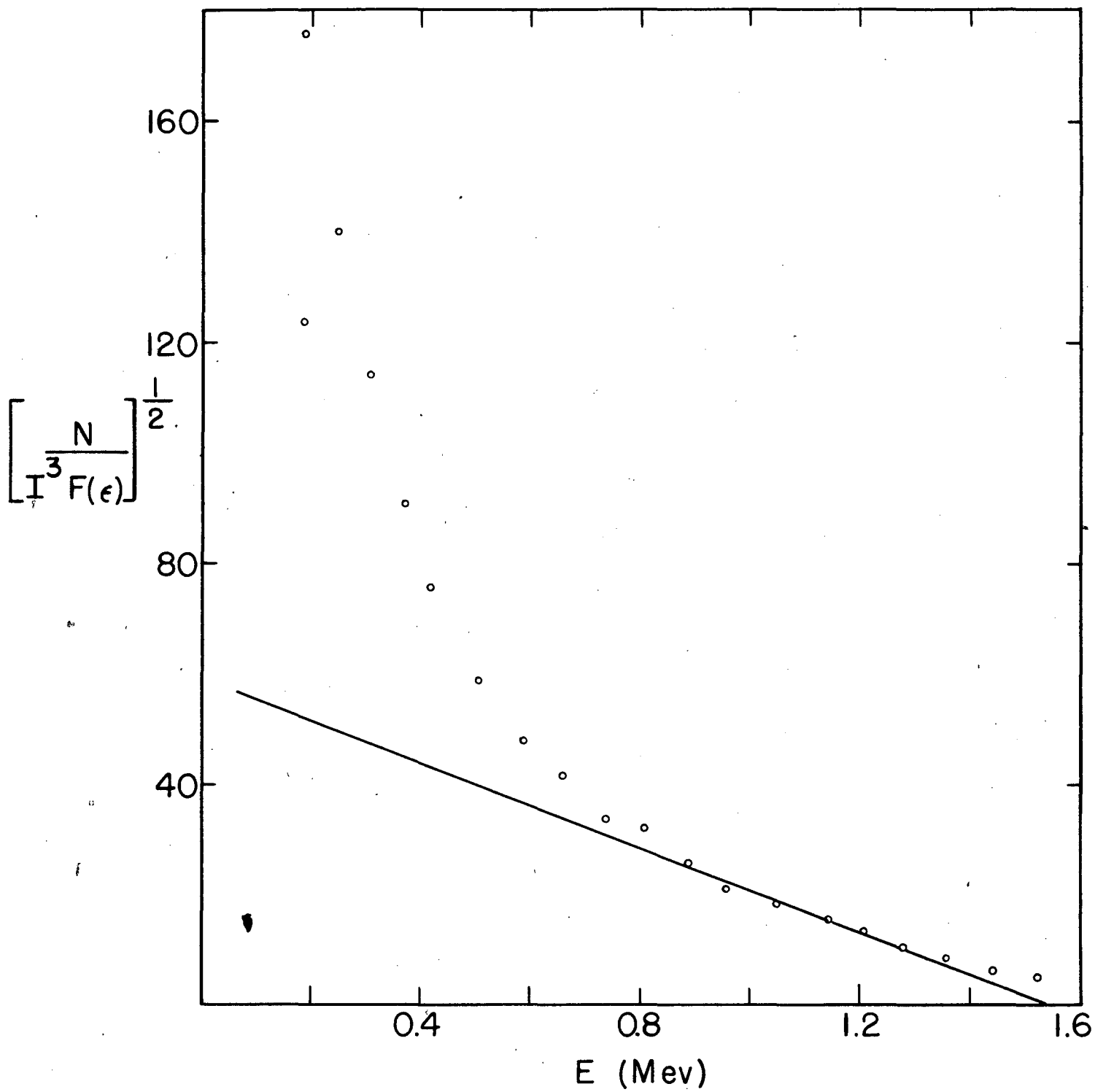


Fig. 6. Aluminum absorption of  $\text{Rb}^{84}$  (A) 1.5-Mev positrons  
(B) 0.37-Mev electrons (C) K x-rays and gamma-rays.

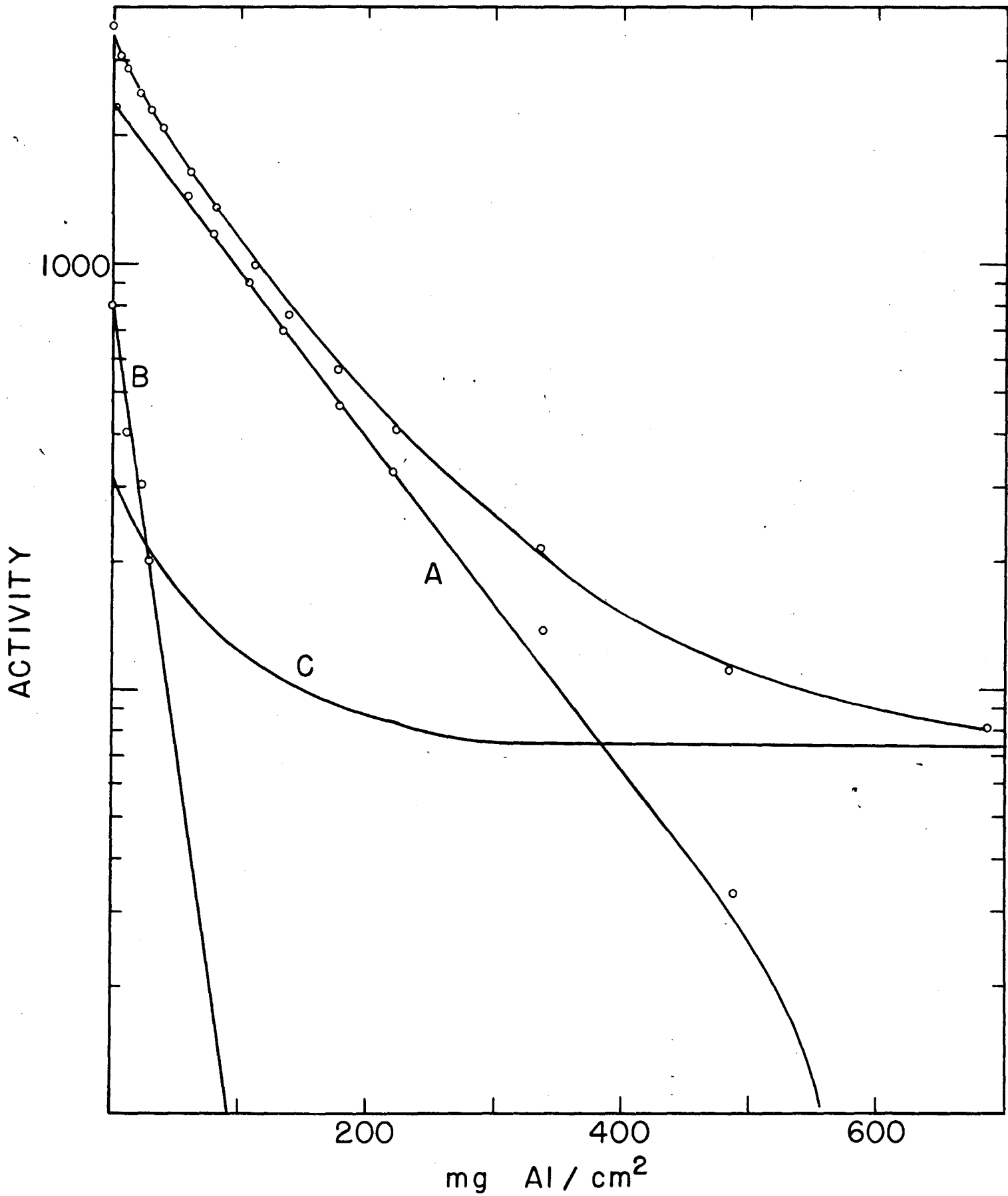
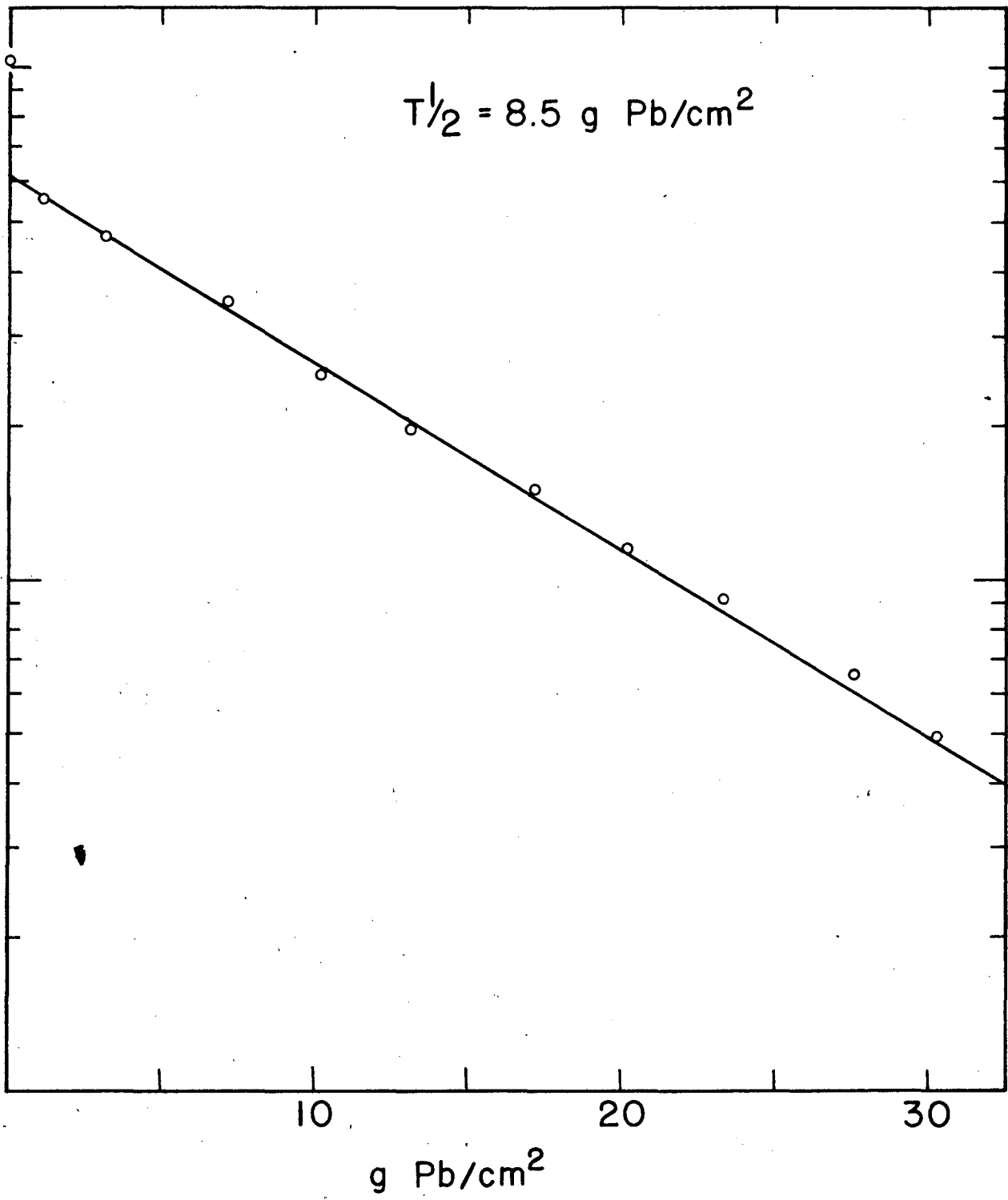




Fig. 7. Lead absorption of  $\text{Rb}^{84}$  gamma-ray  
E = 0.85 Mev.

ACTIVITY



assumed not to contribute to the absorption measurements, since the decay curve indicated fairly pure  $\text{Rb}^{84}$ . The ratio of  $\beta^+:\text{K}:\gamma$  was  $\sim 0.2:1:0.05$ , as estimated from the absorption curve, with the K x-rays assumed to count with 0.5 percent efficiency and the gamma-ray at 0.85 percent efficiency. From this, mode of decay is estimated to be about 85 percent by K-capture.

B. 107-day  $\text{Rb}^{83}$

The amounts of this activity available were so small measurement of its radiation characteristics were not attempted. The ratio of particles: K x-rays: gamma is very roughly 0.1:1:0.5; it is thus indicated that decay is to a large extent by electron capture. No positrons were detected, but the sensitivity for detection was poor.

Nothing can be said concerning the presence or absence of the 113-minute  $\text{Kr}^{83m}$  <sup>6</sup> as a daughter of this activity, as the amounts of  $\text{Rb}^{83}$  present were too small to make experiments designed to detect a krypton daughter practical.

C. 6.3 hr.  $\text{Rb}^{82}$

The radiations of  $\text{Rb}^{82}$  are subject to some uncertainty since this isotope always was mixed with a small amount of the 4.7-hour  $\text{Rb}^{81}$  when prepared for radiation measurements. The energy of the positrons is  $670 \pm 50$  kev, as determined by resolution of a Kurie plot of the mixed positrons of  $\text{Rb}^{82}$  and  $\text{Rb}^{81}$  (Fig. 8). Decay of the spectrum showed an enrichment of the longer-lived  $\text{Rb}^{82}$ .

Aluminum and lead absorption measurements were made on rubidium produced by 18-Mev helium ion bombardment of bromine, in which no transfer line for  $\text{Rb}^{81}$  was observed, and are supposed to be reasonably pure. Lead absorption (Fig. 9) shows two components, a strong gamma at 1.2 Mev (12 grams/cm<sup>2</sup> lead half-thickness) and an additional gamma at about 0.7 Mev in lower intensity. No prominent electron lines could be found associated with  $\text{Rb}^{82}$ .

