# Artificial Radioactive Isotopes of Polonium

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Three new isotopes of polonium have been prepared by bomarding isotope-enriched lead samples with helium ions and bismuth with protons and deuterons. Evidence is given for their assignments as follows:

Isotope	Half-life	Decay	Radiations	Produced by
Po <sup>206</sup>	9 days	$K(90\%)$ , $\alpha(10\%)$	x-rays, $e^-$ , $\gamma 0.8$ Mev,	$\mathrm{Pb}^{204}(\alpha,2n)$
Po <sup>207</sup>	5.7 hours	$K(\sim 100\%), \alpha(0.01\%)$	x-rays, $e^{-(?)}$ , $\gamma 1.3$ Mev, $\alpha 5.1$ Mev	$\mathrm{Pb}^{206}(\alpha, 3n)$
Po <sup>208</sup>	$\sim$ 3 years	α	α5.14 Mev	${ m Pb}^{207}(lpha,3n)\ { m Bi}^{209}(p,2n)\ { m Bi}^{209}(d,3n)$

The 9-day Po<sup>206</sup> has been shown to decay to 6.4-day Bi<sup>206</sup>. No daughter activities of Po<sup>207</sup> and Po<sup>208</sup> have been found. No activity has been found for Po<sup>209</sup>. Also, 110-min.  $F^{18}(\beta^+)$  has been prepared by the reaction  $O^{16}(\alpha, pn)F^{18}$  or  $O^{16}(\alpha, 2n)Ne^{18}\xrightarrow{\text{short}}F^{18}$ .

### INTRODUCTION

ELEMENTS 81 to 84, thallium, lead, bis-muth, and polonium, have many isotopes which are members of the natural radioactive series and whose mass assignments and decay properties are well known.<sup>1</sup> These isotopes all have mass numbers greater than 205 for thallium and 209 for the other three elements. Several additional radioactive isotopes have been prepared by bombardment of the stable isotopes with high energy particles and with neutrons, but knowledge of some of these is much less complete. A thallium activity of 4.23-minute half-life has been assigned to Tl<sup>206</sup> by Broda and Feather, but details of the work have not yet been published.<sup>2</sup> The mass assignments of two other activities are well established: Pb<sup>209</sup>, decays by  $\beta^{-}$ -emission with 3.3hour half-life<sup>3-7</sup> and Tl<sup>204</sup> decays by  $\beta^{-}$ -emission

with 3.5-year half-life.4, 5, 8-10 Of particular importance to the investigation described in this paper is the isotope of bismuth, which decays by orbital electron capture with 6.4-day halflife and which is prepared by deuteron bombardment of lead<sup>4, 5</sup> and by helium-ion bombardment of thallium.11 Fajans and Voigt<sup>5</sup> demonstrated that the yield of the isotope depended only upon the Pb<sup>206</sup> content of the target using 9-Mev deuterons, and therefore that it is Bi<sup>207</sup> or Bi<sup>206</sup>, depending on the reaction involved. More recent work<sup>12</sup> indicates that at this energy both the (d,n) and (d,2n) reactions should take place. If 9-Mev deuterons can produce the (d,2n) reaction on Pb<sup>207</sup> the assignment to Bi<sup>206</sup> is favored since Fajans and Voigt could find no yield of the 6.4-day activity attributable to the Pb<sup>207</sup> contents of the targets. Corson, Mac-Kenzie, and Segrè<sup>13</sup> have shown that Bi<sup>207</sup>

<sup>8</sup> M. L. Pool, J. M. Cork, and R. L. Thornton, Phys. Rev. **52**, 239 (1937).

<sup>&</sup>lt;sup>1</sup> G. T. Seaborg, Rev. Mod. Phys. **16**, 1 (1944). <sup>2</sup> N. Feather, Phys. Rev. **70**, 88 (1946). Note added in proof: This work has been described recently by E. Broda and N. Feather, Proc. Roy. Soc. London A190, 20 (1947). <sup>3</sup> R. L. Thornton and J. M. Cork, Phys. Rev. 51, 383

<sup>(1937).</sup> <sup>4</sup> R. S. Krishnan and E. A. Nahum, Proc. Camb. Phil.

