UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

DISCLAIMER

UCRL 646

UNCLASSIFIED

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

Contract No. W-7405-eng-48

RADIOACTIVE ISOTOPES OF BISMUTH

H. M. Neumann

April 6, 1950

Berkeley, California

Thesis

of

H. M. NEUMANN

to be submitted to the

UNIVERSITY OF CALIFORNIA GRADUATE DIVISION

Berkeley, California

RADIOACTIVE ISOTOPES OF BISMUTH

H. M. Neumann
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

April 6, 1950

INTRODUCTION

Five isotopes of bismuth occur in nature: the single stable isotope, Bi^{209}, and the four radioactive isotopes of mass numbers 210, 211, 212, and 214. Using particle accelerators providing helium ions up to 40 Mev in energy and deuterons up to 20 Mev in energy, it was possible to produce and identify two artificial radioactive isotopes, 6.4-day Bi^{206}[1,2,3] and 12-hr. Bi^{204}.[3] With the advent of the 184-inch Berkeley syncho-cyclotron and its hundreds of Mev energies, it was possible to form highly neutron-deficient bismuth isotopes extending more than ten mass units below stable Bi^{209}. It was observed in early work[4] that several of the isotopes formed at these high energies emit alpha-particles in competition with predominant electron-capture decay. It was known that the heaviest known isotopes (Bi^{210}, Bi^{211}, Bi^{212}, Bi^{214}) are measurably alpha-unstable, but that alpha-emission disappears for lighter isotopes (Bi^{209}, Bi^{206}, Bi^{204}). As part of the study of alpha-decay systematics[5] it seemed worthwhile to investigate the reappearance of alpha-activity at around mass number 200.

It soon became apparent that to make any successful isotopic assignments for these activities it would be necessary to identify all the bismuth isotopes lighter than mass number 204. Chapter I of this thesis is devoted to these investigations, and a discussion of the observed alpha-decay characteristics. Chapter II is devoted to some investigations carried out on heavier bismuth isotopes (mass numbers 207 to 210, inclusive).

CHAPTER I

ISOTOPES LIGHTER THAN MASS 204

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. The 6.4-day and 12-hour components could be resolved fairly easily, but resolution of activities shorter than 12 hrs. was impossible. That a number of components were present was apparent from the alpha-activities observed. 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. Furthermore, attempts of this type only proved the similarities in radiation characteristics of isotopes of similar half-life.

Because of this great complexity another means of identifying the isotopes present was necessary. The method which seemed most applicable in this case was the so-called "milking" procedure, that is, the removal of the daughter activities at periodic intervals.

The number of atoms of the daughter isotope that have grown into a pure parent after the time t is given by $N_2 = N_o \frac{1}{\lambda_1-\lambda_2} (e^{-\lambda_2 t} - e^{-\lambda_1 t})$ where N_o is the number of atoms of parent initially present, and λ_1 and λ_2 are the respective decay constants of parent and daughter. If the daughter is separated periodically at equal time intervals t, $\frac{1}{\lambda_1-\lambda_2} (e^{-\lambda_2 t} - e^{-\lambda_1 t})$ is just a proportionality factor relating N_2 and N_o. Since N_o drops off with the half-life of the parent, N_2 (i.e., the yield of daughter in a separation) must do likewise.

Thus the rate at which the yield of daughter decreases in successive fractions determines the half-life of the parent. This procedure can be extended to chains of activities of any length. In the present case the experiments were restricted to two member chains (Bi-Pb and Pb-Tl) and three member

chains (Bi-Pb-Tl).

For the above considerations to be valid experimentally it is necessary for the parent-daughter separation to be complete, and the length of time necessary to accomplish the separation must be short compared to the period of growth. No really good Bi-Pb separation was found that satisfied both of these conditions. Precipitation of $PbSO_4$ was satisfactory from a purity standpoint, but required too much time. Extraction separations utilizing dithizone did not give sufficient purity. Precipitation of BiOCl could be performed fairly quickly, but did not give complete separation. Even so, this last method seemed to be the best available.

Under the conditions used for the separation, greater than 99 percent of the bismuth was precipitated as BiOCl, while the latter precipitate retained 8 percent of the lead. As an indication of the effect this retention of daughter has on a half-life determination, Fig. 1 illustrates separations done at intervals equal to one-half, once, and twice the half-life of the parent. In practice, the half-life was first roughly determined from the experimental points, the correction then calculated for this half-life and separation interval, and the correction applied to the experimental points to determine the correct half-life. In all experiments involving a Pb-Bi separation this correction was made. Further discussion of yield determination and correction of data will be given, when necessary, in relation to specific isotopes.

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

Fig. 1. Effect of retention of eight percent of daughter with parent. Lines represent actual half-life of parent, circles represent the expected experimental values. Curve A, separations at intervals equal to one-half the half-life of the parent; Curve B, separations at intervals equal to half-life of the parent; Curve C, separations at intervals equal to twice the half-life of the parent.

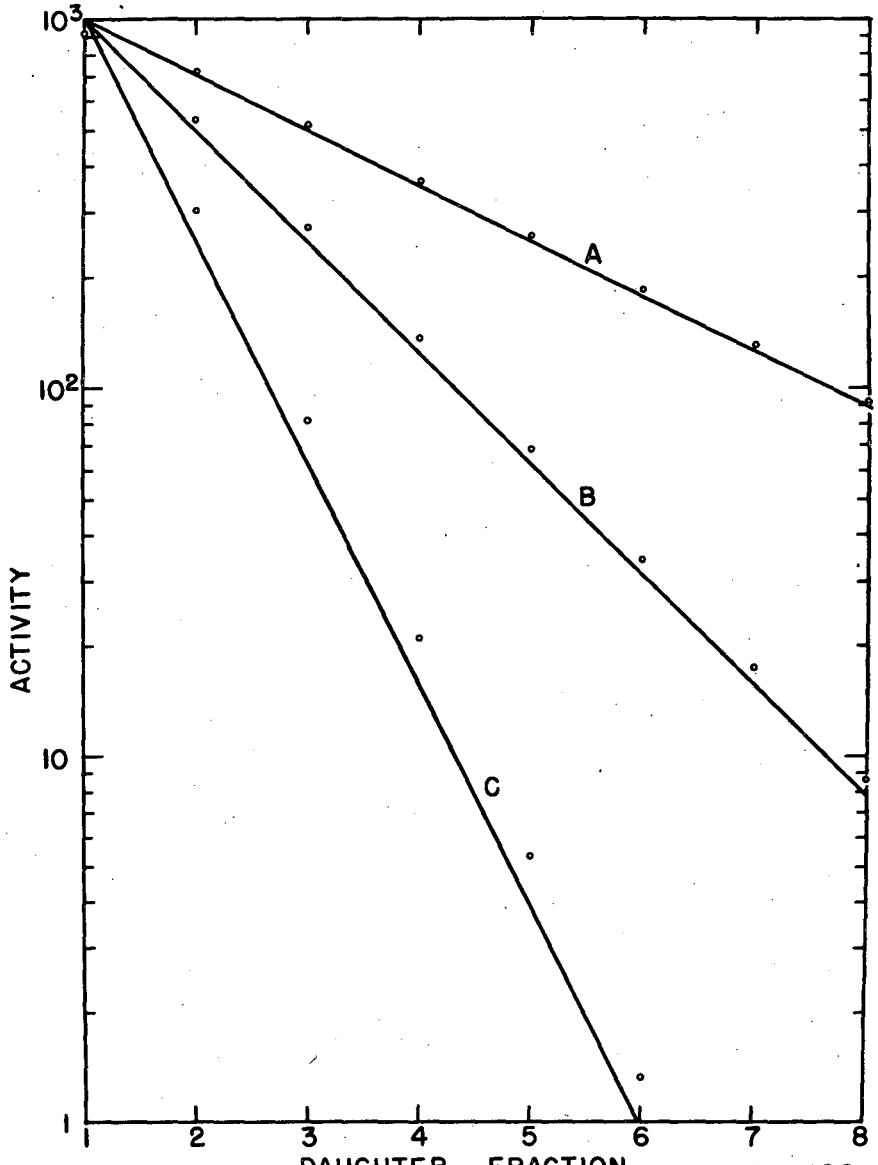


FIG. 1

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 varies with the time of exposure, 10 percent is obtained in 5 min., 25 percent in 15 min., and 65 percent in 45 min. 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 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.[6]

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

Table I

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.	110±20 min.		12±1 hr.
Pb	~25 min.	78±5 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

Since the activities were studied by their electron capture genetic relationships, the activities will be discussed one chain at a time (each identified by its mass number).

Bi^{203}-Pb^{203}

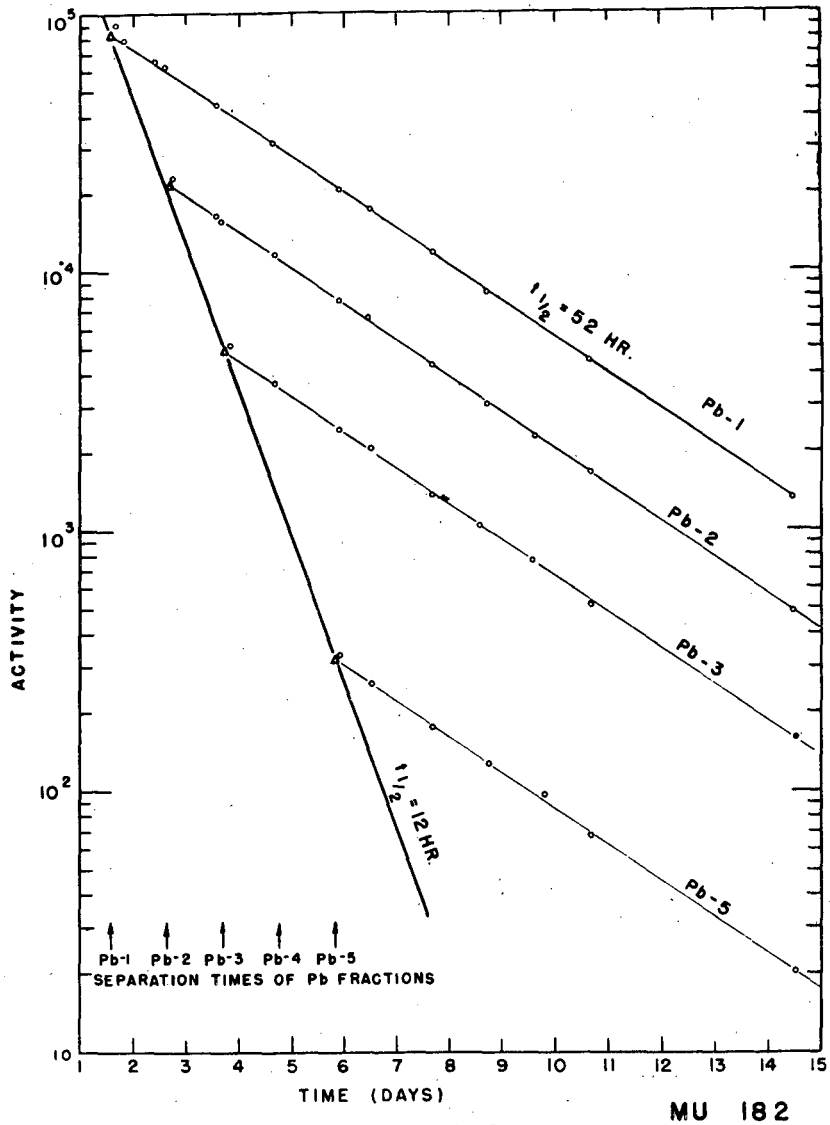
The 52-hr. lead activity was well known prior to this work and was assigned to mass number 203 on the basis of yields from various nuclear reactions.[3,7] The bismuth parent of this lead activity was produced at all energies used (40-Mev protons was lowest energy) as evidenced by the fact that 52-hr. Pb could be removed from a bismuth fraction that had been previously purified of lead. The radiation characteristics of this activity so obtained agreed well with those previously published. Preliminary experiments indicated the life of the parent to be the order of one day.

Two sets of "milkings" were performed to determine the half-life exactly. In the first experiment lead was irradiated for about one hour 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. In removing lead activities from the bismuth fraction the previously described chemistry was employed; that is, bismuth oxychloride was precipitated leaving lead in solution. The lead was then precipitated as $PbCrO_4$. Further purification was obtained by dissolving the chromate by reduction with H_2O_2 in dilute acid, adding inactive bismuth, and repeating the separation. After the second precipitation of $PbCrO_4$ the latter was dissolved and a definite aliquot of the solution evaporated on a stainless steel disk for counting. The decay curves were then followed in order to resolve the 52-hr. period. These decay curves are shown in Fig. 2 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 Fig. 2 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 due to 68-min. Pb^{204m}.

The data shown in Fig. 2 has already been corrected for 8 percent retention of the daughter. In addition to the assumption that this retention is 8 percent in all samples, the implicit assumption is made that if loss of lead occurs during the other operations this loss is the same in all samples.

A second set of "milkings" was performed at 12.5-hour intervals, and the procedure was altered so that the last mentioned assumption was not necessary. Exactly 20 mg. of lead was added prior to the Bi-Pb separation. Following precipitation and separation of BiOCl, $PbCrO_4$ was precipitated from the supernatant. The $PbCrO_4$ was dissolved in dilute H_2SO_4 by reduction with H_2O_2.

Fig. 2. Data showing the genetic relationship between 52-hr. Pb^{203} and its 12-hr. bismuth parent.


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

MU 182

FIG. 2

$PbSO_4$ was then precipitated by evaporation to fumes of SO_3, followed by dilution. The sulfate was filtered through a small filter paper and dried. This served as the sample for counting, and the amount of lead was determined by weight. The yield through the chemical operations was thus known, and no assumption regarding its constancy in all samples was necessary.

