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POLONIUM ISOTOPES PRODUCED WITH HIGH ENERGY PARTICLES

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Berkeley, California

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POLONIUM ISOTOPES PRODUCED WITH HIGH ENERGY PARTICLES*

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September 25, 1950

ABSTRACT

We have investigated four new isotopes of polonium produced from bismuth and lead by high-energy spallation and have studied their decay products, which include three new lead and bismuth activities. The mass assignments are made by the identification of known activities as decay products. Some of the properties of these new isotopes are as follows:

Isotope	Half-life	Mode of Decay	Energy in Mev	
			Alpha Particles	Gamma Rays
Po^{205}	1.5 hr.	EC,α	5.22 ± 0.10	
Po^{204}	3.8 hr.	EC,α	5.37 ± 0.02	
Po^{203}	47 min.	EC		
Po^{202}	52 min.	EC,α	5.59 ± 0.03	
Bi^{205}	14.5 days	EC		0.431, 0.527 0.550, 0.746 1.84
Bi^{202}	95 min.	EC		
Pb^{198}	25 min.	EC		

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

POLONIUM ISOTOPES PRODUCED WITH HIGH ENERGY PARTICLES

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INTRODUCTION

Polonium has eight isotopes, beta stable or with neutron excess, which occur in the natural radioactive series or the artificial neptunium series. Cyclotron-induced transmutations now make possible the production of neutron-deficient polonium isotopes. Templeton, Howland, and Perlman[1] have produced by this means the three isotopes Po^{206}, Po^{207}, and Po^{208}, and Kelly and Segrè[2] have found Po^{209}. The present work was undertaken to extend our knowledge of polonium to the even lighter isotopes which can be made by the 184-inch Berkeley cyclotron. We have observed the radioactivities of the next four isotopes, Po^{205}, Po^{204}, Po^{203}, and Po^{202}. Experiments designed to establish the decay products and mass assignments of these radioactivities led to the discovery of two radioactive species of bismuth and one of lead. Our work was greatly aided by a parallel investigation of bismuth and lead isotopes carried out in this same laboratory by Neumann and Perlman, who have already reported their results.[3]

EXPERIMENTAL METHODS

In most of the experiments a target of natural lead or bismuth was bombarded with particles accelerated in the 184-inch Berkeley cyclotron. Helium ions with lead, or protons or deuterons with bismuth produced good

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[1] Templeton, Howland, and Perlman, Phys. Rev. 72, 758 (1947).

[2] E. L. Kelly and E. Segrè, Phys. Rev. 75, 999 (1949).

[3] H. M. Neumann and I. Perlman, Phys. Rev. 78, 191 (1950).

yields of the polonium isotopes of interest, but also substantial amounts of other spallation and fission products. Particles of various energies were used to improve the relative yield of the isotope desired in a particular experiment. The target was made of metal in the form of strips 0.5 to 2 mm thick; or, when speed in chemical separation was required, bismuth oxide was used. This oxide can be dissolved in acids more rapidly than the metal.

All samples were separated chemically before measurements were made. Polonium was separated from other elements by a procedure using tellurium, with hold-back carriers added for thallium and lead. The tellurium was reduced to the element with stannous chloride, carrying with it the polonium and noble metals. The tellurium was dissolved and precipitated with sulfur dioxide, carrying again the noble metals, but leaving a carrier-free solution of polonium, with 85-95 percent yield. For further purification, polonium was extracted from 6N HCl into a mixture of 20 percent tributyl phosphate and 80 percent dibutyl ether. The extraction coefficient for polonium between the organic and acid layers is about 110. Lead and bismuth daughter activities were removed quantitatively by washing the organic layer with 6N HCl, and purified by precipitation--bismuth as BiOCl and lead as $PbSO_4$ or $PbCrO_4$.

Thallium activities were separated by oxidation of thallium to the thallic state with potassium permanganate, followed by the extraction of thallic chloride with diisopropyl ether saturated with HCl. Occasionally the thallium activities were further purified by evaporation of the ether, reduction of the thallium with hydrogen peroxide, and precipitation of the thallium as Tl_2PtCl_6 in the presence of lead and bismuth hold-back carriers.

Generally, the decay curve of the polonium Geiger activities is so complex that resolution is ambiguous. To avoid this difficulty, the half-lives were determined by separation of the daughter activities at a sequence of equal time intervals, the interval corresponding approximately to the half-life of the parent. The activity $\underline{A}_2$ of the daughter at time $\underline{t}$ is given by:

$$\underline{A}_2 = \underline{C}_2 \frac{\lambda_2\lambda_1 N_o}{\lambda_2-\lambda_1}\left[\underline{e}^{-\lambda_1\underline{t}} - \underline{e}^{-\lambda_2\underline{t}}\right]$$

where $\underline{C}_2$ is the counting efficiency of the daughter, including the geometry factor, λ_2 and λ_1 are the decay constants of daughter and parent, respectively; and $\underline{N}_o$ is the number of atoms of parent at $\underline{t} = 0$. At the start of the time interval, the parent is purified chemically so that the additional term in the general formula does not enter the expression. It will be observed that, if the time $\underline{t}$ is the same for all periods of growth of the daughter before separation from the parent, the exponential terms become a constant factor, and $\underline{A}_2$ is proportional to the disintegration rate ($\lambda_1\underline{N}_o$) of the parent at the beginning of the growth period. Thus, if one plots the logarithm of the initial activity of each daughter fraction against the time of separation, the slope of the line will correspond to the half-life of the parent.

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

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

Table I[a,b]

	198	199	200	201	202	203	204	205	206	207	208
Po					(52m EC,α)	(47m EC)	(3.8h EC,α)	(1.5h EC,α)	9d EC,α	5.7h EC,α	3y α
Bi					(95m EC)	12h EC	12h EC	(14.5d EC)	6.4d EC	long	
Pb	(25m EC)	80m EC	18h EC	8h EC	long	52 hr EC	68m IT	long			
Tl	1.8h EC	7.3h EC	27h EC	72h EC	12d EC						
Hg											

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

[b] H. M. Neumann and I. Perlman, Phys. Rev. 78, 191 (1950).

