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ISOTOPIC ASSIGNMENTS OF BISMUTH ISOTOPES PRODUCED WITH HIGH ENERGY PARTICLES

H. M. Neumann and I. Perlman

November 25, 1949

Berkeley, California

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Isotopic Assignments of Bismuth Isotopes Produced with High Energy Particles

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ABSTRACT

A number of isotopes of bismuth have been identified from the bombardment of lead with high energy deuterons and protons. Evidence for these assignments is based chiefly on the daughter lead and thallium activities resulting from electron-capture decay.

Isotope	Half-life	Mode of Decay	Electron-Capture Decay Products
Bi^{203}	12 hrs.	E.C.	52 hr. Pb
Bi^{201}	118 min.	E.C.	8 hr. Pb 72 hr. Tl
Bi^{201}	62 min.	α, E.C.	
Bi^{200}	35 min.	E.C.	18 hr. Pb 27 hr. Tl
Bi^{199}	25 min.	α, E.C.	80 min. Pb 7 hr. Tl
Bi^{198}	7 min.	α, E.C.	25 min. Pb 1.8 hr. Tl
$Bi^{<197}$	1.7 min.	α, E.C. (?)	

Those isotopes exhibiting alpha-decay have been investigated as to energy and half-life for this mode of decay. A discussion is given of the relationship between half-life and energy for all known alpha-active bismuth isotopes.

INTRODUCTION

Some time ago it was found in this laboratory that the irradiation of lead with high-energy deuterons resulted in the production of highly neutron-deficient bismuth isotopes, some 10-mass units below stable bismuth, and that several of these emit alpha-particles in competition with predominant electron-capture decay.[1] It has long been known that the heavy isotopes of bismuth which are members of the natural radioactive decay series are measurably alpha-unstable, and that alpha-emission effectively disappears in the mass number range below Bi^{211}. This disappearance of alpha-activity with reappearance at around mass number 200 has recently been interpreted in terms of nuclear stability in the region of 126 neutrons and 82 protons superimposed on the general alpha-decay systematics as deduced from nuclei outside of this region.[2] The present communication is concerned with further progress on the isotopic assignments and alpha-energies of these light bismuth isotopes, giving experimental justification for some of the values already summarized in a review article.[3] Further discussion is also given for the alpha-decay half-life and energy relationship for bismuth isotopes.

METHODS

When lead is irradiated with deuterons or protons in the 100-Mev range, the mixture of bismuth isotopes produced is far too complex to allow accurate resolution of the decay curves; and in any case it would be difficult to make isotopic assignments by excitation functions because of the mass number range

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[1] D. H. Templeton and I. Perlman, Phys. Rev. 73, 1211 (1948).

[2] I. Perlman, A. Ghiorso, and G. T. Seaborg, Phys. Rev. (in press, Jan. 1, 1950); ibid., 74, 1730 (1948) and 75, 1096 (1949).

[3] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. 20, 585 (1948).

of the target lead isotopes and the difficulty of separating such reactions as d,8n and d,9n. Furthermore, the short half-lives of most of these bismuth isotopes make it most difficult to attempt to determine distinctive radiation characteristics of the different species and thereby obtain additional aid in the resolution. The alpha-particles are somewhat easier to resolve because of their distinctive energies, but this alone is not of much help in making definite mass assignments. The alpha-branching is too minute to permit isolation of the alpha-decay daughters free of a preponderant amount of thallium and lead activities from the electron-capture decay processes.

The method used for making isotopic assignments of the bismuth isotopes began by purifying the bismuth fraction from the irradiated lead, followed by the removal from this fraction, at short equal intervals, of the lead isotopes which grew from the electron-capture decay. In those cases in which the mixture was fairly simple the lead activities could be resolved, and the rate of decrease of each component in the successive separations indicated the half-life of its bismuth parent. With the genetic relationship between bismuth parent and lead daughter established, it remained to assign mass numbers to the lead isotopes. These were either known or could be determined by observing the known thallium daughters[4] of the lead isotopes.

In most cases the lead fractions removed from the initially purified bismuth fraction were themselves too complex for resolution, and the rate of decrease in growth of each lead component was determined by allowing the lead fraction to stand for definite intervals after which the thallium daughters that had grown in were removed. The amount of a particular thallium component

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[4] D. A. Orth, W. Heiman, L. Marquez, and D. H. Templeton, Phys. Rev. 75, 1100 (1949).

in successive fractions will decrease at a rate corresponding to the half-life of the bismuth grandparent when plotted against the time of lead-bismuth separation. The lead-thallium genetic relationships were determined by removing thallium at periodic intervals from a pure lead fraction. These methods will appear clearer when actual examples are discussed.

The initial bismuth separation from irradiated lead was made in one of two ways. For rapid purification (20 min. - 1 hr., depending on degree of purity demanded) the lead was dissolved in 6M HNO_3; and, after bringing almost to neutrality with sodium hydroxide, the bismuth was plated on a nickel foil upon immersion for several minutes in the warm solution. The nickel foil with bismuth was dissolved in nitric acid, bismuth hydroxide precipitated with ammonia after adding bismuth carrier, and then dissolved in hydrochloric acid. Upon dilution and warming, BiOCl precipitated and this step was repeated several times. The yield of bismuth in the plating step is only about 10% for 5-min. exposure and increases for longer periods. A slower procedure, which removed bismuth from the irradiated lead more nearly quantitatively, began with the dissolution of the target with 6M HNO_3, after which most of the lead was crystallized as $Pb(NO_3)_2$ by the successive evaporation of most of the dilute nitric acid and addition of fuming nitric acid. The bismuth carrier which had been previously added was then precipitated from the nitric acid supernatant solution upon neutralization with ammonia. The bismuth hydroxide was dissolved in hydrochloric acid and the oxychloride precipitated as described above. Since, at the irradiation energies employed, spallation products including thallium, mercury, and gold might be formed in good yield, carriers for these elements were added in all cases before bismuth precipitation as the oxychloride.

In removing lead activities periodically from the purified bismuth fractions the same chemistry was employed; that is, bismuth oxychloride was precipitated leaving lead in solution. When thallium activities were to be removed from a lead fraction, the solution was adjusted to 6M HCl and the thallium extracted as thallic chloride into ether.

In those experiments in which the bismuth alpha-activity was to be measured, the bismuth was plated on nickel foils and these constituted the samples for measurement. For measurement of the short-lived alpha-emitters, the chemistry could be done in ten minutes or less, as no effort was made to remove gold and mercury activities, which also plate on the nickel.

In order to simplify as much as possible the mixtures of activities produced, different proton and deuteron energies were used to confine the reactions to a relatively small mass number region. For example, at 40-Mev proton energy, of the alpha-emitters, only 62-min. Bi^{201} was produced. The electron-capture decay processes were followed with mica end-window Geiger tubes using absorbers to accentuate particular activities. The alpha-particles were measured in standard parallel plate chambers, and the energies were determined with an alpha-particle pulse analyzer.[5]

RESULTS

In Table 1 are summarized the half-lives and genetic relationships of the bismuth, lead, and thallium nuclides pertinent to this study. As no positrons could be measured for any of the species shown, it is assumed that all decay by electron-capture, with the exception of the minor alpha-branching

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[5] 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).

of some of the bismuth isotopes which will be discussed below. Half-life values shown in parentheses in Table 1 had been known previously.