Fig. 8. Kurie plot of the positrons of Rb<sup>81</sup> and  
Rb<sup>82</sup> (A) Rb<sup>81</sup> E max. = 2.92 ( $m_0c^2$ ) or 990 kev  
(B) Rb<sup>82</sup> E max. = 2.30 ( $m_0c^2$ ) or 670 kev.

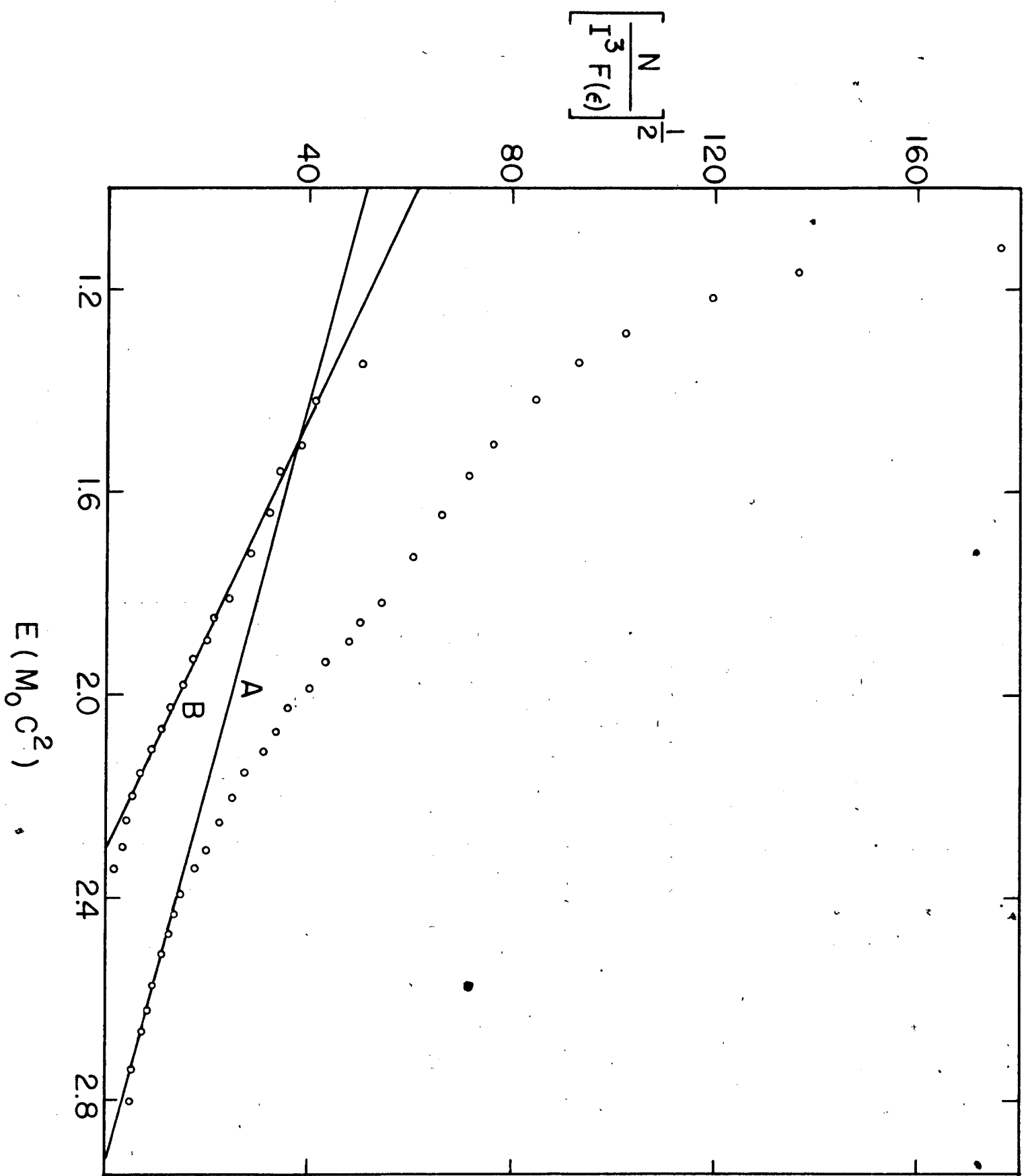
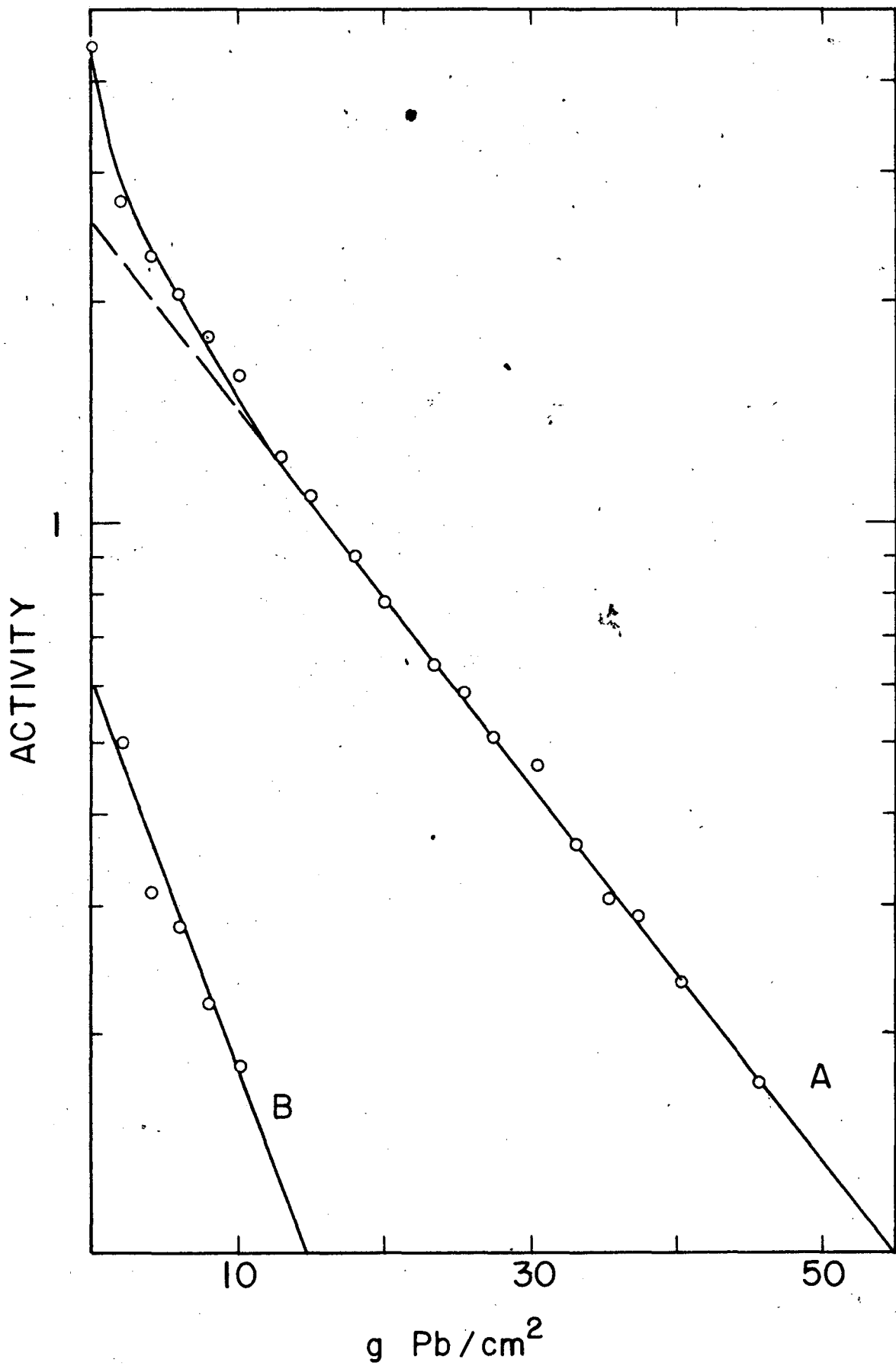


Fig. 9. Lead absorption of  $\text{Rb}^{82}$  gamma-rays

(A) 1.2 Mev ( $T_{1/2} = 12\text{g Pb/cm}^2$ )

(B) 0.7 Mev ( $T_{1/2} = 5.5\text{g Pb/cm}^2$ ).



On the basis of aluminum absorption measurements, (Fig. 10), the ratio of positrons to K x-rays to gammas is 0.1:0.7:1, suggesting that this isotope decays 80-90 percent by electron capture.

D. 4.7 hr. Rb<sup>81</sup>

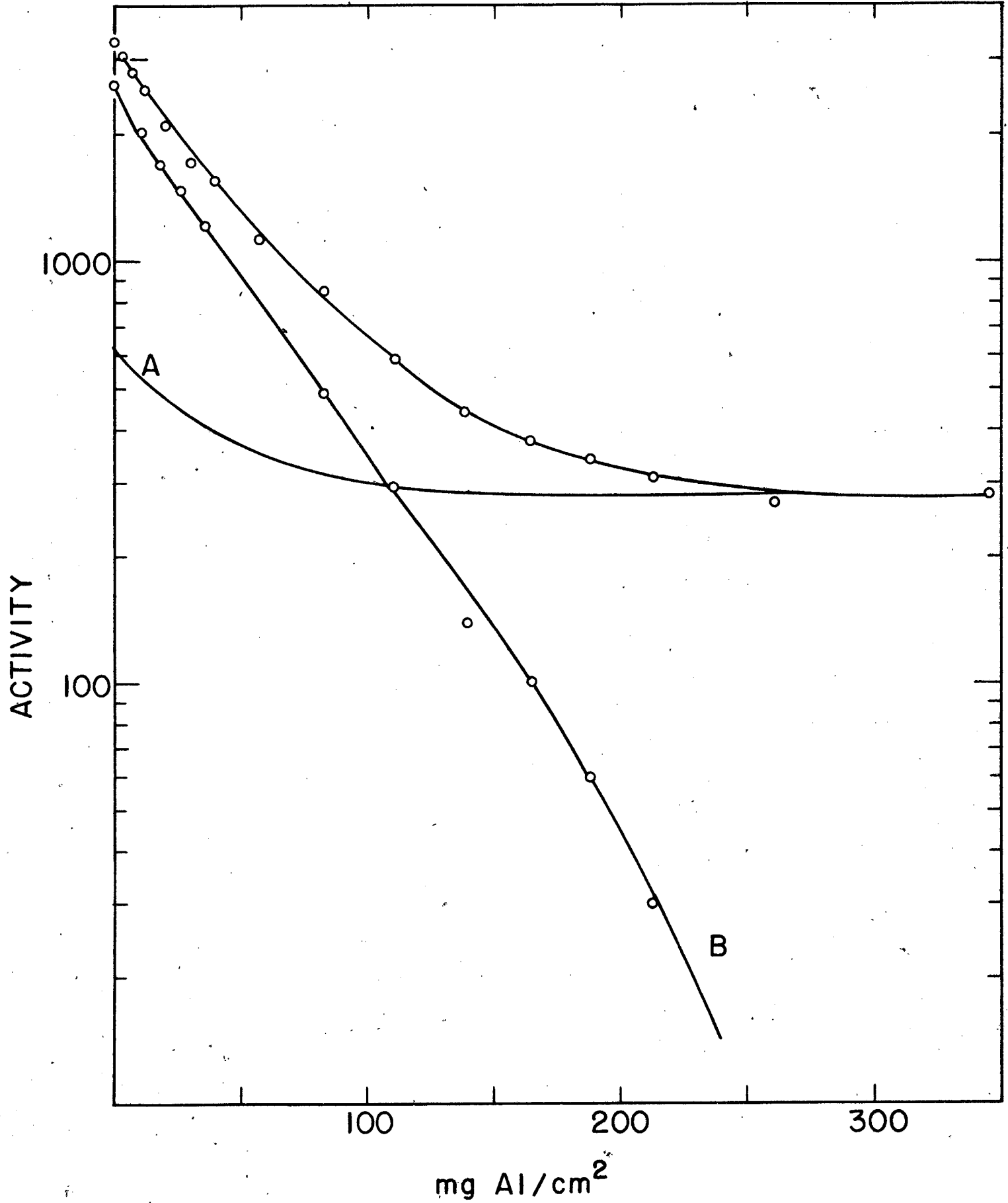
The radiations of Rb<sup>81</sup> are subject to the same uncertainties as those of Rb<sup>82</sup> and are open to the further error of being confused with its 13-sec. Kr<sup>81</sup> daughter.<sup>7</sup> Measurement of the positron and electron energies, originally thought to be associated with Rb<sup>81</sup>, in the double-focusing spectrometer gave  $990 \pm 50$  kev for the range of the positron (Fig. 8) and showed intense K and L conversion lines corresponding to a gamma-ray of  $193 \pm 10$  kev. The energy of this gamma-ray corresponds, within limits of error, to a gamma-ray of 187 kev found by Crentz, Barkas, and co-workers<sup>7</sup> to be associated with a 13-sec. krypton activity produced by bombardment of bromine with protons of low energy. By flaming a strong sample of Rb<sup>81</sup> to drive off the krypton, and observing of the growth of activity of the electron line in the crude spectrometer, Rb<sup>81</sup> was shown to have a krypton daughter with a half-life of  $10 \pm 6$  seconds.