3.8-Hr. Po^{204}

The alpha-decay of polonium produced by high energy irradiation shows periods of about 45 minutes, 1.5 hours, and 4 hours, in addition to longer periods previously identified. To determine the mass assignments of these isotopes, their daughters were identified by the method sketched above. A pure sample of mixed polonium activities was separated from the bombarded target (elapsed time, 2 hours) and the daughter activities separated at intervals of an hour. The decay of the purified bismuth fraction showed a 95-minute period (which will be discussed later), a 12-hour period, and a longer period of 6 to 14 days, in low intensity. Fig. 1 is a plot of the

Fig. 1. Data showing the genetic relationship of 3.8-hr. Po^{204} and 12-hr. Bi^{204}.

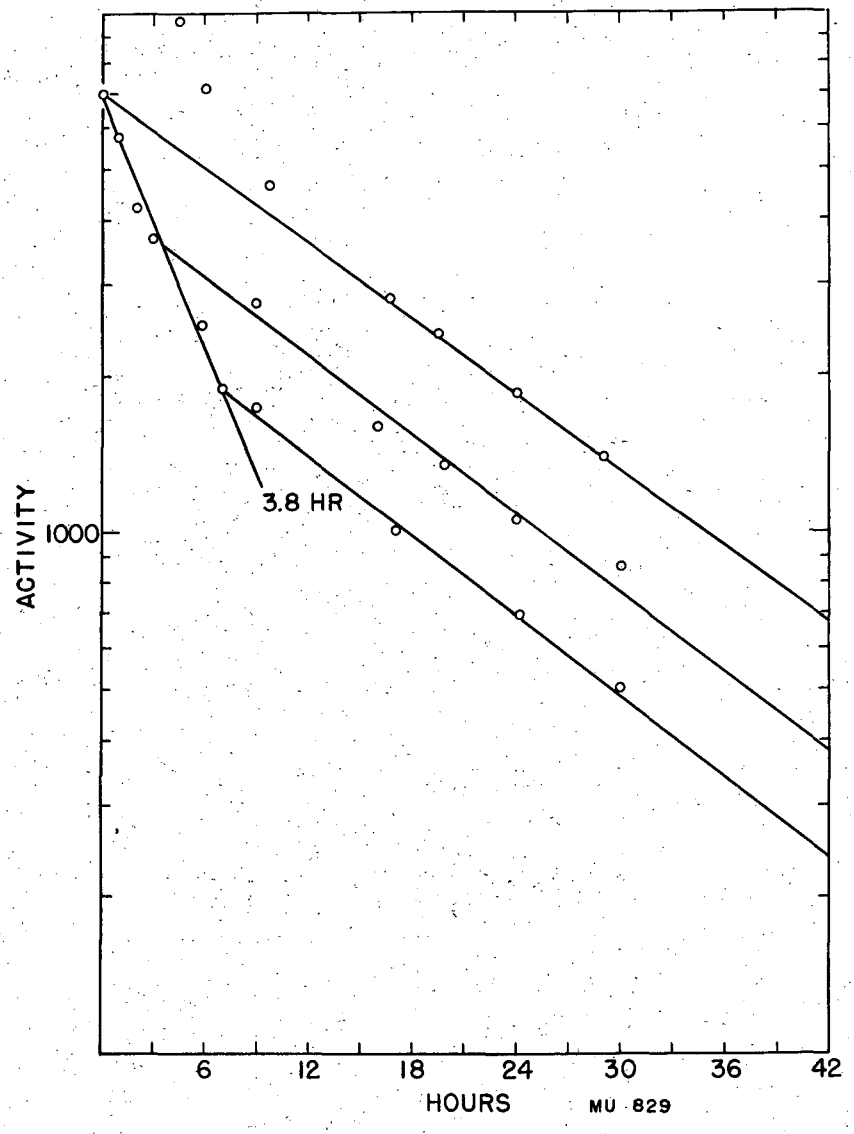


Fig. 1

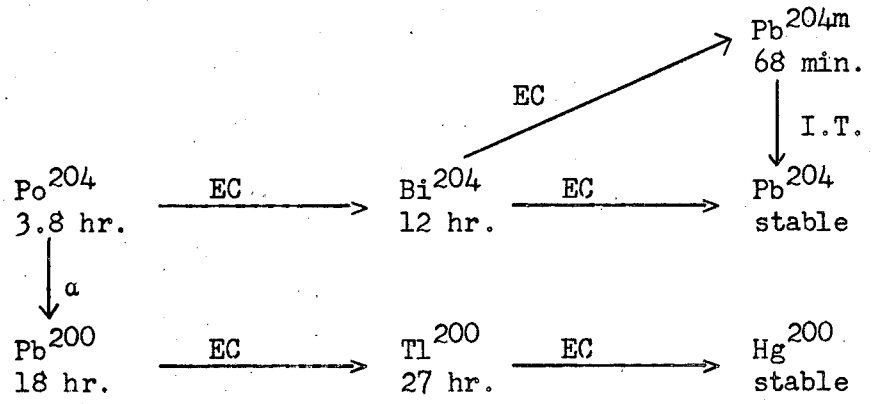
decay of the bismuth fractions for three of the separated samples where the 12-hour period has been extrapolated back to the time of separation. (Only three of the bismuth decay curves are shown, to avoid crowding. Extrapolated points from the decay curves of the other fractions are plotted at the time of separation.) It will be seen from Fig. 1 that the yields at the time of separation determine the half-life of the parent activity of the 12-hour bismuth as 3.8 hours, in agreement with the 3.8-hour value obtained from alpha-decay curves.

