Artificial Radioactive Isotopes of Bismuth and Lead

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Six lead and bismuth activities have been identified from the bombardment of thallium, lead and bismuth with cyclotron deuterons and helium ions, and of lead with pile neutrons.

Isotope	Half-life	Decay	Radiations observed	Produced by
Bi^{204}	12 hours	K	e^-, x-rays, γ	$Tl(\alpha,3n)$ $Pb^{204}(d,2n)$
Bi^{206}	6.4 days	K	e^-, x-rays, γ	Po^{206} K decay $Tl(\alpha,3n)$ $Pb^{206}(d,2n)$ $Pb^{207}(d,3n)$
Bi^{210}	5.0 days	β^-	β^-	$Bi(d,p)$ $Pb^{208}(\alpha,pn)$
Pb^{203}	52 hours	K	e^-, γ	$Tl(d,2n)$ $Pb^{204}(n,2n)$
Pb^{204*}	68 minutes	I.T.	e^-, γ	Bi^{204} K decay $Tl(d,3n)$
Pb^{209}	3.3 hours	β^-	β^-	$Pb(d,p)$ $Pb(n,\gamma)$

No activity has been found which can be attributed to Bi^{205}, Bi^{207}, Pb^{202}, or Pb^{205}, although all of these isotopes must have been produced.

INTRODUCTION

In this laboratory an extensive study is being made of the light radioactive isotopes of lead and neighboring elements.[1,2] In a previous paper[2] three new polonium isotopes have been described. In this paper some results concerning lead and bismuth isotopes are presented.

EXPERIMENTAL

The experimental techniques which were used have been described in a previous paper.[2] The isotopic compositions of the target materials used are listed in Table I. The enriched samples of Pb^{204} and Pb^{206} were prepared in a calutron in this laboratory.[3]

TABLE I. Isotopic compositions of target materials.

Isotope	Ordinary[a] thallium	Ordinary[b] lead	Lead A[c]	Lead B[c]	Bismuth[a]
Tl^{203}	29.1%				
Tl^{205}	70.9				
Pb^{204}		1.5%	27.3%	<0.2%	
Pb^{206}		23.6	32.7	59.7	
Pb^{207}		22.6	13.8	25.2	
Pb^{208}		52.3	26.2	15.1	
Bi^{209}					100%

[a] A. O. Nier, Phys. Rev. **54**, 275 (1938).
[b] A. O. Nier, J. Am. Chem. Soc. **60**, 1571 (1938).
[c] Analyzed by Mr. J. T. Vale of this laboratory.

These materials were bombarded with 20-Mev deuterons and 40-Mev helium ions in the 60-inch cyclotron.[4] Samples of lead A and lead B were bombarded with a mixture of fast and slow neutrons in the heavy-water moderated uranium pile of the Argonne National Laboratory. Each activity described in this paper was identified as to element by chemical separations.

RESULTS

5.0-day Bi^{210}(RaE)

RaE was produced by the reactions:

$$Pb^{208}(\alpha,pn)Bi^{210},\quad \sigma=0.008\times10^{-24}\text{ cm}^2\text{ at 40 Mev},$$
$$Bi^{209}(d,p)Bi^{210},\quad \sigma=0.13\times10^{-24}\text{ cm}^2\text{ at 20 Mev.}$$

[1] J. J. Howland, D. H. Templeton, and I. Perlman, Phys. Rev. **71**, 552 (1947).
[2] D. H. Templeton, J. J. Howland, and I. Perlman, Phys. Rev. **72**, 758 (1947).

[3] We are indebted to Dr. B. J. Moyer, Dr. C. M. Van-Atta, and members of the isotope separation staff for making available these materials, to Dr. E. H. Huffman, Mr. R. C. Lilly, and Miss Dorothy Bockhop for their purification, and to Mr. J. T. Vale for the analyses.
[4] The bombardments were made possible through the cooperation of Dr. J. G. Hamilton, Mr. T. Putnam, and other members of the group that operates the 60-inch cyclotron.