Soc. **36**, 490 (1940). <sup>5</sup> K. Fajans and A. F. Voigt, Phys. Rev. **60**, 619, 626

<sup>(1941).</sup> <sup>6</sup> R. S. Krishnan and E. A. Nahum, Proc. Roy. Soc.

<sup>(</sup>London) A180, 321 (1942). <sup>7</sup> W. Maurer and W. Ramm, Zeits. f. physik 119, 602 (1942).

<sup>&</sup>lt;sup>9</sup> K. Fajans and A. F. Voigt, Phys. Rev. 58, 177 (1940). <sup>10</sup> The mass assignment is deduced as 204 or 206 from published observations. The assignment of the 4.23-min. activity to Tl<sup>206</sup> (reference 2) leaves Tl<sup>204</sup> for the 3.5-year

<sup>&</sup>lt;sup>11</sup> The bombardment of thallium with 40-Mev helium ions will be described in a later paper by the present authors.

<sup>&</sup>lt;sup>12</sup> For example, see footnote in reference (5)-also, unpublished work of the Manhattan Project. <sup>13</sup> D. R. Corson, K. R. MacKenzie, and E. Segrè, Phys.

Rev. 58, 672 (1940).

must have a half-life greater than a month by their failure to detect it as the daughter of the alpha-decay of At<sup>211</sup>. <sup>14</sup> This observation adds further evidence against the Bi<sup>207</sup> assignment for the 6.4-day activity. Additional evidence for the Bi<sup>206</sup> assignment is described below.

The existing literature contains the observation and provisional isotopic assignments of several other light isotopes of thallium, lead, and bismuth.<sup>1</sup> Some of these have been encountered in the present series of bombardments and will be reported in detail in later papers together with additional evidence regarding their assignments.

No isotopes of polonium lighter than 210 have been reported. Corson, MacKenzie, and Segrè<sup>15,16</sup> obtained alpha-active material on bombarding lead and bismuth with 32-Mev helium ions but have made a detailed investigation only of the products from bismuth.<sup>13</sup>

These elements present an attractive field for further investigation in this laboratory since beams of 40-Mev helium ions and 20-Mev deuterons are possible with the 60-inch cyclotron.<sup>17</sup> Particles of these energies produce  $(\alpha,n), (\alpha,2n), (\alpha,3n), (d,p), (d,n), (d,2n), and$ (d,3n) reactions on these elements in high yield. Most of these reactions produce isotopes of lower mass number than the stable ones, and it is these isotopes about which relatively little is known. High intensity neutron sources now available make possible the production of certain long-lived activities which otherwise could not be detected. In addition, gram quantities of various stable isotopes of lead and thallium have been concentrated in the magnetic isotope separators at this laboratory.<sup>18</sup> Bombardment of different isotopic mixtures permits deductions

TABLE I. Isotopic compositions of lead samples.

Sample isotope	Natural lead*	Lead A	Lead B	Lead C	Lead D
204	1.5%	27.3%	<0.2%	<0.2%	Natural uranium
206	23.6	32.7	59.7	1.9	lead, largely
207	22.6	13.8	25.2	7.8	Pb <sup>206</sup> , but analy-
208	52.3	26.2	15.1	90.3	sis not available.

\* A. O. Nier, J. Am. Chem. Soc. 60, 1571 (1938).

concerning the reactions taking place and the mass assignments of the products. The results of this program were desired to facilitate the interpretation of the data from bombardments of elements in this region in the 184-inch cyclotron with deuterons of energies up to 200 Mev and helium ions up to 400 Mev, to be described in later papers.

### EXPERIMENTAL

The isotopic compositions of the lead samples used are listed in Table I, as determined by a Nier type mass spectrometer. These samples were purified by recrystallization as lead chloride. A sample of 1 to 50 mg was spread as a slurry of lead hydroxide on a platinum "interceptor" target, dried under an infra-red heat lamp, and heated for 30 minutes at 650°C. The interceptor, of 1-cm<sup>2</sup> area, receives one-third to one-half of the beam of the 60-inch cyclotron. Ordinary lead was bombarded in the form of metal plates large enough to receive the entire beam and mounted on copper disks. Bismuth was bombarded as the oxide mounted on a similar copper disk.