It will be seen from Table I that the establishment of a 12-hour bismuth activity as the daughter of the 3.8-hour polonium does not give an unambiguous mass number. A similar experiment was done to identify the alpha-decay daughter and fix the mass number. As before, a large sample of purified polonium activities was prepared, from which the bismuth and lead daughter activities were separated at 4-hour intervals. The bismuth activity could be ignored, as none of the bismuth daughters are alpha-emitters, and Po^{200} is presumed to be too short to produce any of the Tl^{200} through an electron capture chain. The lead alpha-daughter was allowed to grow its thallium daughter for about 18 hours; then the thallium was separated. Since the lead fraction had been separated at 4-hour intervals, the activity of the alpha-decay daughter of the 3.8-hour polonium would be decreased by approximately a factor of two between each of the fractions. The time allowed for the thallium to grow was the same for each fraction, so the activity of the lead alpha-daughter will be directly proportional to the activity of its thallium daughter. The principal, and virtually the only activity found was the 27-hr. Tl^{200}. The results are shown in Table II.

Table II

Fraction	27-hr. Tl, counts/minute	
	Observed	Calculated
1	3280	---
2	1740	1600
3	845	780

These experiments have fixed the 3.8-hour polonium at mass 204. Its alpha-decay daughter is the 18-hr. Pb^{200} which was shown by Neumann and Perlman[3] to be the parent of Tl^{200}. The electron capture daughter is Bi^{204}, which has been shown[4] to decay both to Pb^{204} and to Pb^{204m}. These decay relations are:



We have confirmed that Po^{204} gives rise to the 68-min. Pb^{204m}, by an experiment which is described below. Thus these assignments, which are based on the excitation curve for the formation of 27-hour thallium by alpha irradiation of gold[5] are an independent check on the assignment[4] of Pb^{204m}. This isomer is of great interest because of its unique properties as a long-lived highly

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[4]Templeton, Howland, and Perlman, Phys. Rev. 72, 766 (1947).

[5]Orth, Marquez, Heiman, and Templeton, Phys. Rev. 75, 1100 (1949).

excited state of an even-even nucleus.

The energy of the alpha-particles from Po^{204} was determined as 5.37 ± 0.02 Mev by pulse analysis[6] (Fig. 2). The ratio of electron capture disintegrations to alpha disintegrations has not been determined with accuracy. It is estimated as approximately 100 on the assumption that the cross section for the (p,6n) reaction on bismuth is 0.1 barn at its maximum, which is at 70 to 80 Mev.

68-min. Pb^{204m}

The discovery of two 12-hour bismuth isotopes forced a reconsideration of the mass assignment of Pb^{204m}, which was based in part on the observation that the 68-minute lead is a daughter of a 12-hour bismuth. The experiments[4] which showed that this is the case, and that 52-hr. Pb^{203} is not the daughter of the same 12-hour bismuth, were done with bismuth from the alpha irradiation of thallium at an energy which did not produce Bi^{203}. Had these experiments been done on the bismuth produced from lead, both daughters should have been found. The reasons cited[4] for the mass assignment are still valid, though some of them must now be stated in a more involved way to distinguish between the two 12-hour bismuth activities.

To relate Pb^{204m} to Po^{204}, a large quantity of polonium was prepared and purified. Bismuth daughters were separated at 4-hour intervals. Each bismuth fraction was allowed to stand for 24 hours; then lead was separated from the bismuth. The yield of 68-minute activity in each sample is listed in Table III, which shows that the polonium ancestor has a 3.8-hour half-life.

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[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).

Fig. 2. Pulse analysis showing alpha-particle energies for polonium isotopes.

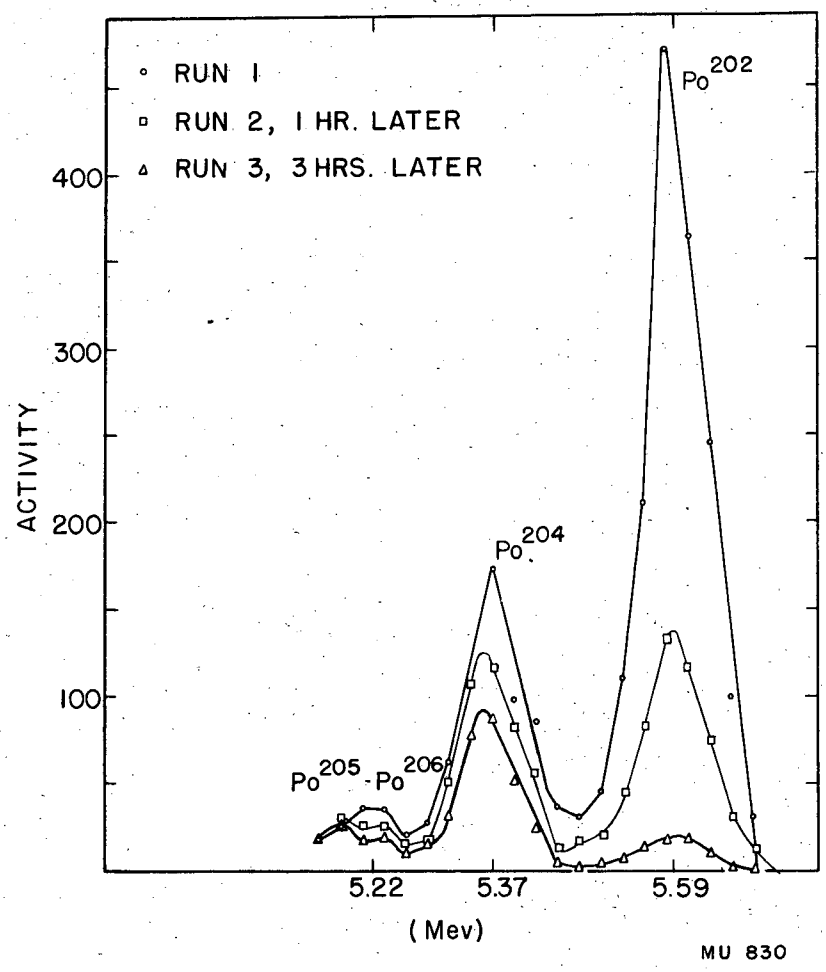

RUN 1
RUN 2, 1 HR. LATER
RUN 3, 3 HRS. LATER
ACTIVITY
400
300
200
100
Po202
Po204
Po205 - Po206
5.22
5.37
5.59
(Mev)
MU 830