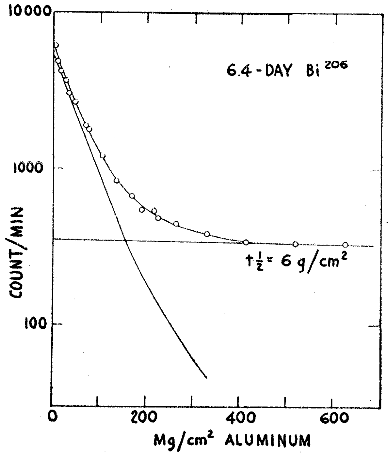

FIG. 1. Aluminum-absorption curve for Bi^{206}.

It was identified by chemical isolation, decay measurement, and observation of the alpha particles of its daughter, Po^{210}. The cross sections are uncertain by 20 percent because of the method used to estimate the intensity of the cyclotron beam. The energy of the incident particles is uncertain by 10 percent, but the targets were thin enough so that there was little degradation of energy.

6.4-day Bi^{206}

The best known light isotope of bismuth is the 6.4-day (orbital electron capture) activity, variously attributed to Bi^{208}, Bi^{207}, or Bi^{206}, and made by bombardment of lead with deuterons.[5, 6] In the previous paper[2] its formation from orbital electron capture decay of a 9-day polonium isotope was described, and evidence was given for the assignment to mass 206. It was also formed directly from lead B by 40-Mev helium ions. If made by the reaction $Pb^{206}(\alpha,p3n)$, the cross section is of the order of 10^{-26} cm². The presence of a considerable amount of 5.0-day Bi^{210} (from Pb^{208}) made this estimate inaccurate.

Bombardment of metallic thallium with 40-Mev helium ions produced Bi^{206} in good yield by one or both of the reactions $Tl^{205}(\alpha,3n)Bi^{206}$ and $Tl^{203}(\alpha,n)Bi^{206}$. By analogy to lead bombardments,[2] the first of these reactions should predominate.

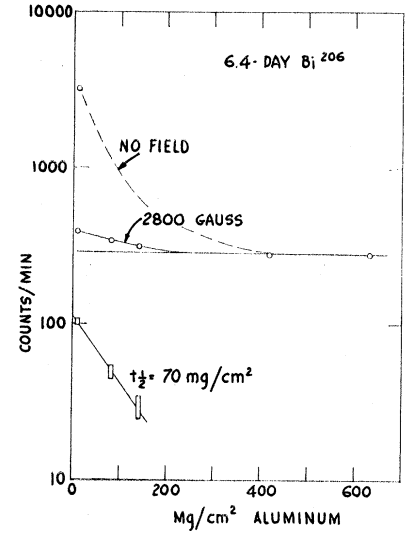

FIG. 2. Aluminum-absorption curve for electromagnetic radiation of Bi^{206}. A magnetic field prevents electrons from being counted.

This activity was also observed whenever lead was bombarded with 20-Mev deuterons.

In Figs. 1 to 4 are shown aluminum- and lead-absorption curves taken with a mica-window brass-wall Geiger counter. In Fig. 2, electrons were deflected with a magnetic field so that the curve represents only the electromagnetic radiations. The energy of the soft component in Fig. 2 is observed as about 14-kev (half thickness 70 mg. Al/cm²) which corresponds approximately to the lead L x-rays (9–15 kev). The counting efficiency for Bi^{206} was calculated to be about 20

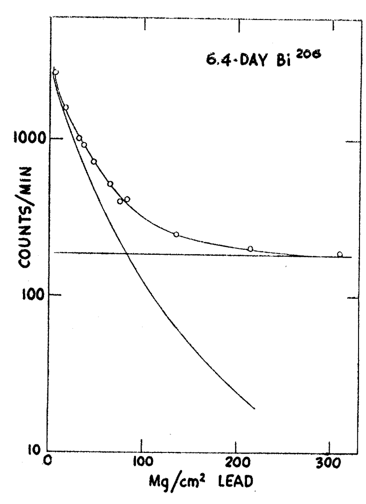

FIG. 3. Lead-absorption curve for Bi^{206}.

[5] R. S. Krishnan and E. A. Nahum, Proc. Camb. Phil. Soc. **36**, 490 (1940).
[6] K. Fajans and A. F. Voigt, Phys. Rev. **60**, 619, 626 (1941).