The various samples were bombarded with 20-Mev deuterons or 40-Mev helium ions in the 60-inch cyclotron, usually for 1 to 4 hours. Immediately after bombardment some fraction or all of the lead oxide was dissolved in acetic acid, the lead metal in nitric acid, and the bismuth oxide in hydrochloric acid. Chemical separations were made after addition of inactive carriers when necessary by some combination of the procedures outlined below. The RaE(Bi<sup>210</sup>) and RaF(Po<sup>210</sup>) always present were very useful for calculating the chemical yields of the various operations. A measured fraction of the solution before chemical separation was evaporated directly on a platinum counting disk.

Polonium was deposited on a silver disk immersed in a 2-M hydrochloric acid solution

<sup>&</sup>lt;sup>14</sup> Element 85 has been named astatine (symbol At) by D. R. Corson, K. R. MacKenzie, and E. Segrè, Nature 159, 24 (1947).
<sup>15</sup> D. R. Corson and K. R. MacKenzie, Phys. Rev. 57,

<sup>&</sup>lt;sup>15</sup> D. R. Corson and K. R. MacKenzie, Phys. Rev. **57**, 250 (1940).

<sup>&</sup>lt;sup>16</sup> D. R. Corson, K. R. MacKenzie, and E. Segrè, Phys. Rev. 57, 459 (1940).

<sup>&</sup>lt;sup>17</sup> 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.

cyclotron. <sup>18</sup> We are indebted to Dr. B. J. Moyer, Dr. C. M. VanAtta, and members of the isotope separation staff for making available the enriched isotopes, to Dr. E. H. Huffman, Mr. R. C. Lilly, and Miss Dorothy Bockhop for their purification, and to Mr. J. T. Vale for the mass spectrometric assays.

free of nitrate. With vigorous stirring a considerable fraction plated onto the silver in a few minutes, but the yield was not quantitative. Neither bismuth nor lead activities were detected on the silver plates. Another procedure was to heat a dry sample in a small crucible which was covered by a platinum plate on which was set a small beaker of cold water. Some of the polonium can be collected on the platinum in this manner.

Bismuth was precipitated as the iodate from 3-M nitric acid solution with an excess of potassium iodate. The precipitate was dissolved in concentrated nitric acid for reprecipitation if desired. The slurry of precipitate was spread on a platinum plate and ignited vigorously in a Bunsen flame to the oxide. This procedure usually removed nearly all of the polonium and resulted in thin, coherent deposits. Another method made use of dithizone (diphenylthiocarbazone) in chloroform which extracts bismuth from aqueous solutions at pH 3.19 The bismuth was recovered by washing the chloroform with a small volume of dilute nitric acid. Bismuth hydroxide or bismuth iodate was then precipitated and spread on a counting disk.

Lead was precipitated by heating a sulfuric acid solution to fumes of SO<sub>3</sub> and diluting with water. The lead sulfate precipitate was dissolved in ammonium acetate solution. Bismuth impurity was precipitated at this point as the hydroxide. Additional bismuth carrier was sometimes added to improve the bismuthactivity separation. The supernatant was made acidic with acetic acid and the lead was precipitated as lead chromate, which, when spread as a slurry, made uniform counting plates. An alternate procedure proved useful for small quantities. Lead was extracted with dithizonechloroform solution from a solution which contained oxalate, cyanide, and hydroxylamine at pH 9. Bismuth had to be removed first as described above. Like bismuth, lead was recovered from the chloroform by an acid wash. Thallium could be recovered from the first aqueous residue.

## Counting Equipment<sup>20</sup>

Electrons and electromagnetic radiation were measured with argon-ethanol filled Geiger tubes having mica windows of about 3-mg/cm<sup>2</sup> thickness. The geometry factor was about 13 percent for the sample position most frequently used. Electromagnetic radiation, positrons, and negative electrons or beta-particles were differentiated with a similar Geiger tube mounted between the poles of an electromagnet. The sample was mounted in such a way that either the undeflected radiation, the positive particles, or the negative particles were counted. This apparatus also served as a crude magnetic spectrometer for electrons. In some cases a silvered glass-wall Geiger tube, of thickness about 50 mg/cm<sup>2</sup>, was also used. Alphaparticles were counted with a Simpson methane proportional counter<sup>21</sup> or in an argon-filled ionization chamber developed in this laboratory. Mixtures of alpha-particles of different energies were analyzed with an alpha-energy analyzer. In this instrument<sup>22</sup> pulses from an argon-filled ionization chamber are amplified and sorted according to size in 48 registering circuits. The pulse resulting from an alpha-particle is recorded on only one of the registers. The ordinal numbers of the registers are a linear function of the amount of ionization produced in the chamber by one alpha-particle and, therefore, over a small range a linear function of the energy of the alpha-particle. A plot of number of alpha pulses recorded against register number is then an alpha particle energy distribution curve (Fig. 3). The energy of an unknown group of particles was estimated by interpolation between alpha-groups of known isotopes which were placed in the ionization chamber at the same time.