Fig. 2

Table III

Fraction	68-min. Pb, counts/min.	
	Observed	Calculated
1	4900	---
2	2200	2350
3	1090	1120

1.5-hr. Po^{205}

A 1.5-hour electron capture activity with associated alpha-particles was resolved from the decay curves of polonium produced by irradiation of lead enriched in mass 204 (27 percent)[7] with 37-Mev helium ions in the 60-inch Crocker Laboratory cyclotron (Fig. 3). The predominant product of 37-Mev helium ion bombardment of an element in this region is known to be the (α,3n) product,[1,2] which in this case is Po^{205}. No lighter polonium should be produced in this irradiation, and since the heavier ones are already known, Po^{205} is the best assignment.

Periodic separations of the bismuth daughters from a large amount of mixed polonium activities at 1.5-hour intervals showed that the yield of longer bismuth periods (i.e., half-lives of the order of a week) indicated a parent with a half-life of 3 to 4 hours. Since the 9-day Po^{206} is known to grow the 6.4-day Bi^{206}, this clearly indicated a second polonium isotope of short half-life growing a longer bismuth. The decay of the bismuth activities showed that the first separation yielded principally a new activity of 14-day half-life,

[7] This lead was enriched with a calutron in Berkeley. We are indebted to Dr. E. H. Huffman, Mr. R. C. Lilly, and Mrs. D. B. Stewart for its purification and to Mr. J. T. Vale for its mass analysis.

Fig. 3. Geiger decay curve of polonium from Pb^{204} with 37-Mev helium ions. A, 5.7-hr. Po^{207}; B, 1.5-hr. Po^{205}.

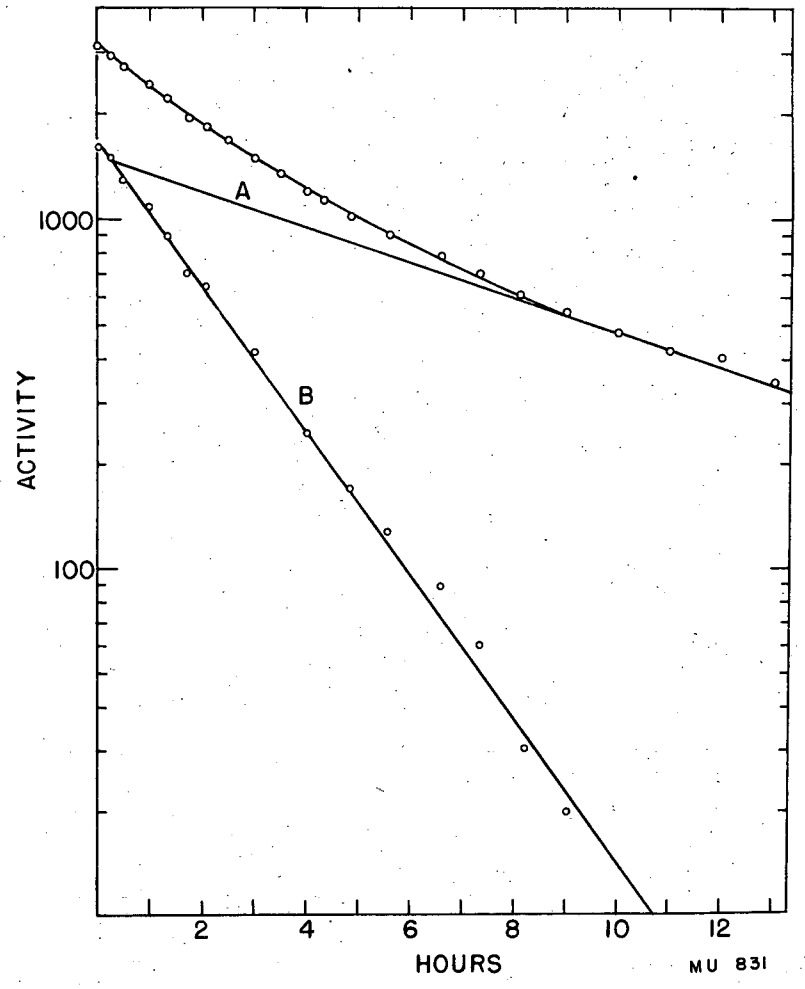


Fig. 3

while the last fractions were almost pure 6.4-day Bi^{206}. After the decay of the Bi^{206}, the yield of the 14-day bismuth corresponded to a parent of 1.5 hours (Fig. 4). This is the half-life of Po^{205}, thus the new bismuth activity is also mass 205. Its radiation characteristics will be described presently.

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

Table IV

Fraction	72-hr. Tl, counts/min. Observed	Calculated
1	650	---
2	300	325
3	160	163
4	85	81

These data confirm the assignment of the 1.5-hour polonium to mass 205. Neumann and Perlman[3] have established the genetic relation between 8-hr. Pb^{201} and 72-hr. Tl^{201}. Barton, Ghiorso, and Perlman[8] showed, subsequent to the above experiments, that 5.5-hr. At^{209} decays both to Bi^{205} and to Po^{209}, thus confirming again the mass assignments. These decay relations are:

[8]Barton, Ghiorso, and Perlman, private communication.

Fig. 4. Yield of 14.5-day bismuth activity plotted to give half-life of polonium parent.