The result of this experiment, corrected for daughter retention, is shown on the right side of Fig. 3. On the left is shown again the result of the first experiment, both corrected and uncorrected. The two values obtained are 12.5 hr. and 11.5 hr. This agreement, and all later experiments, justify the assumption of constancy of yield. The fact that agreement was obtained using intervals varying by a factor of two also indicates there are no serious flaws in the method.

A bismuth activity of 12-hr. half-life had previously been reported as Bi^{204},[3] 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.[3] Recently Karraker and Templeton[8] have prepared a 3.8-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. In addition they found the 12-hr. bismuth with the 52-hr. Pb^{203} daughter, and this pair grew from a polonium of about 48-min. half-life, making a consistent picture if this is assigned to Po^{203}.

Bi^{202}-Pb^{202}-Tl^{202}

The isotope of Bi at mass number 202 could not be identified by the techniques used in these studies because its decay product, Pb^{202}, is undoubtedly very long-lived. Neither Pb^{202} nor 12-day Tl^{202}, which would result from

Fig. 3. Two determinations of the half-life of Bi^{203}.

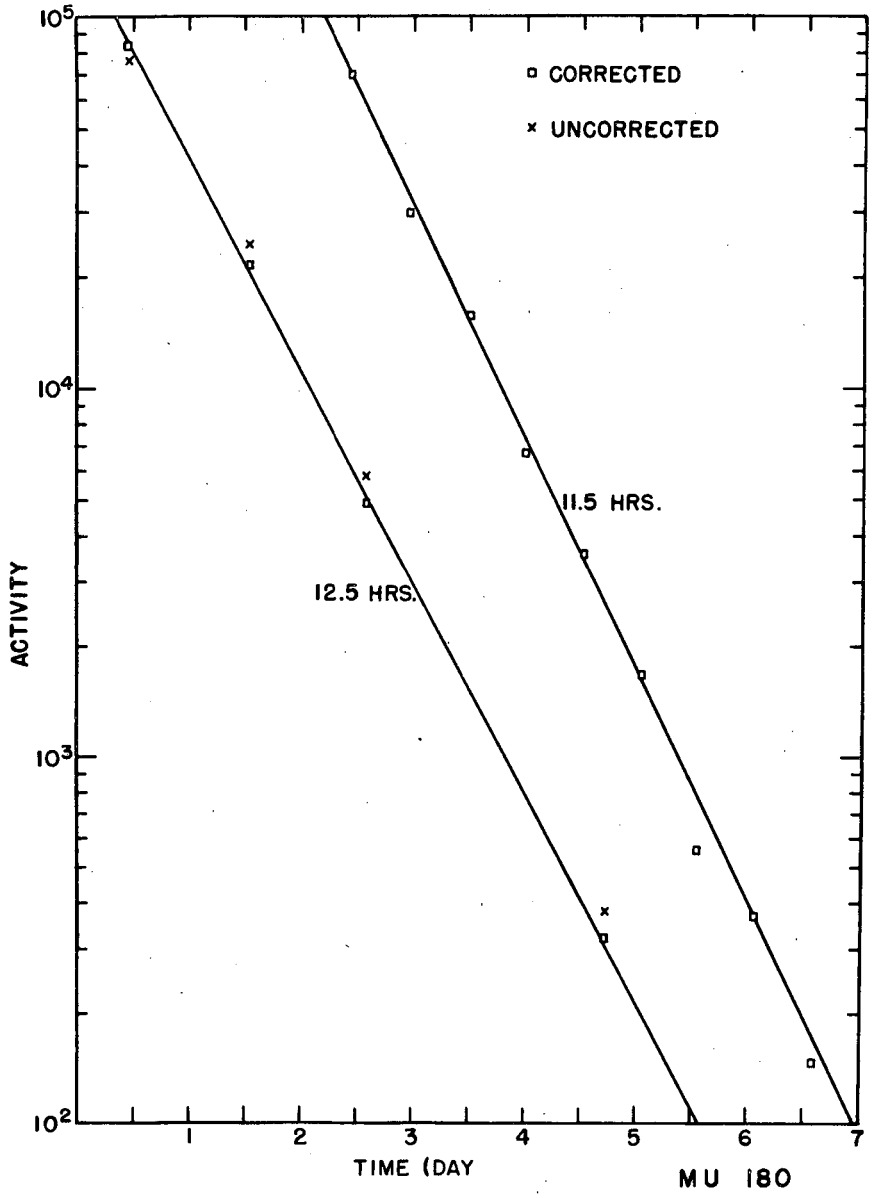

10^5
10^4
10^3
10^2
ACTIVITY
CORRECTED
UNCORRECTED
11.5 HRS.
12.5 HRS.
1
2
3
4
5
6
7
TIME (DAY
MU 180

FIG. 3

electron-capture decay of Pb^{202}, were observed in these experiments.

Templeton, Howland, and Perlman[3] set a minimum half-life of 500 years for Pb^{202} on the basis of the estimated yield from a thallium irradiation with 20-Mev deuterons. From the work of Karraker and Templeton[8] Bi^{202} is a 95-min. activity decaying by electron capture. In the present experiments the combined lead decay products from several bismuth fractions were examined for the growth of the 12-day thallium. 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 at 950 years.

Bi^{201}-Pb^{201}-Tl^{201}

Attempts to apply the method used for Bi^{203}-Pb^{203} for other isotopes of Bi proved difficult since the decay curve of the Pb daughter fraction was generally dominated by 68-min. Pb^{204m} and 52-hr. Pb^{203}, each growing from its respective 12-hr. parent. The way out of this difficulty was to allow each lead daughter fraction to decay for a definite period and then remove the thallium granddaughters that had grown in. Since the activity in the thallium fraction depends on the amount of lead parent present at its separation from bismuth, the activities of the successive thallium fractions will drop off with the half-life of the bismuth grandparent.

The lifetime of the intermediate lead can be determined in exactly the same way by purifying Pb and then removing Tl at periodic intervals.

The chemical separation of lead and thallium was accomplished by adjusting the lead solution to 6N HCl and extracting the thallium into diethyl ether as $TlCl_3$. Prior to the extraction a few drops of $KMnO_4$ solution were added to insure the oxidation of thallium to the thallic state. Thallium does not extract in the thallous state. Normally the aqueous solution was about 10 ml in volume, and it was twice extracted with equal volumes of ether. The ether layers were

then combined and washed with 10 ml 6N HCl. The ether was evaporated to a small volume, and then the remainder evaporated on a stainless steel disk for counting. In the early experiments no attempt was made to determine the chemical yield, but the yields were assumed constant. Occasional accidental losses during the evaporation step made some means of measuring the overall yield desirable. This was accomplished by adding a definite amount of the β^- active 3-yr. Tl^{204} as a tracer prior to the Pb-Tl separation. In the later experiments this method was applied advantageously.

The activities corresponding to Bi^{198}, Bi^{199}, and Bi^{200} might be determined by applying the "milking" method since Tl^{198}, Tl^{199}, and Tl^{200} were known.[9] Preliminary experiments indicated these activities could be detected by the two-step "milkings," and in addition a 72-hr. Tl activity was detected. As previously mentioned the 12-day Tl^{202} was not detected. The most likely assignment for the 72-hr. activity was then Tl^{201}.

By separations at appropriate intervals the 72-hr. activity could be obtained in essentially pure form.

The decay of this activity is shown in Fig. 4. Some of the radiation characteristics 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. Fig. 5 is the absorption curve taken in lead, resolved into components 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. There is apparently no electromagnetic radiation more energetic than the 210-kev γ-ray. The absorption in aluminum is shown in Fig. 6, resolved

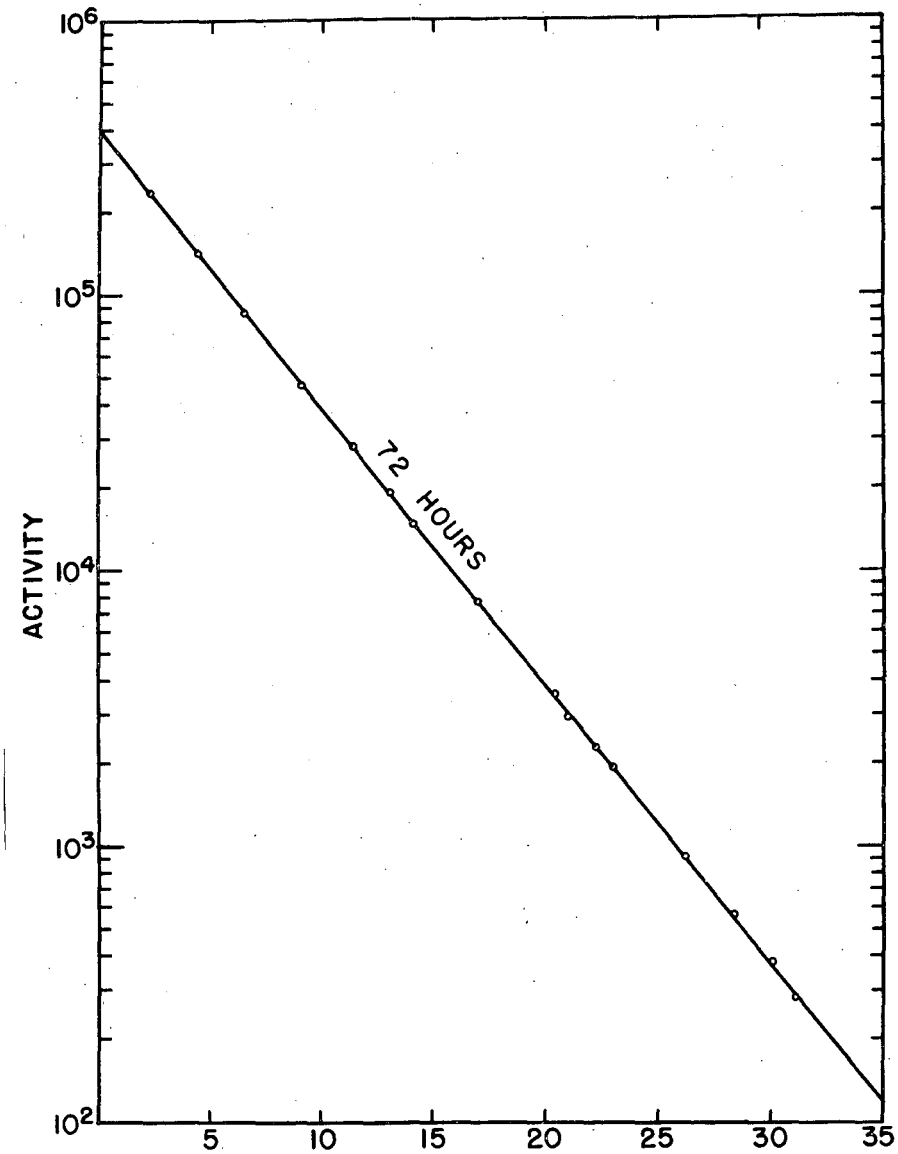

10^6
10^5
10^4
10^3
10^2
ACTIVITY
72 HOURS
5
10
15
20
25
30
35
TIME (DAYS)
MU 181

FIG. 4

Fig. 5. Lead absorption curve for Tl^{201} showing K x-rays (A) and a 210-kev gamma-ray (B).

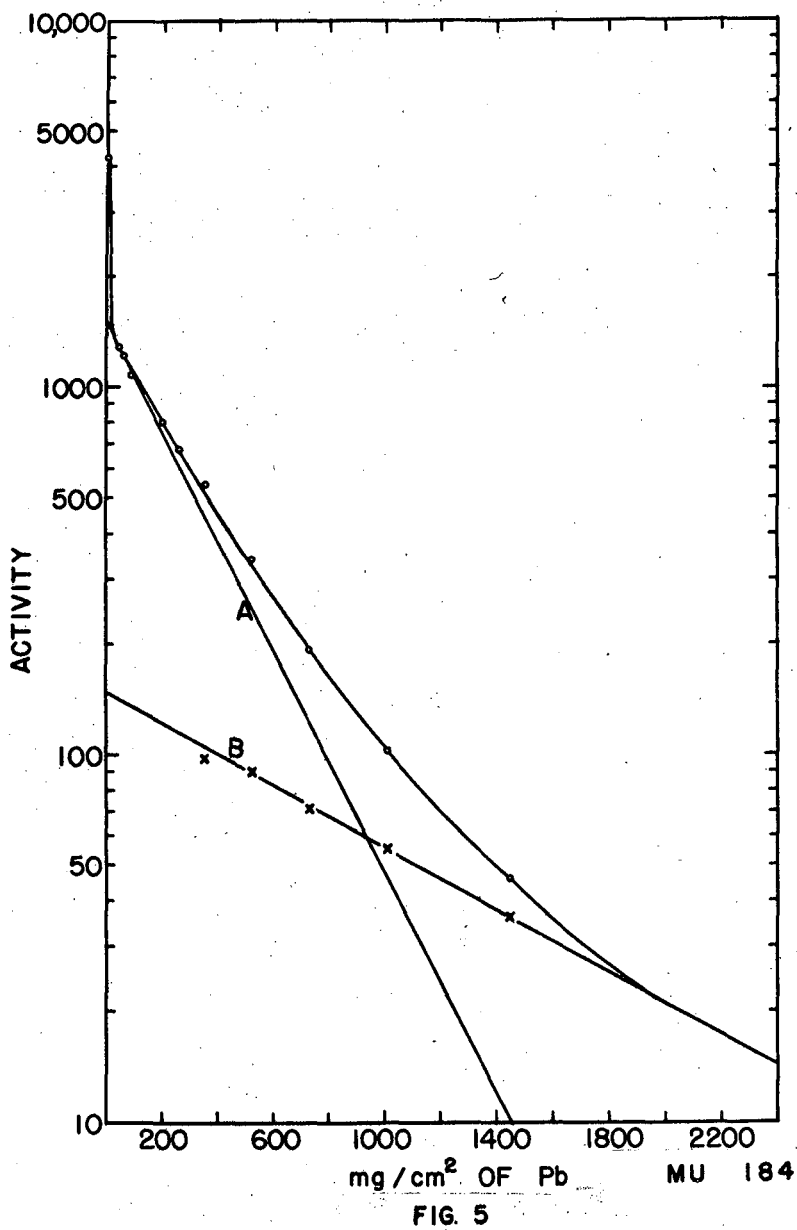

10,000
5000
1000
500
100
50
10
ACTIVITY
A
B
200
600
1000
1400
1800
2200
mg / cm² OF Pb
MU 184

FIG. 5

Fig. 6. Aluminum absorption curve for Tl^{201} showing total electromagnetic radiation (A), K x-rays (B), L x-rays (C), and electrons (D).