In the same bombardments that Crentz, Barkas, and co-workers observed the 13-sec. Kr<sup>81</sup>, they also observed a 55-sec. krypton isomeric transition with a strong conversion electron corresponding to a gamma of 127 kev. These activities were produced by bombarding bromine (masses 79 and 81) with protons of 5-6 Mev energy, and both isotopes are thought to be products of p,n reactions. Since electrons from the 127-kev gamma were not observed by beta-ray spectrometer measurements, it is thought likely that the 55-sec. krypton lies at mass 79. If the 55-sec. krypton does lie at mass 81, the amount formed by decay of Rb<sup>81</sup> must be less than 5 percent of 13-sec. activity.



Fig. 10. Aluminum absorption of  $\text{Rb}^{82}$

- (A) K x-rays and gamma-rays
- (B) positrons, E max. = 670 kev.



Absorption data were taken on  $\text{Rb}^{81}$  samples prepared by bombardment of bromine with 100-Mev helium ions. A mass spectrographic transfer plate showed that the ratio of mass 81 to mass 82 was approximately 4. The data presented are corrected for the presence of the  $\text{Rb}^{82}$ .

Absorption in lead (Fig. 11) shows a gamma-ray of 0.95 Mev ( $9.5 \text{ g/cm}^2$  of lead half-thickness). Absorption in aluminum (Fig. 12) has given the positron to K x-ray to gamma ratio as 0.1:1:0.4, allowing an estimate of 90 percent decay by K-electron capture. Half of the x-rays observed were assumed to be due to the krypton daughter. The number of conversion electrons from the 13-second krypton daughter is approximately three times the number of positrons of the rubidium parent.

Fig. 11. Lead absorption of  $\text{Rb}^{81}$  gamma-ray  
E = 0.95 Mev.

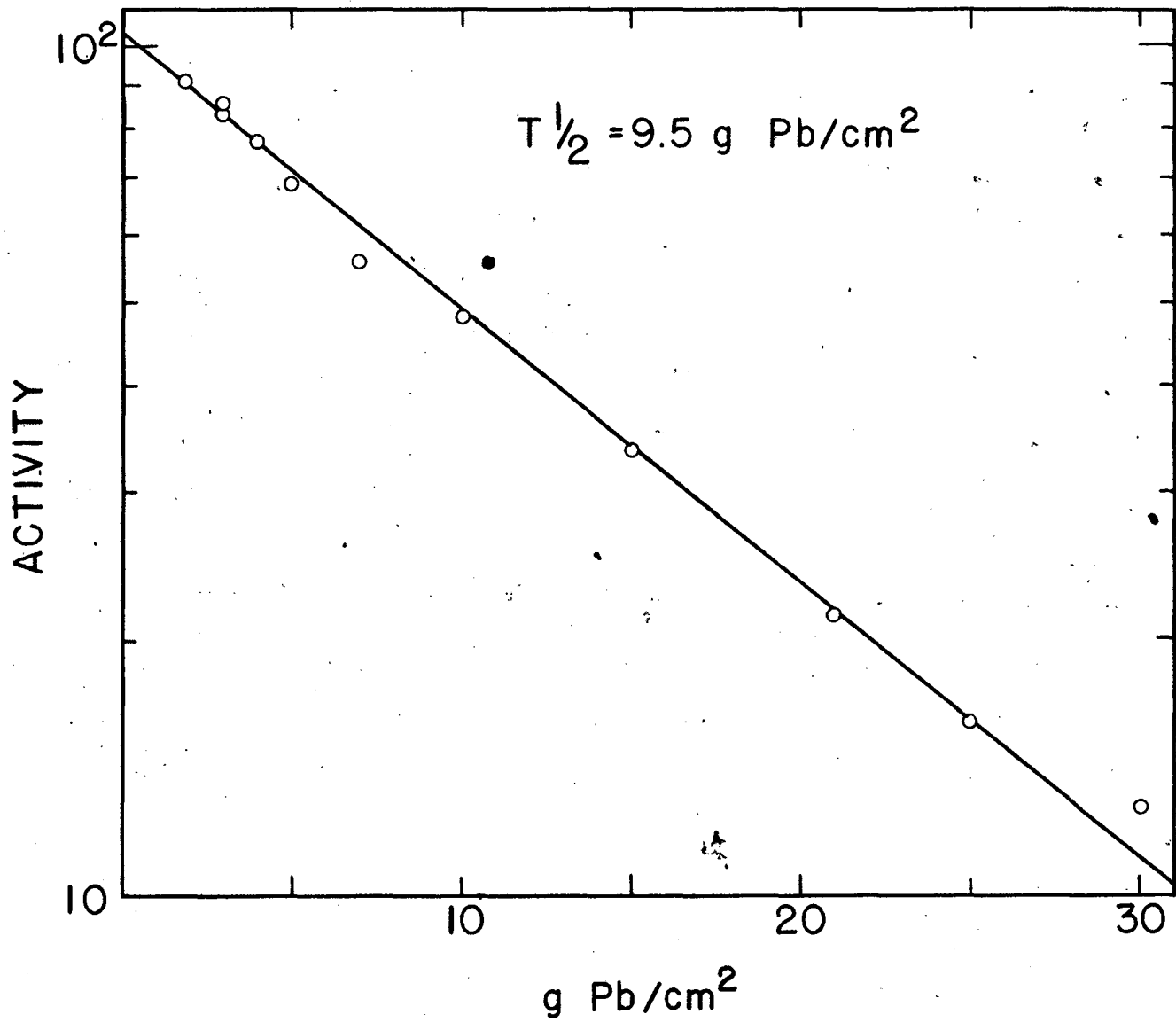
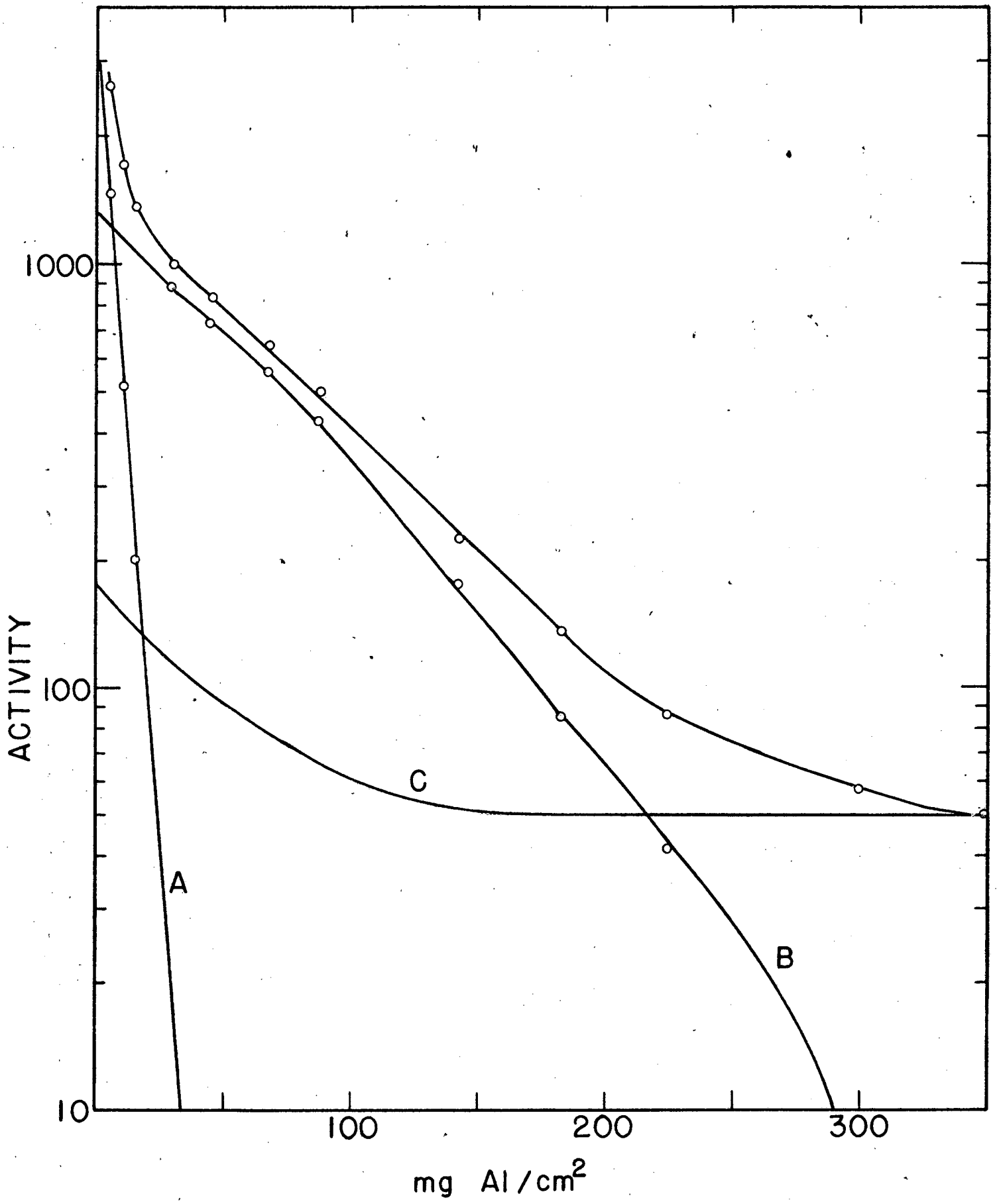


Fig. 12. Aluminum absorption of  $\text{Rb}^{81}$  and  $\text{Kr}^{81}$

- (A)  $\text{Kr}^{81}$  electrons  $E = 173 \text{ kev}$
- (B)  $\text{Rb}^{81}$  positrons  $E \text{ max.} = 990 \text{ kev}$
- (C) K x-rays and gamma-rays.



## CHAPTER II

## RADIOACTIVE ISOTOPES OF POLONIUM, BISMUTH, AND LEAD

## I. INTRODUCTION

Polonium has many isotopes which occur in the natural radioactive series, in the mass ranges of beta-stability and of neutron excess. Cyclotron-induced transmutations now make possible the production of neutron-deficient polonium isotopes. Templeton, Howland, and Perlman<sup>8</sup> have produced by this means the three isotopes  $\text{Po}^{206}$ ,  $\text{Po}^{207}$ , and  $\text{Po}^{208}$ , and Kelley and Segre<sup>9</sup> have found  $\text{Po}^{209}$ . The present work was undertaken to extend our knowledge of polonium to the even lighter isotopes whose production was made possible by the successful completion of the 184-inch Berkeley cyclotron. We have observed the radioactivities of the next four isotopes,  $\text{Po}^{205}$ ,  $\text{Po}^{204}$ ,  $\text{Po}^{203}$ , and  $\text{Po}^{202}$ . Experiments designed to establish the decay products and mass assignments of these radioactivities led to the discovery of two radioactive species of bismuth and one of lead. Our work was greatly aided by a parallel investigation of bismuth and lead isotopes carried out in this same laboratory by Neumann and Perlman, who have already reported some of their results.<sup>10</sup>

## II. EXPERIMENTAL METHODS

In most of the experiments a target of natural lead or bismuth was bombarded with particles accelerated in the 184-inch Berkeley cyclotron. Helium ions with lead, or protons or deuterons with bismuth produced good yields of the polonium isotopes of interest, but also substantial amounts of other spallation and fission products. Particles of various energies were used to improve the relative yield of the isotope desired in a particular experiment. The target was made of metal in the form of strips 0.5 to 2 mm. thick; or, when speed in chemical separation was required, bismuth oxide was used.



This oxide can be dissolved in acids much more rapidly than can the metals.

All samples were separated chemically before measurements were made. Polonium was separated from other elements by a procedure using tellurium, with hold-back carriers added for thallium and lead. The tellurium was reduced to the element with stannous chloride, carrying with it the polonium and noble metals. The tellurium was dissolved and precipitated with sulfur dioxide, carrying again the noble metals, but leaving a carrier-free solution of polonium, with 85-95 percent yield. For further purification, polonium was extracted from 6N hydrochloric acid into a mixture of 20 percent tributyl phosphate and 80 percent dibutyl ether. The extraction coefficient for polonium between the organic and acid layers is about 110. Lead and bismuth daughter activities were removed quantitatively by washing the organic layer with 6N hydrochloric acid, and purified by precipitation - bismuth as  $\text{BiOCl}$  and lead as  $\text{PbSO}_4$  or  $\text{PbCrO}_4$ .

Thallium activities were separated by oxidation of thallium to the thallic state with potassium permanganate, and the extraction of thallic chloride with diisopropyl ether saturated with hydrochloric acid. Occasionally the thallium activities were further purified by evaporation of the ether, reduction of the thallium with hydrogen peroxide, and precipitation of the thallium as  $\text{Tl}_2\text{PtCl}_6$  in the presence of lead and bismuth hold-back carriers.

Generally, the decay curve of the polonium Geiger activities is so complex that resolution is ambiguous. The approach most frequently used involves the separation of the daughter activities at equal time intervals, the specific time interval chosen by consideration of the approximate half-life of the parent. The activity of the daughter at a time  $t$  is given by:

$$A_2 = C_2 \frac{\lambda_2 \lambda_1 N_0}{\lambda_2 - \lambda_1} \left[ e^{-\lambda_1 t} - e^{-\lambda_2 t} \right]$$

where  $A_2$  is counting rate of daughter;  $C_2$  is counting efficiency of daughter;  $\lambda_2$  is the decay constant of daughter;  $\lambda_1$  is the decay constant of parent; and

No is the number of atoms of parent at  $t = 0$ . At the start of the time interval, the parent is purified chemically so the additional term in the general formula does not enter into the expression. From this expression it will be observed that if the time  $t$  is the same for all growth periods of the daughter before separation from the parent, the exponential terms become a constant factor, and  $A_2$  is proportional to the activity ( $\lambda_1 N_0$ ) of the parent at the beginning of the growth period. Thus, the slope of the activity of the daughter vs. time of separation will give the half-life of the parent.