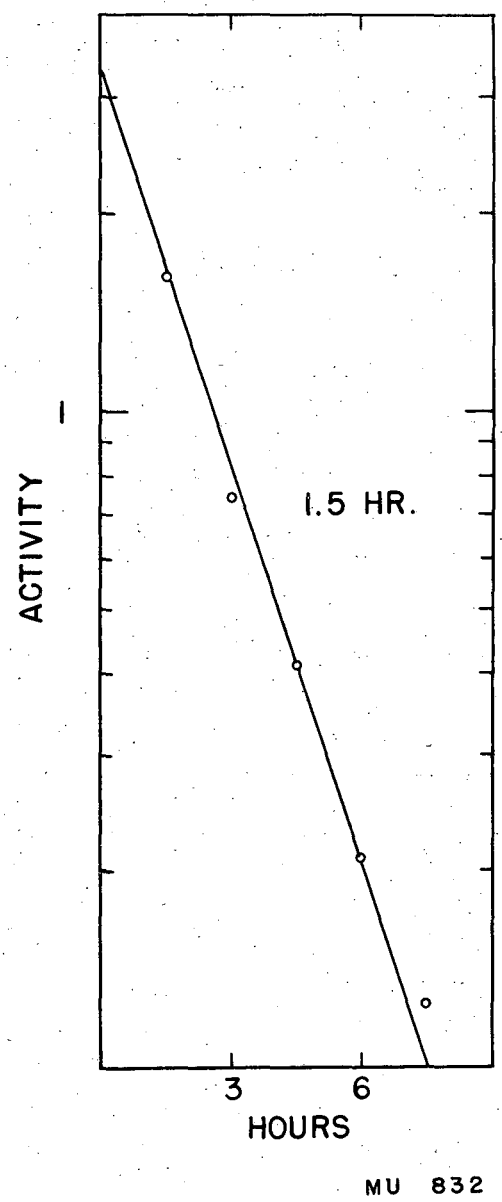
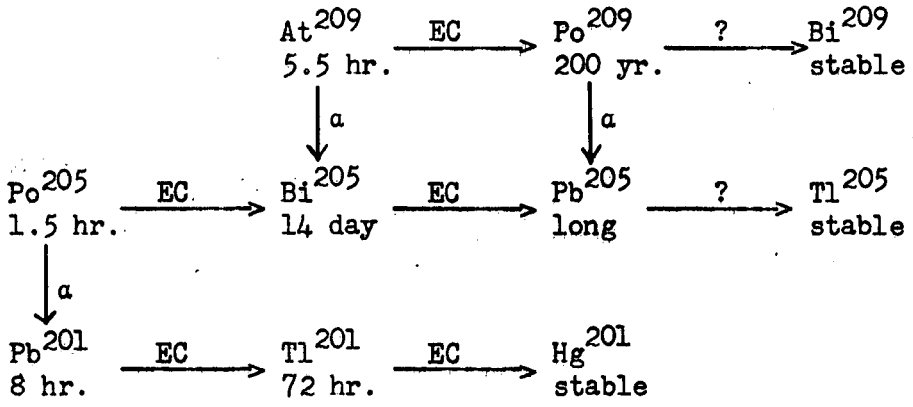


Fig. 4



The energy of the alpha-particles of Po^{205} was determined as 5.22 ± 0.10 Mev by following the decay of peaks obtained with the alpha pulse analyzer. The peak corresponding to Po^{206} (5.22 Mev)[9] decayed about 20 percent in four hours. Neighboring peaks due to Po^{208} and Po^{210} prevented greater accuracy in the energy estimate.

The ratio of electron capture disintegrations to alpha disintegrations is estimated as 400 by counting both the alpha-particles and the gross Geiger count, and making the crude assumption of one Geiger count (at 100 percent geometry) per electron capture event.

14.5-day Bi^{205}

The half-life of Bi^{205} produced by the decay of Po^{205} was found to be 14.5 days (Fig. 5). Aluminum absorption curves showed that about 15 percent of the Geiger counts of this isotope are due to electromagnetic radiation. Lead absorption measurements showed a gamma-ray of 1.7 Mev (half-thickness 14.5 g/cm^2). With a beta spectrometer we found conversion electrons corresponding to 1.84 Mev for this gamma-ray, as well as others for gamma-rays of 431, 527, 550 and 746 Kev. The existing data are insufficient to establish the decay scheme.

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[9] A. Ghiorso, private communication.

Fig. 5. Decay of 14.5-day Bi^{205}.

