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Rept. submitted for pub. in
Inorganic and Nuclear Chemistry.

UCRL-11148

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
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AEC Contract No. W-7405-eng-48

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ABSTRACT

The half-lives of isomers of At^{208}, At^{206}, and At^{204} produced in alpha particle bombardments of bismuth were investigated by periodically separating and analyzing the polonium daughters.

Only the shorter-lived "isomer" of each astatine isotope was observed; i. e. 9 min At^{204}, 33 min At^{206}, and 1.6 hr At^{208}. It was found that the previously assigned longer-lived isomers of At^{208}, At^{206}, and probably At^{204} were illusions due to chemical effects.

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I. INTRODUCTION

Early bombardments of bismuth with high energy alpha-particles from the 184-inch cyclotron produced a 25-min isotope which was assigned to At^{204}, a 2.7-hr isotope assigned to At^{206} and a 6.3-hr isotope assigned to At^{208}; all decayed by electron-capture and were detected by the growth of known polonium daughters.[1] In a different type of experiment there was produced a 1.7-hr astatine activity[2] which decayed by α-emission and electron capture and this was also assigned to At^{208}. The 2.7-hr period for At^{206} and 6.3-hr period for At^{208} were apparently confirmed[3] in experiments similar to those first used.

More recent bombardments of gold with carbon ions[4, 5] have produced a 9-min α-emitter assigned to At^{204} and a 30-min alpha-emitter assigned to At^{206}. In the carbon ion bombardments no evidence was found for longer-lived isomers of At^{204} and At^{206}. A special effort was made to detect the longer-lived isomers.[5] For At^{206} an upper limit of 0.07 for the ratio of the yield of 25-min period to that of the 9-min period was set, and for At^{204} the upper limit for the ratio of the yield of the 2.7-hr period to that of the 30-min period was 0.04. Based upon these observations the question has been raised whether different methods of production of odd-odd astatine isotopes may yield different isomers.[4, 5]

The initial object of this work was to determine if the 9-min At^{204} and 30-min

At^{206} are made in an alpha-particle bombardment of bismuth, and to use different techniques for showing the existence of the 25-min At^{204} and 2.7-hr At^{206} isomers. The decay of At^{208} was also followed carefully.

II. EXPERIMENTAL METHODS AND RESULTS

A. Target Preparation and Bombardment

The target was a piece of fused Bi_2O_3 made by melting Bi_2O_3 powder in a platinum boat heated by high frequency induction. In position, it presented a plane 2.7 cm x 0.6 cm perpendicular to the beam and a thickness of 0.4 cm through which the beam traversed. In order to reproduce the conditions of the earlier experiments, the 330 MeV alpha-particles from the 184-inch cyclotron were slowed by a tungsten absorber so that the energy range within the target was 200-100 MeV. The irradiation time was usually two hours. These conditions were varied somewhat in different experiments but not in any essential manner.

After bombardment, the Bi_2O_3 target was wrapped in platinum foil containing a narrow slit and was heated by high-frequency induction. The Bi_2O_3 melted, and the astatine vaporized onto a cold platinum plate about 1 cm above the slit. This plate then served as a source for astatine in the experiments which followed. In some experiments, a cold glass plate was used as collector.

The astatine activities in question decay largely by electron capture and in the absence of information on distinctive γ-rays, their existences could best be shown by observing the growth of the respective polonium daughters through measuring their well-characterized alpha particles. As in the previous experiments, polonium fractions were removed periodically and subjected to alpha-energy analysis using an ionization chamber coupled to a multi-channel pulse height discriminator. The decrease in intensity of a particular alpha group in such successive "milkings" should reproduce the decay of the astatine parent.

In the early experiments, the polonium fractions were removed using

solution chemistry. Several different techniques were used in the present study and since these variations in method were instrumental in clearing up the problem, the results of each will be described separately.

B. Recoil Collecting Experiment

In the recoil milking process, a negative potential of 300 volts is applied to a collector plate held about 1/8 inch away from the source plate containing the astatine isotopes. When the astatine isotopes undergo electron-capture, the resulting charged polonium atoms are ejected onto the collector plate. The efficiency for this type of collection is low but apparently quite uniform. By changing the collector plates every 25 min, or other intervals, the growth rate of the polonium isotopes could be followed, and thereby the half-life of the astatine parent determined. Some astatine is also collected because it is ionized by the radio-activity of the source, but much of this was removed by flaming the platinum collector plates at the end of collection.