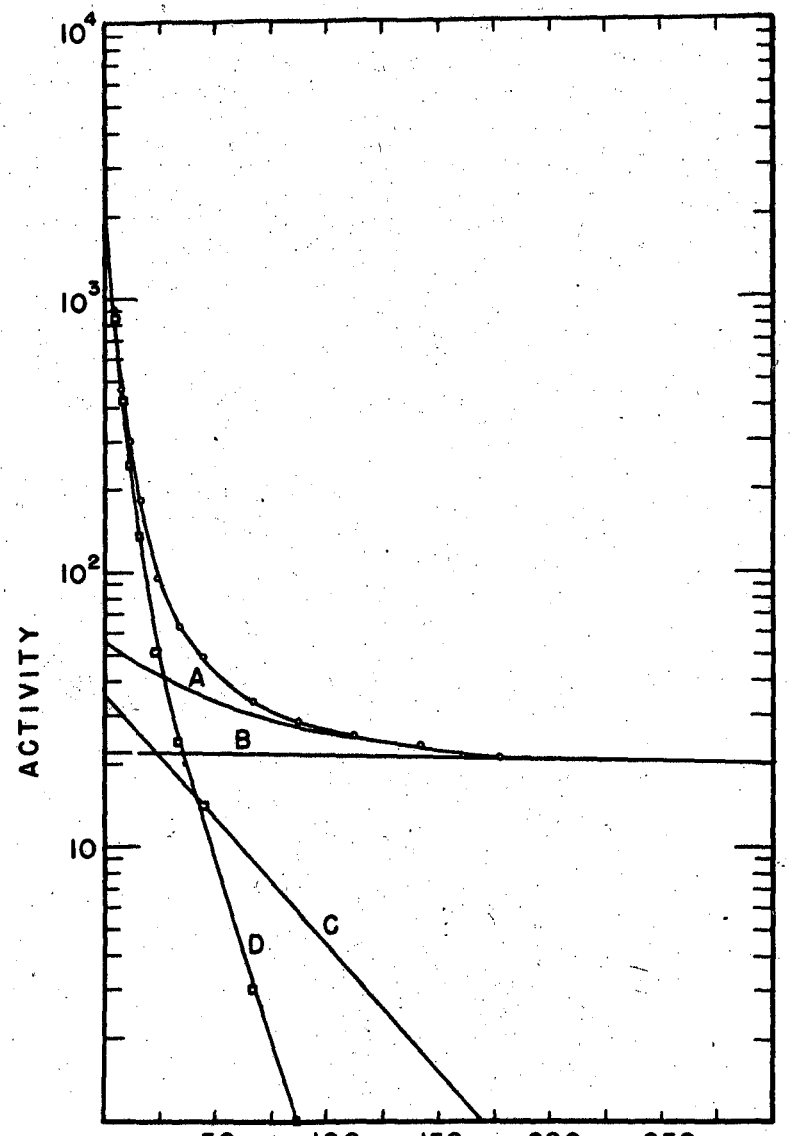

10^4
10^3
10^2
10
ACTIVITY
A
B
C
D
50
100
150
200
250
mg/cm² OF Al
MU 187

FIG. 6

into electron, L x-ray, and K x-ray components.

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 percent for the counting yield after correcting for geometry loss, the indicated internal conversion is 60 percent. 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.

The use of the double "milking" procedure to determine the half-life of Bi^{201} led to results suggesting a value of about 90 min. However, from other considerations, which will be discussed later in detail, the 62-min. alpha-emitter seemed best assigned to mass number 201, and consideration of its half-life and alpha-decay energy made it seem likely that the 62-min. activity should decay predominantly by electron capture. 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}.

Several experiments were performed in an attempt to show that the growth of Tl^{201} proceeded from two bismuth parents. The results are shown in Fig. 7. The separation intervals used were variously 50 min., 60 min., and 2.5 hours. The correction to be applied for daughter retention is difficult to evaluate,

Fig. 7. Four determinations of the bismuth ancestor of 72-hr. Tl^{201}. Solid lines are the best 90-min. line for each experiment. Dotted lines are the best two-component curves fitting the data.

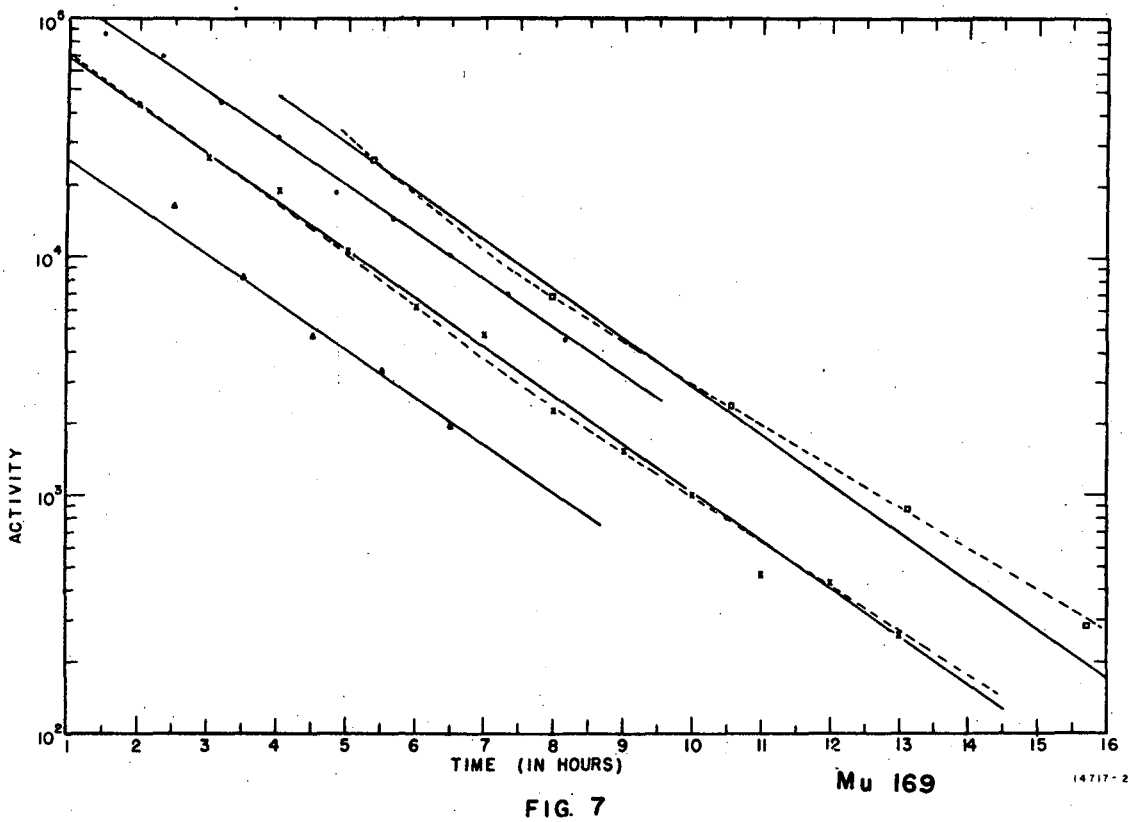


FIG. 7

since it would be dependent on the number and lives of the parents. In fact, the 8 percent retention factor has the unfortunate property of straightening an actual two component curve, making it even more difficult to observe the complex nature of the curve. The correction that has been applied is that for a 90-min. parent. For each experiment the best 90-min. line has been drawn, and for the two experiments in which the separations were carried out at later times, when presumably the 1-hr. component had largely decayed, the possible two-component curve is indicated by a dotted line. Of these two the experiment employing a 2.5-hr. "milking" interval indicates the longer component to be about 110 min.

Since in the method employed all separations are made from the same parent solution, systematic errors are very possible. It is possible that all of these have not been sufficiently compensated for in our corrections, and that the observed half-life will be dependent on the "milking" interval. It will be noticed that in the two experiments on Bi^{203}, a longer half-life (by 10 percent) accompanied the longer separation interval.

To eliminate the possibility of systematic error an experiment of the following type was performed. Bismuth was purified from the target in the usual manner, and then divided into six equal portions. Lead was removed successively from these portions at hourly intervals, only one lead separation being made from each portion. Thallium was later removed as usual from the lead fractions. The activity now should increase in successive thallium fractions, and plotting the activity of the latter against time should retrace the growth of the lead daughter in the bismuth parent. The lead daughter was known to have an 8-hr. half-life, so that theoretical curves for the growth of this activity from various parents could be constructed (Fig. 8). The curves are all normalized to intersect at the time of the first separation, and the experimental results are normalized so that the first point is at this intersection. This is a relatively insensitive method, and requires somewhat more confidence in the individual points than is

Fig. 8. Growth of 8-hr. Pb^{201} from its bismuth parent, the data being obtained from yields of thallium daughter. The curves are the theoretical growth curves of an 8-hr. activity from parents of the indicated lives.

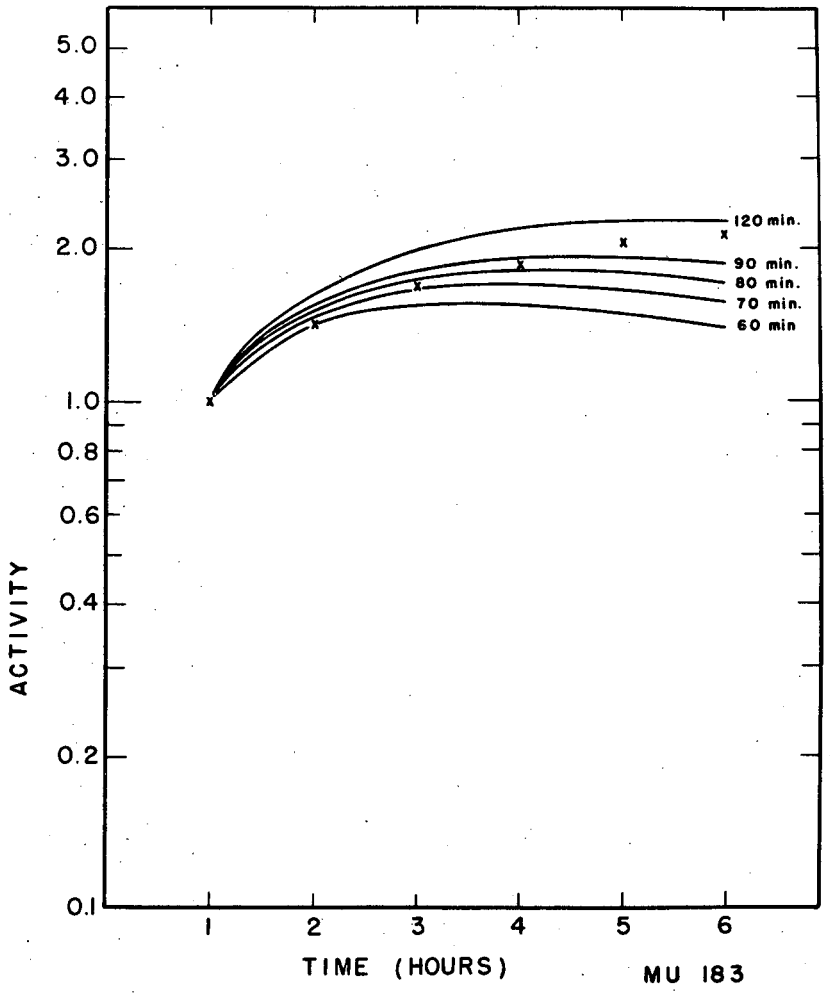

5.0
4.0
3.0
2.0
1.0
0.8
0.6
0.4
0.2
0.1
ACTIVITY
120 min.
90 min.
80 min.
70 min.
60 min
1
2
3
4
5
6
TIME (HOURS)
MU 183

FIG. 8

justified. The data fits the 90-min. curve if an experimental error of 15 percent is allowed, and the error may well be this large. The definite trend of the experimental points away from any of the theoretical curves would be the behavior expected for growth from two isomers, and the result is certainly consistent with this hypothesis.

The inherent inaccuracies of these experiments make it possible to fit the data equally well to a single isotope for Bi^{201} of 90-min. half-life, or to two isomers of 1- and 2-hr. half-lives.

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 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. 9 (a). 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. 9 (c), showing again that the 27-hr. thallium and 7-hr. thallium lie at lower mass numbers. Finally, as shown in Fig. 9 (b), 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 the 68-min. 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. 9 (a), (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

Fig. 9. Activities resulting from irradiation of lead with 40-Mev protons.

(a) Bismuth alpha-activity 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).