In the identification of alpha-decay daughters, the thallium electron-capture daughter of the lead alpha-decay daughter was usually separated for measurement, rather than the lead activity itself. This procedure was followed since a greater degree of purity was attained in the thallium separations than in the lead separations; furthermore, the decay curve of the thallium activities is simpler than the decay curve of the lead activities, since no similar half-lives are found and no daughters are growing.

Isotopes of interest in this paper are shown in Table II, where isotopes enclosed in parentheses are isotopes identified by this work, and stable isotopes are shaded.

Table II<sup>6,10</sup>

	198		200		202	203	204	205	206		208
Po					(52m EC, a)	(48m EC, a)	(3.8h EC, a)	(1.5h EC, a)	9d a, EC	5.7h a, EC	3y a
Bi					(95m EC)	12h EC	12h EC	(14d EC)	6.4d EC		
Pb	(~25m EC)	~80m EC	18h EC	8h EC	long	52 hr EC	68m IT				
Tl	1.8h EC	7.3h EC	27h EC	72h EC	~12d EC						
Hg											

## III. RESULTS

A. 3.8-hr. Po<sup>204</sup>

The alpha-decay of polonium produced by high energy bombardment shows periods of about 45-min., 1.5-hrs., and 4-hrs. half-life, in addition to longer periods previously identified. To determine the mass assignment of these isotopes, the method sketched above was used. A pure sample of mixed polonium activities was separated from the bombarded target (elapsed time, 2 hours) and the daughter activities separated at intervals of an hour. The decay of the purified bismuth fraction showed a 95-minute period (which will be discussed later), a 12-hour period, and a longer period of 6 to 14 days, in low intensity. Fig. 13 is a plot of the decay of the bismuth fractions for three of the separated samples where the 12-hr. period has been extrapolated back to the time of separation. (Only three of the bismuth decay curves are shown, to avoid crowding. Extrapolated points from the decay curves of the other fractions are plotted at the time of separation.) It will be seen from Fig. 13 that the yields at the time of separation determine the half-life of the parent activity of the 12-hr. bismuth as 3.8 hrs., in agreement with the 3.8-hr. value obtained from alpha-decay curves.

It will be seen from Table II that the establishment of a 12-hr. bismuth activity as the daughter of the 4-hr. polonium does not give an unambiguous mass number. A similar experiment was done to identify the alpha-decay daughter and fix the mass number. As before, a large sample of purified polonium activities was prepared, from which the bismuth and lead daughter activities were separated at 4-hr. intervals. The bismuth activity could be ignored, as none of the bismuth daughters are alpha-emitters, and Po<sup>200</sup> is presumed too short to produce any of the Tl<sup>200</sup> through an electron-capture chain. The lead alpha-daughter was allowed to grow its thallium daughter for about 18 hours, then the thallium was separated. Since the lead fraction had been separated at 4-hr. intervals,

Fig. 13. Data showing the genetic relationship  
of 3.8-hr.  $\text{Po}^{204}$  and 12-hr.  $\text{Bi}^{204}$ .

ACTIVITY

1000

3.8 HR

6

12

18

24

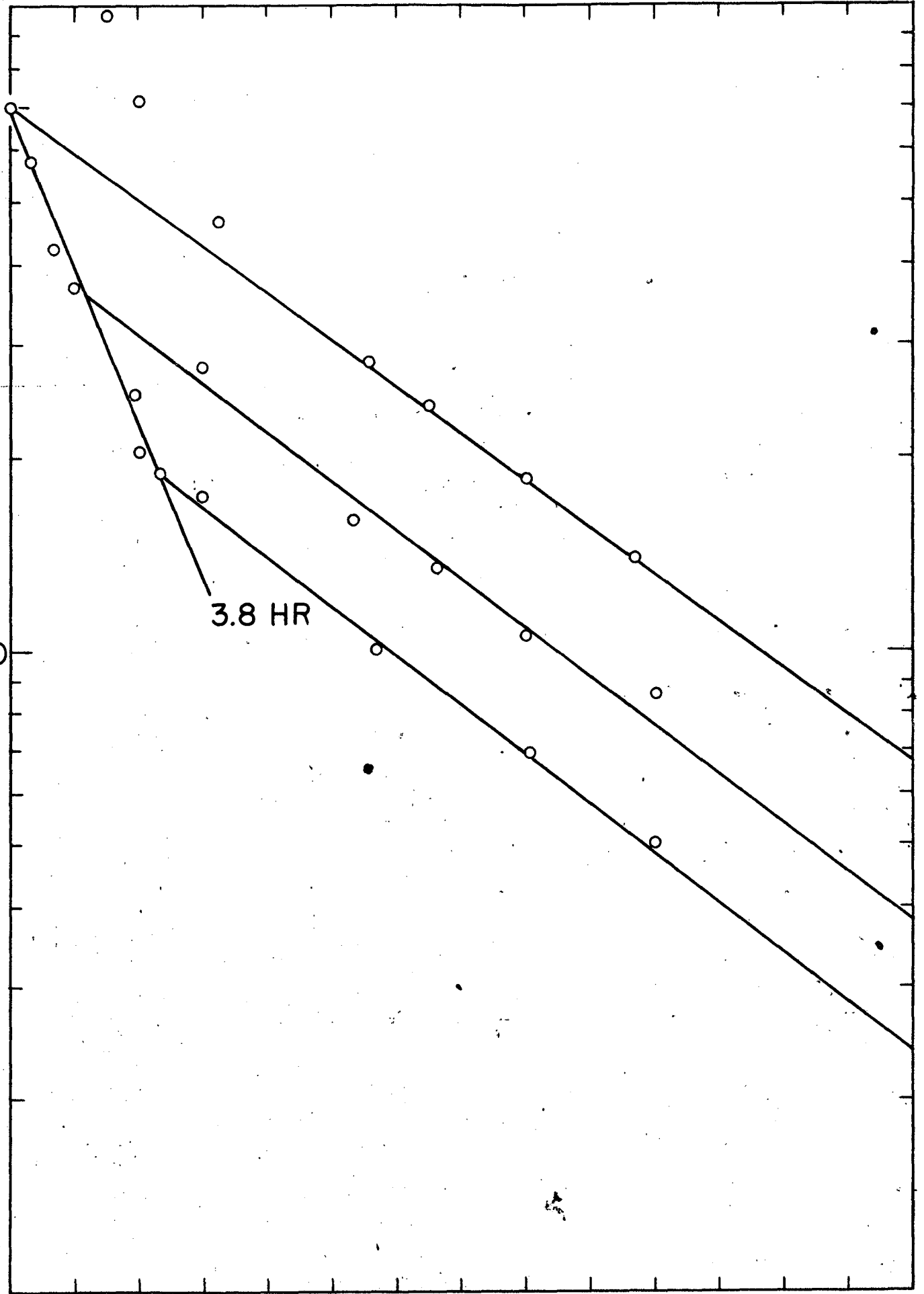
30

36

42

HOURS

14705-1



the activity of the alpha-decay daughter of the 3.8-hr. polonium would be decreased by approximately a factor of 2 between each of the fractions. The time allowed for the thallium to grow was the same for each fraction, so the activity of the lead alpha-daughter will be directly proportional to the activity of its thallium daughter. The principle, and virtually the only activity found was the 27-hr. Tl<sup>200</sup>. The results are shown in Table III.

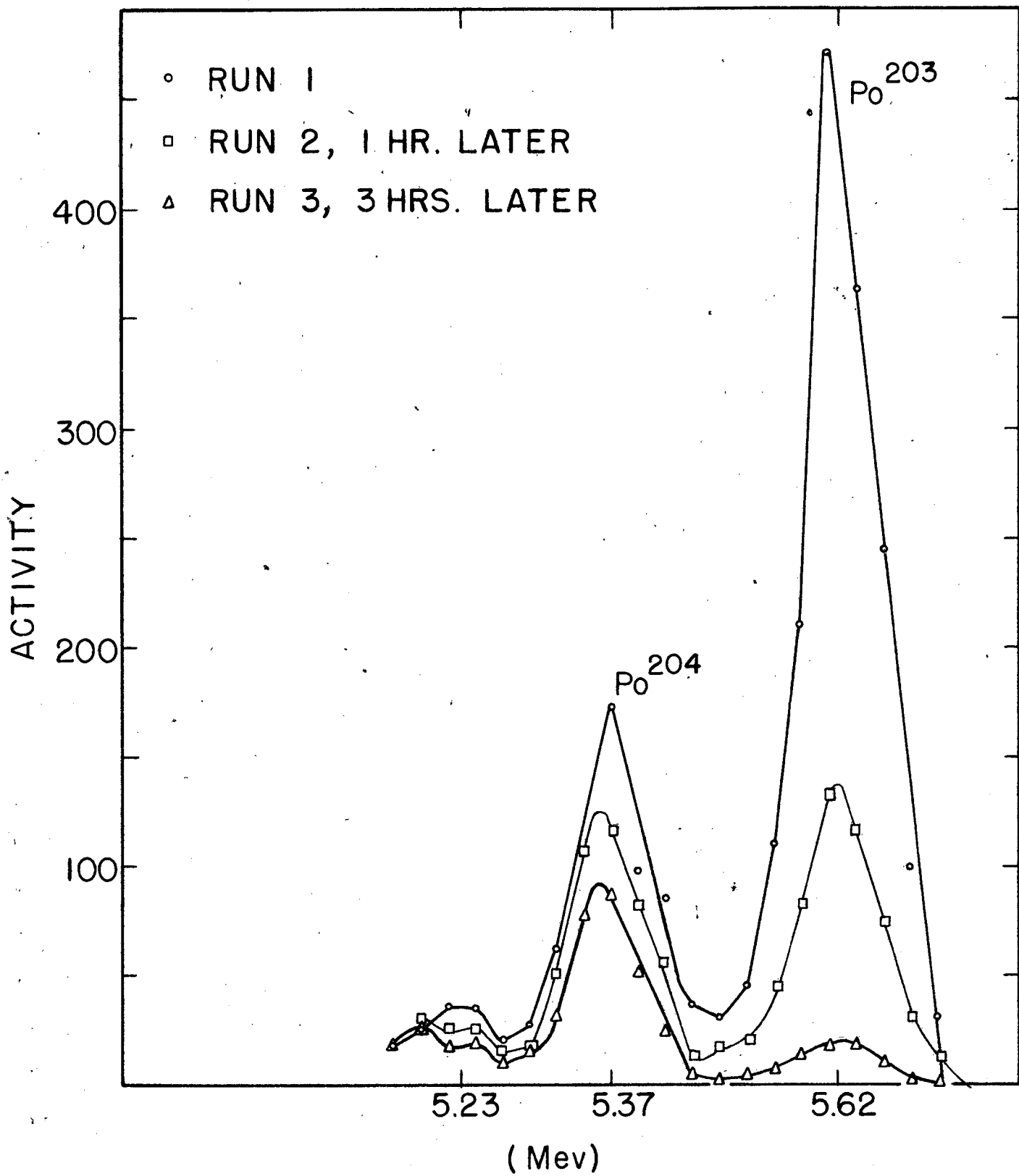
Table III

Fraction	27-hr. Tl A in c/m	Calc. (on basis of 3.8 hr. half-life)
1	3280	---
2	1740	1600
3	845	780

This experiment has fixed the 4-hr. polonium at mass 204 and shown its alpha-decay daughter to be the 18-hr. Pb<sup>200</sup> whose genetic relation to the 27-hr. Tl<sup>200</sup> has been shown by Neumann and Perlman.<sup>10</sup> All of these assignments depend on that of the 27-hr. thallium which was assigned to mass 200 by means of the excitation curve for its production by alpha irradiation of gold.<sup>11</sup>

The energy of the alpha-particles from Po<sup>204</sup> is  $5.37 \pm 0.05$  Mev, as determined by a pulse analyzer<sup>12</sup> (Figure 14). No attempt was made to determine the other radiation characteristics of the isotope because of the large amounts of other activities present in the samples. The ratio of electron-capture disintegrations to alpha-disintegrations is estimated as 1800 by counting the alpha-particles to determine the alpha-disintegrations and counting the 1.2-Mev gamma rays of the separated 12-hr. bismuth daughter. For this calculation the 12-hr. Bi<sup>204</sup> was assumed to emit one hard gamma per disintegration.

Fig. 14. Pulse analysis showing alpha-particle energies for polonium isotopes.





B. 1.5-hr. Po<sup>205</sup>

A 1.5-hr. electron-capture activity with associated alpha-particles was resolved from the decay curve of polonium produced by bombarding lead enriched in mass 204 (27 percent)<sup>13</sup> with 37-Mev helium ions in the 60-inch Crocker Laboratory cyclotron (Figure 15). The predominant product of 27-Mev helium ion bombardment of an element in this region is known to be the  $\alpha, 3n$  product,<sup>9</sup> which in this case is Po<sup>205</sup>. No lighter polonium should be produced in this irradiation, and since the heavier ones are already known, Po<sup>205</sup> is the best assignment.

Periodic separations of the bismuth daughters from a large amount of mixed polonium activities at 1.5-hour intervals showed that the yield of longer bismuth periods (i.e., half-lives of the order of a week) indicated a parent with a half-life of 3 to 6 hrs. Since the 9-da Po<sup>206</sup> is known to grow the 6.4-da Bi<sup>206</sup>, this clearly indicated a second polonium isotope of short half-life growing a longer bismuth. The decay of the bismuth activities showed that the first separation yielded principally a new activity of 14-days half-life, while the last fractions were almost pure 6.4-da Bi<sup>206</sup>. After the decay of the Bi<sup>206</sup>, the yield of the 14-da bismuth corresponded to a parent of 1.5 hours (Figure 16). This is the half-life of Po<sup>205</sup>, thus the new bismuth activity is also mass 205. Its radiation characteristics will be described presently.