# **Cross Section Measurements**

Cross sections were calculated directly only for thin samples mounted on the small inter-

<sup>&</sup>lt;sup>19</sup> Cf. E. B. Sandell. Colorimetric Determination of Traces of Metals (Interscience Publishers, Inc., New York, 1944), pp. 72, 157, 282.

<sup>20</sup> The authors are indebted to Mr. H. P. Robinson, who was responsible for the electronic equipment, and to Mr. R. A. James and Mr. L. O. Morgan who designed the magnetic counter.

 <sup>&</sup>lt;sup>21</sup> J. A. Simpson, Jr., Phys. Rev. 70, 117 (1946).
 <sup>22</sup> A. Ghiorso, A. H. Jaffey, H. P. Robinson, and B. Weissbourd, An Alpha Pulse Analyzer Apparatus (Plutonium Project Record, Vol. 14B, 17.3 (1946)) (to be issued).



FIG. 1. Decay of gamma-ray of  $Po^{207}$ , counted through 0.46 g/cm<sup>2</sup> aluminum.

ceptor targets. It was assumed that the beam intensity was not diminished appreciably by the thin sample. The total beam current to the target was integrated by the cyclotron instruments. The number of atoms bombarded was calculated from a gravimetric or colorimetric determination of material in a measured fraction of the solution dissolved after bombardment. The number of atoms transmuted was estimated from counting data for known fractions of this same solution.

### RESULTS

#### 140-Day Po<sup>210</sup>

In every case in which lead was bombarded with 40-Mev helium ions  $Po^{210}(RaF)$  was produced by the reaction  $Pb^{208}(\alpha,2n)Po^{210}$ . Its identity was checked by the alpha-pulse analyzer and by decay measurements. It constituted almost all of the alpha-active material formed when lead *C* (Table I) was bombarded. It was also formed indirectly by decay of 5-day  $Bi^{210}(RaE)$  from  $Pb^{208}(\alpha,pn)Bi^{210}$ , but this contribution was small and was corrected for in all calculations.

Three new activities were observed which followed Po<sup>210</sup> quantitatively in all chemical separations and which were separated from bismuth, lead, and thallium.

# 5.7-Hour Po<sup>207</sup>

Bombardment of lead C yielded a 6-hour polonium Geiger activity. It was produced in



FIG. 2. Aluminum absorption curve for 5.7-hour Po<sup>207</sup>.

much better yield from lead B, and its half-life was observed as  $5.7\pm0.1$  hours for 8 half-lives by counting its gamma-radiation (Fig. 1). An aluminum-absorption curve is shown in Fig. 2. The soft component is probably a mixture of Lx-rays and conversion electrons. No observations were made on the isolated polonium with a magnetic field, but the original mixture was shown to contain no positron activity of half-life longer than two hours. The gamma-ray has a halfthickness in lead of 12 g/cm<sup>2</sup>, corresponding to 1.3 Mev. These activities are assumed to be due to an orbital electron capture forming an unidentified bismuth isotope.

A weak alpha-activity of half-life about 6 hours was observed in the polonium from lead B and lead D. In other bombardments it was masked by large yields of other alpha-emitters. With the pulse-analyzer the 6-hour decay was shown to be part of a peak at 5.14 Mev, and not of the Po<sup>210</sup> peak at 5.30 Mev. Thus the energy of the alpha-particles is about 5.1 Mev.