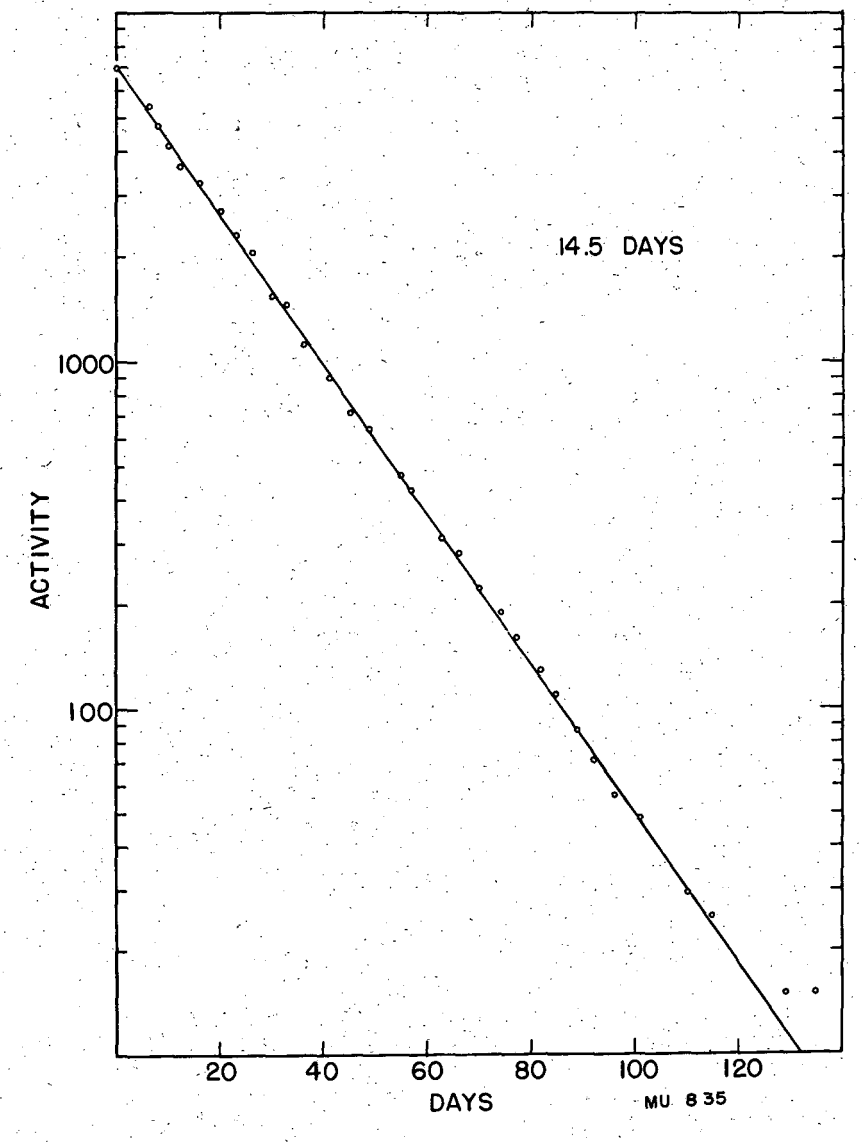


Fig. 5

47-min. Po^{203}

Repetition of the experiment described above--separation of the 12-hour bismuth daughters from a large sample of polonium at regular intervals--indicated that two polonium parents were producing 12-hour bismuth. The significant difference from earlier experiments was that the polonium was purified fairly rapidly, so that the daughter separations were started within an hour of the end of the bombardment. The data indicated another polonium isotope of a shorter half-life than Po^{204}. It must be at mass 203, since 52-hr. Pb^{203} appeared in the decay curves of the first bismuth fractions, but not in later ones.

The half-life of the Po^{203} was determined by preparing a large sample of polonium, purifying quickly after bombardment, and separating the bismuth daughters at half-hour intervals. The bismuth was allowed to decay for 24 hours, then the lead activity produced by bismuth decay was separated from the bismuth fraction. Since the lead daughters were allowed to grow into each bismuth fraction for the same period, the activity of 52-hr. Pb^{203} grown in was directly proportional to the initial activity of Bi^{203} in each fraction. The data plotted in Fig. 6 determine the half-life as 47 ± 5 minutes.

52-min. Po^{202} and 95-min. Bi^{202}

In addition to the other bismuth activities mentioned above, we observed a 95-minute activity among the bismuth daughters (Fig. 7) of polonium produced in bombardments at fairly high energy (protons of more than 70 Mev on bismuth, or helium ions of more than 120 Mev on lead). The yield of this 95-minute activity as a function of separation time showed that its polonium parent has a half-life of 52 ± 5 minutes (Fig. 8).

Fig. 6. Yield of 52-hr. Pb^{203} showing a 47-minute polonium parent.

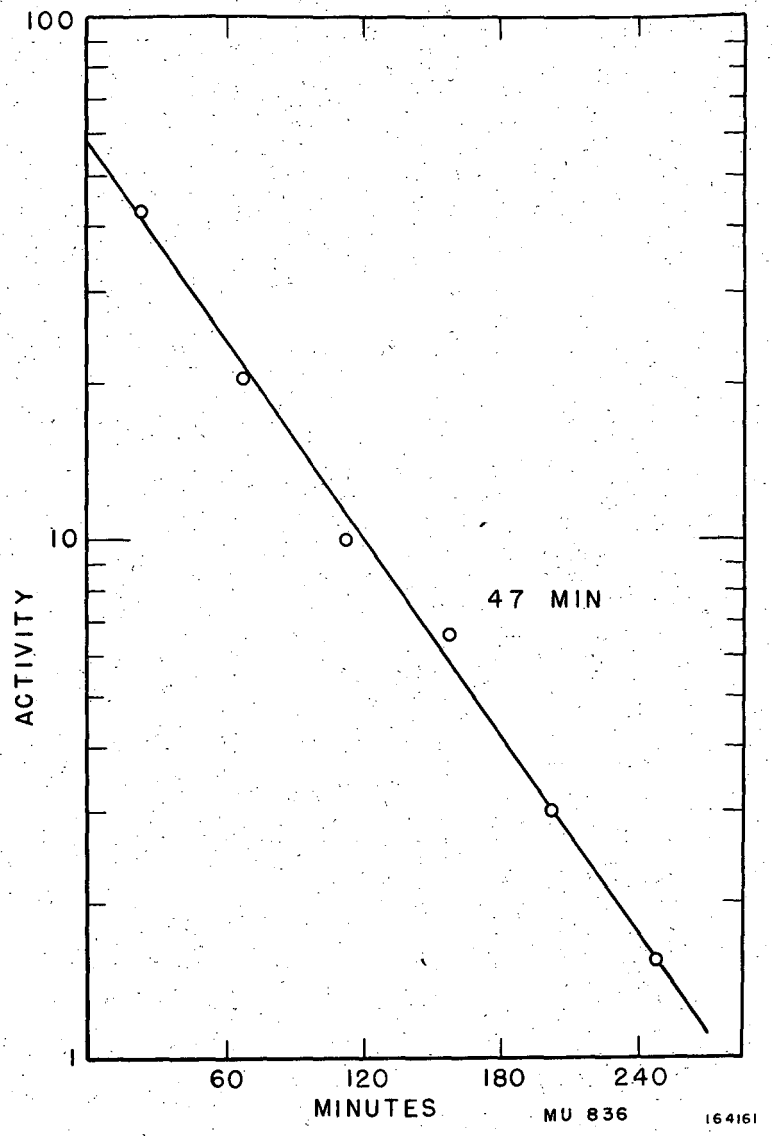


Fig. 6

Fig. 7. Decay of bismuth daughter activities.
A, 12-hr. Bi^{203} and 12-hr. Bi^{204}; B, 95-min. Bi^{202}.