The alpha-decay daughter of Po<sup>205</sup> was identified in a manner similar to that of Po<sup>204</sup>. A large quantity of polonium activity was prepared, purified, and the lead alpha-decay daughter activity separated at 1.5-hr. intervals. After 16 hours, the thallium daughters of the lead were separated from each sample. The thallium samples were exclusively the 72-hr. Tl<sup>201</sup>, since none of the lighter polonium isotopes were produced in this particular bombardment. The results are shown in Table IV.

Fig. 15. Geiger decay curve of polonium

- (A) 5.7-hr.  $\text{Po}^{207}$
- (B) 1.5-hr.  $\text{Po}^{205}$ .

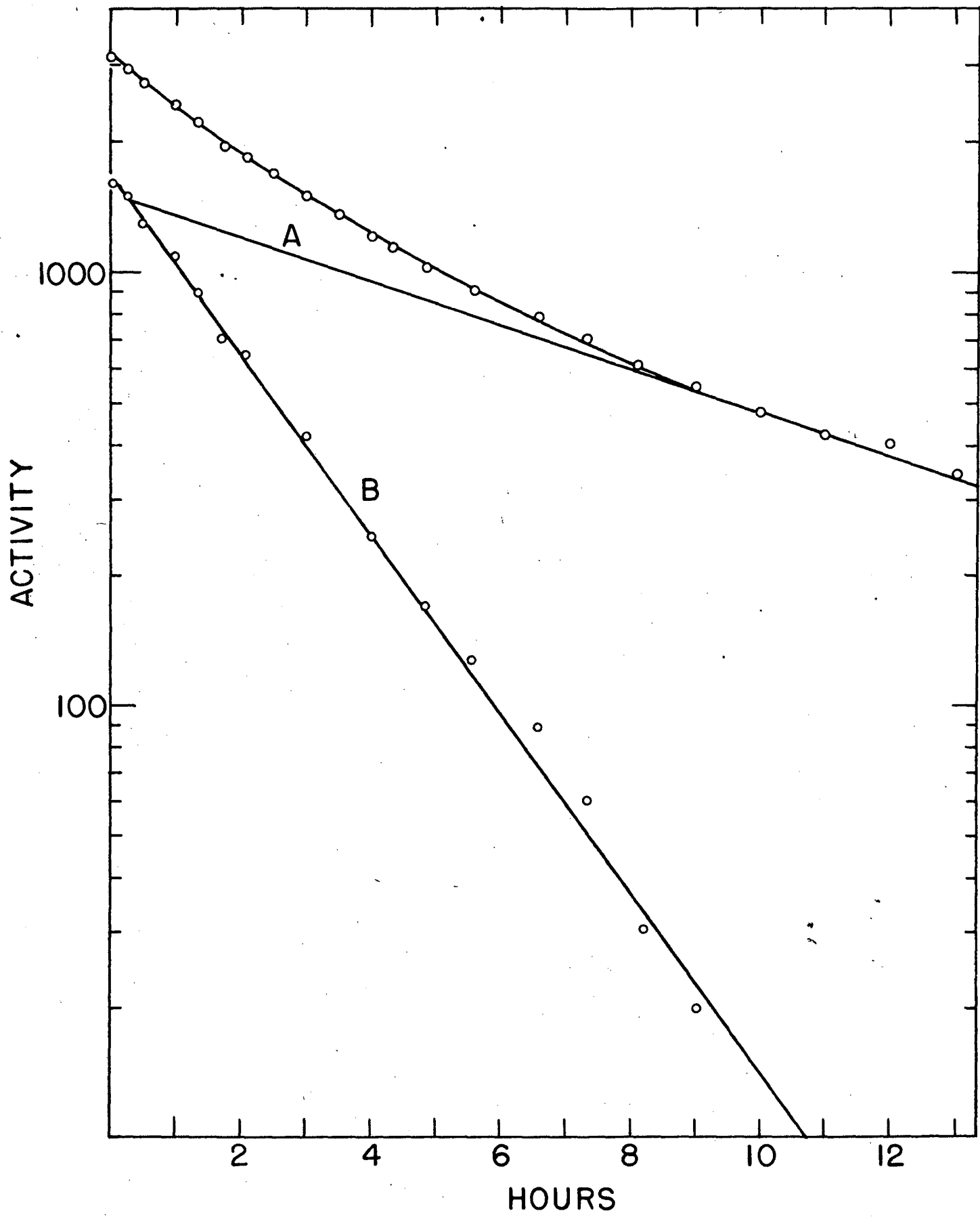
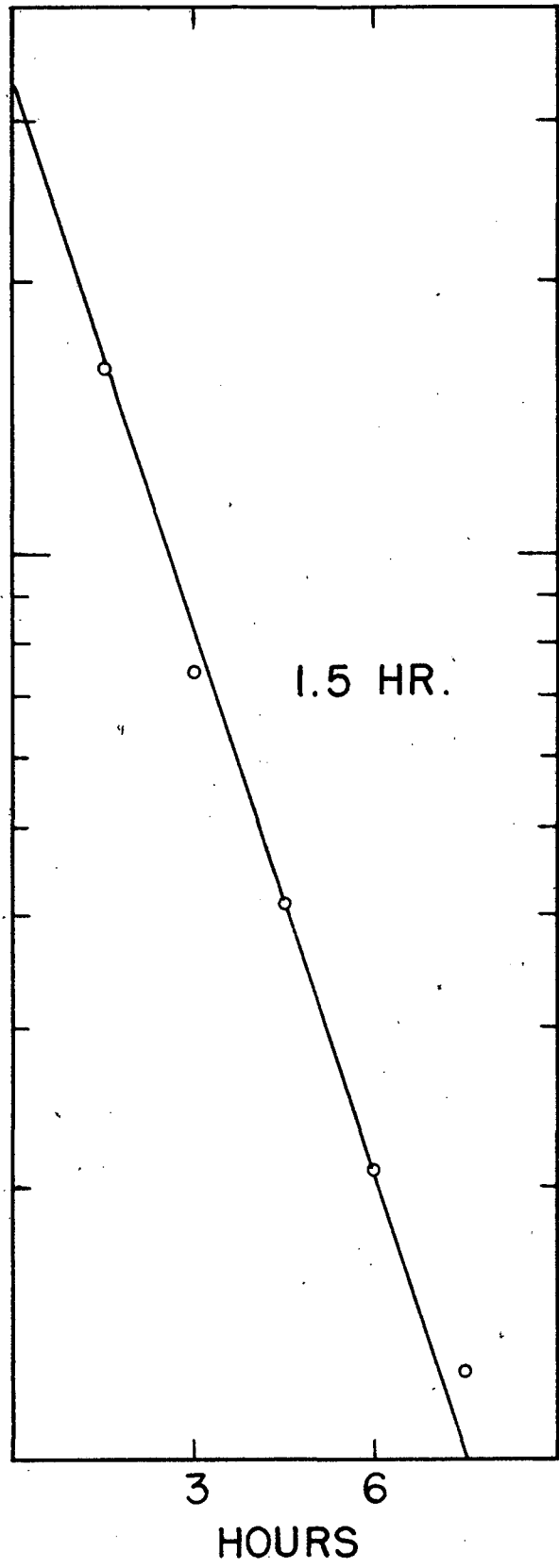


Fig. 16. Yield of 14.5-day bismuth activity plotted  
to give half-life of polonium parent.

ACTIVITY



HOURS

Table IV

Fraction	Activity 72-hr. Tl <sup>201</sup>	Calc.
1	650	---
2	300	325
3	160	163
4	85	81

The results confirm the mass assignment of the 1.5-hr. Po to mass 205. Since the genetic relation between the 8-hr. Pb<sup>201</sup> and the 72-hr. Tl<sup>201</sup> has been established,<sup>6</sup> the 8-hr. Pb<sup>201</sup> is the alpha-decay daughter of the 1.5-hr. polonium activity.

The energy of the alpha-particles was determined as  $5.2 \pm 0.1$  Mev on the alpha pulse analyzer,<sup>12</sup> by following the decay of the peak heights over a period of 4 hours (Figure 17). The peak corresponding to Po<sup>206</sup> (5.20 Mev) decayed about 20 percent over the period of observation, corresponding to a half-life of the order of 1 to 3 hours for an additional component in the peak.

The ratio of electron-capture disintegrations to alpha-disintegrations is estimated to be about 200, as determined by counting the alpha-particles from the parent and counting the separated 14-day Bi<sup>205</sup> daughter. This value is quite uncertain, due to the difficulty in resolving the alpha-decay curve. The counting efficiency of the bismuth daughter was assumed to be 10 percent. This assumption will be discussed in connection with Bi<sup>205</sup>.

C. 48-min. Po<sup>203</sup>

Repetition of the experiment described before - separation of the 12-hr. bismuth daughters from a large sample of polonium at regular intervals, indicated that two polonium parents were producing 12-hr. bismuth (Figure 18). The

Fig. 17. Pulse analysis showing alpha-particle  
energy for  $\text{Po}^{205}$ .

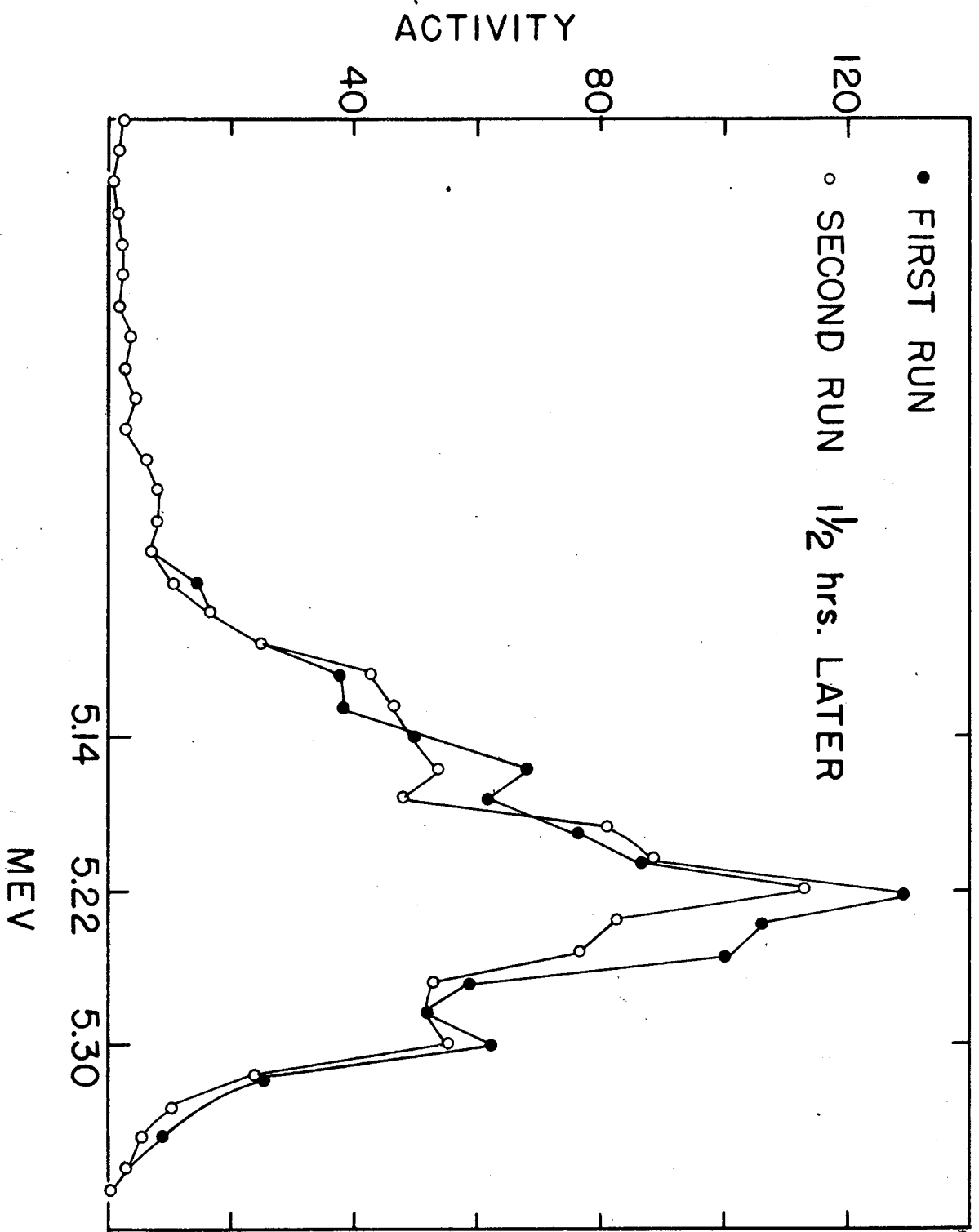
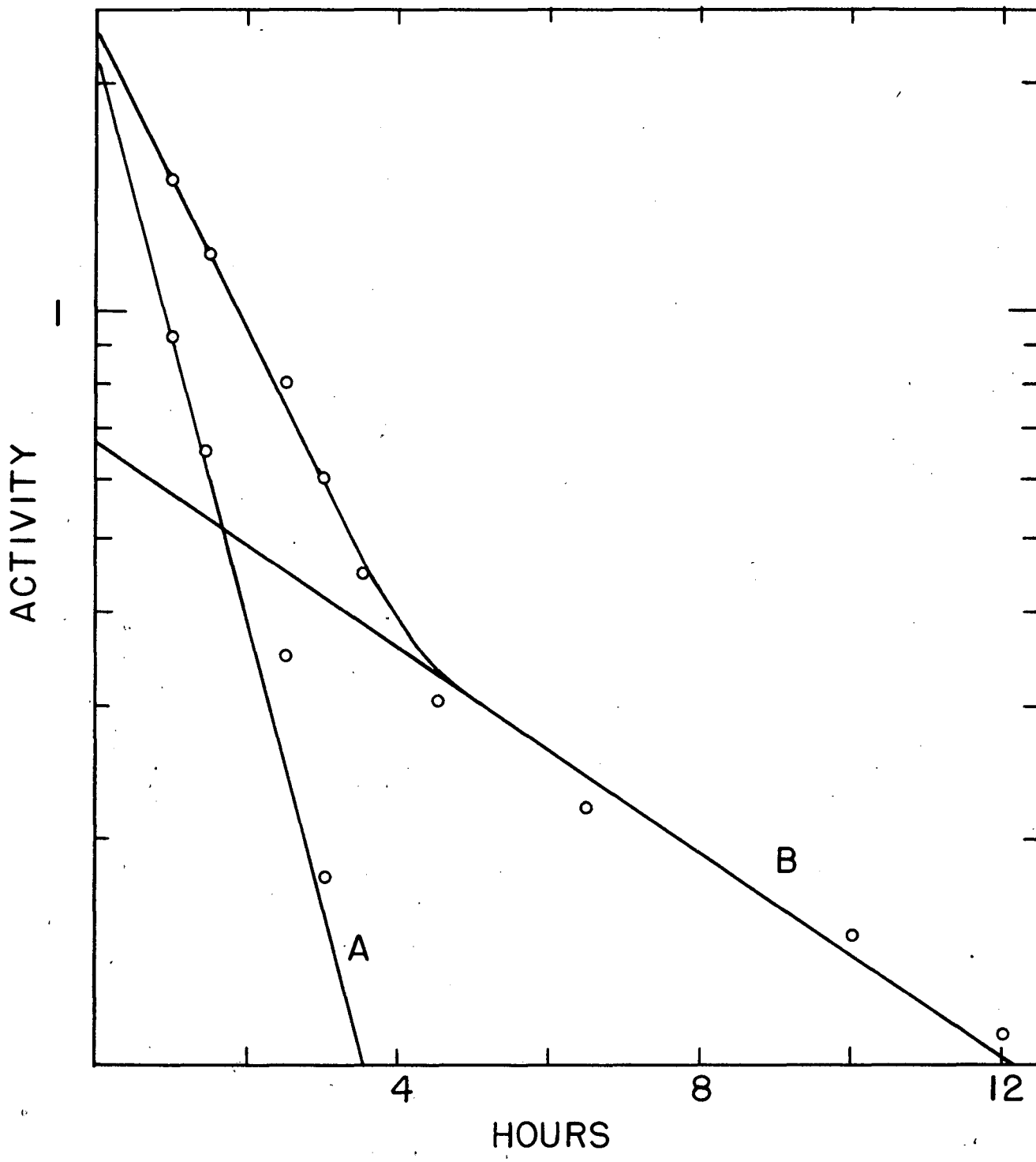