Table 1

Genetic Relationships and Half-Lives of the Bismuth, Lead, and Thallium Nuclides

A / Z	198	199	200	201		202	203
Bi	7±1 min.	25±5 min.	35±5 min.	62±8 min.	118±10 min.		12±1 hr.
Pb	~25 min.	~80 min.	18±3 hr.	8±2 hr.		(very long)	(52 hr.)
Tl	(1.8 hr.)	(7 hr.)	(27 hr.)	72±3 hr.		(12 day)	stable

12-hr. Bi^{203}.— No attempt was made to resolve this period directly out of the bismuth fraction nor to determine radiation characteristics, since it has the same half-life as Bi^{204} which was always present. However, the half-life was readily discerned by periodically removing the lead fraction; and upon resolution of 52-hr. Pb^{203}, it was found that its yield decreased with a half-life of 12 ± 1 hrs. The radiation characteristics of the 52-hr. lead agreed well with those previously published, and a discussion of the assignment of this 52-hr. period to Pb^{203} is given in an earlier paper.[6]

The results of one of the experiments which proved the genetic relationship between 52-hr. Pb^{203} and a 12-hr. bismuth activity is shown in Figure 1. Lead was irradiated for about 1 hr. with 60-Mev protons and a bismuth fraction was purified 4 hours later. At exact intervals of 25.5 hours (arbitrary choice) lead activities which grew from the bismuth were removed

[6] D. H. Templeton, J. J. Howland, and I. Perlman, Phys. Rev. 72, 766 (1947).

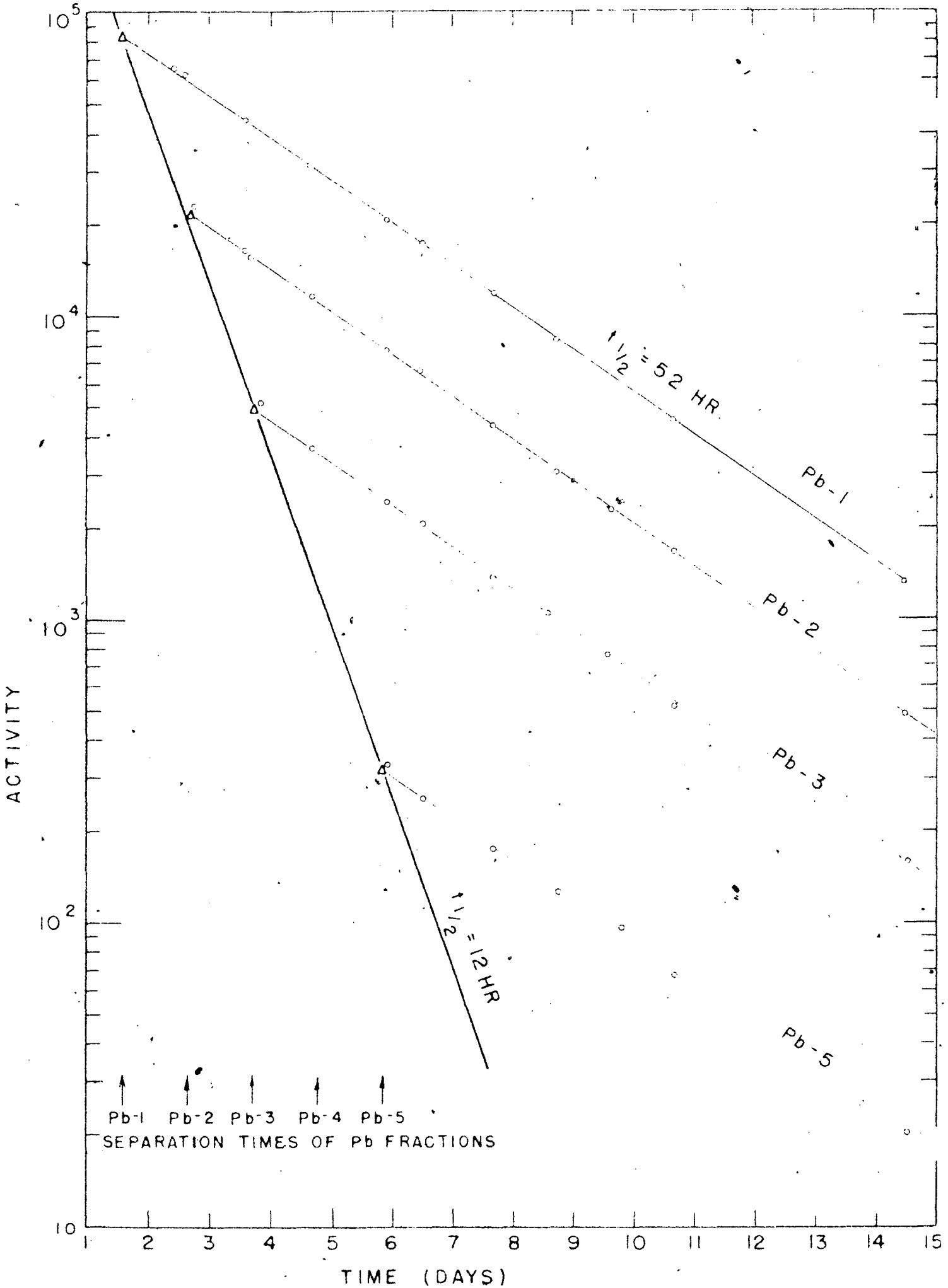
ACTIVITY
TIME (DAYS)
$t_{1/2}$ = 52 HR
$t_{1/2}$ = 12 HR
Pb-1
Pb-2
Pb-3
Pb-5
Pb-1 Pb-2 Pb-3 Pb-4 Pb-5
SEPARATION TIMES OF Pb FRACTIONS

Figure 1: Data showing the genetic relationship between 52-hr. Pb^{203} and its 12 hr. bismuth parent.

and their decay curves followed in order to resolve the 52-hr. period. These decay curves are shown in Figure 1 with each vertical arrow indicating the time of separation for the particular lead sample. The 52-hr. components extrapolated to time of separation (points indicated by triangles) define a line which is the decay curve of the parent, shown in Figure 1 by the heavy line with half-life of about 12 hours. The small amount of a shorter-lived component noted in the lead decay curves before tailing into 52 hours is principally due to 68-min. Pb^{204m}.

A bismuth activity of 12-hrs. half-life had previously[6] been reported as Bi^{204}, but it is fairly certain that the two 12-hr. activities are different isotopes. In the assignment of the 12-hr. bismuth to Bi^{204}, Tl was irradiated with 38-Mev helium ions, the bismuth fraction was separated, and the 68-min. lead was found to grow from a 12-hr. bismuth parent.[6] Recently Karraker and Templeton[7] have prepared a 4-hr. polonium activity assigned to Po^{204} which, in its electron-capture branching, decays successively through a 12-hr. bismuth and 68-min. lead in agreement with the mass assignment already cited.[6] In addition they found the 12-hr. bismuth with the 52-hr. Pb^{203} daughter, and this pair grew from a polonium isotope of about 37-min. half-life making a consistent picture if this is assigned to Po^{203}.