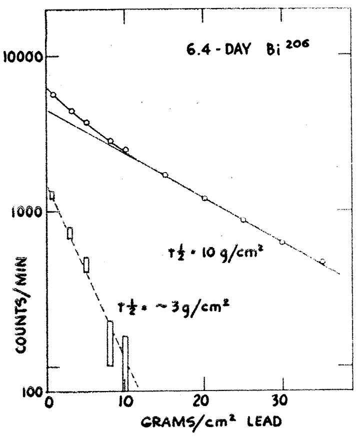

FIG. 4. Lead-absorption curve for Bi^{206} gamma-rays.

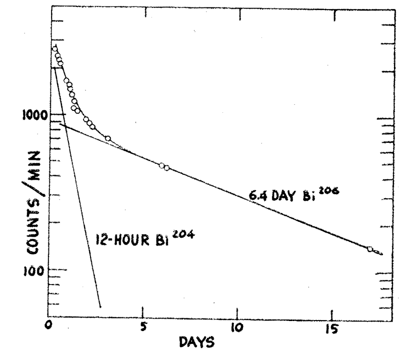

FIG. 5. Decay curve for Bi^{204} and Bi^{206} produced by helium-ion bombardment of thallium.

percent[7] from yield data for Po^{206} in helium ion bombardments.[2] This calculation is based on the assumption that the cross section for $Pb^{204}(\alpha,2n)Po^{206}$ is the same as that for Pb^{208}-$(\alpha,2n)Po^{210}$, and on the known counting efficiency and geometry for Po^{210} alpha particles. The relative counting efficiencies of Po^{206} and Bi^{206} are known from the growth and decay curves. From the yield of Bi^{206} so obtained, a 1 percent counting efficiency for the L x-rays is obtained, if there is one per disintegration. The K x-rays were not definitely observed, but the counting procedures were not very sensitive for differentiating these radiations from the gamma-rays.

The lead absorption curve for the gamma-rays, shown in Fig. 4, gives an energy of 1.1 ± 0.1-Mev (half-thickness 10 g Pb/cm^2). There appears also to be a softer gamma-ray of about 0.4-Mev energy (half-thickness 3 g Pb/cm^2). Fajans and Voigt[6] have reported 1.1 Mev for the gamma-energy based on lead absorption.

Most of the observed counts are due to conversion electrons. The magnetic counter used as a crude spectrometer showed the presence of electrons and no positrons. The spectrum is complex but could not be resolved into its components. The maximum intensity of radiation corresponded to an electron of about 0.3 Mev and one or more weaker lines of higher energy were also present. The aluminum absorption curve for the electrons (Fig. 1) is not simple enough for accurate estimates of the ranges of the individual components. The end-point of about 350 mg/cm^2 of aluminum ($\sim$0.85 Mev) is somewhat higher than the values 0.30 ± 0.02 and 0.2 g/cm^2 observed by Fajans and Voigt[6] and Krishnan and Nahum,[5] respectively.

[7] This counting efficiency is for samples mounted on silver with no correction for back scattering or for window thickness (3 mg/cm^2 mica).

12-hour Bi^{204}

An activity of half-life 12 ± 1 hours was observed in the bismuth fraction isolated from thallium bombarded with 40-Mev helium ions (Fig. 5) and lead A (enriched Pb^{204}) with 20-Mev deuterons. A weak activity of 10–16 hour half-life was observed in ordinary lead bombarded with deuterons.

The yield of 12-hour Bi was at least 15-fold greater from lead A than from natural lead. Since the Pb^{204} content of lead A is 18 times that of ordinary lead, while no other lead isotope is similarly enriched, it is concluded that the 12-hour bismuth is formed in appreciable yield only from Pb^{204}. The bombardments were monitored by the yield of Bi^{206}.

The mass assignment of the 12-hour bismuth may be deduced by the same sort of argument as those used in the previous paper[2] for polonium isotopes. Since 20 Mev deuterons produce the activity from Pb^{204} in good yield, it must be Bi^{203}, Bi^{204}, or Bi^{205}. Since 40 Mev helium ions produce it from thallium, it is probably Bi^{204} or heavier. Since it is not produced in good yield from Pb^{206}, Pb^{207}, or Pb^{208} with 20-Mev deuterons, it cannot be Bi^{205} or heavier. Therefore it has been assigned Bi^{204}.