Fig. 18. Yield of 12-hr. bismuth plotted to show  
half-lives of two polonium parents

(A) 45-min.  $\text{Po}^{203}$

(B) 3.8-hr.  $\text{Po}^{204}$ .



significant difference from earlier experiments was that the polonium was purified fairly rapidly, so that the separations were started within an hour of the end of bombardment. The data indicated another polonium isotope of a shorter half-life than  $\text{Po}^{204}$ , decaying to a 12-hr. bismuth. Since the 52-hr.  $\text{Pb}^{203}$  appeared in the decay curve of the first bismuth fractions, but could not be identified in later fractions, the shorter isotope must be at mass 203.

The half-life of the  $\text{Po}^{203}$  was determined by preparing a large sample of polonium, purifying quickly after bombardment, and separating the bismuth daughters at half-hour intervals. The bismuth was allowed to decay for 24 hours, then the lead activity produced by bismuth decay was separated from the bismuth fraction. Since the lead daughters were allowed to grow into each bismuth fraction for the same period, the activity of 52-hr.  $\text{Pb}^{203}$  grown in will be directly proportional to the activity of 12-hr.  $\text{Bi}^{203}$  in each fraction. Thus a log of the activity of 52-hr.  $\text{Pb}^{203}$  vs. time plot gives the half-life of  $\text{Po}^{203}$  directly. The value thus determined is  $44 \pm 8$  min. (Figure 19). The half-life adopted, 48 minutes, was obtained by following decay of alpha-particles on the pulse analyzer.

This assignment was confirmed by showing a genetic relation between the 80-min.  $\text{Pb}^{199}$  and a 48-min. polonium parent, where the isotope measured was, as before, the 7.3-hr.  $\text{Tl}^{199}$  daughter of  $\text{Pb}^{199}$ , and not the lead activity itself. A large amount of polonium activities was prepared, purified, and the lead alpha-decay daughters separated at 40-min. intervals. After allowing the thallium daughters to grow into the lead fraction for 15 minutes, the thallium daughters were separated. The resolved intensities of the 7.3-hr.  $\text{Tl}^{199}$ , extrapolated to time of isolation, are shown in Table V.

Fig. 19. Yield of 52-hr.  $\text{Pb}^{203}$  showing a 44-min.  
polonium parent.

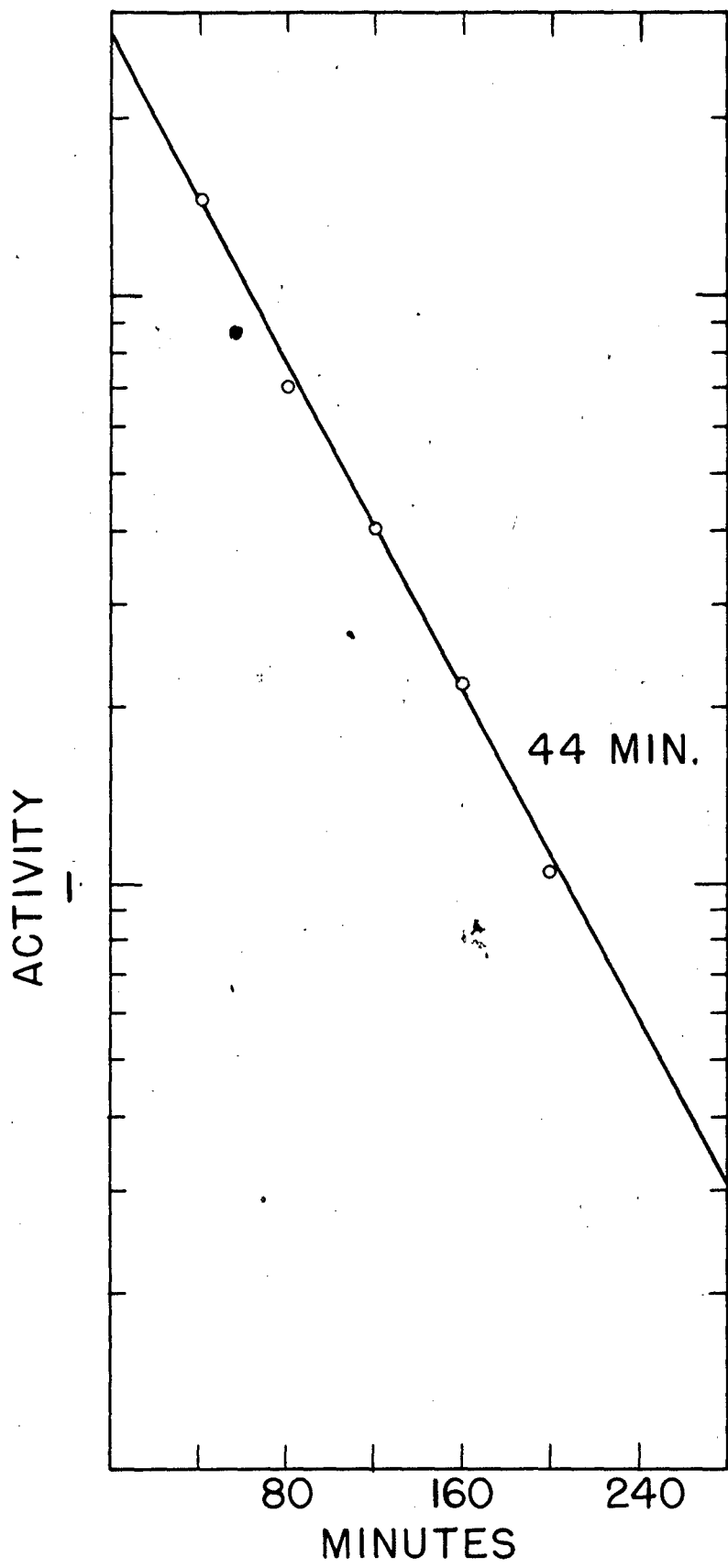


TABLE V

Fraction	Activity of 7.3-hr. Tl	Calc. for 48-min. half-life
1	4700	4400
2	2490	--
3	1450	1410
4	680	800

The energy of the alpha-particles from  $\text{Po}^{203}$  is 5.62 Mev, as determined by pulse analysis (Figure 14). Since  $\text{Po}^{203}$  and  $\text{Po}^{202}$ , which will be discussed presently, have practically the same half-life, the alpha-particles were assigned to  $\text{Po}^{203}$  on the basis of the ratio of electron-capture decay to alpha-decay for each isotope, as described below.

The EC/ $\alpha$  disintegration ratio for each isotope was determined by allowing the bismuth and lead daughters to grow from a large sample of polonium activities. The lead and bismuth daughters were separated simultaneously, and the lead fraction separated from the bismuth. The lead fraction was allowed to grow its thallium daughter activities. The thallium activities were separated after a measured interval and the decay of the thallium fraction followed on a Geiger counter. Decay of an aliquot of the bismuth was also followed, and the remainder of the bismuth allowed to grow the 52-hr.  $\text{Pb}^{203}$ . From the amount of the 7.3-hr.  $\text{Tl}^{199}$  activity it was possible to calculate the number of atoms of  $\text{Po}^{203}$  which had undergone decay by alpha-disintegration during the growth period; and from the amount of 52-hr.  $\text{Pb}^{203}$  activity the number of atoms of  $\text{Po}^{203}$  which had undergone decay by electron capture could be determined. The ratio of these gave a EC/ $\alpha$  ratio for  $\text{Po}^{203}$  of about 100, assuming equal counting efficiencies for  $\text{Tl}^{198}$  and  $\text{Pb}^{203}$ . The EC/ $\alpha$  for  $\text{Po}^{202}$  was determined by calculating the number of alpha-disintegrations from the amount of  $\text{Tl}^{198}$  activity, and calculating

the number of electron-capture disintegrations from the amount of Bi<sup>202</sup>. The EC/a ratio determined was about 10<sup>4</sup>.

Since Po<sup>203</sup> has approximately one hundred times as many alpha-particles as Po<sup>202</sup>, Po<sup>203</sup> has been assigned the single observed group of alpha-particles.

D. 52-min. Po<sup>202</sup>

In experiments where the bismuth daughters were separated from polonium parents prepared in fairly high energy (>80-Mev protons or >120-Mev helium ions) bombardments, a 95-min. bismuth activity was observed in the bismuth fraction, in addition to other activities mentioned previously. A plot of its yield vs. time of separation showed that its polonium parent was a 52 ± 5 min. activity (Figure 20).

Separation of the thallium daughters of lead produced by alpha-decay showed the presence of the 1.8-hr. Tl<sup>198</sup> in the thallium fraction. The experiment to identify the alpha-decay daughter was done in exactly the same manner as the experiment to detect the alpha-decay daughter of Po<sup>203</sup>. As before, the interval between separations was 40 minutes. The results are shown in Table VI.