Yield data presented below for different isotopic mixtures show that the 5.7-hour isotope is made chiefly from Pb<sup>206</sup> by 40-Mev helium ions. It is not made in appreciable yield from Bi<sup>209</sup> by 20-Mev deuterons. If the  $(\alpha,3n)$  and (d,3n) reactions are prolific while the  $(\alpha,4n)$  and (d,4n) are not at these energies, the isotopic assignment must be Po<sup>207</sup>.

Estimating the Pb<sup>206</sup>( $\alpha$ ,3*n*)Po<sup>207</sup> cross section as  $1-2 \times 10^{-24}$  cm<sup>2</sup>, the counting efficiency of Po<sup>207</sup> must be 3-5 percent. Since this is higher



FIG. 3. Pulse analysis curve of Po<sup>208</sup>, Po<sup>210</sup> mixture, with standard Th<sup>230</sup> plate also in ionization chamber.

than expected for L x-rays, some of the soft component is attributed to conversion electrons. This counting efficiency corresponds to an alpha-branching ratio of  $5-8\times10^{-5}$ , or  $\sim0.01$ percent. Thus the half-life for alpha-decay is several years, as expected for an alpha particle of 5.1 Mev in this region.

No daughter of this isotope has been observed. The alpha-branching is so small that observation of its daughter could not be expected in our experiments. The bismuth daughter of the electron capture must be short (less than 15 min.) or very long (many years). In particular, it cannot be the 6.4 day Bi<sup>206</sup>. From analogy to known isotopes of other odd elements, Bi<sup>207</sup> is expected to be rather long-lived.

### 3-Year Po<sup>208</sup>

Pulse analysis of the long-lived alpha-particles obtained from lead A, lead B, and lead D showed in addition to those of Po<sup>210</sup> a new group of particles at 5.14 Mev (Fig. 3). Bombardment of bismuth oxide with 20-Mev deuterons produced the same activity in good yield. Lofgren<sup>23</sup> obtained a small amount of long-lived polonium alpha-activity by bombardment of bismuth metal with 15-Mev protons accelerated in the 37-inch frequency-modulated cyclotron. We showed by pulse analysis that it had the same range as this new isotope and that it was free of Po<sup>210</sup>.

Repeated alpha-pulse analyses for three months showed less than 10 percent decay in the new peak, while that of  $Po^{210}$  decayed about 35



FIG. 4. Decay of Po<sup>208</sup>, corrected for Po<sup>210</sup> by means of pulse analysis.

percent as expected for its 140-day half-life. Direct decay measurements for ten months, corrected for  $Po^{210}$  from pulse-analysis data, showed decay of the new polonium isotope with about a 3-year half-life (Fig. 4). A large sample of this isotope after five months had so little Geiger activity associated with it, if sufficient mica was interposed to stop the alpha-particles, that the half-life of the lead daughter, if it emits x-rays, must be longer than 100 years.

From arguments presented below, based on yields from the different isotopic mixtures, the isotope is assigned to  $Po^{208}$ .

### 9-Day Po<sup>206</sup>

Bombardment of lead B with 40-Mev helium ions produced a small yield Geiger activity in



FIG. 5. Growth and decay of Po<sup>206</sup>, Bi<sup>206</sup> mixture. The solid line is the theoretical curve obtained by subtraction of the 6.4-day line from the 9-day line.

<sup>&</sup>lt;sup>23</sup> We wish to thank Dr. E. J. Lofgren for his cooperation in providing us with this sample.

the polonium fraction whose half-life was variously observed over short time intervals as 12 to 20 days. With lead A the yield was much better, and the anomalous decay rate was explained. Figure 5 shows the decay of a sample of polonium purified after the 5.7-hour activity was dead. An initial growth was observed, followed by decay, which suggests that the observed count is due to a parent and daughter of comparable half-life. Bismuth separated from purified polonium a few days old was found to decay with about a 6-day half-life and was concluded to be identical with the well-known 6.4-day Bi<sup>206</sup>. The observed data are best fitted by assuming the half-life of the parent to be 9 days, and the counting efficiency of the bismuth to be about 1.3 times that of the polonium. If  $C_0$  is the initial counting rate of Po, k the ratio of counting efficiency of Bi to Po, and  $\lambda_1$  and  $\lambda_2$  the decay constants of Po and Bi, respectively, the total counting rate at time t for the Po, plus the Bi which has grown in, is:

$$C = C_0 [1 + k\lambda_2/(\lambda_2 - \lambda_1)] e^{-\lambda_1 t} - C_0 [k\lambda_2/(\lambda_2 - \lambda_1)] e^{-\lambda_2 t}.$$

The two terms of this expression, when plotted on semilog graph paper, are straight lines of slope corresponding to the two half-lives (Fig. 5).