<u>Bi^{202}</u>.— This isotope of bismuth could not be identified by the techniques used in these studies because its decay product, Pb^{202}, has never been seen and is undoubtedly very long-lived. A minimum half-life of 500 years was set on the basis of the estimated yield from a thallium irradiation.[6] From work of Karraker and Templeton[7] Bi^{202} is a 2-hr. activity decaying by

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[7] D. G. Karraker and D. H. Templeton (private communication).

electron-capture. In the present experiments the combined lead decay products from several bismuth fractions were examined for the growth of 12-day Tl^{202} from electron-capture decay of Pb^{202}. No activity was found, and from the estimated amount of Bi^{202} which should have been formed, a lower limit for the half-life of Pb^{202} may be set which is about the same as that mentioned above.

<u>Bi^{201} isomers</u>.-- The assignment of a pair of isomers to Bi^{201} came about through an apparent discrepancy in the assignment of an alpha-decay period and electron-capture decay period to the same isotope. In a previous study[1] a weak alpha-activity of 1-2 hr. half-life was reported in the bismuth fraction from the irradiation of lead with 100-Mev deuterons. In the present studies the half-life has been more accurately determined as 62 min., and excitation functions showed it to be the heaviest of the several bismuth alpha-emitters. Through knowledge of the characteristics of Bi^{202}, Bi^{203}, and Bi^{204}, it was deduced that this activity belongs to rare alpha-branching in the decay of Bi^{201}. It should therefore be possible to identify the lead and thallium daughters; and by the methods already described for identifying the bismuth parent, their yields should decrease with the half-life of the parent. When this experiment was done, a new decay chain consisting of an 8-hr. lead and 72-hr. thallium was indeed found. The thallium in particular was best assigned to Tl^{201}, because Tl^{198}, Tl^{199}, and Tl^{200} have been assigned[4] to other activities, while Tl^{202} is not only assigned to a different activity, but would be blocked by long-lived Pb^{202}. However, the half-life for the Bi^{201} parent in the first experiments turned out to be about 90-min. rather than 62 min., the half-life of the alpha-activity thought to be Bi^{201}. The most promising solution at present is to assume that there are independently decaying isomers, one of about 1-hr. half-life which has

measurable alpha-branching, and another of about 2-hrs. half-life which only exhibits electron-capture decay. It would not be easy to resolve accurately two such components even if they alone were present, but in this case the bismuth fraction itself is hopelessly complex, and one must resort to periodic removal of lead, and from this, thallium, to obtain the progressive decrease in yield of 72-hr. Tl^{201}. The inaccuracies inherent in such a procedure make it possible to fit the data equally well to a single component for Bi^{201} of 90-min. half-life, or two components of 1- and 2-hr. half-lives.

One experiment yielded results which give somewhat more substance to the postulate of isomers for Bi^{201}. A sample of lead irradiated with 100-Mev protons had its bismuth fraction removed some 5 hrs. after irradiation, and from this a lead fraction was removed at 155-min. intervals for the next 13 hours. Each lead fraction was allowed to decay a definite period of time (36 hrs.) and thallium was removed, its decay followed, and the 72-hr. component resolved. When the yields of 72-hr. Tl^{201} were extrapolated back to the respective times of separation, the half-life for Bi^{201} defined by these points turned out to be 118 min. rather than 90 min. No other experiments were carried out in which the bismuth decay was followed for such a long period of time, and presumably in this experiment the 1-hr. component had largely decayed allowing the detection of the 2-hr. component. While the assumption of isomers for Bi^{201} explains the discrepancies between the decay rates of the alpha-particles and the other radiation, and fits the decay data obtained through the 72-hr. Tl^{201} as well as any other assumption, this assignment cannot be considered as definitely proved.

The association of 72-hr. thallium with the 62-min. bismuth alpha-emitter comes about also from excitation function measurements. Here too,

the assignments are not unique, but the observations are in accord with the most likely assignments made according to the previous discussion. When lead was irradiated with 40-Mev protons, the only bismuth alpha-emitter found was the 1-hr. period shown in Fig. 2a. Purification of the bismuth had taken place less than 1 hr. after irradiation so that the 25-min. period would have been noted had it been produced. The thallium fraction which was removed from lead, which in turn came from the bismuth fraction, showed only the 72-hr. period as seen in Fig. 2c, showing again that the 27-hr. thallium and 7-hr. thallium lie at lower mass numbers. Finally, as shown in Fig. 2b, the lead fraction removed from the bismuth fraction could be resolved into three components: 52-hr. Pb^{203} in good yield, an 8-hr. period attributable to Pb^{201}, and a 68-min. period which is probably Pb^{204m}. The 72-hr. thallium growing from the 8-hr. lead would not affect appreciably the decay curve of the 52-hr. Pb^{203}. The curves of Fig. 2a, b, c are therefore consistent with the assignment of the 1-hr. alpha-emitter to Bi^{201} and the 72-hr. thallium to Tl^{201} coming via 8-hr. Pb^{201}.

The establishment of the half-life for the Pb^{201} parent of the 72-hr. Tl^{201} resulted from a separate experiment in which the genetically related pair 18-hr. Pb^{200} and 27-hr. Tl^{200} was also observed. Following the irradiation of lead with 180-Mev deuterons, the bismuth fraction was purified and allowed to stand for an hour to grow lead daughters. The lead fraction from this was then purified and after 8 hours the thallium was removed, with further thallium separations at succeeding 8-hr. intervals. These thallium fractions were followed, resolved into 72- and 27-hr. components, and the yields of each plotted according to the successive times of separation from the parent lead fraction. Aside from the first thallium sample which contained in addition some 7-hr. Tl^{199} from Pb^{199} still present, the thallium decay

Figure 2: Activities resulting from irradiation of lead with 40-Mev protons.

(a) Bismuth alpha-activity decay curve.

(b) Electron-capture activities of lead formed by decay of bismuth.

(c) Electron-capture activity of thallium formed by decay of lead activities shown in (b).