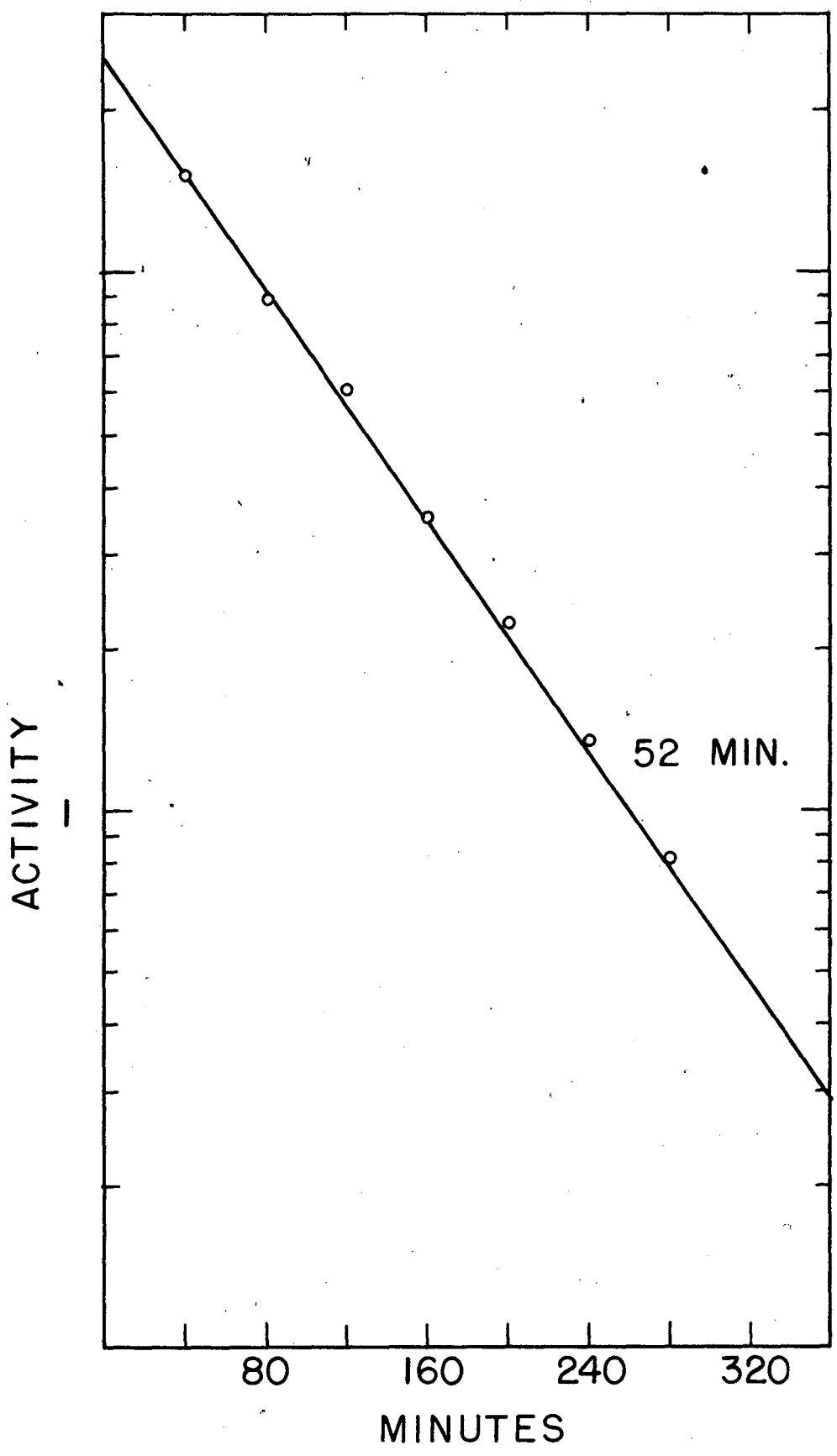
Table VI

Fraction	Activity	Calc. for 52-min. half-life
1	320	--
2	160	175
3	100	100

The same experiment gave a confirmation of earlier results on Po<sup>203</sup>.

Fig. 20. Yield of 95-min. Bi<sup>202</sup> showing a 52-min. polonium parent.





The occurrence of two isotopes of polonium with virtually the same half-lives complicates the determination of alpha-particles energies for each individual isotope. No alpha-particles which could be attributed to  $\text{Po}^{202}$  were found by pulse analysis. It is doubtful that they would be seen unless they were significantly different in energy from  $\text{Po}^{203}$ .

The partial alpha-half-lives of  $\text{Po}^{202}$  and  $\text{Po}^{203}$  calculated from these branching ratios show an unexpected deviation from the trend predicted on the basis of alpha-decay systematics.<sup>14</sup> The expected trend below mass 205 would be to shorter alpha-half-lives and higher alpha-decay energies with decreasing mass number. The experiments to establish the branching ratios of  $\text{Po}^{202}$  and  $\text{Po}^{203}$  are sufficiently difficult that the probability of error is quite large, and for this reason no significance is attached to the deviations from the expected trend.

E. 25-min.  $\text{Pb}^{198}$

The lead alpha-decay daughter of  $\text{Po}^{202}$  was identified by establishing a genetic relation between the 1.8-hr.  $\text{Tl}^{198}$  and its lead parent. This was done in the manner described before; a large sample of lead activities was prepared by bombarding thallium with protons of 120 Mev, the lead separated and purified, and the thallium daughters separated at 30-minute intervals. The resolved yields of the 1.8-hr. thallium show a half-life of  $25 \pm 10$  minutes for its lead parent. The 25-min.  $\text{Pb}^{198}$  is, of course, the alpha-decay daughter of  $\text{Po}^{202}$ . Neumann and Perlman<sup>10</sup> have observed a lead activity directly which had an apparent half-life of 25 minutes, which supports the value determined.

F. 95-min.  $\text{Bi}^{202}$

The 95-min.  $\text{Bi}^{202}$  daughter of  $\text{Po}^{202}$  has been found in bismuth fractions separated from polonium prepared by bombarding bismuth of energies greater than

60 Mev (Figure 21). Since it was always obtained with large quantities of other bismuth activities, absorption data were considered unreliable. No positrons or alpha-particles were found associated with it, although if the electron capture to alpha-decay ratio is greater than  $10^5$ , it is doubtful that alpha-decay would have been detected.

The possibility of the 95-min. bismuth being an isomer of  $\text{Bi}^{203}$  has been ruled out, since it is not detected in bismuth formed by bombarding lead enriched in mass 204 (27 percent)<sup>13</sup> with 18-Mev deuterons. Since the  $d,3n$  reaction is known to take place with high yield at this energy,<sup>9</sup> it is presumed that the 95-min. bismuth activity is not mass 203. Bismuth of mass 203 was formed in this bombardment, as lead separated from bismuth after a period of growth showed the 52-hr.  $\text{Pb}^{203}$  in good yield.

#### G. 14.5-day $\text{Bi}^{205}$

This activity has been previously mentioned as produced by decay of the 1.5-hr.  $\text{Po}^{205}$  (Figure 22). Aluminum absorption curves show that about 15 per cent of the gross Geiger counts of this isotope are due to electromagnetic radiation (Figure 23). Lead absorption shows the presence of a hard gamma of 1.7-Mev energy (14.5 g/Pb half-thickness)(Figure 24). Addition gamma-rays of 431, 550, 527, and 746 kev were identified on a beta-ray spectrometer by their conversion electrons. Conversion electrons from the hard gamma gave a value of 1.84 Mev, in substantial agreement with the value given by absorption measurements.

The following tentative decay scheme has been assumed for  $\text{Bi}^{205}$  on the basis of the data (Figure 25).

Fig. 21. Decay of bismuth daughter activities

(A) 12-hr. Bi<sup>203</sup> and 12-hr. Bi<sup>204</sup>

(B) 95-min. Bi<sup>202</sup>.

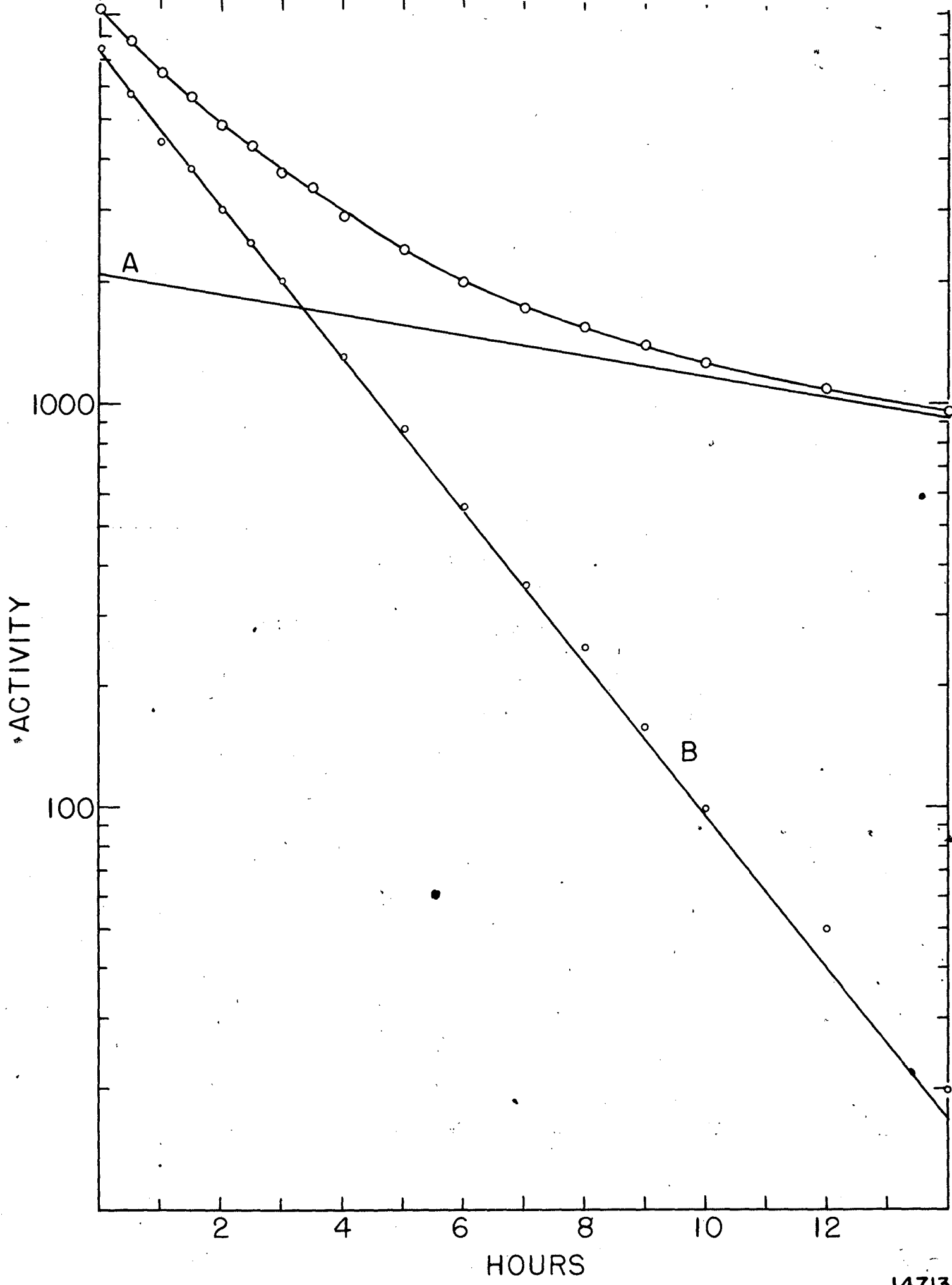


Fig. 22. Decay of 14.5-day Bi<sup>205</sup>.

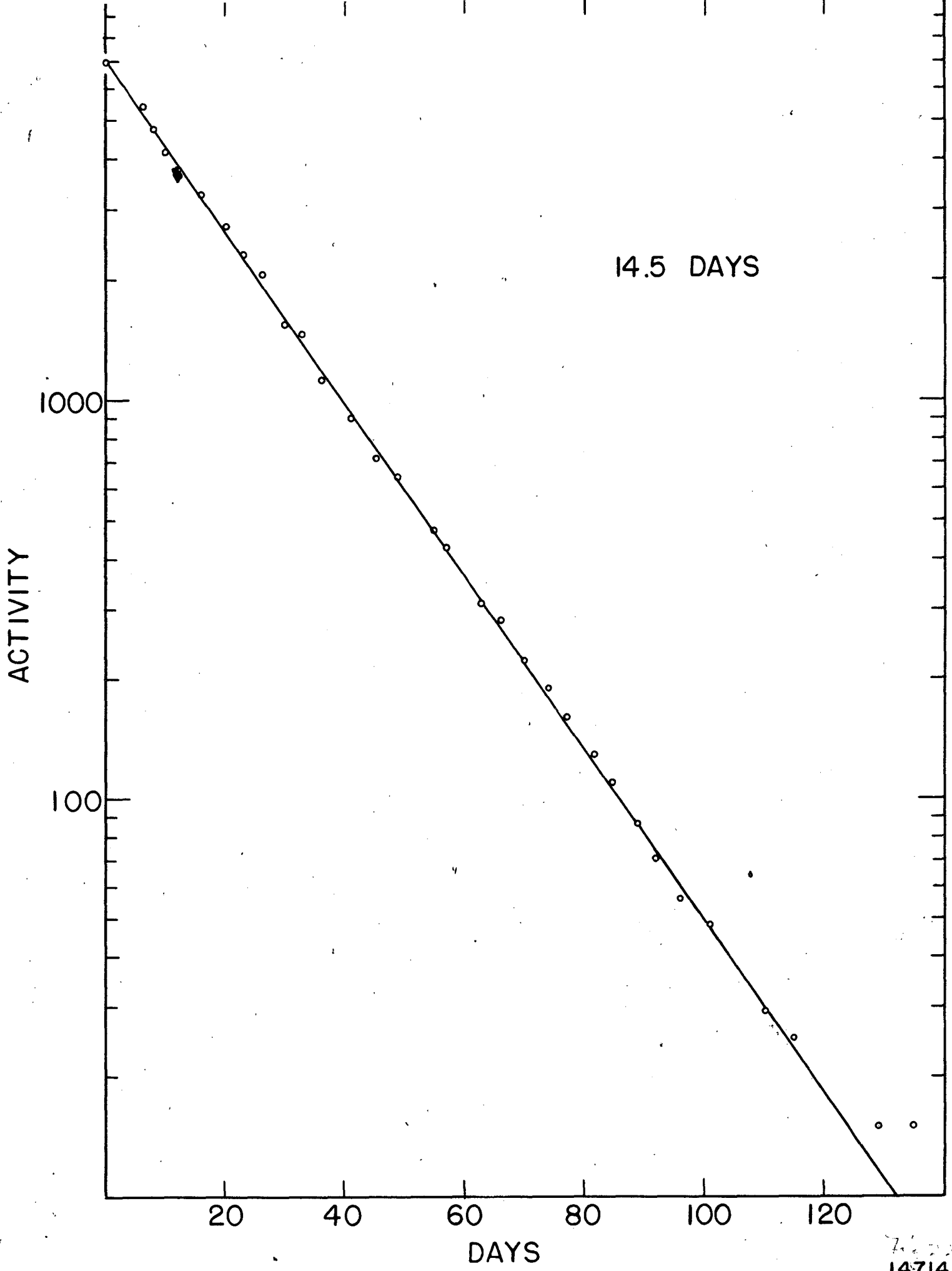


Fig. 23. Aluminum absorption of Bi<sup>205</sup>

- (A) X-rays and gamma-rays
- (B) electrons.