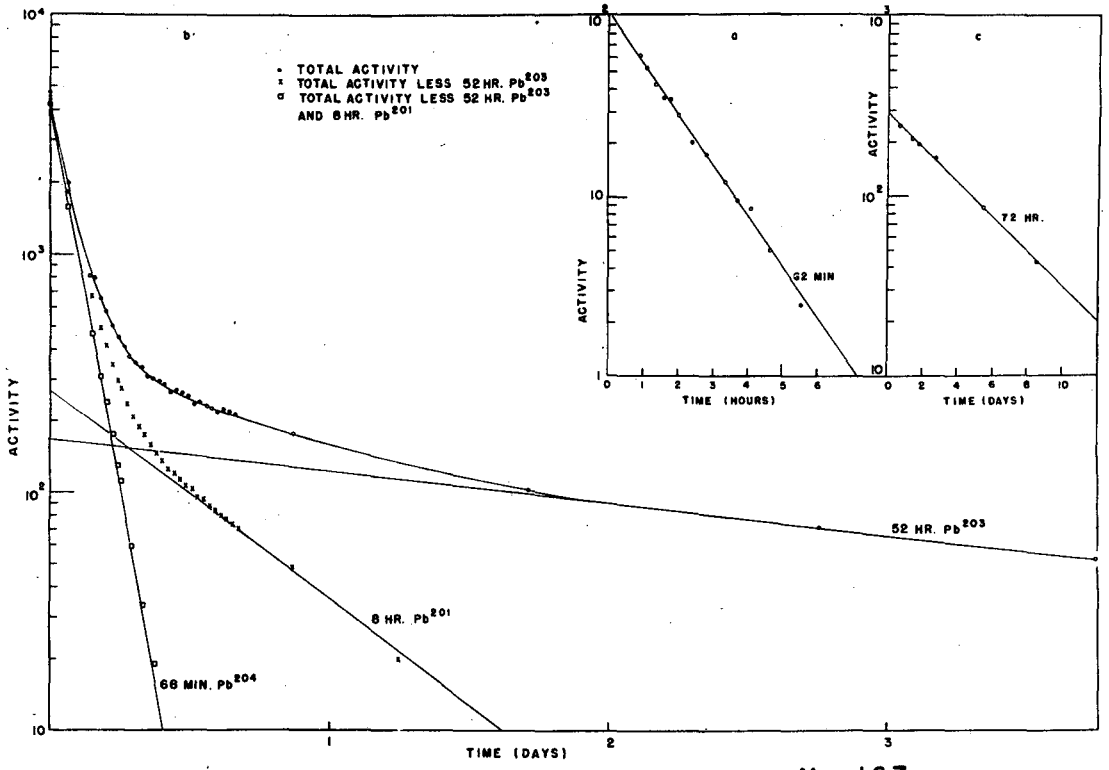

b
• TOTAL ACTIVITY
x TOTAL ACTIVITY LESS 52 HR. Pb203
o TOTAL ACTIVITY LESS 52 HR. Pb203 AND 8 HR. Pb201
a
62 MIN
c
72 HR.
ACTIVITY
TIME (HOURS)
TIME (DAYS)
52 HR. Pb203
8 HR. Pb201
66 MIN. Pb204

FIG. 9

Mu 167

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 curves could be resolved into only the two components. These data are shown in Fig. 10 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 hours.

Bi^{200}-Pb^{200}-Tl^{200}

The assignment of this isotope is based upon the assignment[9] 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 in the presence of large amounts of the 72-hr. period. Fig. 11 shows the decay of a thallium sample prepared in the following way: lead was irradiated with 150-Mev protons, bismuth was removed, the daughter lead was separated from the bismuth parent after a 1-hr. growth period, the lead fraction was then allowed to decay for 35 hours, following which thallium was removed. The decay of the thallium was followed, both without absorbers and through 11.5 g/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

Fig. 10. Half-lives of lead parents of 72-hr. Tl (o) and of 27-hr. Tl (□). Data obtained from yields of respective Tl daughters.

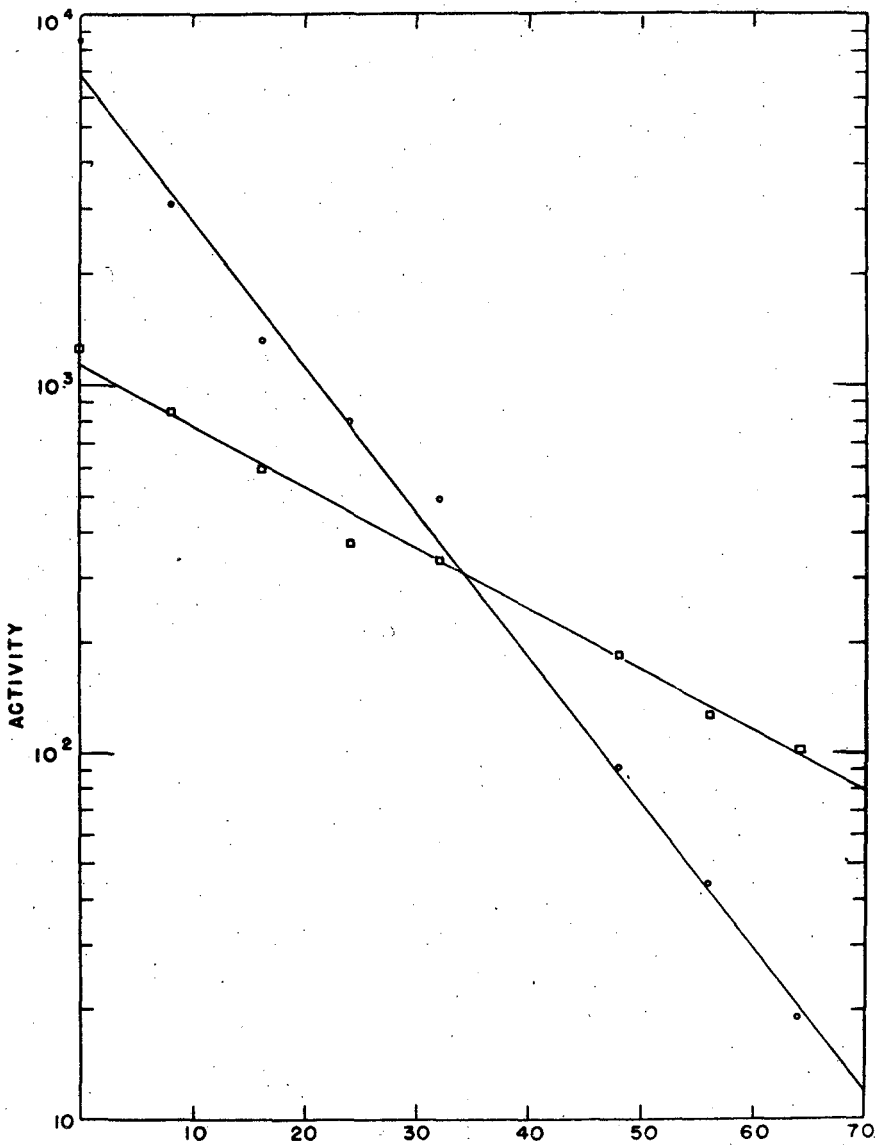


FIG. 10

Fig. 11. Decay curves of 27-hr. Tl^{200} and 72-hr. Tl^{201} mixture.

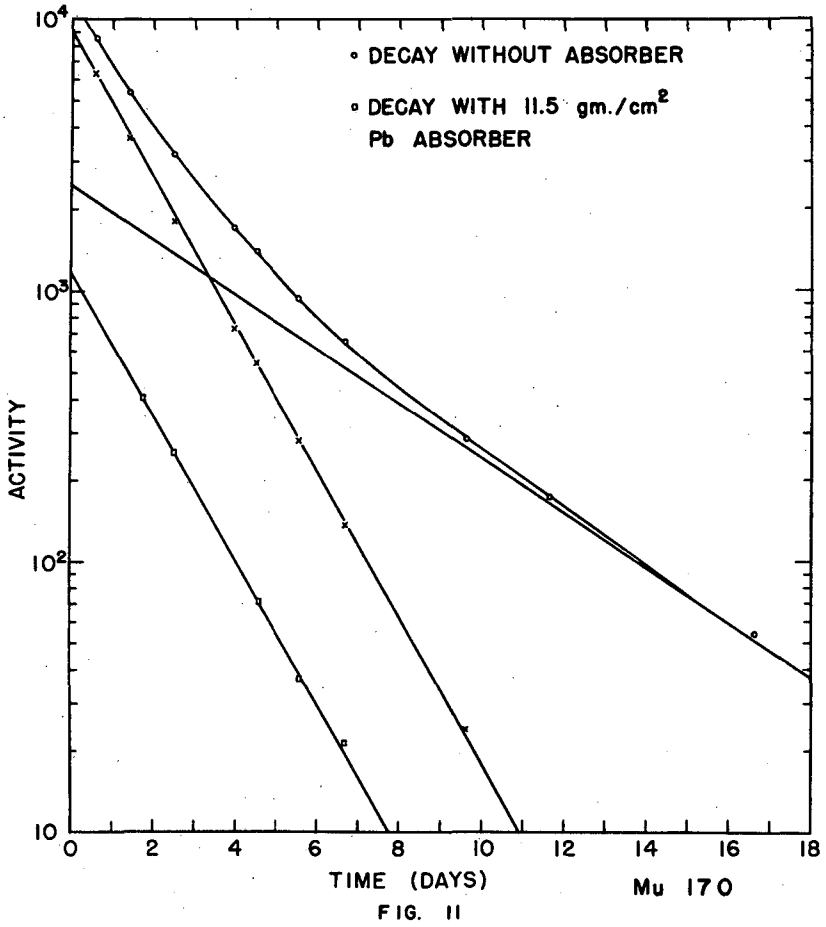

DECAY WITHOUT ABSORBER
DECAY WITH 11.5 gm./cm²
Pb ABSORBER
ACTIVITY
TIME (DAYS)
Mu 170

FIG. 11

abundance. Gold when irradiated with 38-Mev helium ions should produce the 27-hr. period by the (a,n) reaction according to earlier work;[9] 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 conversion 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. 10 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. 2 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 g/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. 12 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}.

Fig. 12. Data showing genetic relationship between 27-hr. Tl^{200} and a 35-min. bismuth.

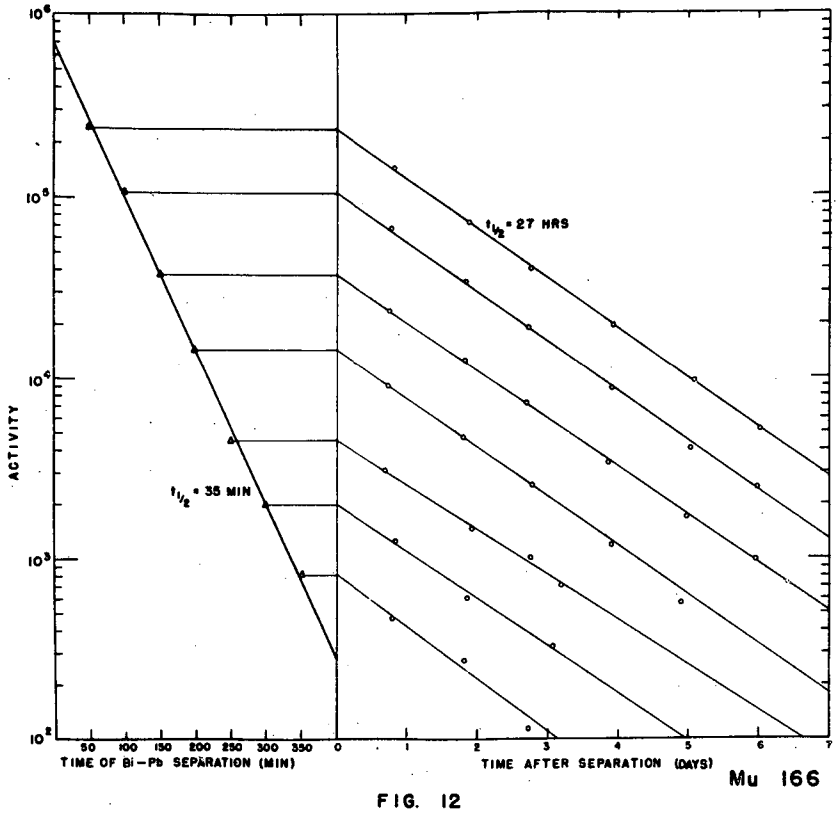

ACTIVITY
10^6
10^5
10^4
10^3
10^2
$t_{1/2}$ = 27 HRS
$t_{1/2}$ = 35 MIN
50 100 150 200 250 300 350
TIME OF Bi–Pb SEPARATION (MIN)
0 1 2 3 4 5 6 7
TIME AFTER SEPARATION (DAYS)
Mu 166

FIG. 12

Bi^{199}-Pb^{199}-Tl^{199}

The assignment of the decay chain at mass number 199 rests on the assignment of the 7-hr. Tl^{9} produced by alpha-particle bombardment of gold. The method of determining the lives of the Bi and Pb activities is the same as that already discussed.

In order to determine the half-life for Pb^{199}, a Pb fraction was obtained by decay of Bi, and subjected to successive removals of thallium at 60-min. intervals, and the yields of the 7-hr. component were used to define the half-life of the Pb^{199} parent. The results of two such experiments are shown in Fig. 13. The first, in which the yields of Tl were monitored with Tl^{204} tracer, gives a value of 79 min. The second, in which yields were not monitored (note the one obviously low point), gives a value of 77 min. This is excellent agreement, and the 78-min. average is taken as the best value.

To determine the life of the Bi ancestor, Pb was milked at 20-min. and 10-min. intervals in various experiments. These lead fractions were allowed to decay for 4 hours and thallium then removed. The 4-hr. growth period was chosen since a 7-hr. activity growing from a 78-min. parent will be at a maximum at this time. The 27-hr. and 72-hr. activities do not reach a maximum until about 30 hours, so that although all three thallium activities are present, there is considerable enrichment of the 7-hr. activity. This is illustrated in Fig. 14. In such a sample a 160-kev conversion electron, as determined by the crude β-ray spectrometer, appeared as the most characteristic radiation of the 7-hr. activity. The 7-hr. Tl^{199}, prepared by alpha-particle bombardment of gold, also showed this characteristic electron, indicating the activities are identical.