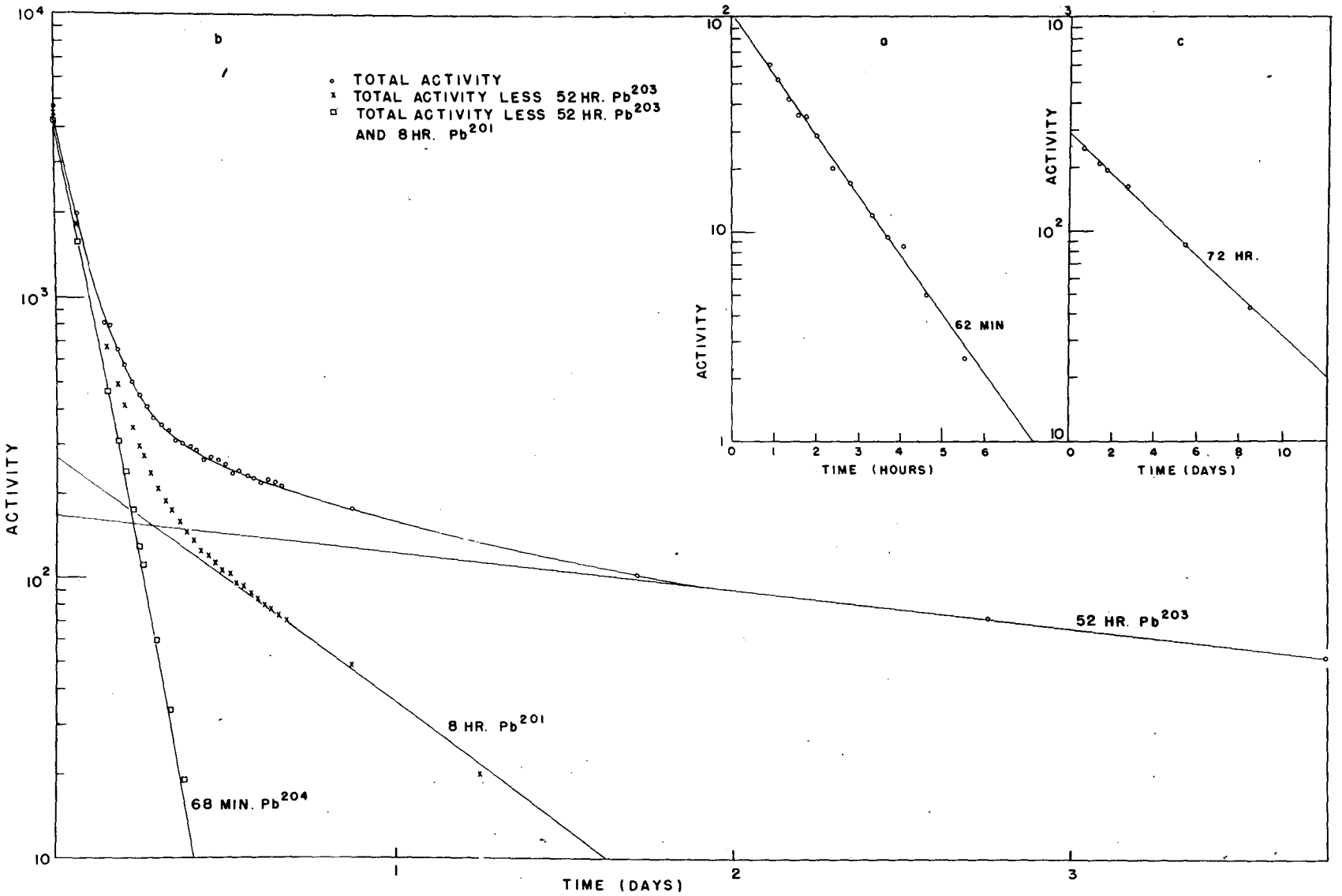
b
TOTAL ACTIVITY
TOTAL ACTIVITY LESS 52 HR. Pb203
TOTAL ACTIVITY LESS 52 HR. Pb203 AND 8 HR. Pb201
ACTIVITY
TIME (DAYS)
52 HR. Pb203
8 HR. Pb201
68 MIN. Pb204
a
62 MIN
TIME (HOURS)
c
72 HR.
TIME (DAYS)

FIG. 2

curves could be resolved into only the two components. These data are shown in Fig. 3 from which it may be seen that the lead parent of 72-hr. Tl^{201} has a half-life of about 8 hours, while that of the 27-hr. Tl^{200} is about 18 hrs.

Some of the radiation characteristics of 72-hr. Tl^{201} were determined by absorption methods and by means of a crude beta-ray spectrometer. In a sample of purified thallium in which the decay showed an essentially straight 72-hr. decay period, the beta-ray spectrometer showed a strong conversion line at 130 kev and a weaker one at 190 kev which correspond to the K- and L-lines of an approximately 210-kev γ-ray. Absorption curves taken with lead could be resolved into a component of 710 mg/cm^2 and 200 mg/cm^2 which are taken to be the 210-kev γ-ray and 70-kev K x-ray. Within the accuracy limits, the resolution is not unique but can be made consistent with the observed electron lines. The degree of conversion of the 210-kev γ-ray cannot be known better than the counting yield of the 210-kev γ-rays in the argon-filled counters, and this is poorly known. Assuming 0.3% for the counting yield after correcting for geometry loss, the indicated internal conversion is 60%. Both K and L x-rays have been resolved roughly in conformity with the decay scheme of K-capture to a 210-kev state whose γ-ray is largely converted in the K-shell. Because of uncertainties in counting efficiencies of electromagnetic radiation, it is not possible to say whether or not all of the K-capture processes go to the 210-kev state nor to give an accurate conversion coefficient for the γ-ray.

<u>35-min. Bi^{200}</u>.— The assignment of this isotope is based upon the assignment[4] of the 27-hr. thallium to Tl^{200}. The decay sequence starts with 35-min. Bi^{200}, the daughter of which is an 18-hr. lead activity. The distinguishing property of 27-hr. Tl^{200} is hard gamma-radiation which is totally absent from the 72-hr. Tl^{201}, making it possible to measure the 27-hr. period

Figure 3: Half-lives of lead parents of 72-hr. Tl (◎) and of 27-hr. Tl (▣). Data obtained from yields of respective Tl daughters.