The flamed plates were placed in ionization chambers, and the characteristic alpha-groups measured.

Figures 1a-1c show the alpha-pulse analyses for three collection periods started 25 min, 125 min, and 10 hr after irradiation respectively. The Po^{210} peak comes from the decay of the well known 8.3-hr At^{210}. In Fig. 2 are shown the intensities of the Po^{210} and Po^{204} peaks for a series of 25 minute collections; the intensities of the Po peaks were corrected to the mid-points of the collection periods. A least squares fit gave a line corresponding to a half-life of 8.9 ± 0.2 min for At^{204}. Unless indicated the standard deviations are smaller than the size of the points. Errors due to varying collection efficiency or varying retention of

Po following flaming are not included in the standard deviations but are reflected by the scattering of the points.

There was no evidence for a 25-min period for At^{204}. From the upper limit of the Po^{204} intensity taken from the graph shown in Fig. 1b an upper limit for the mass of the 25-min period relative to the 9-min period could be set at 0.4% for a saturation bombardment. This result differs clearly from the early experiments.

A similar analysis for the At^{206} half-life is shown in Fig. 3. The indicated standard deviations do not include small errors for differing collection times. With this exception the points are plotted in the same manner as in Fig. 2. The best straight line fit for the intensities of the Po^{206} peaks gave a half-life of 32.8 ± 0.5 min for the At^{206} parent. The maximum amount of a 2.7-hr activity for At^{206} (corrected for bombardment to saturation) was set as 1% of the mass of the 33-min activity. The yield curve of Po^{206} showed a small tail at long time intervals, even longer than could correspond to 2.7-hr period for the parent. This is believed to be a consequence of the low collection efficiency whereby most of the Po^{206} remains on the plate and a small fraction is ionized and collected at a rate unrelated to the decay of the At^{206} parent.

The half-life for the parent of Po^{208} is displayed in Fig. 4. The points are plotted in the same manner as in Fig. 3. A value of 1.6 hr was obtained and again no evidence was found for a longer-lived isomer. In this case an upper limit for the mass of a 6.3-hr isomer relative to that of a 1.6-hr activity (corrected to the end of a saturation bombardment) was 3.2%.

Some further information was obtained by isolating a pure astaine fraction 10 hours after the end of bombardment. This was done by gently heating a source plate which had been used up to that time for collecting recoils. After all of the

astatine activities had decayed the remaining polonium alpha groups were analyzed. The most significant finding was an even smaller upper limit (1%) for the amount of a 6.3-hr isomer of At^{208} which could have been present.

With the thought that the recoil efficiency for different astatine isomers might be different, it appeared advisable to use an astatine-polonium separation process which would not distinguish between isomers.

C. Selective Vaporization

It is well known that if a platinum plate containing previously volatilized polonium and astatine is heated to temperatures up to 450°C an astatine-polonium separation can be effected. At such temperatures the astatine yield is not quantitative, but that which volatilizes is quite free of polonium. It was found that in the region of 420° - 450°C about 50% of the astatine could be volatilized and collected with less than 1% of the polonium. The half-life of the astatine parent of each polonium activity can be determined by volatilizing the astatine onto a fresh plate at intervals and measuring the residual polonium activities. The yield of astatine volalitized need not be known provided that there is present an astatine isotope of known half-life to permit normalization. Since the amount of astatine became attenuated by successive volatilizations in addition to the decay, it was the practice to prepare three astatine sources, to use one for a few transfers, and then to switch to a second. Also the analysis was made more precise by removing residual astatine by heating to a higher temperature.

The time intervals chosen for these experiments were too long to obtain information on At^{204}. For At^{208} and At^{206}, only periods of 1.63 ± 0.03 hr and 30 min respectively were found. Upper limits of ~1% could be set for the existence of longer-lived isomers.

D. Solution Chemistry

In view of the failure to see the earlier reported forms of At^{204}, At^{206} and At^{208} it became likely that the type of chemistry previously employed introduced fictitious half-lives. For this reason, experiments were conducted duplicating the early ones in the type of chemistry employed.

An astatine sample obtained by heating the Bi_2O_3 target was removed from the catcher plate with hydrochloric acid and extracted into a 2% solution of tributyl phosphate in diisopropyl ether. This was then contacted periodically with 2M nitric acid - 4M hydrochloric acid aqueous solution which removes the polonium. These polonium samples were then subjected to alpha pulse analysis. Two aqueous contacts of the organic phase separated by 8 min were made at each milking. The relative amounts of polonium in the two contacts indicated that at least 85% of the extractable polonium was removed in each contact or ~97% in each milking.