Fig. 15 shows three determinations of the life of Bi^{199} by obtaining the yield of 7-hr. Tl^{199} at the time of Pb-Tl separation. The first experiment was performed using a "milking" interval of 20 min., and the procedure was that

Fig. 13. Half-life of lead parent of 7-hr. Tl^{199} as obtained from yields of the latter. Experiment A, 79 min.; Experiment B, 77 min.

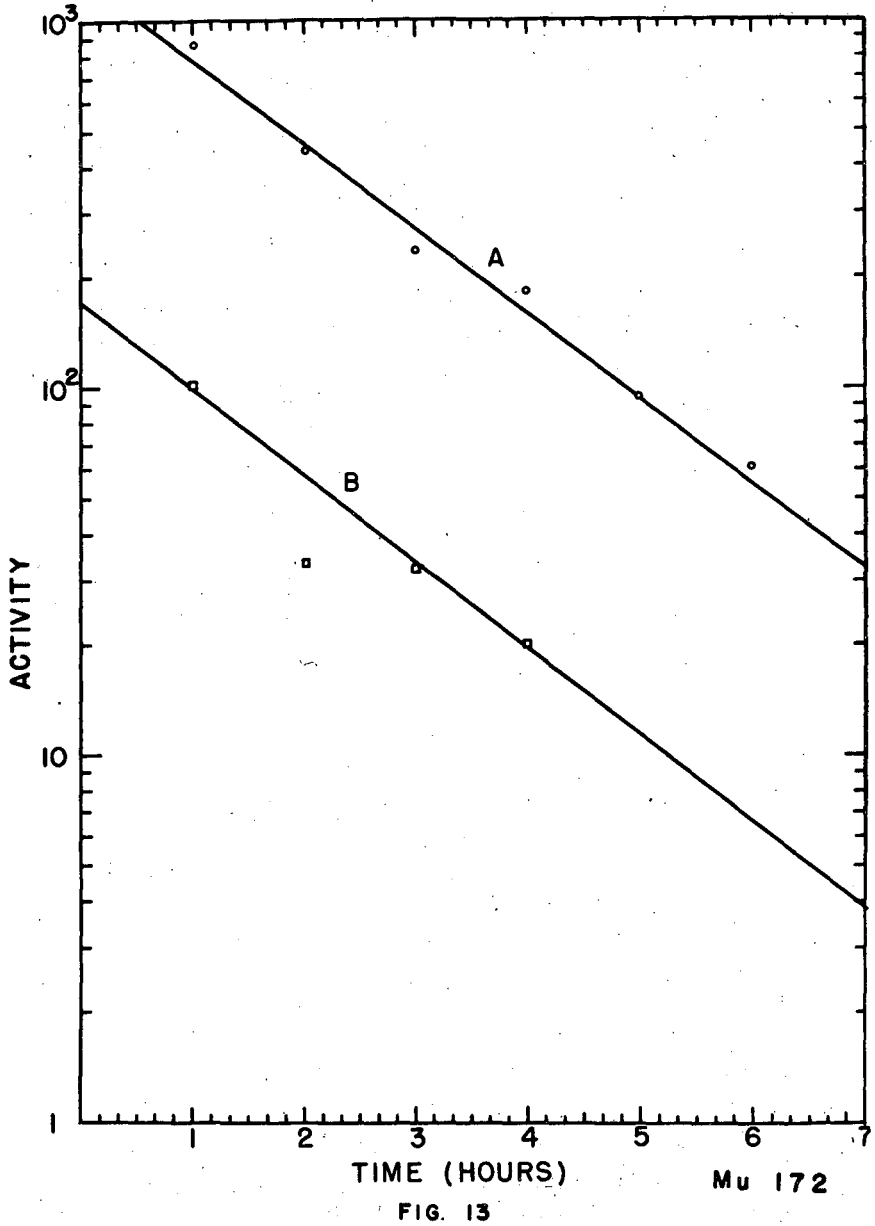


FIG. 13

Fig. 14. Decay curve of a mixture of 7-hr. Tl^{199} (A), 27-hr. Tl^{200} (B), and 72-hr. Tl^{201} (C).

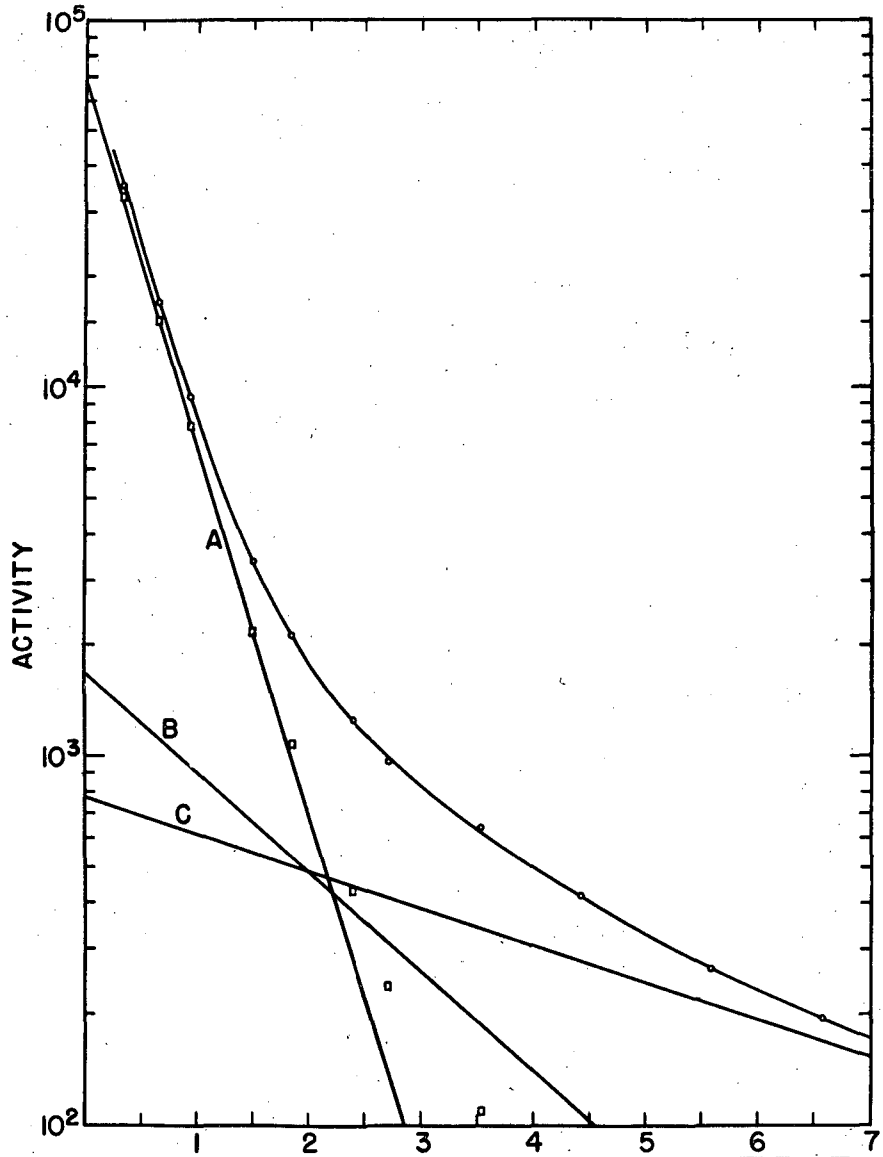


FIG. 14

Fig. 15. Three determinations of the half-life of Bi^{199}. Experiment A, 25 min.; Experiment B, 22 min.; Experiment C, 22 min.

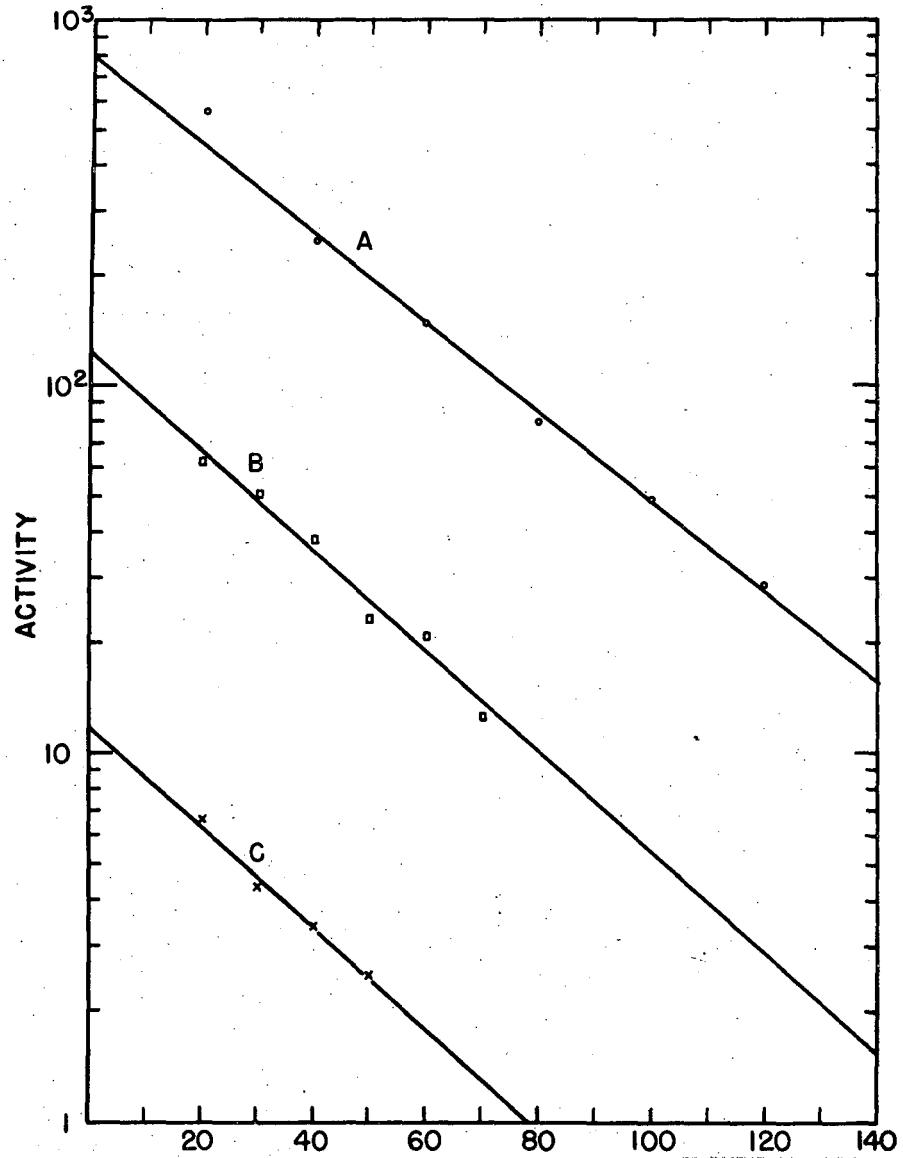


FIG. 15

already described. The second and third were done at 10-min. intervals. This latter interval did not allow time for the usual BiOCl scavenge as part of the lead purification, and for this reason one has somewhat less confidence in the result even though the thallium fractions were of sufficient purity to allow easy resolution of the 7-hr. activity.

The three experiments give, respectively, values of 25 min., 22 min., and 22 min. The 25 min. value is preferred both because of the mentioned experimental disadvantages in the last two determinations, and because of the observation of a 25-min. alpha-emitting period.

Bi^{198}-Pb^{198}-Tl^{198}

It had been previously observed that a 1.8-hr. thallium[9] results from the irradiation of gold with 38-Mev helium ions, and that the most likely assignment for this activity is Tl^{198}. An attempt was made to see if this thallium activity could be linked through electron-capture processes to one of the light bismuth alpha-emitters, in particular that reported[4] with a 9-min. period.

For this purpose two similar experiments were performed. In each case a 10-min. irradiation of lead with 150-Mev protons was followed by rapid bismuth isolation (about 25 min.), and lead removal from the bismuth fraction at 10-min. intervals. As previously mentioned the use of this interval necessitated the omission of the BiOCl scavenge, resulting in greater possibility of bismuth contamination of the lead fraction. Traces of this impurity might also extract with the thallium, and be observed in the thallium decay curve.

After allowing each lead fraction to decay for 50 min., thallium was isolated and the decay of the latter followed in an attempt to resolve any 1.8-hr. component present. Fig. 16 shows the early decay of the first thallium fraction obtained, and indicates the small amount of activity attributable to 1.8-hr. Tl. The resolution of the complex decay was made by first subtracting

Fig. 16. Decay curve of thallium fraction containing 1.8-hr. Tl^{198}. Components resolved from the curve are A, 1.8-hr. Tl^{198}; B, 7-hr. Tl^{199}; C, 27-hr. Tl^{200}; D, 72-hr. Tl^{201}; E, bismuth impurity.