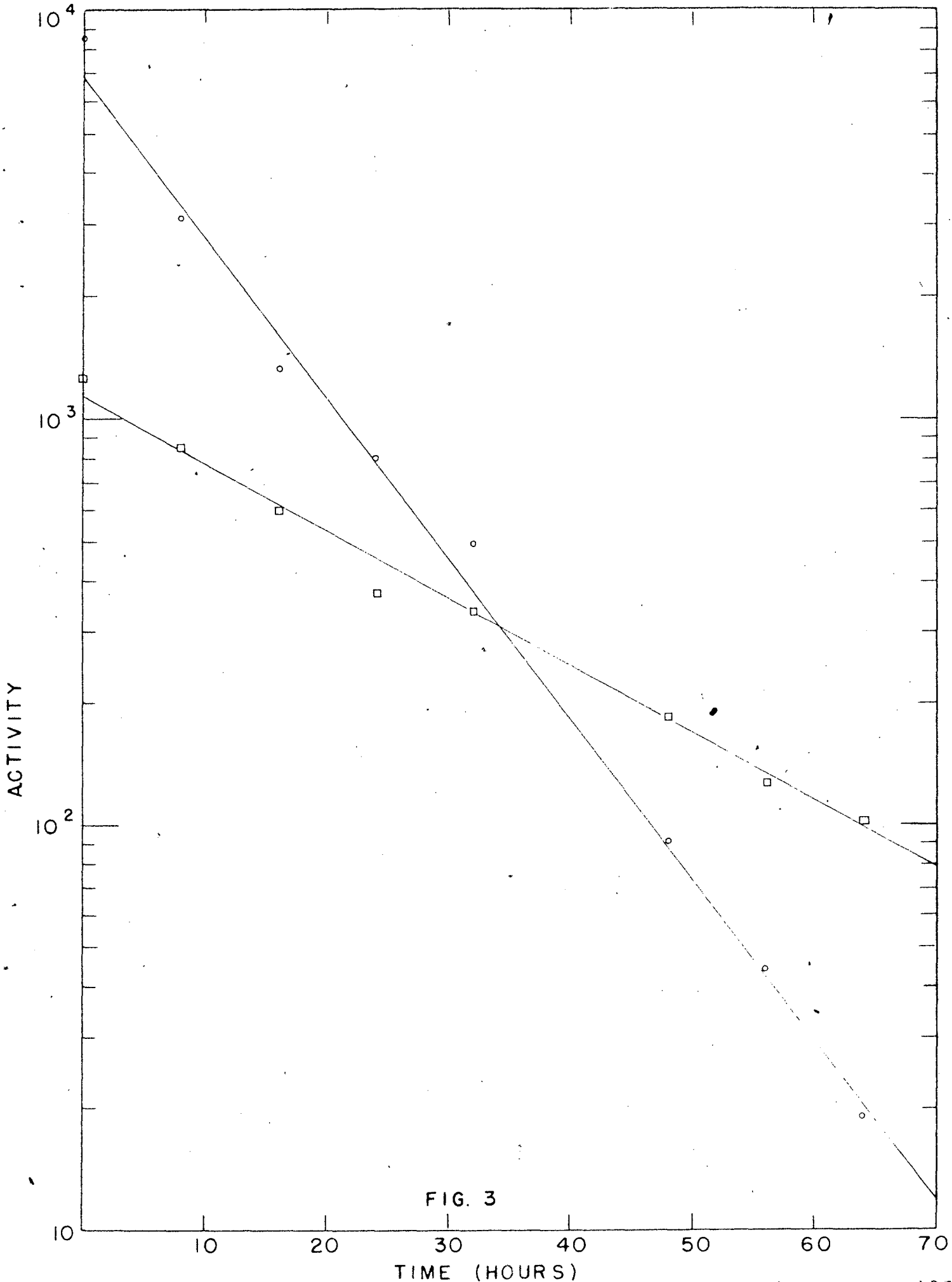

FIG. 3

in the presence of large amounts of the 72-hr. period. Following the irradiation of lead with 150-Mev protons, bismuth was removed, and the daughter lead separated from the bismuth parent after a one-hour growth period. The lead fraction was then allowed to decay for 35 hrs., following which thallium was removed. The decay of the thallium was followed, both without absorbers and through 11.5 gm/cm^2 lead with 234 mg/cm^2 beryllium to absorb secondary electrons created in the lead absorber. The decay curve taken without absorbers could be resolved into 27- and 72-hr. periods, while that taken through lead showed only the 27-hr. half-life. Lead absorption curves showed the presence of a γ-ray of 1.6 Mev, and a softer one of about 300-400 kev in about equal abundance. Gold when irradiated with 38-Mev α-particles should produce the 27-hr. period by the (α,n) reaction according to earlier work;[4] and when this experiment was tried, the 27-hr. period did show up. Furthermore, its lead absorption curve was identical with that of the 27-hr. period obtained from bismuth decay. A group of electrons of ~350 kev in low abundance is also found associated with Tl^{200}.

The genetic relationship between the 27-hr. thallium and a lead parent of 18-hr. half-life was shown in the same experiment that the 8-hr. lead was linked with the 72-hr. thallium (see Fig. 3 and its explanation in the previous discussion). The establishment of a 35-min. bismuth as the parent of the 18-hr. Pb^{200} and 27-hr. Tl^{200} was accomplished in a manner somewhat analogous to that already described in relation to Fig. 1 for the 12-hr. Bi^{203} - 52-hr. Pb^{203} pair. In the present case lead was irradiated with 150-Mev deuterons for 45 min., the bismuth fraction purified 70 min. after the bombardment, and the lead fraction removed at 50-min. intervals thereafter. After allowing each lead sample so obtained to decay for 25 hours, thallium fractions were removed and their decay periods followed. By taking the decay

curves through lead absorbers (4 gm/cm^2), only the 27-hr. thallium appeared so that there was no problem of resolution. The yields of 27-hr. components, extrapolated back to times of separation, define the decay curve of the bismuth parent. This is shown in Fig. 4 in which the abscissa has a split time scale in order to accommodate the 35-min. period and 27-hr. period on the same graph.

No alpha-activity accompanying the 35-min. Bi^{200} could be identified. As will be shown, there is alpha-activity of 25-min. half-life which has been associated with Bi^{199}, and the apparent absence of a separate group for Bi^{200} may only mean that the alpha half-life of Bi^{200} is 10 times greater than that of Bi^{199}.

<u>25-min. Bi^{199}</u>.-- A bismuth of this period was first noted through its alpha-emission,[1] and has now been assigned to Bi^{199} through its genetic relationship to 7-hr. Tl^{199} by way of electron-capture decay. The method of determining these assignments is similar to others already discussed. In this case the best data were obtained from the irradiation of lead with 180-Mev deuterons for 15 min., after which bismuth was purified (time-1 hr.) and the lead fraction removed from it at 20-min. intervals. These lead fractions were allowed to decay for 4 hrs. and thallium then removed. The thallium decay curves contained the 72-hr. Tl^{201}, 27-hr. Tl^{200}, as well as 7-hr. Tl^{199} so that it was necessary to resolve the decay curves. When this was done the 7-hr. Tl^{199} yields, extrapolated to points of lead separation, defined a rather good 25-min. curve for the Bi^{199}. The probable error of the determination in this experiment was only a couple of minutes, but the agreement between a number of separate experiments was not this close.

In order to determine the half-life for Pb^{199}, one of the lead fractions was subjected to successive removals of thallium at 60-min. intervals, and the