As in the early work with solution chemistry, we detected substantial amounts of Po^{206} and Po^{208} being apparently produced by longer-lived astatine parents. That these longer-lived astatine half-lives are ficticious is vividly illustrated by comparing Fig. 5 and 6. Figure 5 shows the alpha spectrum of polonium isotopes which grew into an astatine sample purified from polonium by vaporization about 22 hours after the end of bombardment. Figure 6 shows a similar spectrum in which the At was separated from the Po by solution chemistry. The results of a number of such experiments with solution chemistry was that the apparent half-life of the astatine parent is more closely connected with the interval of milking the polonium than with the true half-life. This idea is developed further in the following section.

III. DISCUSSION

The reason for the previous findings of the longer-lived isomers is very likely due to some effects in the wet chemistry procedure. The polonium in the organic solution seems to be stored partly in a form in which it can be directly extracted into aqueous solution and partly in a form in which it cannot be directly extracted. The observed results might be explained qualitatively in terms of a slow reaction rate for the transition between the two forms. If one assumes constant values for the transition reaction rates, it is possible to obtain apparent half-lives for the astatine parents equal to the periods between milkings, as found in the early studies. The forgoing explanation is considerably oversimplified in that the chemical composition of the organic solution (and hence the reaction rates) may change with time. Indeed in the present work it was found that the proportion of extractable polonium varied during the course of our experiment.

FOOTNOTES AND REFERENCES

* Work done under the auspices of the U. S. Atomic Energy Commission.

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FIGURE CAPTIONS

Fig. 1. Alpha spectra of polonium recoil daughters separated from astatine by recoil collection. a-The polonium growth started 25 min after the end of bombardment. The growth period was 24 min. The data was recorded 5 min after the end of the growth period. The counting period was 50 min.

b. The polonium growth started 2 hours 5 min after the end of bombardment. The growth period was 24 min. The data was recorded 6 min later. The counting period was 250 min.

c. The polonium growth started 10 hours 4 min after the end of bombardment. The growth period was 5 hours 46 min. The data was recorded 3 days later. The counting period was 1000 min. Zero events are not recorded in the figure.

Fig. 2. Decay curve of At^{204} determined by recoil collection and alpha pulse analysis of Po^{204}. For comparison the corresponding curve for 8.3 h At^{210} is shown.

Fig. 3. Decay curve of At^{206} determined by recoil collection and alpha pulse analysis of Po^{206}. For comparison the corresponding curve for 8.3 h At^{210} is shown. The length of the various collections is indicated by the spacing between points. The points were corrected to a 25 min collection time and plotted at the midpoint of the collection period.

FIGURE CAPTIONS (con't)

Fig. 4. Decay curve of At^{208} determined by recoil collection and alpha pulse analysis of Po^{208}. For comparison the corresponding curve for 8.3 h At^{210} is shown. The length of the various collections is indicated by the spacing between points. The points were corrected to a 25 min collection time and plotted at the midpoint of the collection period.

Fig. 5. Alpha spectrum of polonium isotopes separated from astatine by selective vaporization. The polonium growth started 22.5 hours after the end of bombardment. The growth period was 6.0 hours. The data was recorded 50 hours later. The counting period was 1280 minutes. Zero counts are indicated by a vertical arrow.

Fig. 6. Alpha spectrum of polonium isotopes separated from astatine by solvent extraction. The polonium growth started 20.6 hours after the end of bombardment. The growth period was 3.03 days. The data was recorded 2 days later. The counting period was 970 min. Zero counts are indicated by a vertical arrow.