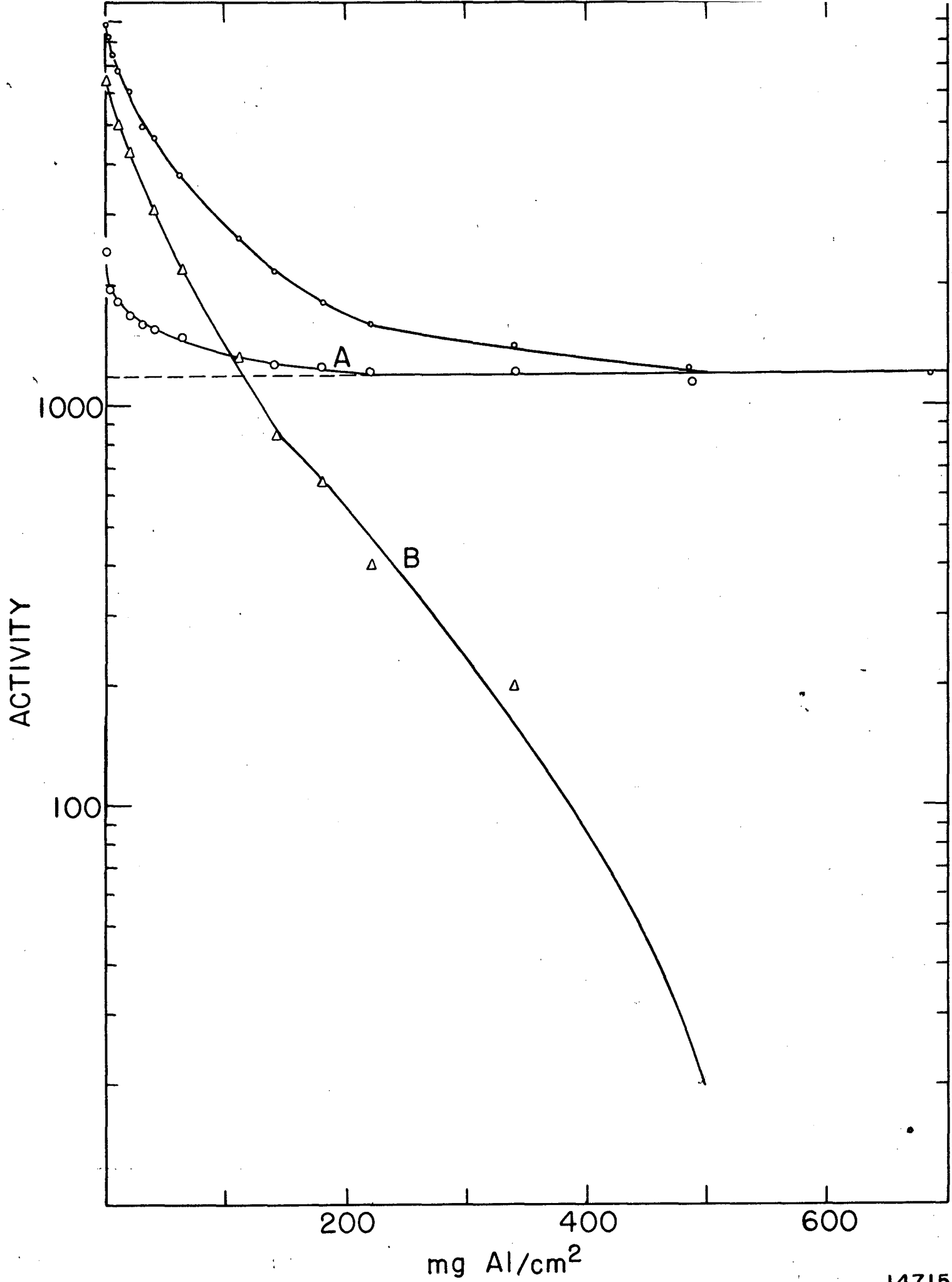


Fig. 24. Lead absorption of Bi<sup>205</sup> gamma-ray  
E = 1.84 Mev.

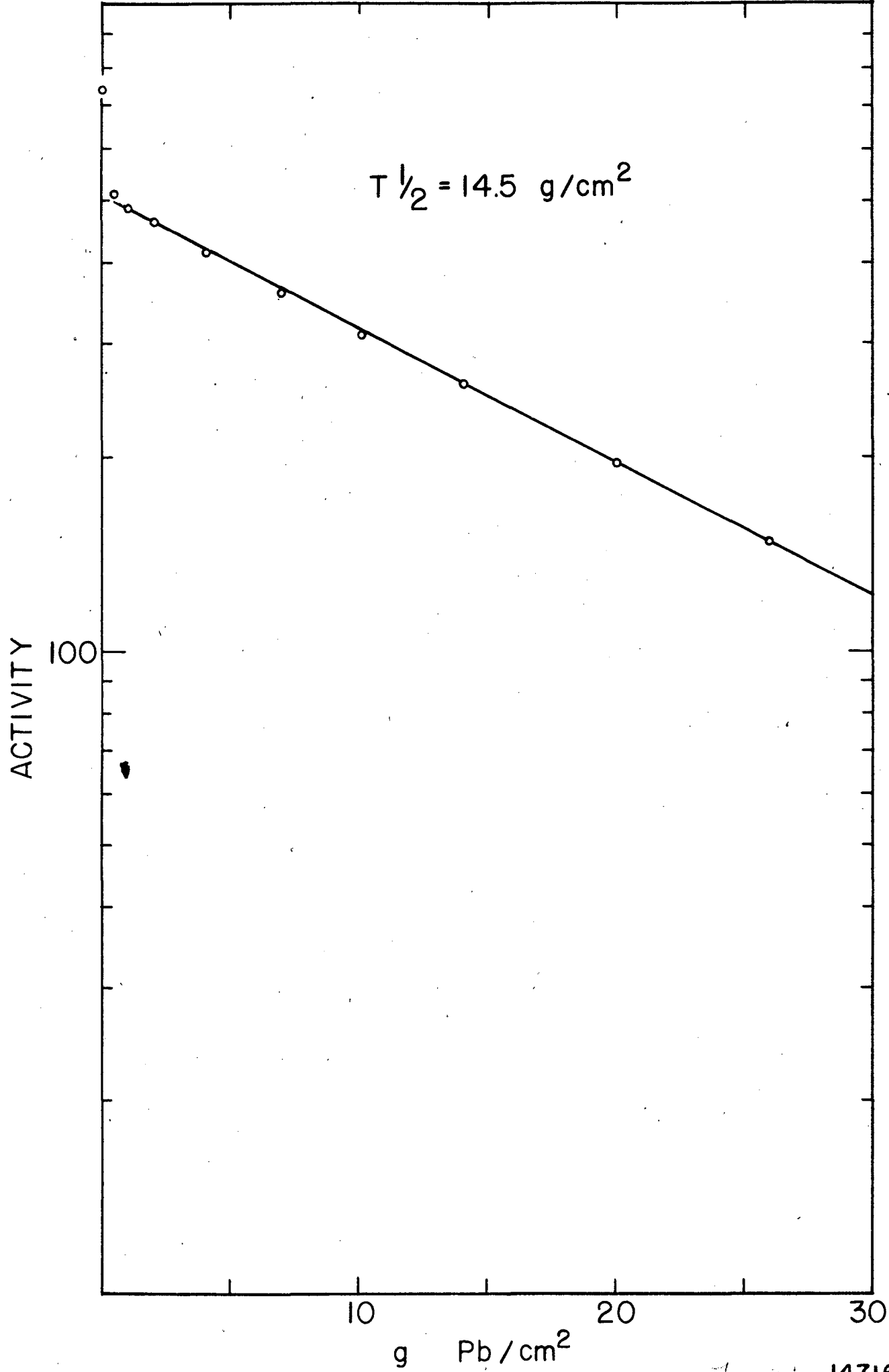
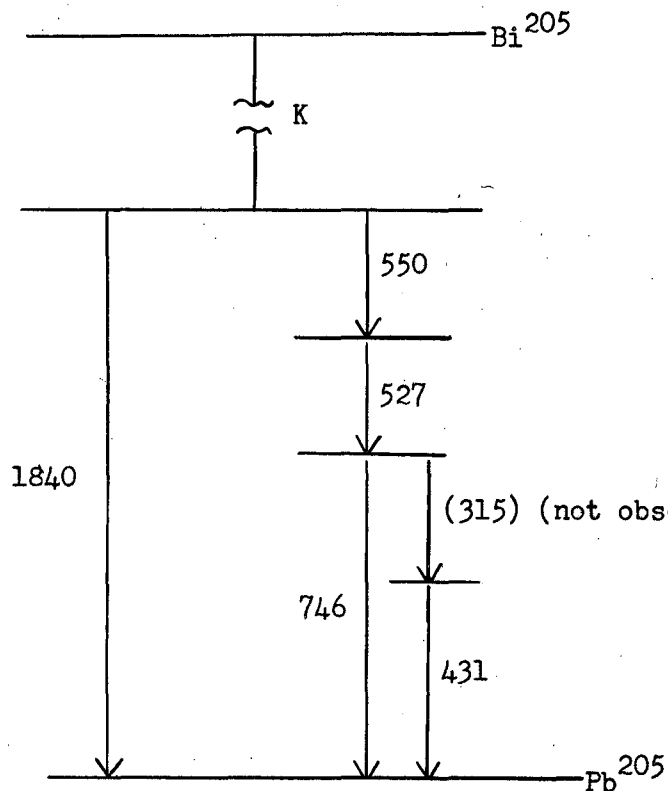


Figure 25

Proposed Decay Scheme of  $\text{Bi}^{205}$



Assuming counting efficiencies of one percent per Mev for the gamma rays, the absolute counting efficiency for the gamma-rays in argon-filled Geiger tubes is estimated at about 1.8 percent. Since there were about 6 times as many electrons counted as gamma-rays, the over-all counting efficiency is estimated as about 10 percent.

H. 68-min.  $\text{Pb}^{204m}$

The discovery of two 12-hr. bismuth isotopes forced a reconsideration of the mass assignment of the 68-min.  $\text{Pb}^{204m}$  which was based in part on the observation that the 68-min. Pb is a daughter of a 12-hr. Bi. A large quantity of polonium was prepared, purified, and the bismuth daughters separated at 4-hr. intervals. Each bismuth fraction was allowed to stand for 24 hours,

then lead was separated from the bismuth. The intensities of the resolved  $68\text{mPb}^{204\text{m}}$  activity are shown in Table VII. These data show that the 68-min. Pb is a descendant of  $\text{Po}^{204}$ , and therefore was correctly assigned.

Table VII

Fraction	Act. $68\text{mPb}$	Calc. (on 3.8 hr. $T_{1/2}$ )
1	4900	
2	2200	2350
3	1090	1120

This work was carried out under the auspices of the U. S. Atomic Energy Commission.

REFERENCES

<sup>1</sup>L. G. Lewis and R. J. Hayden, U. S. Atomic Energy Commission Declassified Document MDDC-1556, (Sept. 19, 1947); R. J. Hayden, Phys. Rev. 74, 650 (1948).

We are indebted to Dr. A. J. Dempster, Dr. M. G. Ingraham, and Dr. R. J. Hayden for information and advice on experimental techniques.

<sup>2</sup>F. L. Reynolds, D. G. Karraker, and D. H. Templeton, Phys. Rev. 75, 313 (1949).

<sup>3</sup>W. C. Barber, Phys. Rev. 72, 1156 (1947).

<sup>4</sup>N. Svartholm and K. Seigbahn, Arkivfur Matematisk Astronomi och Fysik 33A, 1-28 (1946).

<sup>5</sup>J. O. Hancock and J. C. Butler, Phys. Rev. 57, 1088 (1940).

<sup>6</sup>G. T. Seaborg and I. Perlman, Rev. Mod. Phys. 20, 585 (1948).

<sup>7</sup>E. Crentz, L. A. Delsasso, R. B. Sutton, M. G. White, and W. H. Barkas, Phys. Rev. 58, 481 (1940); also W. H. Barkas, E. C. Crentz, L. A. Delsasso, and R. A. Sutton, Phys. Rev. 57, 1087 (1940).

<sup>8</sup>D. H. Templeton, J. J. Howland, and I. Perlman, Phys. Rev. 72, 758 (1947).

<sup>9</sup>E. L. Kelly and E. Segre, Phys. Rev. 75, 999 (1949).

<sup>10</sup>H. M. Neumann and I. Perlman, Phys. Rev. (in press).

<sup>11</sup>D. A. Orth, L. Marquez, W. J. Heiman, and D. H. Templeton, Phys. Rev. 75, 1100 (1949).

<sup>12</sup>A. Ghiorso, A. H. Jaffey, H. P. Robinson, and B. Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, "The Transuranium Elements: Research Papers," Paper No. 16.8 (McGraw-Hill Book Co., Inc., New York, 1949).

<sup>13</sup>We are indebted to Dr. E. H. Huffman, Mr. R. C. Lilly, and Mrs. D. B. Stewart for purification of the lead, and Mr. J. T. Vale for the mass spectrometric assays.

<sup>14</sup>Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).