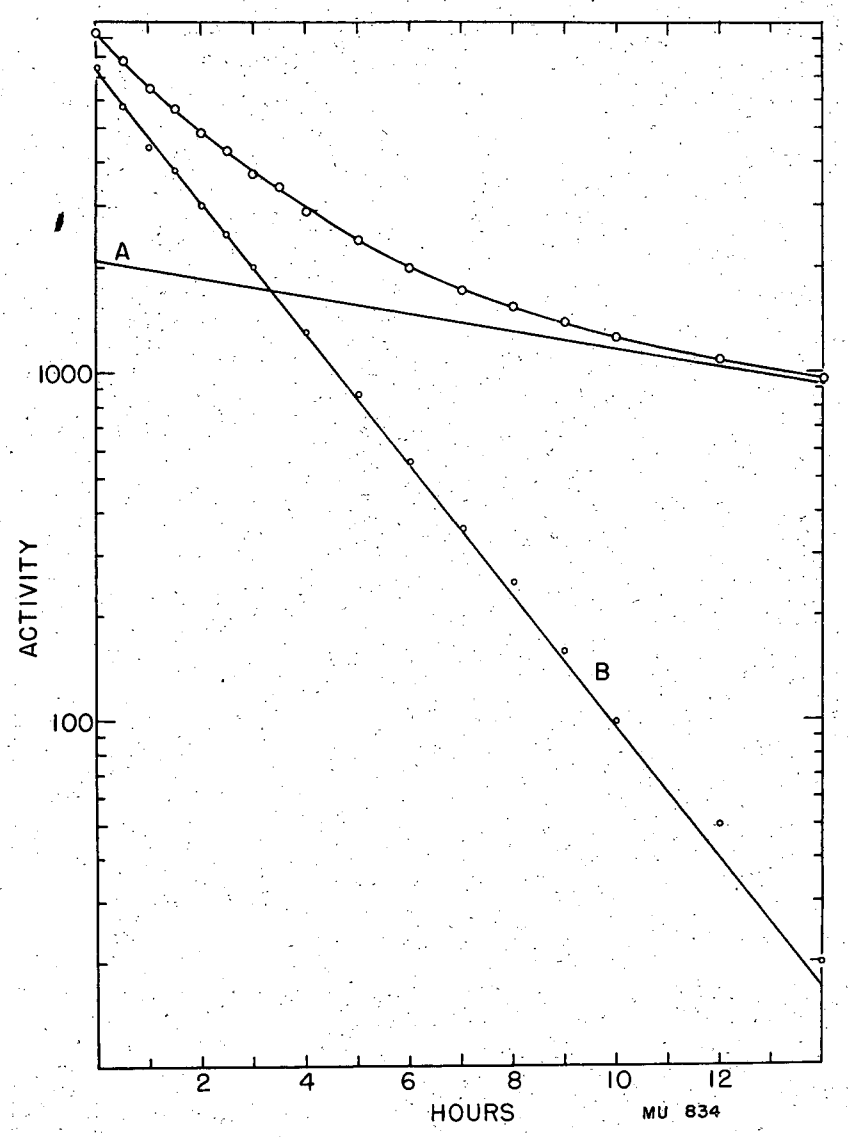


Fig. 7

Fig. 8. Yield of 95-min. Bi^{202} showing a 52-minute polonium parent.