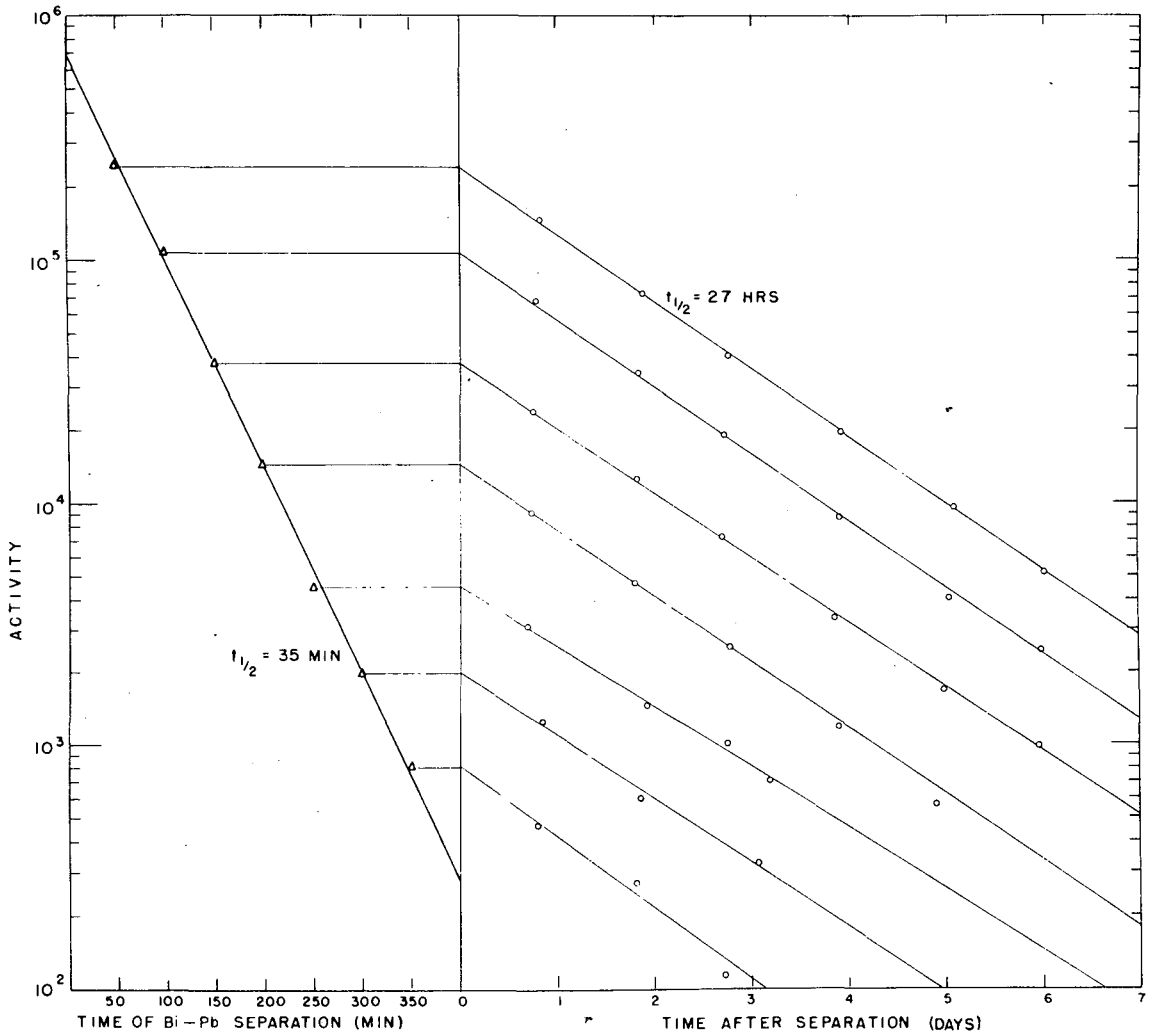
ACTIVITY
10^6
10^5
10^4
10^3
10^2
$t_{1/2}$ = 35 MIN
$t_{1/2}$ = 27 HRS
50 100 150 200 250 300 350
TIME OF Bi – Pb SEPARATION (MIN)
0 1 2 3 4 5 6 7
TIME AFTER SEPARATION (DAYS)

FIG. 4

Figure 4: Data showing genetic relationship between 27-hr. Tl^{200} and a 35-min. bismuth.

yields of the 7-hr. component were used to define the half-life of the Pb^{199} parent. A value of ~80 min. was obtained for the half-life of Pb^{199} in this manner.

<u>7-min. $Bi^{198(?)}$.</u>-- It had been previously reported[4] that a 1.8-hr. thallium results from the irradiation of gold with 38-Mev alpha-particles, and that the most likely assignment for this activity is Tl^{198}. An attempt has been made to see if this thallium activity can be linked through electron-capture processes to one of the light bismuth alpha-emitters, in particular that previously reported[1] with a 9-min. period. A short irradiation (10 min.) of lead with 120-Mev protons was followed by rapid bismuth isolation (25 min.), and lead removal from the bismuth fraction at ten-minute intervals. Each of these lead fractions was allowed to stand 53 min. after which thallium was isolated. The decay of these fractions was followed in an attempt to resolve any 1.8-hr. component present. About all that can be said is that the thallium fractions contained a small amount of activity shorter than 7 hrs. and entirely consistent with being 1.8 hrs., and this short-lived activity disappeared rapidly in successive fractions. When the short-lived thallium was resolved from the complex decay curve with 1.8-hr. half-life, the yields of this component indicated a half-life for the bismuth ancestor of 7 min.

One of the early lead fractions itself was followed and showed a component of about 25-min. half-life which is probably Pb^{198}, since a similar activity was prepared by Karraker and Templeton[7] from protons on thallium and this was shown to decay to 1.8-hr. Tl^{198}.

ALPHA-DECAY PROPERTIES

The earlier work[1] on these alpha-emitting bismuth isotopes showed four periods: 2 min., 9 min., 27 min., and 1-2 hrs. of which the shortest was not proved to be bismuth. It was recognized that the half-lives were those of the electron-capture decay and the alpha-particles arose from rare alpha-branching. In addition the energies could be but poorly determined, and the particles from the three longer periods were thought to lie in the range 5.2-5.8 Mev. Better energy values and decay periods have now been obtained with some excitation function data identifying these alpha-periods with some of the electron-capture activities already discussed. Table 2 summarizes the information that has thus far been obtained.

Table 2

Alpha-decay Properties of Neutron Deficient Bismuth Isotopes

Mass Number	Half-life	α-particle Energy (Mev)	"Threshold" (protons on natural lead)	ratio EC/α	α-decay half-life
Bi^{201}	62 min.	5.15	<40	3×10^4	2.6 yrs.
Bi^{199}	25 min.	5.47	50-60	8×10^3	140 days
Bi^{198}	7 min.	5.83	60-80	2×10^3	10 days
$Bi^{<198}$	1.7 min.	6.2	<180		

The 1-hr. alpha-emitter can be prepared free of shorter periods by choice of irradiation energy. Using deuterons of 60 Mev, only a 62-min. alpha-emitter was in evidence and this was found in low yield. The alpha-particle energy determined by an ionization chamber with pulse height analyzer was 5.15 ± 0.06 Mev. The activity also appeared with 40-, 50-, and 60-Mev protons, and with 60-Mev protons just a trace of the 25-min. alpha-emitter

began to appear. These facts place the 62-min. alpha-emitter at a higher mass number than the others. By eliminating bismuth isotopes of mass number 202 or greater it would seem reasonable to assign this period to Bi^{200} or Bi^{201}. Bi^{200} decay has already been shown rather definitely to be governed by a 35-min. half-life (see previous discussion), and the most consistent picture would be to assign the 62-min. activity to an isomeric state of Bi^{201} as already discussed.

It has already been mentioned that no alpha-activity has been found which could be attributed to Bi^{200}. In the experiment mentioned just above with the 60-Mev deuterons on lead in which only the 62-min. alpha-emitter was seen, it was found that thallium fractions from bismuth decay showed comparable amounts of 72-hr. Tl^{201} and 27-hr. Tl^{200}. This means that Bi^{200} was present in good yield but has no detectable alpha-particles, for any present would have gone down with a half-life of 35 min.

The best half-life for the period previously reported[1] as 27 min. was obtained by following the alpha-group on the alpha-particle pulse analyzer. This turned out to be 25 min. which is within the range of values found for the electron-capture half-life for Bi^{199}. This assignment also conforms with the appearance of these alpha-particles at a proton threshold energy of 60 Mev, some 20 Mev higher than the threshold for the activity assigned to Bi^{201}. The alpha-particle energy was found to be 5.47 ± 0.06 Mev.