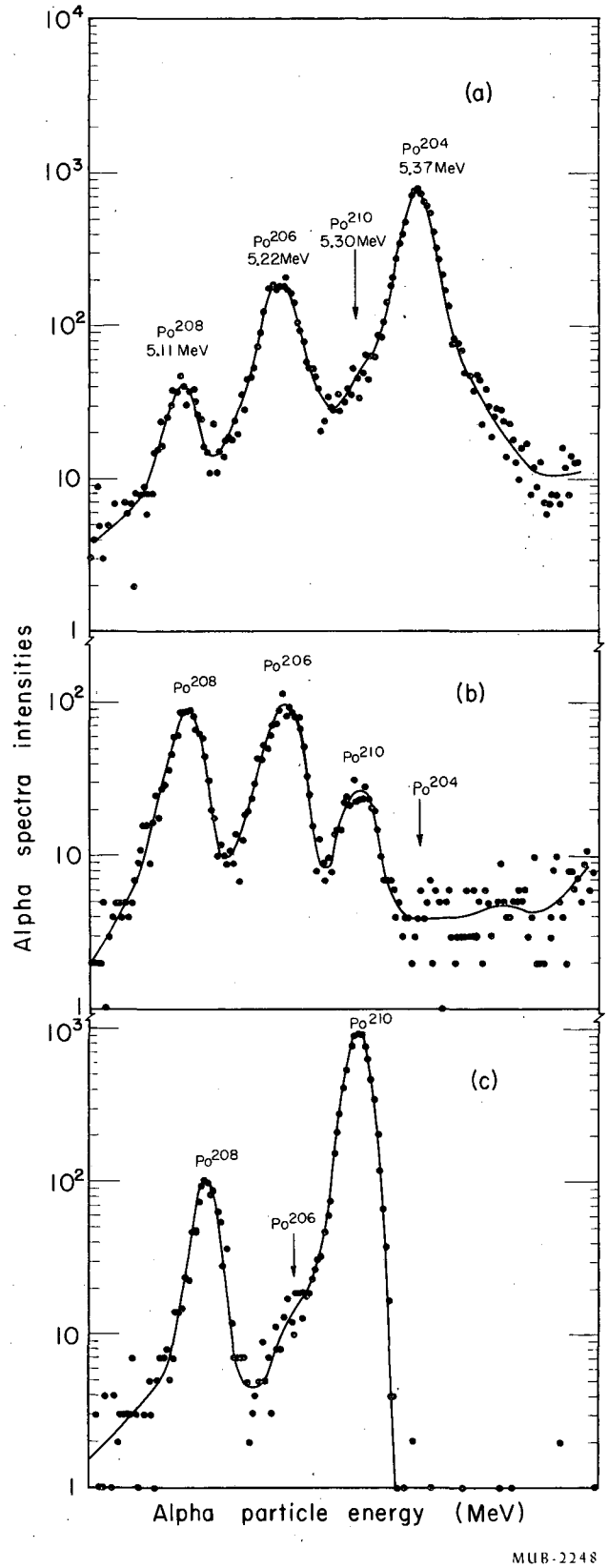
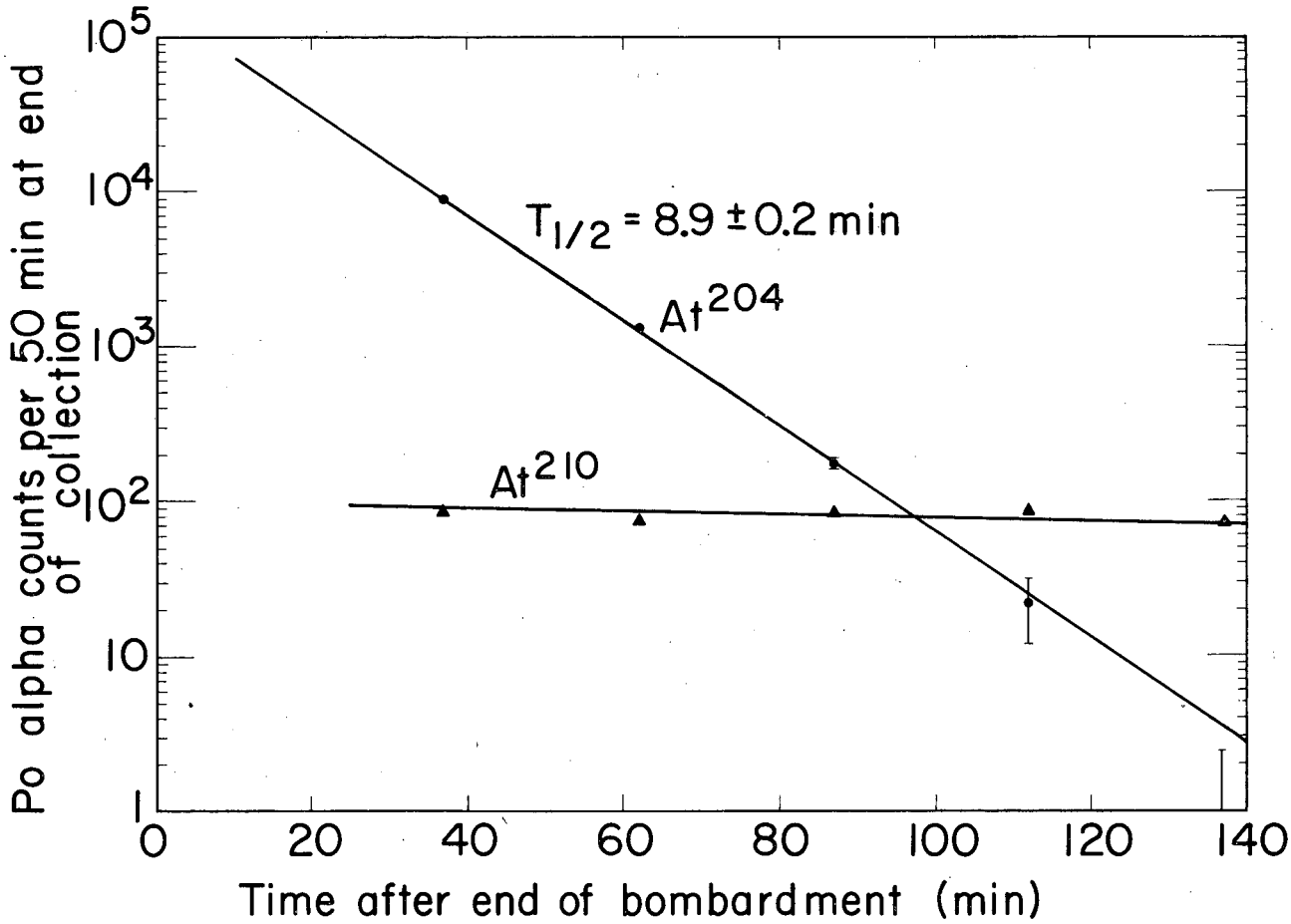