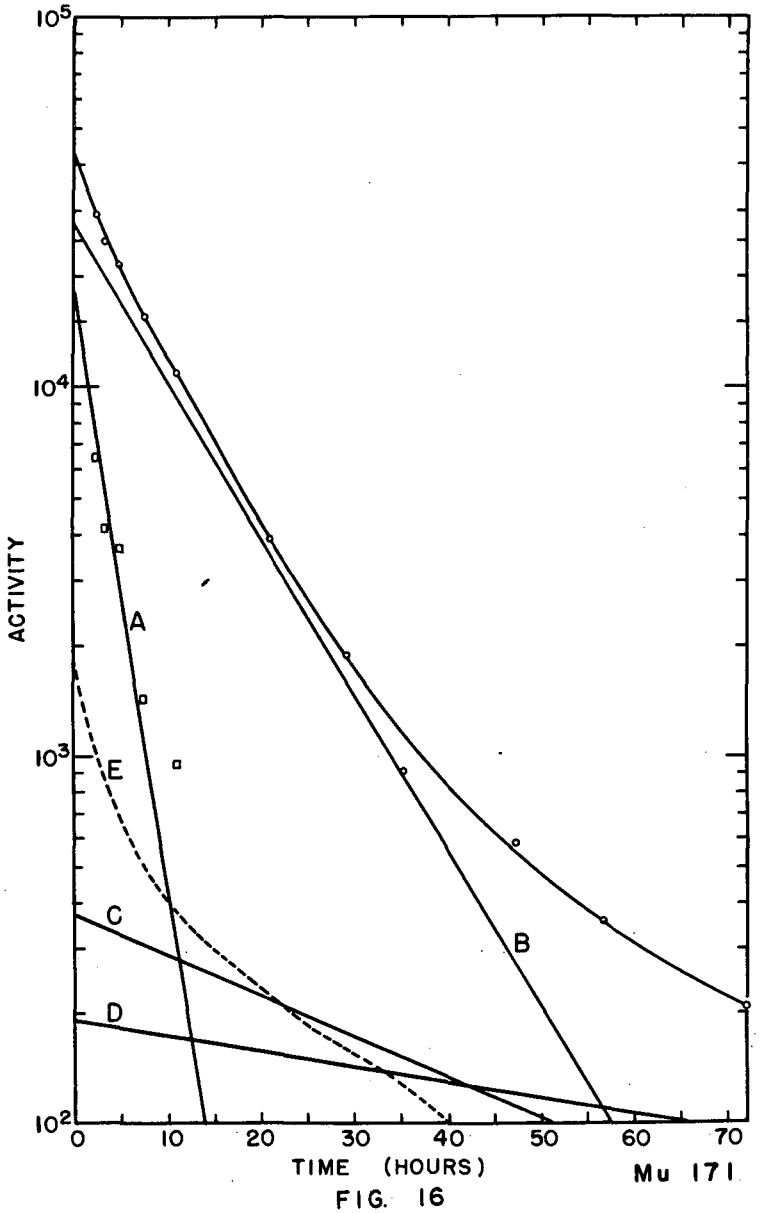

10^5
10^4
10^3
10^2
ACTIVITY
A
B
C
D
E
0
10
20
30
40
50
60
70
TIME (HOURS)
Mu 171

FIG. 16

the 72-hr. and 27-hr. activities, and then drawing the best 7-hr. line although the points indicated the presence of an activity intermediate between 7 hr. and 27 hr. The difference between the 7-hr. line and the actual points was taken as a measure of the bismuth impurity in the sample. The dotted curve in the figure is the decay of the bismuth parent mixture superimposed on the figure in such a position that it represents the contribution of the impurity to the decay curve. The remaining activity was assumed to be 1.8-hr. Tl^{198}.

The half-life values for the bismuth ancestor as obtained by following the yield of 1.8-hr. Tl are shown in Fig. 17. The respective values are 7 min. (curve A) and 6 min. (curve B). For comparison purposes, curve C is the decay of one of the alpha-activities as followed on the alpha-pulse analyzer. The half-life obtained by this method is 7 min.

One of the early lead fractions itself was followed and showed a component of about 25-min. half-life. This activity is probably Pb^{198}, since a similar activity was prepared by Karraker and Templeton[8] from protons on thallium and this was shown to decay to 1.8-hr. Tl^{198}.

Alpha-decay Properties

The earlier work[4] on these alpha-emitting bismuth isotopes showed four periods: 2 min., 9 min., 27 min., and 1-2 hours 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 be 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 II summarizes the information that has been obtained.

Fig. 17. Half-life of Bi^{198}: curves A (7 min.) and B (6 min.) obtained from yields of 1.8 Tl^{198} in "milking" experiments; curve C is the decay of the 5.83-Mev alpha-particle group observed by pulse analysis.

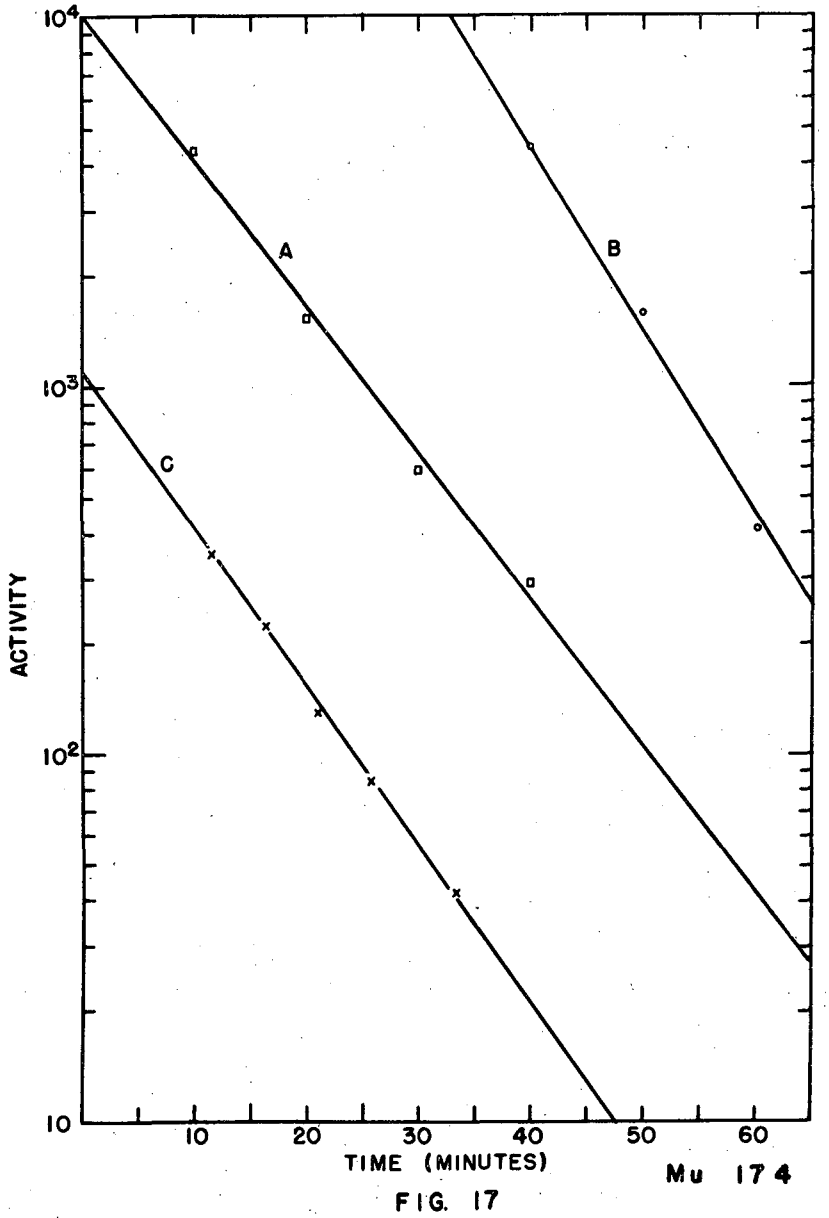

10^4
10^3
10^2
10
ACTIVITY
A
B
C
10
20
30
40
50
60
TIME (MINUTES)
Mu 174

FIG. 17

Table II

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
$Bi^{\leq 197}$	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. It is very unlikely that 40-Mev protons on Pb^{204} (the lightest target isotope) could produce isotopes lighter than Bi^{200}. Isotopes of mass number 203 or greater can be eliminated because of their known characteristics. Bi^{200} decay has already been shown rather definitely to be governed by a 35-min. half-life, so this period is either Bi^{202} or Bi^{201}. 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[4] 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}. The threshold for appearance of Bi^{199} would be expected to be about 55 Mev, some 20 Mev higher than the threshold for the activity assigned to Bi^{201}, and this is the observed result. 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 previously reported[4] as 9 min. appeared. The best half-life obtained by following the alpha-group on the alpha-pulse analyzer was 7 min., as was shown in Fig. 17. Since its order of appearance in the excitation function 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 (Fig. 18) 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 II. 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. 19 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

Fig. 18. Early alpha-decay curve for bismuth, showing a 1.7-min. activity.

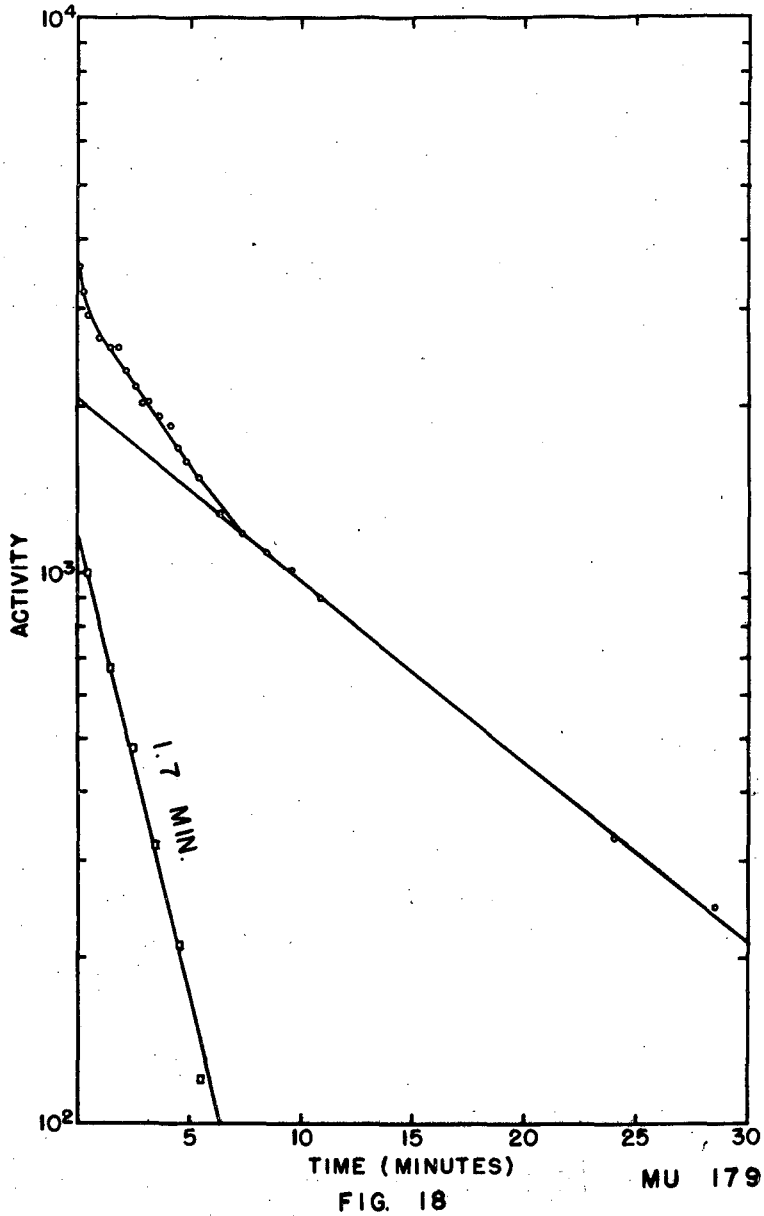


FIG. 18

Fig. 19. 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.

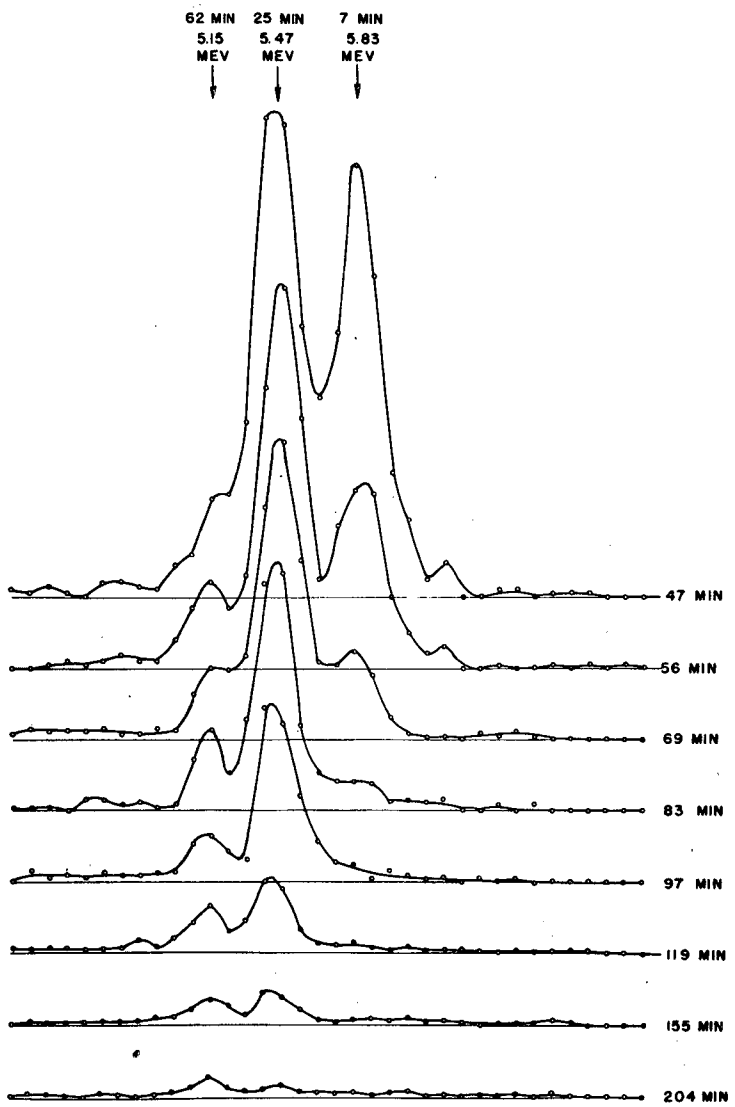

62 MIN
5.15
MEV
25 MIN
5.47
MEV
7 MIN
5.83
MEV
47 MIN
56 MIN
69 MIN
83 MIN
97 MIN
119 MIN
155 MIN
204 MIN
ALPHA PARTICLE ENERGY (MEV)

FIG. 19

Mu 168

the alpha-particle energy analysis was made. 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. 19 that the 25 min. and 7 min. periods were obtained.