At higher bombardment energies (100 Mev for good yield) the shorter period appeared which was called 9 min. in the earlier report.[1] The best half-life obtained by following the alpha-group on the alpha pulse analyzer was 7 min., which agrees with the half-life ascribed to Bi^{198} through its electron-capture relationship with 1.8-hr. Tl^{198}. Since its order of appearance in the excitation function also is in harmony with the assignment

of such a low mass number, it is probable that these alpha-particles of 5.83 ± 0.06 Mev should be assigned to Bi^{198}.

Finally, the 1.7-min. alpha-activity has been shown to be bismuth, insofar as chemical plating on nickel foil is a criterion, and the alpha-particle energy was determined as 6.2 ± 0.1 Mev. It is probable that the mass number is still lower than Bi^{198} and has been so indicated in Table 2. No attempt has been made to measure the electron-capture decay sequence of this nuclide, because the short half-life would make it most difficult to perform the necessary chemical separations.

In Fig. 5 is shown a series of alpha-particle pulse analyzer curves taken on the bismuth fraction from the irradiation of lead with 180-Mev deuterons. The abscissa gives the alpha-energy or, as the data is taken, the channels of the pulse height discriminator, while the ordinates indicate the number of counts registered in each channel normalized to the same counting period. The time indicated for each curve is that from the end of irradiation to the time that the alpha-particle energy analysis was made. It is seen, first of all, why in the previous study[1] it was not possible to obtain a good half-life for the 5.15-Mev group by following the gross decay. It is only when the bombardment energy is lowered to the point at which this is the only alpha-group formed that it is possible to obtain the 62-min. period. It can be seen that the decay periods for the 5.47-Mev and 5.83-Mev groups may readily be determined by following each peak separately, and it was by data of the type shown in Fig. 5 that the 25-min. and 7-min. periods were obtained. From Fig. 5 it may be seen in a qualitative fashion that the 5.83-Mev group disappears first so that in the interval 1-2 hrs. after bombardment, the 5.47-Mev group is the principal one present, and that the 5.15-Mev group only begins to appear distinctly after about half of the 5.47-Mev group has decayed.

Figure 5: Successive alpha-energy analyses of bismuth activities produced by irradiation of lead with 180-Mev deuterons. The time to the right of each curve indicates interval between end of irradiation and mid-time of analysis. Original data normalized to equal counting time.

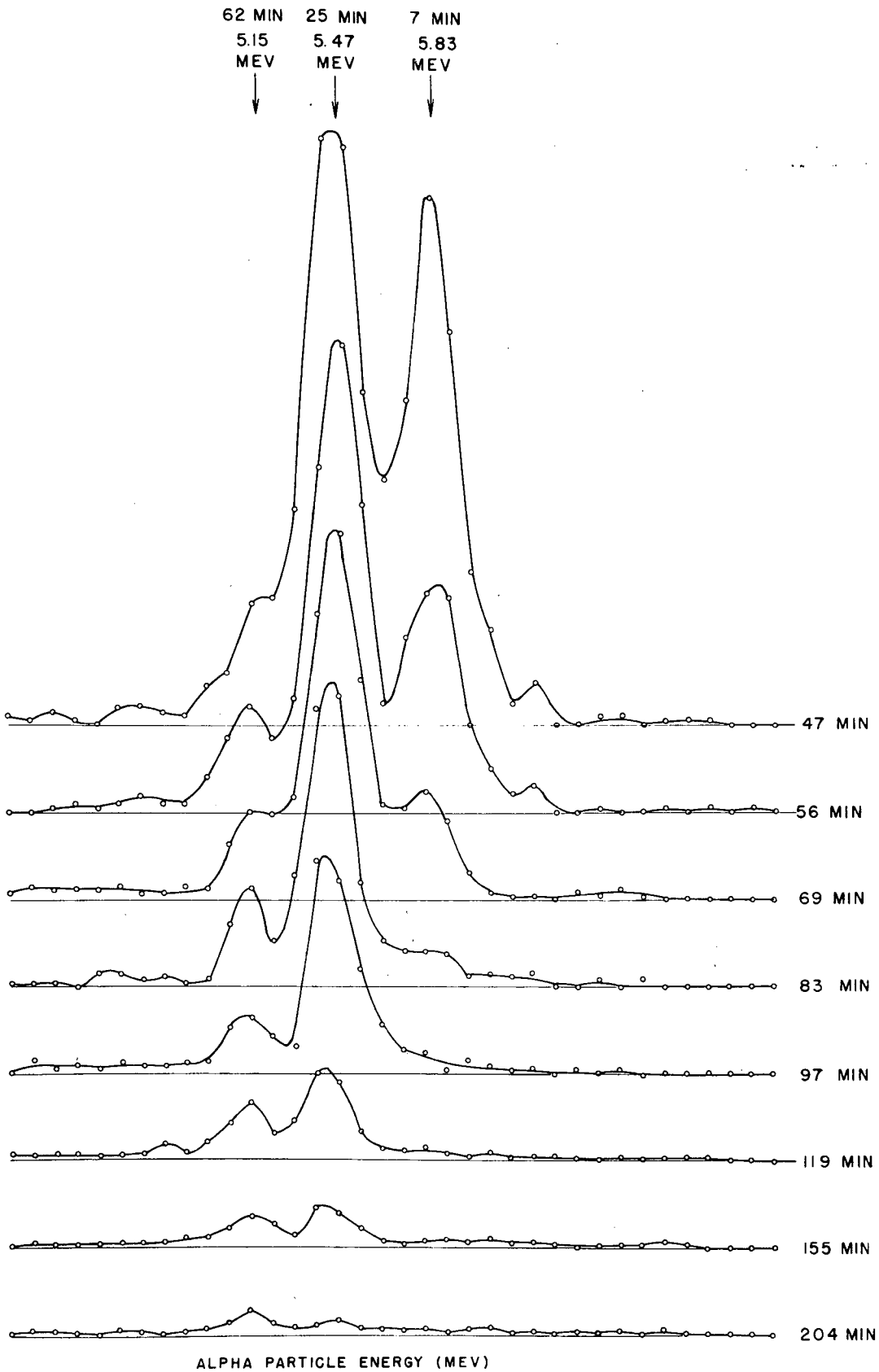
62 MIN
5.15
MEV
25 MIN
5.47
MEV
7 MIN
5.83
MEV
47 MIN
56 MIN
69 MIN
83 MIN
97 MIN
119 MIN
155 MIN
204 MIN
ALPHA PARTICLE ENERGY (MEV)

FIG. 5

Half-life and energy relationship.— One of the major points of interest in these artificially produced bismuth alpha-emitters is their relation in the systematics of alpha-decay. Reference is made to the paper[2] in which the trends in alpha-decay properties are discussed. In particular, alpha-emission in these bismuth isotopes is interpreted as a resumption of alpha-emission of the heavy bismuth isotopes, which is interrupted by the unfavorable energetics toward alpha-emission caused by crossing the region of 126 neutrons.