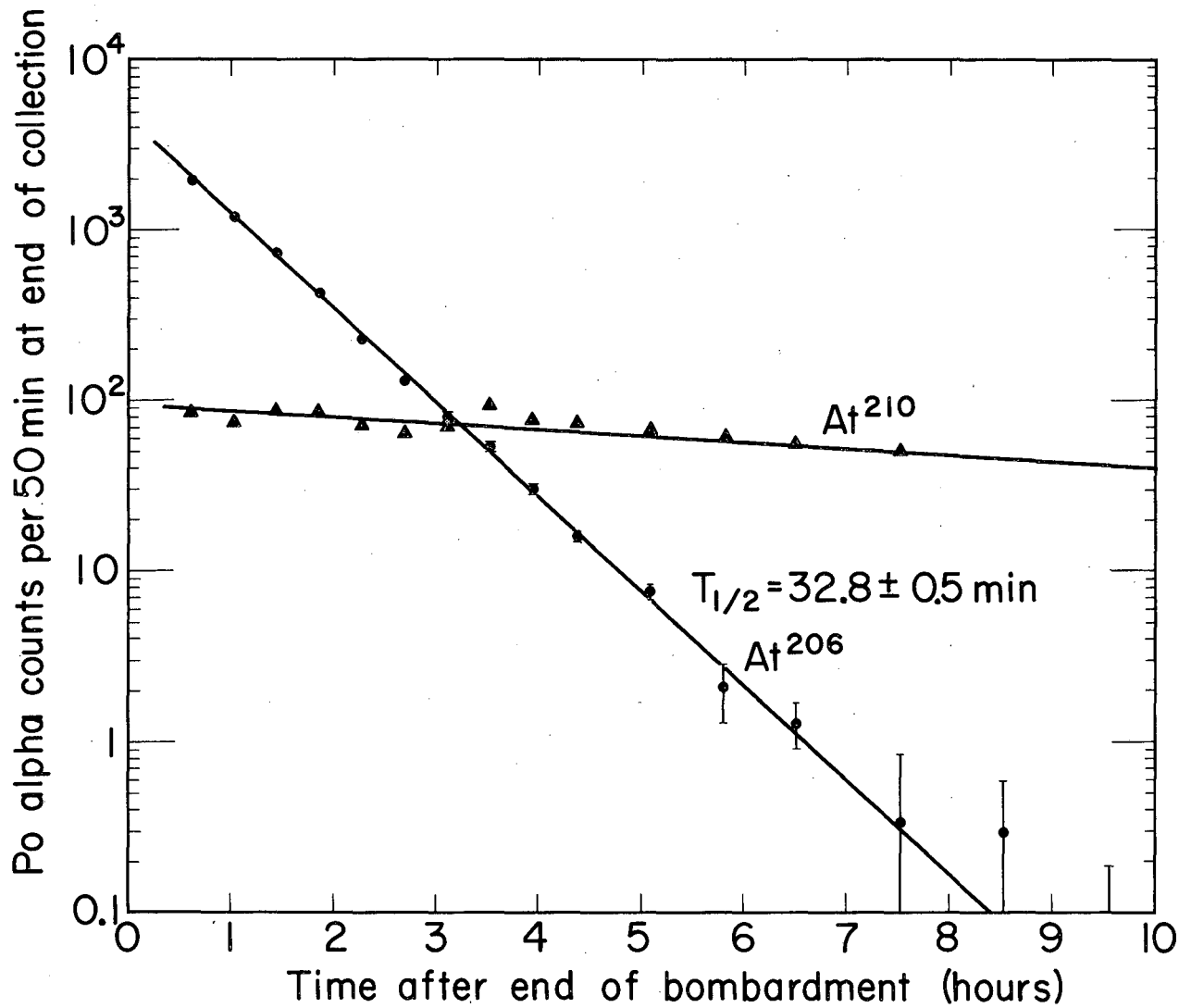