The gross alpha-decay of another portion of the same bismuth fraction is shown in Fig. 20. This particular curve resolved into components of 7 min., 24 min., and 60 min. The arrows indicate times corresponding to the pulse analyses of Fig. 19. The decay curves ordinarily obtained were not this good, in that the 1-hr. period was usually less well defined. 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. value consistently.

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[5] 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[5] 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

Fig. 20. Complex alpha-decay curve of bismuth activities produced by irradiation of lead with 180-Mev deuterons. The arrows indicate times corresponding to the pulse analyses of Fig. 19. The curve has been resolved into three components of the following half-lives: 7 min. (A), 24 min. (B), and 60 min. (C).

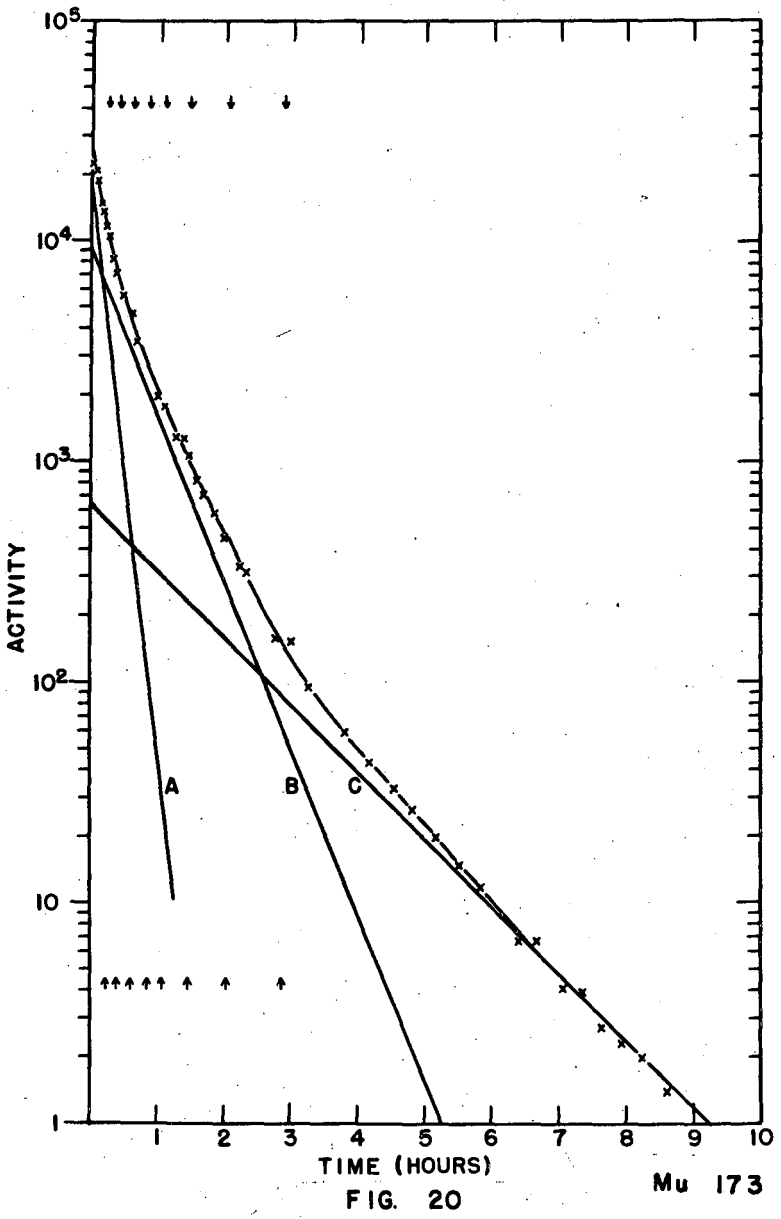


FIG. 20

others) in a broad region can be described by the same function for nuclear radius, $r = 1.48A^{1/3} \cdot 10^{-13}$ 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 percent in argon-filled Geiger tubes, corrected to 100 percent geometry.

The alpha-decay branching ratios estimated for Bi^{198}, Bi^{199}, and Bi^{201} are shown in Table II. Fig. 21 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,[5] it is only possible to guess at the effect of nuclear radius. About all that can be said is that values lower by about 10 percent from those given by $r = 1.48A^{1/3} \cdot 10^{-13}$ cm. seem to be required.

Fig. 21. 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 $1.48A^{1/3} \cdot 10^{-13}$ cm.

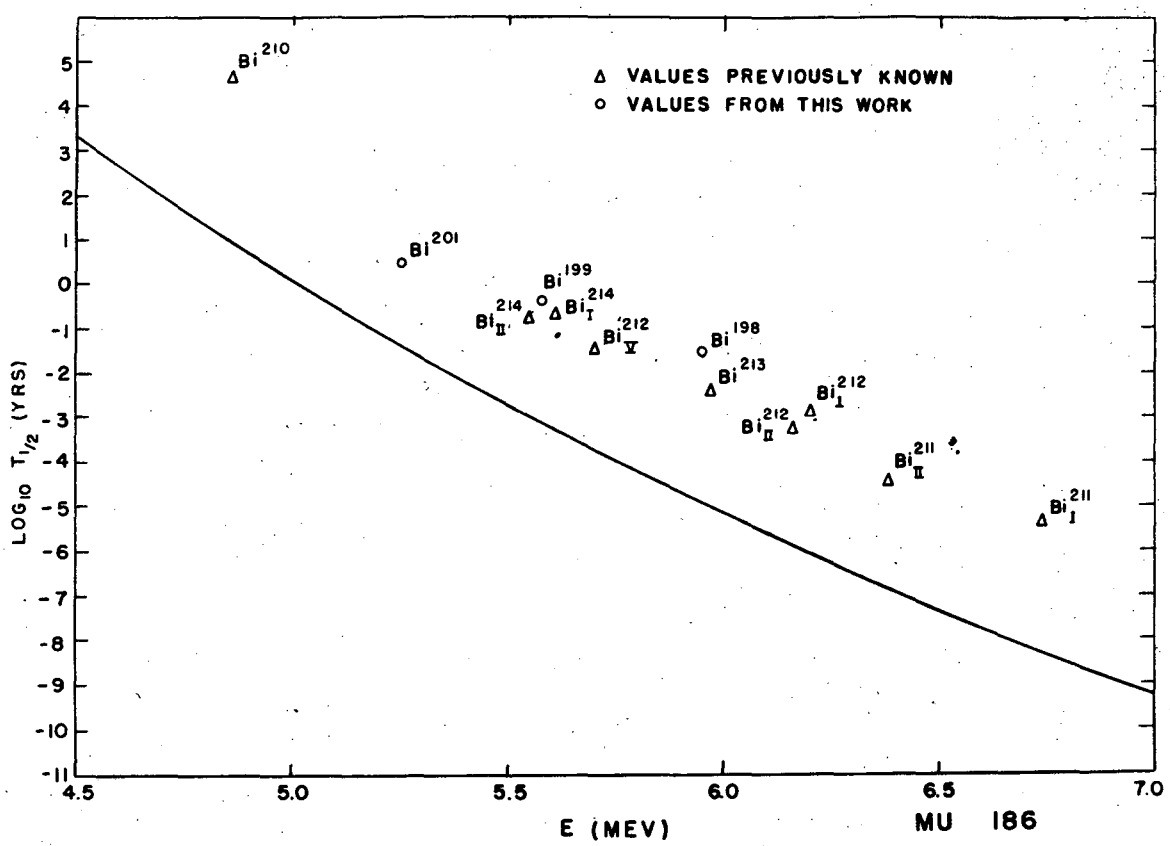

Δ VALUES PREVIOUSLY KNOWN
o VALUES FROM THIS WORK
Bi 210
Bi 201
Bi 199
Bi II 214
Bi I 214
Bi V 212
Bi 198
Bi 213
Bi I 212
Bi II 212
Bi II 211
Bi I 211
LOG10 T1/2 (YRS)
E (MEV)
MU 186

FIG. 21

Chapter II

ISOTOPES OF MASS NUMBERS 207 TO 210

Bi^{209}

Although the radioactivity of Bi^{209} is not detectable, this isotope is certainly alpha-unstable. Failure to observe the alpha-particles sets a minimum half-life of about 10^{13} years. (This half-life corresponds to a specific activity of 0.4 disintegrations per minute per milligram.) By referring to Fig. 21 it will be noticed that the alpha-half-lives for bismuth are abnormally long by factors of 10^2 to 10^4. If we assume the half-life to be 10^{13} years and that the life is abnormally long by as much as a factor of 10^6 on account of increased stability due to 126 neutrons, the alpha-decay energy is 4.0 Mev. This provides us with an upper limit that will be useful in later considerations.

Bi^{208}

No definite activity has ever been observed that could be attributed to Bi^{208}. Workers studying the (γ,n) reaction on Bi^{209} have certainly produced this isotope, since they detect the neutrons formed in the reaction, but they detect no activity with a life between the limits of a few minutes and a few months.[10]

This isotope is unstable with respect to electron-capture decay to Pb^{208}, probably by about 2 Mev. The decay energy for this transition is equal to the difference in the alpha-decay energies of Po^{212} (8.9 Mev) and At^{212} (estimated to be 7.4 Mev)[5] plus the electron-capture energy for At^{212} decaying to Po^{212} (energy unknown).

Since the electron-capture energy of Bi^{208} might be well over 2 Mev, the possibility of its being short-lived was investigated. For this purpose lead oxide was bombarded for two minutes with 18-Mev deuterons. The oxide was

dissolved and the bismuth chemically separated by the nickel-plating method described earlier. Counting of the bismuth began within five minutes after the end of bombardment, and no activity shorter than 12 hr. was detected. This negative result placed an upper limit of 30 seconds on the half-life if Bi^{208} were actually short-lived.

Irradiations of bismuth were attempted using neutrons produced by 18-Mev deuteron bombardment of beryllium. No chemistry was performed, and the bismuth was counted within a few seconds after bombardment. Again no activity attributable to Bi^{208} was observed. It is possible, however, that the neutron energy was insufficient to cause the (n,2n) reaction, or that the cross section for the reaction was too low to allow detection of the product.

If not short-lived, and the evidence indicates it is not, the life must be greater than 200 years. Bombardment of lead with deuterons yields a long-lived product after the decay of 6.4-day Bi^{206} and 14-day Bi^{205},[8] but this activity may be Bi^{207} or Bi^{208} or both. The limiting value for the half-life is based on the assumption that all of this activity is Bi^{208}. Since there is evidence that Bi^{207} is actually present in the mixture (see the section on Bi^{207}), the life is probably much longer.

No detectable alpha-activity would be expected from Bi^{208}. The best evidence available indicates that Bi^{208} and Bi^{209} have similar alpha-energies. This conclusion follows from the fact that the neutron-binding energies of Bi^{209} and Tl^{205}, as measured by the threshold for the (γ,n) reaction, are respectively 7.45[10] and 7.48[11] Mev. We would then expect the alpha-energy of Bi^{208} to also be less than 4.0 Mev, and its partial alpha-half-life to be correspondingly long.

If we take 4.0 Mev as the upper limit to the alpha-energy, we can also state that Bi^{208} will not be capable of beta-emission to Po^{208}. The other data necessary

for this calculation are: alpha-decay energy of Po^{208}, 5.2 Mev, and the beta-energy of Tl^{204}, 0.8 Mev. The result is that Po^{208} must be unstable by at least 0.4 Mev with respect to Bi^{208}.

Bi^{207}

Preparation of Bi^{207} by direct formation in bombardment is complicated by the accompanying formation of Bi^{208}. The product resulting from deuteron bombardment of lead is an example.

Decay of astatine, however, provides a way of preparing Bi^{207} free of Bi^{208}. A large source of At^{211} (about 10^9 disintegrations/minute) was prepared by helium ion bombardment of bismuth, followed by volatilization of the astatine from the target metal. The At^{212} (0.25-sec. half-life) had already decayed away before volatilization, hence the Bi^{208} interference was eliminated. After allowing the At^{211} source to decay a number of days, chemistry was performed on the remaining activity, and bismuth was isolated.

The Bi^{207} activity was small, and was due chiefly to conversion electrons. The absorption of the electrons in aluminum (Fig. 22) is almost identical to that of the long-lived activity produced from lead with deuterons. Sufficient activity from the latter source was available to examine with a crude beta-ray spectrometer. The spectrum is very complex, with the predominant line at about 860 kev and less prominent lines at lower energies. This unusual spectrum accounts for the appearance of the absorption curve, which looks very much like that of a beta-emitter.

Estimates of the half-life will depend on what is assumed as the counting efficiency for this activity. If the counting efficiency is 10 percent, the life is 10 years. However, the Bi^{207}-Bi^{208} mixture prepared from lead has shown no decay over a period of two years, and a life greater than 10 years is indicated. The discrepancy could be explained by a higher counting efficiency for

Fig. 22. Aluminum absorption curve for Bi^{207}.

The components are: A, total electromagnetic radiation; B, gamma and K x-rays; C, L x-rays; and D, electrons.