One of the generalizations that came from the study of alpha-decay systematics[2] was that the half-life decay energy relationships for the even-even nuclei were in quantitative agreement with alpha-decay theory, while other types deviated in the direction of prohibition in alpha-decay. The general cause for the prohibition is attributed to the existence of odd nucleons and is only slightly effected by spin change in the particle emission. One of the parameters in alpha-decay theory which cannot be evaluated independently to the precision required is the nuclear radius, and a significant point is that the even-even nuclei (and presumably the others) in a broad region can be described by the same function for nuclear radius, $r = 1.48\ A^{1/3} \cdot 10^{-13}$ cm. However, there are even-even nuclides which show forbidden alpha-decay if the radius is described by this function, and the obvious explanation is that the nuclear radius is smaller than would be calculated in this manner. Such nuclides are those which have 126 neutrons and less or those which decay into or through the region of 82 protons. The position of the bismuth alpha-emitters is somewhat difficult to assess since the quantitative treatment is complicated by the necessity of estimating the degree of prohibition due to odd nucleons. However, the degree of prohibition for the bismuth nuclides Bi^{214} to Bi^{210} is such that one must attribute part of the effect to abnormally low nuclear radius. This would be explainable

by nuclear radius change for decaying through lead (82 protons) and in some cases through 126 neutrons.

It would be of interest to compare the new light bismuth isotopes with the heavier ones to see if any trend in nuclear radius can be inferred. Unfortunately, one can have little confidence in the estimation of the alpha-decay half-lives for these new light bismuth isotopes, since they must be calculated from extremely rare alpha-branching of electron-capture processes. The number of electron-capture events for each alpha-emission must be calculated from the yield of the thallium decay product (from two electron-capture events), since it is not possible to resolve the decay curves of the bismuth fraction itself. Furthermore, one is faced with the general unsatisfactory situation of estimating numbers of electron-capture events. For present purposes we have attempted to resolve the K x-rays as the common denominator in all of the electron-capture processes and to assume one K x-ray per disintegration. This assumption could give low values if there is L-capture, and high values if there are K internal-conversion events. The counting yield for mercury K x-rays (70 kev) was taken to be 0.3% in argon-filled Geiger tubes, corrected to 100% geometry.

The alpha-decay branching ratios estimated for Bi^{198}, Bi^{199}, and Bi^{201} are shown in Table 2. Figure 6 shows a plot of the half-lives of bismuth isotopes with reference to a calculated line which would apply to these isotopes if their decay were unprohibited by the nuclear type (odd nucleons), and if the nuclear radius in this region could be described by the above mentioned expression. The highly forbidden nature of all of these alpha-emitters makes it seem likely that all have low nuclear radii. Since the prohibition due to odd nucleons is known to vary considerably,[2] it is only possible to guess at

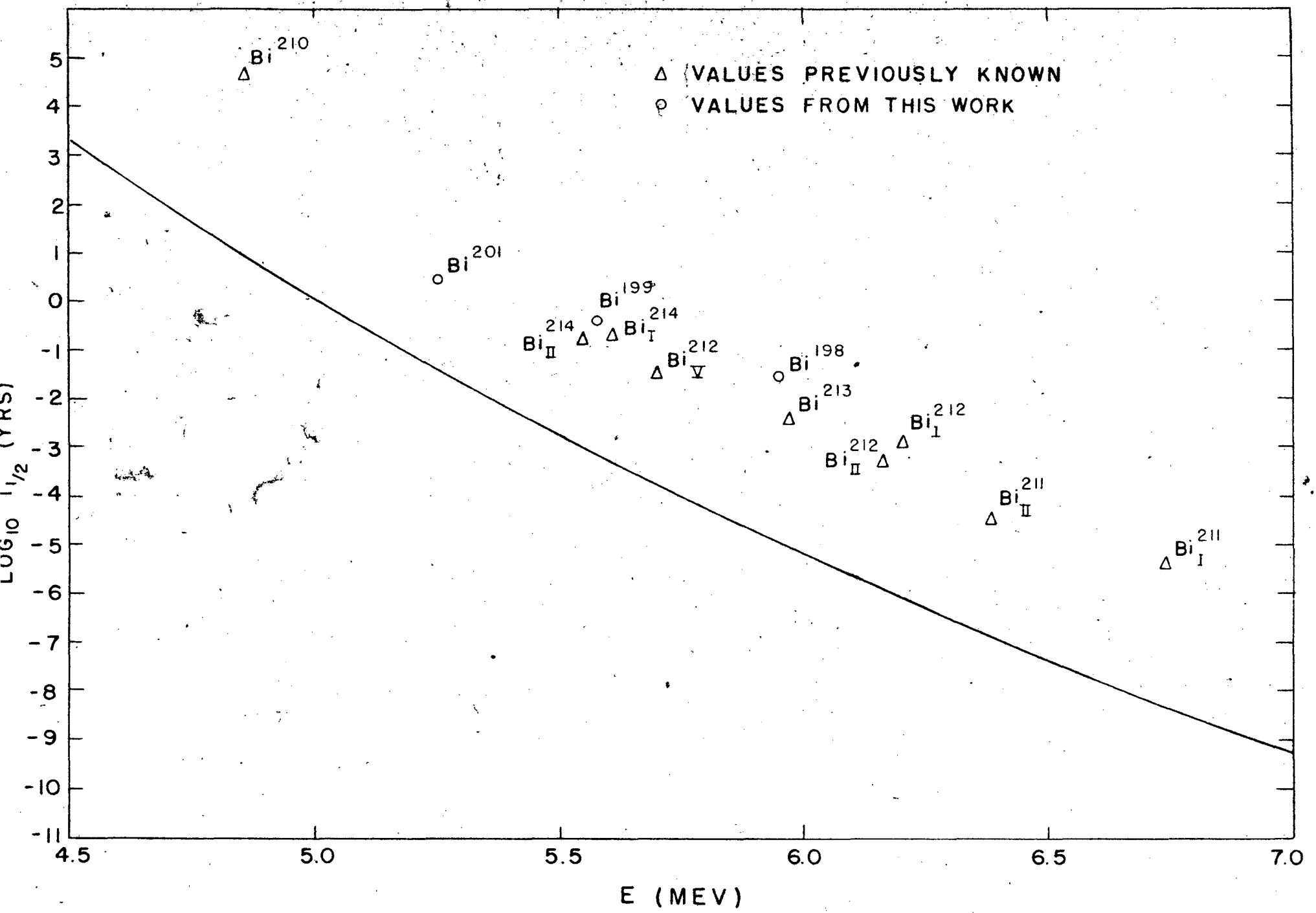
Δ VALUES PREVIOUSLY KNOWN
○ VALUES FROM THIS WORK
Bi 210
Bi 201
Bi 199
Bi II 214
Bi I 214
Bi V 212
Bi 198
Bi 213
Bi I 212
Bi II 212
Bi II 211
Bi I 211
LOG10 T1/2 (YRS)
E (MEV)
5
4
3
2
1
0
-1
-2
-3
-4
-5
-6
-7
-8
-9
-10
-11
4.5
5.0
5.5
6.0
6.5
7.0

FIG. 6

14274-1

Figure 6: Partial alpha half-life vs. alpha-decay energy. The curve gives values calculated for transitions unprohibited by nuclear type and assuming the nuclear radius equals $1.48\ A^{1/3} \cdot 10^{-13}$ cm.

the effect of nuclear radius. About all that can be said is that values lower by about 10% from those given by $r = 1.48\ A^{1/3} \cdot 10^{-13}$ cm seem to be required.

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