(a)
Po204
5.37MeV
Po210
5.30MeV
Po206
5.22MeV
Po208
5.11 MeV
(b)
Po208
Po206
Po210
Po204
(c)
Po210
Po208
Po206
Alpha spectra intensities
Alpha particle energy (MeV)
MUB-2248

Fig. 1



MUB-2249

Fig. 2



MUB-2250

Fig. 3.

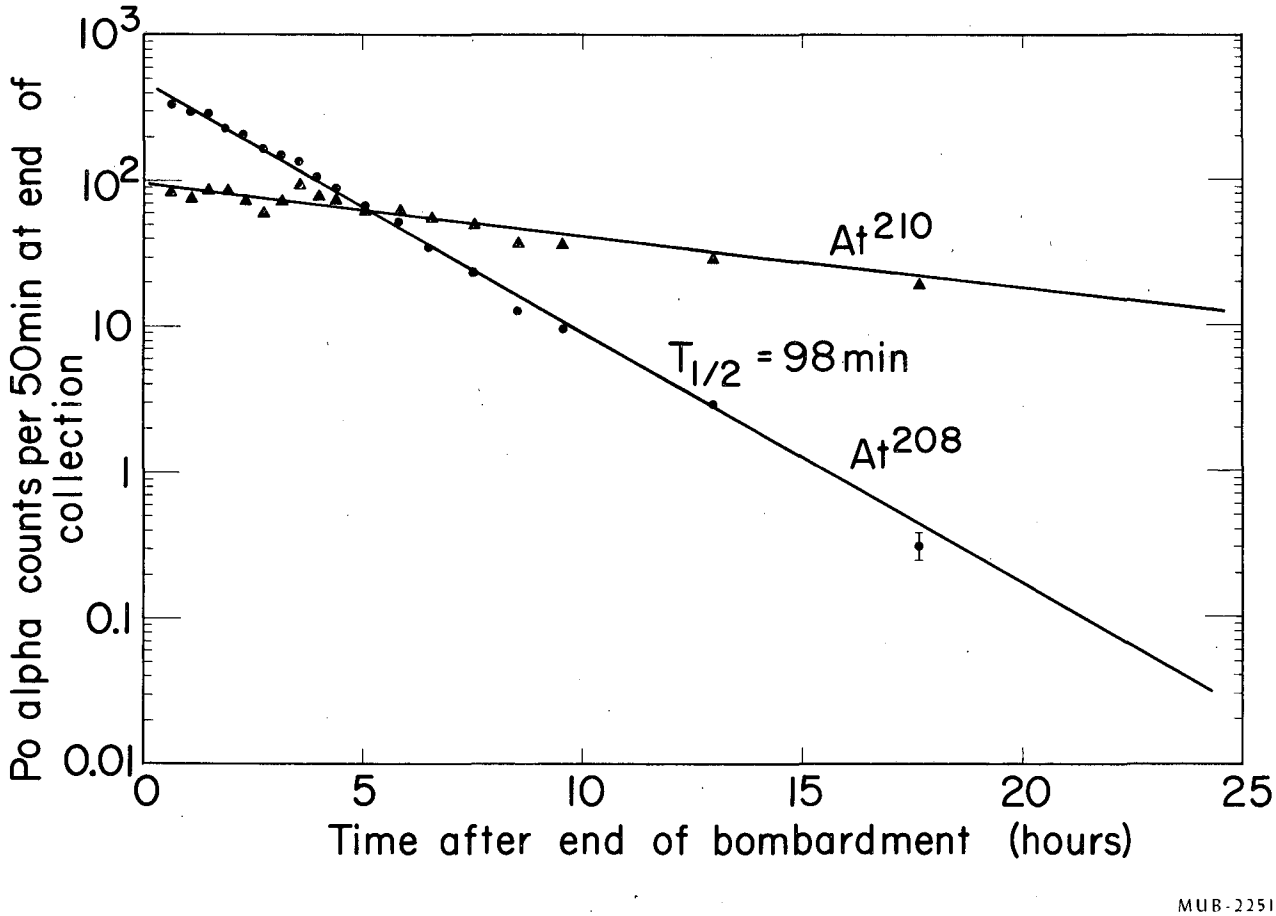

Po alpha counts per 50min at end of collection
10^3
10^2
10
1
0.1
0.01
0
5
10
15
20
25
Time after end of bombardment (hours)
At^210
T1/2 = 98 min
At^208
MUB-2251

Fig. 4.