In Fig. 6 is shown an aluminum-absorption curve for the radiations. The soft component has two definite groups of electrons of ranges estimated as approximately 100 and 300 mg/cm<sup>2</sup>



FIG. 6. Aluminum absorption curve for Po<sup>206</sup>.



FIG. 7. Alpha-decay curve for polonium from lead A, showing 9-day Po<sup>206</sup>.

aluminum, corresponding to 400 and 800 kev. The gamma-ray has a half-thickness in lead of  $7.8\pm0.5$  g/cm<sup>2</sup>, corresponding to about 800 kev. There is also a weak gamma-component of 1 g/cm<sup>2</sup> half-thickness (~300 kev). The two electron groups probably are due to partial conversion of the two gamma-rays, the discrepancy in energies being less than the experimental uncertainty. The x-rays make only a small contribution to the observed counting rate, and they have not been resolved in our absorption measurements.

No positrons have been observed in mixtures which contained Po<sup>206</sup>. Therefore the decay is by orbital electron capture, since a bismuth daughter is produced.

In Fig. 7 is shown the alpha-decay curve for the polonium from lead A. In addition to 7-hour (At<sup>211</sup> contamination  $+Po^{207}$ ) and >150-day (Po<sup>210</sup>+Po<sup>208</sup>) periods a component of about 9-day half-life was observed. Pulse analysis failed to resolve this peak from those at 5.14 and 5.30 Mev. The energy is therefore  $5.2 \pm 0.1$  Mev. The yield of this alpha-activity from Pb<sup>204</sup> was 0.095 that of Po<sup>210</sup> from Pb<sup>208</sup> in the same bombardment. If the alpha-activity is due to branching decay of Po<sup>206</sup> and if the cross sections for  $Pb^{204}(\alpha,2n)Po^{206}$  and  $Pb^{208}(\alpha,2n)Po^{210}$  are equal, the decay of Po<sup>206</sup> must be 10 percent alpha and 90 percent electron capture. This corresponds to a counting efficiency of about 18 percent for the gamma-rays, x-rays, and electrons of Po<sup>206</sup>, and 23 percent for Bi<sup>206</sup>, for samples mounted on

silver with no correction for back scattering or for window ( $\sim 3 \text{ mg/cm}^2$ ) and air absorption.

If the 9-day alpha-activity is assigned to Po<sup>205</sup> the yield is too low, and for Po<sup>204</sup> too high, on the basis of the arguments outlined below concerning  $(\alpha, 3n)$  and  $(\alpha, 4n)$  yields. The 90-day half-life for alpha-decay, calculated from the above branching ratio, agrees better with the observed energy of the alpha-particle than the 9-day value does.

The daughter of the alpha-decay has not been observed. It is not the 52-hour lead, whose assignment to Pb<sup>203</sup> will be discussed in a later paper. If it is Pb<sup>202</sup> decaying to 13-day Tl<sup>202</sup>, the half-life of Pb<sup>202</sup> must be longer than a few weeks. Other experiments also have indicated that Pb<sup>202</sup> is relatively long-lived.24

## Yields

In Table II are listed the ratios of the yields of the polonium isotopes from lead B to those from lead C, together with ratios of isotope concentrations in the two lead samples. From these data it is evident that Po210 was made predominantly by the reaction  $Pb^{208}(\alpha, 2n)Po^{210}$  and that the 5.7-h polonium was made mostly from Pb<sup>206</sup>. The 3-year polonium was made from Pb<sup>207</sup> or to some extent from all three lead isotopes. The concentration of Pb<sup>204</sup> in these samples was so low that it had no effect. The 9-day polonium yield was about 100 times higher from lead Athan from lead B or lead C, and therefore this isotope is made from Pb<sup>204</sup>.

The cross section for the reaction Pb<sup>208</sup>- $(\alpha, 2n)$ Po<sup>210</sup> with 40-Mev helium ions was calculated as  $0.4 \times 10^{-24}$  cm<sup>2</sup>.

