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Ernest O. Lawrence Radiation Laboratory

NUCLEAR SPECTROSCOPIC STUDIES OF POLONIUM-204

Berkeley, California

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Jiann-min An

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(M. S. Thesis)

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Contents

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ABSTRACT

Sources of Polonium-204 were derived from the electron capture decay of Astatine-204 made by bombarding Au^{197} with C^{12} in the Heavy Ion Linear Accelerator at Berkeley. Physical and chemical methods were adjusted to make the sources as pure as possible for the measurements.

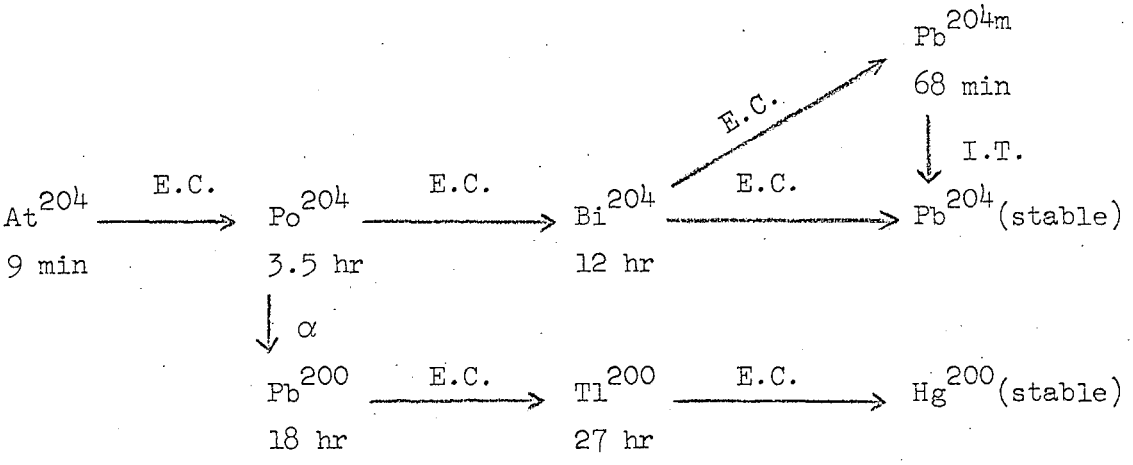
Using scintillation crystals coupled to a 400 channel pulse-height analyzer, an investigation of some of the nuclear radiations of Po^{204} has been made. Gamma-rays from Po^{204} were identified by their half-life; their energies and relative intensities were carefully determined and the alpha branching ratio was measured.

I. INTRODUCTION

In the course of research on the radioactivity of various uranium and thorium minerals, Pierre and Marie Curie discovered polonium in 1898. This was the first new element to be discovered as a result of its radio-activity. They found that polonium behaved similarly to bismuth to form an insoluble sulphide compound and subsequently were able to separate them through different volatility by vacuum sublimation. In the early days, the work on polonium was complicated by the extremely small quantities available and sometimes by incomplete separation of radio-active impurities, so that it was not until 1906 that the identification of polonium was definitely established. However, the chemistry of polonium has been placed on a sound basis only within the last fifteen years.

Polonium-210 is a natural product in the uranium-radium decay chain. Trace amounts of polonium were usually obtainable from aged radon tubes and from aged radium salts. Now, with the intense neutron fluxes in nuclear reactors, polonium can be economically prepared by the thermal neutron capture of bismuth, $Bi^{209}(n,\gamma)\ Bi^{210} \xrightarrow{\beta} Po^{210}$. This process is currently used for the production of milligram quantities of polonium. Templeton, Howland and Perlman[1] have produced Po^{206}, Po^{207} and Po^{208} by bombarding lead or bismuth in a cyclotron with 20-MeV deuterons or 40 MeV helium ions in 1947. Kelly and Segrè[2] have found Po^{209}. By using higher energy beams in the 184-inch cyclotron at Berkeley, Karraker and Templeton[3] have produced the lighter isotopes of Po^{205}, Po^{204}, Po^{203} and Po^{202}. Through improved measurements Karraker, Ghiorso and Templeton[4] have reported Po^{200} and Po^{201}. Still lighter polonium isotopes have also been studied. The isotopes with mass number below 200 had been studied by Swedish scientists of the Nobel Institute with heavy-ion bombardments and in Uppsala with proton bombardments of bismuth.[5]