Since the cross section for $Tl^{203}(\alpha,4n)$ might possibly be high enough to account for the observed yield of the 12 h Bi, the assignment Bi^{203} is a remote possibility. However, the assignment of the 52 h Pb activity to Pb^{203} is quite definite, and this activity is not a daughter of the 12 h Bi.

Krishnan and Nahum[5] have reported a very weak 18±2-hour activity in the bismuth from lead bombarded with 9 Mev deuterons. We have not observed this activity unless it is the same as this 12-hour isotope.

In Fig. 6 is shown an aluminum absorption curve for a mixture of the 12-hour and 6.4-day bismuth activities. Subtraction of the contribution due to Bi^{206} calculated from the curves in Figs. 1 and 5 gives a curve for the 12-hour activity alone. About 7 percent of the counts are due to the gamma-rays, but there are insufficient data for an estimate of the energy.

The magnetic counter showed that only about 10 percent of the counts are due to electromagnetic radiations. About 7 percent are due to the gamma-rays, and so, like Bi^{206}, very little of the soft component is due to L x-rays. Examination of the charged particles with the magnetic counter showed negative electrons of energy about 200 kev, and a smaller number with energies as high as 750 kev. The latter value is uncertain because of the presence of a considerable amount of Bi^{206}. No positrons were observed. The absorption curve for these particles (Fig. 6) has a soft component of range 30 to 50 mg/cm² Al, (~0.2 Mev) and a harder component of range 300 mg/cm² Al or more (~0.8 Mev).

When bismuth containing the 12-hour activity was purified, allowed to stand, and then separated from added inactive lead, the lead was found to contain the 68-minute period attributed to Pb^{204*}.

In a series of such lead separations from aliquots of a bismuth fraction, the yield of 68-minute lead fell off with an 11±2 hours half-life. Thus 68-minute lead is undoubtedly the daughter of the 12-hour bismuth by orbital-electron capture. The yield of lead was about 4 counts for each 100 counts of 12-hour bismuth present. Because of the low yield, the growth of lead daughter is not apparent on the bismuth-decay curves. The 68-minute lead activity is discussed in detail below.

68-minute Pb^{204*}

A one-hour lead activity has been produced by bombardment of lead with fast neutrons (68-minutes,[8] 80-minutes[9]), and thallium with deuterons (65±5 minutes[6]). Fajans and Voigt[6] reported the yield from thallium with 9-Mev deuterons as 1/150 that of 52-hour lead. Krishnan and Nahum[5] did not find the activity in similar deuteron bombardments.

It was not found in 9-Mev deuteron bombardment of lead.[6] Using ordinary lead, uranium lead, and thorium lead, Maurer and Ramm[8] showed that the activity is made from Pb^{204} by neutrons of 0.8-Mev and higher energy, and that the cross section with thermal neutrons is small. The cross section for formation from Pb^{206} is very small. From these facts they deduced that the activity must be either Pb^{204*} or Pb^{205}. The low yields from thallium plus deuterons, from lead plus thermal neutrons, from lead plus deuterons, and from Pb^{206} plus fast neutrons were cited as evidence against the Pb^{205} assignment.

In the present work the activity was produced from thallium with 20 Mev deuterons, probably by both the reactions $Tl^{203}(d,n)Pb^{204*}$ and $Tl^{205}(d,3n)Pb^{204*}$.

Helium ion bombardment (40 Mev) of thallium produced the activity as the daughter of Bi^{204}, formed by $Tl^{203}(\alpha,3n)Bi^{204}$. There is no evidence for its direct formation by $Tl^{203}(\alpha,p2n)$-

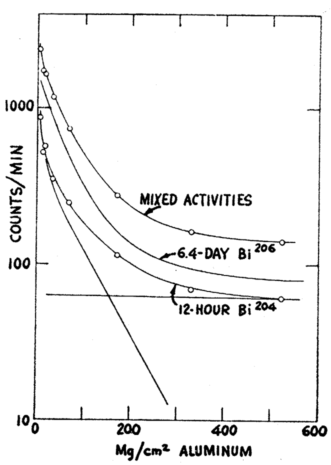

FIG. 6. Aluminum-absorption curve for Bi^{204} and Bi^{206} mixture.