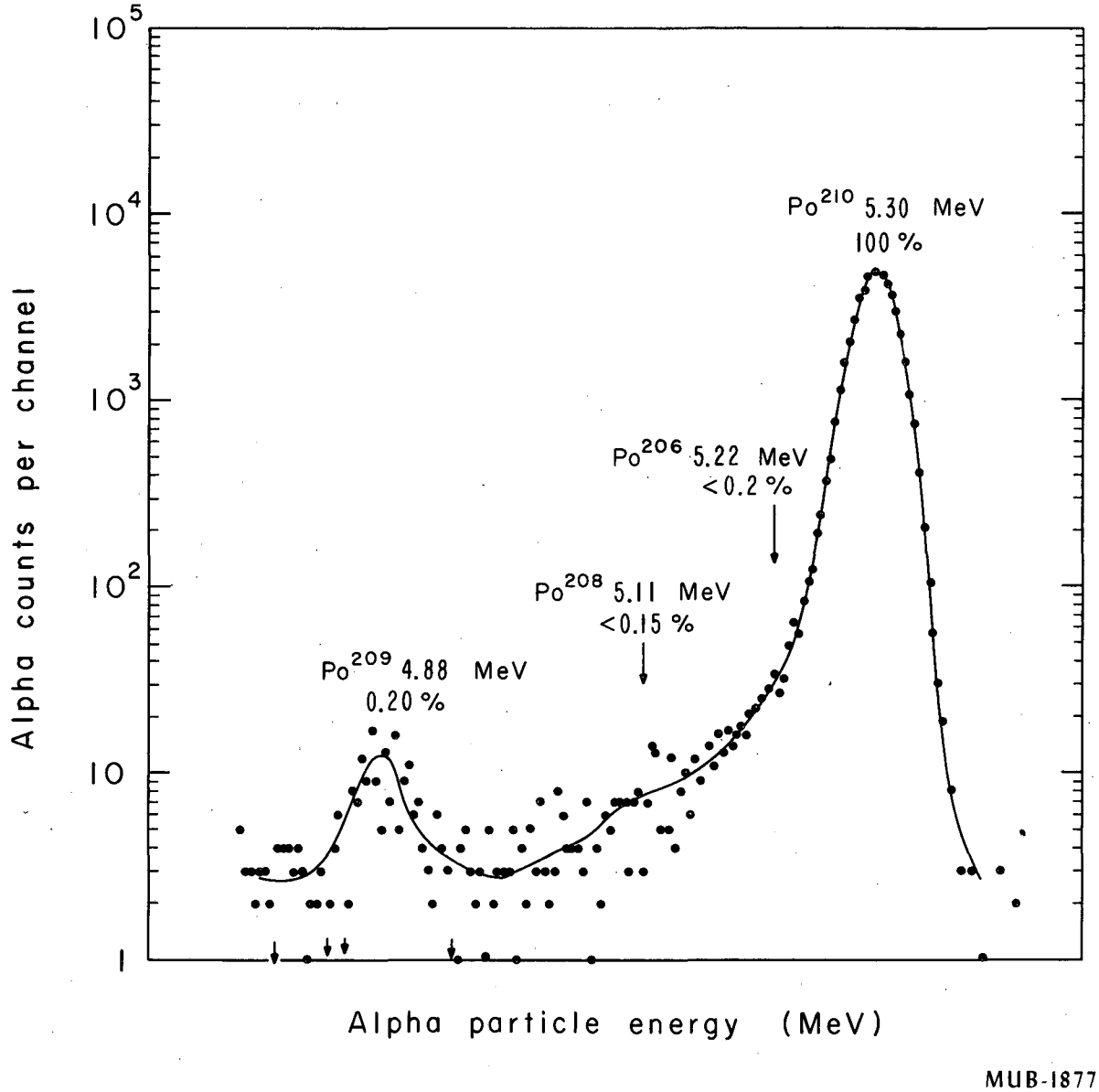

10^5
10^4
10^3
10^2
10
1
Alpha counts per channel
Po^{210} 5.30 MeV
100 %
Po^{206} 5.22 MeV
<0.2 %
Po^{208} 5.11 MeV
<0.15 %
Po^{209} 4.88 MeV
0.20 %
Alpha particle energy (MeV)
MUB-1877

Fig. 5.

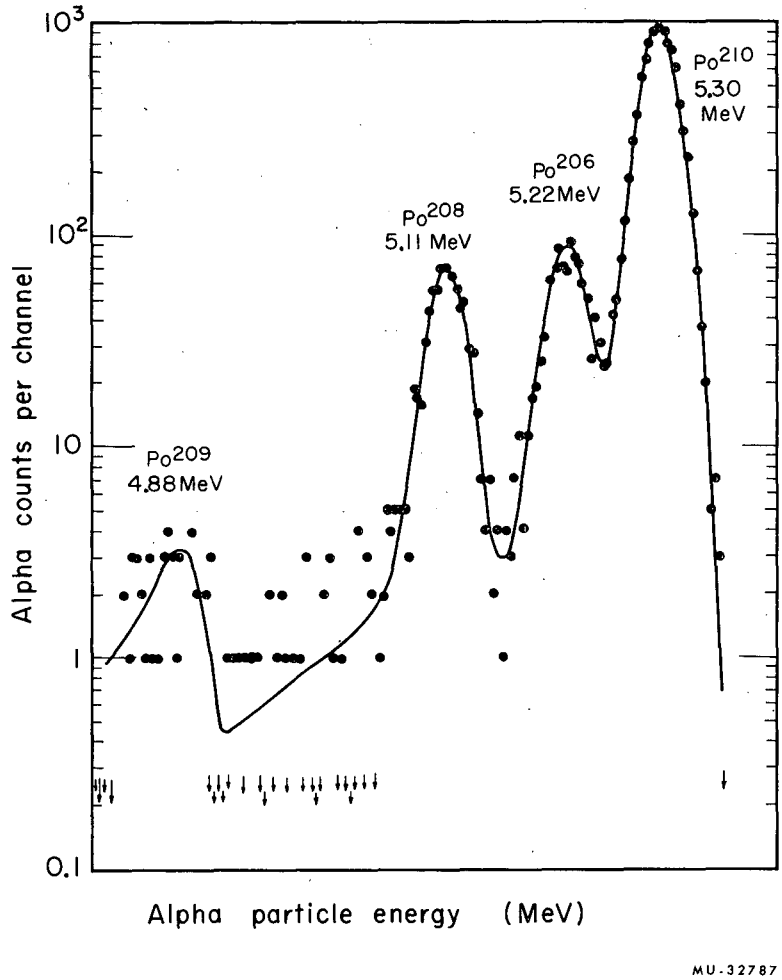

10^3
10^2
10
1
0.1
Alpha counts per channel
Alpha particle energy (MeV)
Po^{209}
4.88 MeV
Po^{208}
5.11 MeV
Po^{206}
5.22 MeV
Po^{210}
5.30
MeV
MU-32787

Fig. 6.

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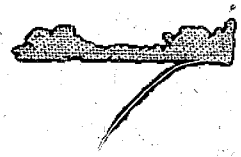
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