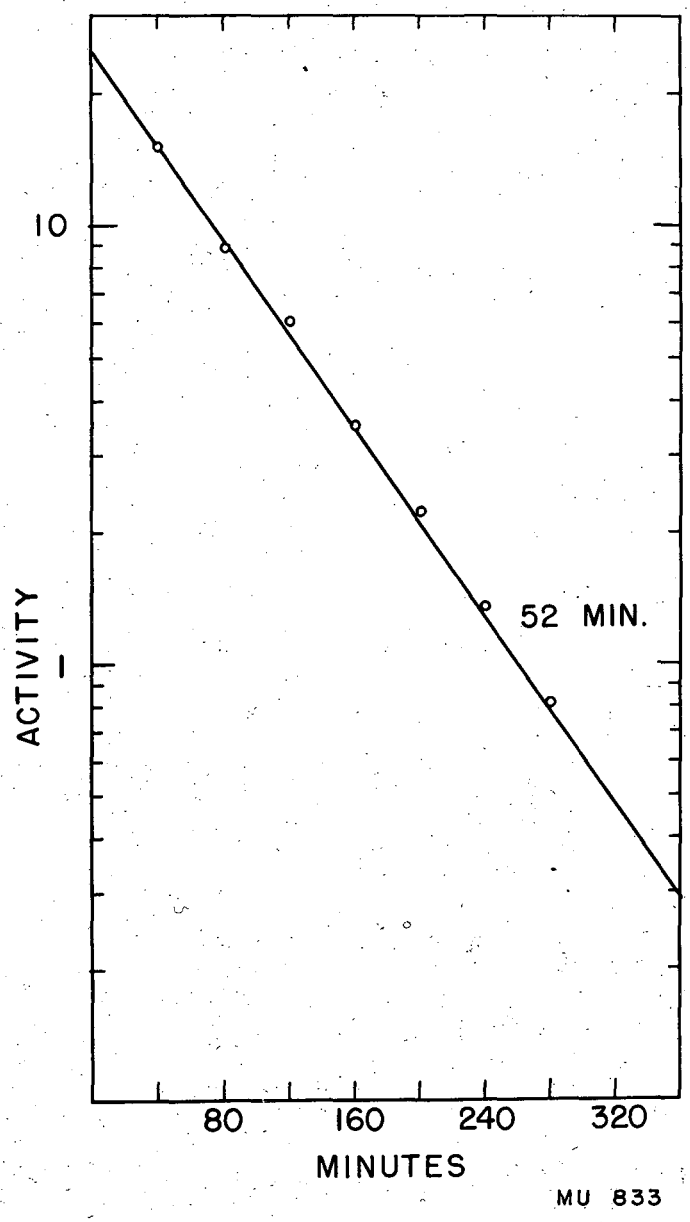


Fig. 8

If this polonium parent were the Po^{203} described above, then the new bismuth activity would be an isomer of Bi^{203}. In all attempts to milk Pb^{203} from this bismuth the yield corresponded to a bismuth parent of 12-hour half-life. However, this result does not rule out isomerism; if the ratio of isomeric transition to electron capture for the upper state is equal to $(t_2-t_1)/t_1$, where t_1 and t_2 are the respective half-lives of the upper and lower states, the daughter is produced at a rate corresponding exactly to decay of a single parent of half-life t_2.

The 95-minute activity was not detected in bismuth formed by irradiation of lead, containing 27 percent Pb^{204}, with 18-Mev deuterons from the 60-inch cyclotron.[7] Since the (d,3n) reaction is known to take place with high yield at this energy on bismuth,[2] it is expected to do the same on Pb^{204}. Bismuth of mass 203 was formed in this experiment, because lead separated from bismuth after a period of growth showed Pb^{203} in good yield. Therefore, it was presumed that the 95-minute activity is not Bi^{203}. If the assignment were 201 or 200, known lead and thallium activities should have been found as daughters. Thus 202 is the best assignment.

Additional evidence for the assignment to mass 202 was obtained from an excitation experiment, which at the same time identified the alpha-particles of 5.59 ± 0.03 Mev energy (Fig. 2) with Po^{202}. The electrostatically deflected proton beam from the 184-inch cyclotron was directed on a target composed of copper absorbers with bismuth foils interposed. From each foil the polonium was separated and purified. Pulse analysis of an aliquot of the polonium gave the yield of the 48-minute alpha-activity. After a period of about two hours, the bismuth daughters were separated from the remainder of the polonium. Decay measurements on an aliquot of this bismuth gave the relative yield of

Bi^{202} and thereby the yield of Po^{202}. The remainder of the bismuth was allowed to decay approximately a day, after which lead was separated and purified. The relative yield of Pb^{203}, and thereby of Po^{203}, was determined by following decay of this lead fraction. The resulting data are listed in Table V.

Table V

Total Absorber Thickness (g/cm^2)	Proton Energy[b] (Mev)	Relative Yield[a] 95-min. Bi	Relative Yield[a] Pb^{203}
82.76	(90)	0.93	0.12
84.74	(80)	1.01	0.22
85.68	(75)	1.10	0.22
86.59	(70)	1.14	0.36
88.34	(60)	≤ 1.3[c]	4.5

[a] In arbitrary units, proportional to the yield of 48-minute alpha activity in the same foil.

[b] Uncertain because the initial energy is not known exactly, and because of straggling in the absorbers.

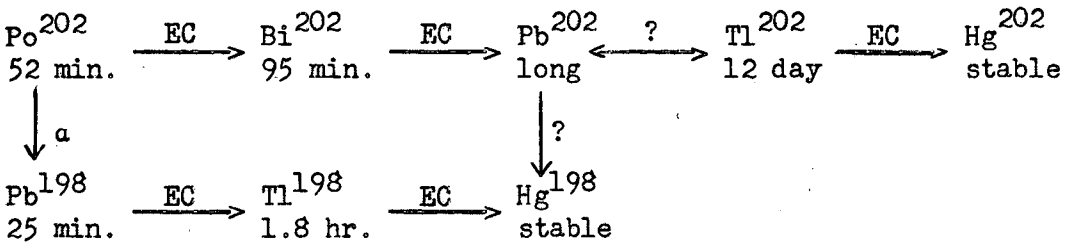
[c] Not detected.

From these data it is clear that the alpha-particles belong to the parent of the 95-minute bismuth, and that the mass assignment is less than 203. The variation of the yields from one foil to another is not presented here because of uncertain chemical yields in the isolation of the polonium; however, these data are in agreement with a mass difference of one unit for the 95-minute bismuth and Pb^{203}.

No alpha-particles were observed which could be attributed to Po^{203}. However, if the electron capture to alpha disintegration ratio were ten times larger than that of Po^{202}, it is doubtful if the alpha-particles could have been observed. From the energies of the alpha-particles of the other polonium isotopes, a reasonable prediction is that the alphas of Po^{203} have an energy of 5.4 ± 0.1 Mev.

No reliable data are available concerning the energies of the electrons and gamma-rays of Bi^{202}, since our samples were never pure. No positrons nor alpha-particles were found associated with it. If the alpha-decay to electron capture decay ratio is less than 10^{-5}, it is doubtful that alpha-decay would have been detected.

Identification of the lead alpha-decay products of Po^{202} and Po^{203}, or of the thallium daughters of these leads, failed for lack of sufficient radiochemical purity of the daughter samples. The half-life of Pb^{198} was determined by an experiment described below. Thus the decay of Po^{202} is presumed to be:



The long-lived Pb^{202} is still undetected.

25-min. Pb^{198}

The half-life of Pb^{198} was estimated by establishing a genetic relation between the 1.8-hr. Tl^{198} and its lead parent. This was done in the manner described before; a large sample of lead activities was prepared by bombarding

thallium with protons of 120 Mev, the lead separated and purified, and the thallium daughters separated at 30-minute intervals. The resolved yields of the 1.8-hour thallium show a half-life of 25 ± 10 minutes for its lead parent. Neumann and Perlman[3] have observed directly a lead activity which had an apparent half-life of 25 minutes, in agreement with our value.

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