[8] W. Maurer and W. Ramm, Zeits. f. Physik **119**, 602 (1942).
[9] H. DeVries and G. Diemer, Physica **6**, 599 (1939).

Pb^{204*}. The activity was observed in 20-Mev deuteron bombardment of enriched Pb^{204} by the mechanism $Pb^{204}(d,2n)Bi^{204} \xrightarrow{12h\ K} Pb^{204*}$. There is no evidence of its direct production by $Pb^{204}(d,d)Pb^{204*}$.

Our pile-neutron bombarded lead was examined too late for detection of a 68-minute period.

The properties of the radiations have been described by previous workers.[6,8] We have observed gamma-rays, negative electrons of several hundred-kev energy, and no positrons, in accord with the published data. With our counters, about 7 percent of the counts are due to gamma-rays, which is about the same as the ratio for Bi^{204}. Because of this fact it seems improbable that the counting efficiencies of the two activities are very much different, and so only about 4 percent of the disintegrations of Bi^{204} produce 68-minute Pb^{204*}. The other disintegrations presumably give stable Pb^{204}.

3.3-hour Pb^{209}

There is general agreement that a 3.3-hour lead β^--emitter produced by deuteron bombardment of lead[5,6,10] and by neutrom bombardment of lead and bismuth[8] is due to Pb^{209}. Maurer and Ramm[8] have summarized the properties of the radiations and the arguments for the mass assignment.

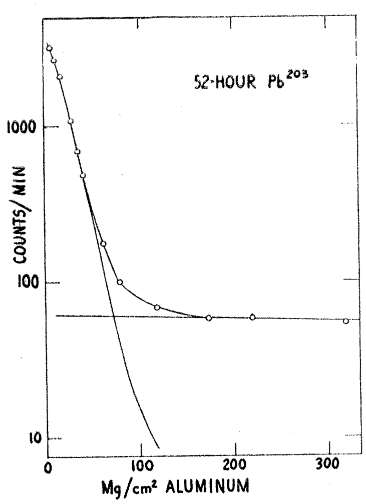

FIG. 7. Aluminum-absorption curve for Pb^{203}.

[10] R. L. Thornton and J. M. Cork, Phys. Rev. **51**, 383 (1937).

In the bombardments of the various lead samples with 40-Mev helium ions, the lead fractions showed a weak 3.3-hour activity mixed with other periods, all of which could be explained as contamination by the bismuth and polonium activities which are formed in much higher yield. The amount of Pb^{209} could be accounted for by a reasonable estimate of the neutron flux in the neighborhood of the target, using 10^{-27} cm^2 for the capture cross section of Pb^{208} for thermal neutrons.[8] There is no evidence for the $Pb^{208}(\alpha,n2p)Pb^{209}$ reaction, but if it occurs the cross section is less than 10^{-29} cm^2.

The 3.3-hour activity was also observed, as expected, in all 20-Mev deuteron bombardments of lead. In the pile-neutrom bombardments, the samples were not received until after this activity had decayed too much for observation.

52-hour Pb^{203}

A 52-hour lead activity has been prepared by deuteron bombardment of thallium[5,6,11,12] and by neutron bombardment of lead.[5,8] Maurer and Ramm[8] bombarded ordinary lead, thorium lead, and uranium lead with 5.3-Mev and 15.2-Mev neutrons. The 52-hour activity was produced only with ordinary lead and 15.2-Mev neutrons, and they concluded it to be formed by the reaction $Pb^{204}(n,2n)Pb^{203}$. Krishnan and Nahum[5] reported the formation of the activity with both "fast" and "slow" neutrons. The activity was not observed in 9-Mev deuteron bombardments of lead.[5,6] The assignment has also been suggested as Pb^{204*}, Pb^{205}, and Pb^{206*}.