For the bombardment of bismuth oxide with 20-Mev deuterons, cross sections were calculated as follows:

${\operatorname{Bi}}^{{209}}(d,p){\operatorname{Bi}}^{{210}},$	$\sigma = 0.13 \times 10^{-24} \text{ cm}^2$ ;
${ m Bi}^{209}(d,n){ m Po}^{210},$	$\sigma = 0.036 \times 10^{-24} \text{ cm}^2$ ;
$Bi^{209}(d.3n)Po^{208}$ .	$\sigma = 1.1 \times 10^{-24} \text{ cm}^2$ .

The absolute accuracy is rather uncertain, but the relative values should be correct within a few percent. The ratio  $\sigma(d,p)/\sigma(d,n) = 3.6$  agrees

TABLE II. Relative yields from lead B and lead C plus 40-Mev helium ions; and relative concentrations of lead isotopes.

Isotope	Yield $B$ /yield $C$	Isotope	% <b>B</b> /%C
Po <sup>210</sup>	0.16	Pb <sup>208</sup>	0.17
3-v. Po	$\sim 3.5$	$Pb^{207}$	3.20
5.7-hr. Po	20.0	$\mathrm{Pb^{206}}$	31.00

well with a reasonable extrapolation of the available data at lower energies.25-28 The absolute values reported by Cork and co-workers<sup>26-28</sup> appear to be inconsistent with this research and with each other.

# **Mass Assignments**

It has been generally observed for uranium and neighboring elements that the cross sections for a particular type of reaction such as  $(\alpha, 2n)$ of nuclei with high energy particles are of comparable magnitude for neighboring isotopes.<sup>29</sup> If we assume this generalization for the leadpolonium region, we can deduce the mass assignments of the three new polonium isotopes. The  $(\alpha,n)$  reaction is known to be poor for Pb<sup>207</sup>- $(\alpha, n)$ Po<sup>210</sup>. The cross section for Pb<sup>208</sup> $(\alpha, 2n)$ Po<sup>210</sup>, is about 0.4×10<sup>-24</sup> cm<sup>2</sup>. Leininger and Segrè<sup>30</sup> using the same energy helium ions have shown that for  $Bi^{209}(\alpha, 3n)At^{210\text{short}}Po^{210}$ , the yield is even higher than for  $Bi^{209}(\alpha, 2n)At^{211}$ .

The 9-day polonium is 206 or lighter, since it is made in good yield from Pb<sup>204</sup>. But its daughter the 6.4-d. Bi, is 206 or 207.5 Therefore both must be 206, which is confirmed by other arguments stated above for the bismuth isotope.

The low yield of  $Po^{206}$  from lead B shows that the cross section for the reaction  $Pb^{206}(\alpha, 4n)Po^{206}$ is very small. The 5.7-hour isotope must be Po<sup>207</sup> or Po<sup>208</sup>, since it is made in good yield from Pb<sup>206</sup>. If Po<sup>208</sup>, its yield should depend more on the Pb<sup>207</sup> than it does. The (d,3n) cross section for 20 Mev deuterons is known to be high for heavier elements, and it is expected to be high for bismuth. The very low yield of 5.7-hour

<sup>30</sup> R. F. Leininger and E. Segrè, Private Communication

<sup>&</sup>lt;sup>24</sup> Bombardment of thallium with 20-Mev deuterons should produce  $Pb^{202}$  by the reaction  $TI^{203}(d,3n)Pb^{202}$ . In such a bombardment, to be described in a later paper, we failed to observe 13-day Tl<sup>202</sup>.

 <sup>&</sup>lt;sup>25</sup> D. G. Hurst, R. Latham, and W. B. Lewis, Proc. Roy. Soc. (London) 174, 126 (1940).
 <sup>26</sup> J. M. Cork, J. Halpern, and H. Tatel, Phys. Rev. 57, 348, 371 (1940).

 <sup>&</sup>lt;sup>26</sup> J. M. Cork, Phys. Rev. **70**, 563 (1946).
 <sup>28</sup> H. E. Tatel and J. M. Cork, Phys. Rev. **71**, 159 (1947).
 <sup>29</sup> Unpublished work of the Manhattan Project.

polonium from bismuth plus deuterons makes the 208 assignment unsatisfactory. Therefore the isotope must be Po<sup>207</sup>.