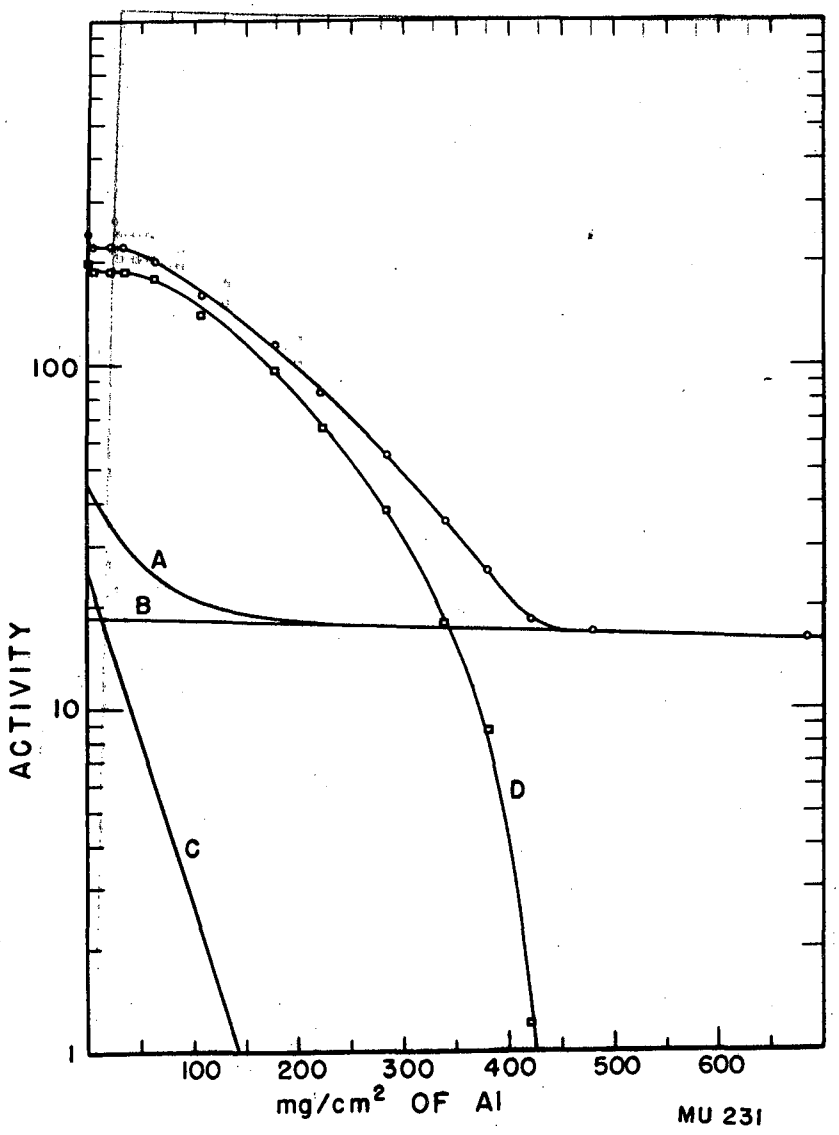

ACTIVITY
100
10
1
A
B
C
D
100
200
300
400
500
600
mg/cm² OF Al
MU 231

FIG. 22

Bi^{207}, or the presence of comparable amounts of Bi^{208}. It would seem quite certain that the life of Bi^{207} is in the range 10-100 years.

Bi^{210} and Bi^{210m}

The well-known 5.0-day beta-emitting Bi^{210} (RaE) occurs naturally as a member of the uranium series. Broda and Feather[12] discovered that this isotope exhibits rare alpha-branching (10^{-5}-10^{-6} percent) by detecting the 4-min. Tl^{206} daughter resulting from the alpha-decay.

In an attempt to produce long-lived Bi^{208}, bismuth metal was irradiated with pile neutrons in the hope that Bi^{208} would be formed by an (n,2n) reaction from the relatively small number of fast neutrons. A long-lived activity indeed was observed, but it proved to be an alpha-emitter rather than the expected electron-capture activity. The alpha-particle energy was 5.03 ± 0.05 Mev as determined in an ionization chamber coupled to a pulse height analyzer. This value for the energy is one of the arguments against the assignment to Bi^{208} since, as pointed out, we would expect the alpha-energy to be less than 4.0 Mev.

The alpha-activity was observed by allowing the irradiated bismuth to stand for six months until all of the 5-day RaE had decayed, after which it was rigorously purified, first from the great alpha-activity of Po^{210} present and then from all other possible alpha-activities. To reduce any Geiger-counter activity that might be present, purification designed to separate from all other elements was performed. Special precautions were taken to remove silver and iron, which were known to be contaminants, in addition to heavy elements. The bismuth finally reached a constant specific activity of 1.9 ± 0.1 alpha-disintegrations per minute per milligram and 2.0 ± 0.2 Geiger counts per minute per milligram.

The alpha-emitter is assigned to an isomer of Bi^{210} because of the identification of a thallium daughter with the properties of Tl^{206}. The daughter was separated from a solution of the parent by precipitation of thallous chloroplatinate.

The thallium was counted within two minutes after separation and exhibited the decay of Fig. 23. The half-life obtained was 4.2 ± 0.5 min. in agreement with the reported value for Tl^{206}, 4.23 min.[2]

Within the experimental uncertainty of about 10 percent, then, all of the radioactivity of the sample might be explained by the alpha-particles of Bi^{210m} in equilibrium with its 4-min. beta-emitting daughter Tl^{206}. There was available insufficient activity to determine the beta-particle energy precisely, but absorption in aluminum indicated a maximum energy of 1.5 ± 0.3 Mev. Through a factor of ten the absorption curve looks like that reported by Krishnan and Nahum[1] for Tl^{206}.

The excess Geiger activity was quite real, however. It was electromagnetic in character, and a crude lead absorption of the few counts available indicated an energy in the range 30-400 kev. This radiation could conceivably be x-rays from Bi^{208}. If Bi^{208} were present one might expect some conversion electrons to be seen. Because of the low specific activity, self-absorption effects are large in samples with moderate counting rates (~400 counts/min.), and counting errors are large in samples with small self-absorption. Thus the failure to observe conversion electrons does not mean they cannot be present.

The disconcerting aspect of the assignment of the 5.03-Mev alpha-emitter to Bi^{210m} lies in the consideration of the lifetimes toward different modes of decay. If it is a metastable state of Bi^{210}, it should also decay to the ground state and directly to Po^{210} by beta-emission. Indeed, if we accept the data to be discussed below, this nucleus should also be unstable with respect to Pb^{210} (RaD). It is possible to calculate these decay energies from the alpha-energies of the two states of Bi^{210} and known beta-energies, and to set some lower limits on the half-lives for the different processes.

Let us first consider the isomeric energy; that is, the difference between Bi^{210m} and RaE, the ground state. This can be obtained by comparing the

Fig. 23. The decay curve of the 4.2-min. thallium resulting from the decay of its long-lived Bi^{210m} parent. The circles are the experimental decay points, and the squares are the values obtained by subtracting the residual activity (background and parent contamination) from the experimental decay curve.

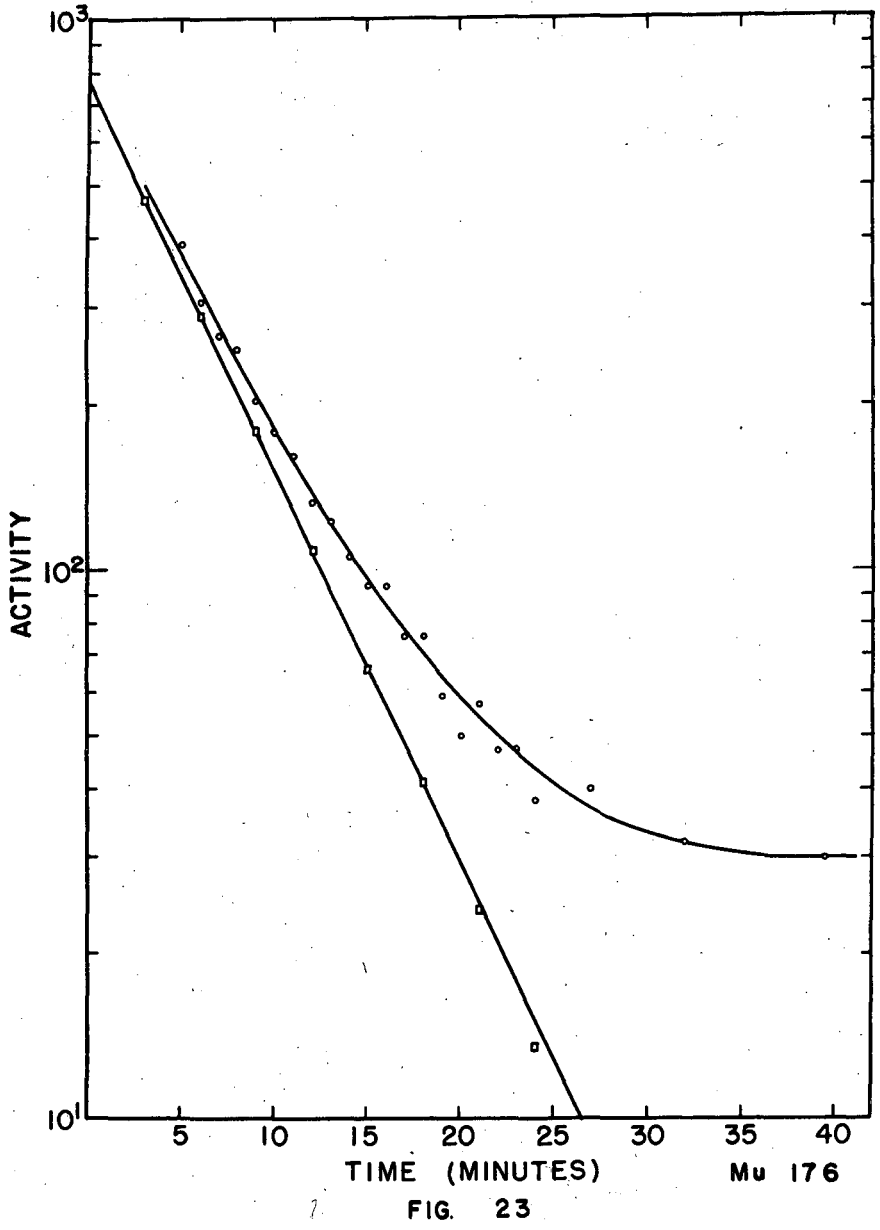


FIG. 23

alpha-energies of the two states, making the assumption that both alpha-transitions go to the ground state of Tl^{206}. The alpha-decay energy for Bi^{210m} as determined in the present study is 5.12 ± 0.05 Mev. That for RaE can be calculated by closing a cycle involving the beta-decay energies for RaE and Tl^{206} and the alpha-decay energy of Po^{210}. Broda and Feather[12] made this calculation and arrived at the decay energy 4.86 Mev for the alpha-decay of RaE. In re-evaluating the data of Krishnan and Nahum[1] and of Fajans and Voight[2] for the beta-energy of Tl^{206}, 1.63 ± 0.10 Mev seemed to be the best value. This revises the alpha-energy of RaE to 4.94 ± 0.10 Mev. The energy between the isomeric states of Bi^{210} would then be 0.18 ± 0.11 Mev. The unobserved beta-decay energy is accordingly 1.35 Mev and the electron-capture energy to Pb^{210}, 0.1 Mev.

The alpha-half-life for Bi^{210m} is not known other than that it must be greater than 25 years, based on observation of the alpha-activity over a period of 15 months. From the failure to find Po^{210}, which would result from both the isomeric transition and beta-decay, growing into the sample during a 63-day growth period, it is possible to say that these decay modes are at least 2000 times slower than the alpha-decay and therefore have minimum half-lives of 5×10^4 years.

It is obvious that the most stringent requirement in explaining the long half-lives will be demanded by the isomeric transition. Using the expression and conventions given by Segre and Helmholz[13] relating decay constant to the order of the transition and the transition energy, it would appear impossible to explain the $>5 \times 10^4$ yr. half-life if the transition energy is as great as 0.18 Mev even if one assumes a fifth order transition. However, if the transition energy is 0.07 Mev, the calculated half-life can become sufficiently long.

If our calculations are correct, it is apparent that this isomer cannot exist as a member of the naturally-occurring uranium series. As a demonstration of this fact, bismuth was separated from 100 grams of pitchblende, the 5-day

activity was allowed to decay out, and the sample was then repurified and examined for presence of the 5.03-Mev alpha-emitter. A negative result was obtained.

ACKNOWLEDGMENTS

I wish to express my gratitude to Professor I. Perlman for his guidance of this work, and to Professor G. T. Seaborg for his interest. I would like to thank Mr. J. T. Vale, Mr. T. M. Putnam, Jr., Mr. G. B. Rossi, and the crews of the 60-inch and 184-inch cyclotrons for the irradiations which made these studies possible. I also wish to thank Mr. Albert Ghiorso for his aid in determining the alpha-particle energies, and Mrs. Elinor Potter for her assistance in counting samples.

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

REFERENCES

[1]R. S. Krishnan and E. A. Nahum, Proc. Camb. Phil. Soc. 36, 490 (1940).

[2]C. Fajans and A. F. Voight, Phys. Rev. 60, 619, 626 (1941).

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

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

[5]I. Perlman, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 77, 26 (1950).

[6]Ghiorso, Jaffey, Robinson, and 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).

[7]W. Mauer and W. Ramm, Zeits. f. Physik 119, 602 (1942).

[8]D. G. Karraker and D. H. Templeton, private communication.

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

[10]McElhinney, Hanson, Becker, Duffield, and Diven, Phys. Rev. 75, 542 (1949).

[11]Hanson, Duffield, Knight, Diven, and Palevsky, Phys. Rev. 76, 578 (1949).

[12]E. Broda and N. Feather, Proc. Roy. Soc. (London) A190, 20 (1947).

[13]E. Segrè and A. C. Helmholz, Rev. Mod. Phys. 21, 271 (1949).