We did not observe this activity in any 20-Mev deuteron bombardment of lead, including bombardment of enriched Pb^{204}. This fact makes the Pb^{205} assignment very unlikely. It was observed in lead A (27.3 percent Pb^{204}) bombarded with pile neutrons but not in lead B (<0.2 percent Pb^{204}), confirming the conclusion of Maurer and Ramm[8] that it is made only from Pb^{204}. The samples were placed in the pile at a point where both fast and slow neutrons were present. This activity was also produced in good yield by bombardment of thallium with 20-Mev deuterons, by the reaction $Tl^{203}(d,2n)Pb^{203}$. These experi-

[11] K. Fajans and A. F. Voigt, Phys. Rev. **58**, 177 (1940).
[12] R. S. Krishnan and E. A. Nahum, Proc. Roy. Soc. (London), **A180**, 321 (1942).

ments confirm the assignment of this activity to Pb^{203}.

The absence of the activity in lead plus 20-Mev deuteron bombardments indicates that Bi^{203} is moderately long-lived, because it should have been produced, and it almost certainly decays by electron capture to Pb^{203}.

An aluminum-absorption curve for this activity (Fig. 7) agrees very well with that published by Maurer and Ramm.[8] The soft component was shown to be mostly a line of negative electrons of energy $\sim$150 kev by use of the magnetic counter. The resolution of this counter is not good enough to show the fine structure reported for these electrons.[13]

The published values[5, 6, 8] for the maximum electron energies from aluminum absorption vary from 330 to 500 kev, but our data with the magnetic counter indicate a very low abundance of such harder electrons. Possibly some of this harder radiation is due to x-rays.

Our lead-absorption curves are in agreement with published data[6, 8] but are not extensive enough for resolution of the gamma-ray components.

Other bismuth isotopes

We have found no activities corresponding to Bi^{205} or Bi^{207} which should be formed from lead by 20-Mev deuterons. These isotopes are expected to be longer lived than the adjacent even-mass isotopes, and they may be very much longer.

Pb^{205} and Pb^{202}

In the bombardment of thallium with 20-Mev deuterons both Pb^{205} and Pb^{202} should have been produced in good yield. No activities were found which could be attributed to these isotopes. Pb^{202} should decay by electron capture to 13-day Tl^{202},[5, 6, 8] which was not observed in the bombardment. Therefore Pb^{202} is long-lived. One expects that Pb^{205} also is long-lived. If either isotope has a counting efficiency the same as Pb^{203}, its half-life must be greater than 500 years.

Other lead activities

Several other periods have been reported as belonging to lead isotopes, but none of these has yet been confirmed by independent workers. None of these activities was specifically looked for in the experiments reported here.

Krishnan and Nahum[5] reported an intense 10.25 ± 0.25-minute positron activity in lead separated from thallium which had been bombarded with 9-Mev deuterons. The activity was not produced with 7-Mev deuterons. Fajans and Voigt[6] made several attempts to find this activity by the same method without success. Maurer and Ramm[8] could not produce it with fast neutrons on lead.

Pool, Cork and Thornton[14] have reported weak 5-minute and 1.5-hour activities in lead bombarded with fast neutrons. DeVries and Diemer[9] concluded by chemical separations that the former period was not lead or thallium, but some impurity, and found 80-minutes for the latter. Bretscher and Cook[15] later reported the formation of a 4.6-minute thallium activity by fast neutron bombardment of lead. Probably in both cases the longer period is a mixture of 68-minute and 3.3-hour lead.

Waldman and Collins[16] have reported a very weak 1.6 ± 0.2-minute activity produced from lead by x-rays of energy greater than 0.65 Mev. It was not found by Maurer and Ramm[8] using fast neutrons on lead.

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[13] A. L. Lutz, M. L. Pool, and J. D. Kurbatov, Phys. Rev. **65**, 61 (1944).

[14] M. L. Pool, J. M. Cork, and R. L. Thornton, Phys. Rev. **52**, 239 (1937).

[15] E. Bretscher and L. G. Cook, Nature **146**, 430 (1940).

[16] B. Waldman and G. B. Collins, Phys. Rev. **57**, 338 (1940).