The 3-year polonium, made in good yield from Pb<sup>207</sup>, must be Po<sup>208</sup> or Po<sup>209</sup>. The high yield of bismuth plus 20 Mev deuterons confirms this restriction. But the low yield from lead C makes the 209 assignment unsatisfactory, for the reaction  $Pb^{208}(\alpha, 3n)Po^{209}$  should have a high cross section. The observed yield from lead B corresponds to  $\sigma = 0.81 \times 10^{-24}$  cm<sup>2</sup>. This is in agreement with  $Pb^{206}(\alpha, 2n)Po^{208}$  ( $\sigma = 0.4 \times 10^{-24} \text{ cm}^2$ ) and  $Pb^{207}(\alpha, 3n)Po^{208}$  ( $\sigma = 3 \times 10^{-24} \text{ cm}^2$ ).

No activity has been observed which can be assigned to Po<sup>209</sup>. This isotope should be produced in high yield by the reactions  $Bi^{209}(d,2n)Po^{209}$  and  $Pb^{208}(\alpha, 3n)Po^{209}$ . If its half-life is short, it must be less than 20 min. If it is long-lived and emits alpha particles of about the same energy as Po<sup>208</sup>, it would escape detection if its half-life were 10 years or more.

Some of the alpha particles attributed to Po<sup>208</sup> may have come from Po<sup>209</sup>, but because of the yield arguments stated above most of them must belong to Po<sup>208</sup>. If Po<sup>209</sup> is long-lived and emits x-rays, its half-life must be greater than 1000 years.

### 110-minute $F^{18}$

In the various bombardments of lead oxide with 40-Mev helium ions a Geiger activity was observed of half-life  $107 \pm 5$  minutes. About 1 percent of the radiation penetrated 2  $g/cm^2$  of lead. With the magnetic counter positions were detected which decayed with a half-life of 115  $\pm 10$  min. Chemical separations showed that it was not polonium, bismuth, lead, or thallium. The yield was very erratic when samples were evaporated from acid solution. The activity was carried by a lanthanum fluoride precipitate. This activity is believed to be due to F<sup>18</sup>, formed by

the reaction  $O^{16}(\alpha, pn)F^{18}$  or indirectly by  $O^{16}(\alpha, 2n) Ne^{18 \text{short}} F^{18}$ . The half-life of  $F^{18}$  has been reported variously as 107 to 120 minutes, and it is known to be a positron emitter.<sup>31-39</sup>

### 7.5-hour At<sup>211</sup>

In the polonium from lead A, two long-range alpha-peaks were observed with the pulseanalyzer. Both decayed with a  $7.5 \pm 1$  hour halflife, and their energies are in agreement within experimental error with the reported values 5.94 Mev<sup>13</sup> and 7.44<sup>40</sup> Mev for At<sup>211</sup> and Po<sup>211</sup>, respectively. The yields were in the ratio 44/56in agreement with the ratio 40/60 reported<sup>13</sup> for At<sup>211</sup> and Po<sup>211</sup> in equilibrium. Astatine would have plated on metallic silver and would not have been separated from the polonium.

The observed yield corresponded to an impurity of a few parts per million of bismuth in the lead sample, from which it would be made by the reaction  $\operatorname{Bi}^{209}(\alpha, 2n)\operatorname{At}^{211}$ . The first component in Fig. 7 is largely due to this activity, and prevented observation of Po<sup>207</sup> in this case. In other bombardments pulse analysis showed that At<sup>211</sup> was absent.

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 <sup>34</sup> T. Yasaki and S. Watanabe, Nature 141, 787 (1938) (half-life 120 min.).
 <sup>35</sup> M. L. Durgidger, Phys. Rev. 57, 1086 (1040)

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<sup>36</sup> S. B. Welles, Phys. Rev. 59, 679 (1941) (half-life 114 min.). <sup>37</sup> R. S. Krishnan, Nature **148**, 407 (1941) (half-life

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<sup>&</sup>lt;sup>31</sup> A. H. Snell, Phys. Rev. 51, 143 (1937) (half-life  $112 \pm 4$  min.).