Polonium-204 has been observed and reported by different workers.[3,6] The decay relations of Po^{204} are as follows:



In the present investigation, Po^{204} was produced by bombarding Au^{197} with C^{12} in the Heavy Ion Linear Accelerator (Hilac). The polonium samples used in this measurement were chemically purified from bismuth and lead by the method developed by Kimura[7] and co-workers. The gamma rays from the polonium samples were measured with the recently developed germanium crystal detectors. It was the prime objective of the present study to provide information for the further study of the decay scheme of Po^{204}.

II. EXPERIMENTAL METHOD

A. Preparation of Targets

The target consisted of a gold foil in front of which there was interposed an aluminum foil as absorber. By using different thickness of absorber[11] in the beam of particles from the accelerator, we could reduce the energy of the particles to some desired value before it reached the gold foil. From known[11] range-energy relationships we could choose the proper thickness of the gold foil and hence to control the energy band to which the target atoms were exposed. Hoff, Asaro and Perlman[12] have given the relative yields for astatine isotopes produced in carbon ion bombardments of gold. With those data together with the excitation functions for (C^{12}, xn) reactions on Au^{197}, [13] the desired sources could be prepared without difficulty.

B. Bombardment Characteristics

Using the branching ratios and half-lives of different astatine isotopes, Thomas, Gordon, Latimer and Seaborg[13] have calculated the cross sections and given the excitation functions for (C^{12}, xn) reactions on Au^{197} which are shown in Fig. 1. In the present work, our interest is in the 5n reaction. However, the energy range which would give the maximum 5n reaction is not suitable for this bombardment, because a substantial amount of At^{205} and its undesirable daughter Po^{205} would be produced. From the literature we know that Po^{205} has a half-life of 1.5 hours; Po^{204} 3.5 hours; Po^{203} 42 min; Po^{202} 43 min. Therefore we chose the beam energy of 90 MeV to 100 MeV in order to avoid the 4n reaction. The Po^{203} and Po^{202} produced in this bombardment would decay much faster than the Po^{204} and hence would not disturb the experiment. This energy range did give very satisfactory results. We did not observe any radiation from the contaminant Po^{205}.

Bombardments were carried out with Berkeley Heavy Ion Linear Accelerator (Hilac) which accelerates heavy ions to 10.4 MeV per nucleon. The carbon ion beam intensities used were in the range of 2 to 3 microamperes. The entire assembly served as a Faraday cup for monitoring the beams. It was cooled to avoid melting the target. The bombarding times ranged from 20 to 60 min.

C. Methods of Separation and Chemical Purification

1. Vaporization Method

Following the bombardment, the target holder was removed from the Hilac and disassembled under a hood. The gold foil was removed and transferred into a glove box and placed inside a small quartz cup which was heated with an induction heating coil. The gold foil was melted and the volatile species, astatine, polonium and others were collected on platinum plate cooled with dry ice. This platinum plate was removed from the glove box and placed on a hot plate which was preheated to a temperature of about 380°-390° C. A bottomless aluminum cup was placed over the sample and part of the activity from the first platinum plate was vaporized onto a second platinum plate which was in turn cooled by dry ice. At this temperature only astatine was vaporized and left polonium and bismuth on the first platinum plate. No polonium or bismuth activities were observed initially on the second platinum plate following the second vaporization. Twenty minutes after the second vaporization the second platinum plate was flamed slightly or heated on the hot plate at a temperature not higher than 400° C. By this means we got rid of the astatine activity and left its polonium daughters on the platinum plate. Twenty minutes after the second vaporization most of the At^{204} has decayed away, but At^{205} with a half-life of 25 min would still remain in substantial amount. This is a way to hold down the contaminant polonium-205. This platinum plate was kept waiting for 5 hours for all the polonium-203, polonium-202 and most of the polonium-205 to decay away. Then a third vaporization was made at a temperature of 440° C. At this temperature most of the polonium would be vaporized away and bismuth and lead would stay. The polonium activity was collected onto

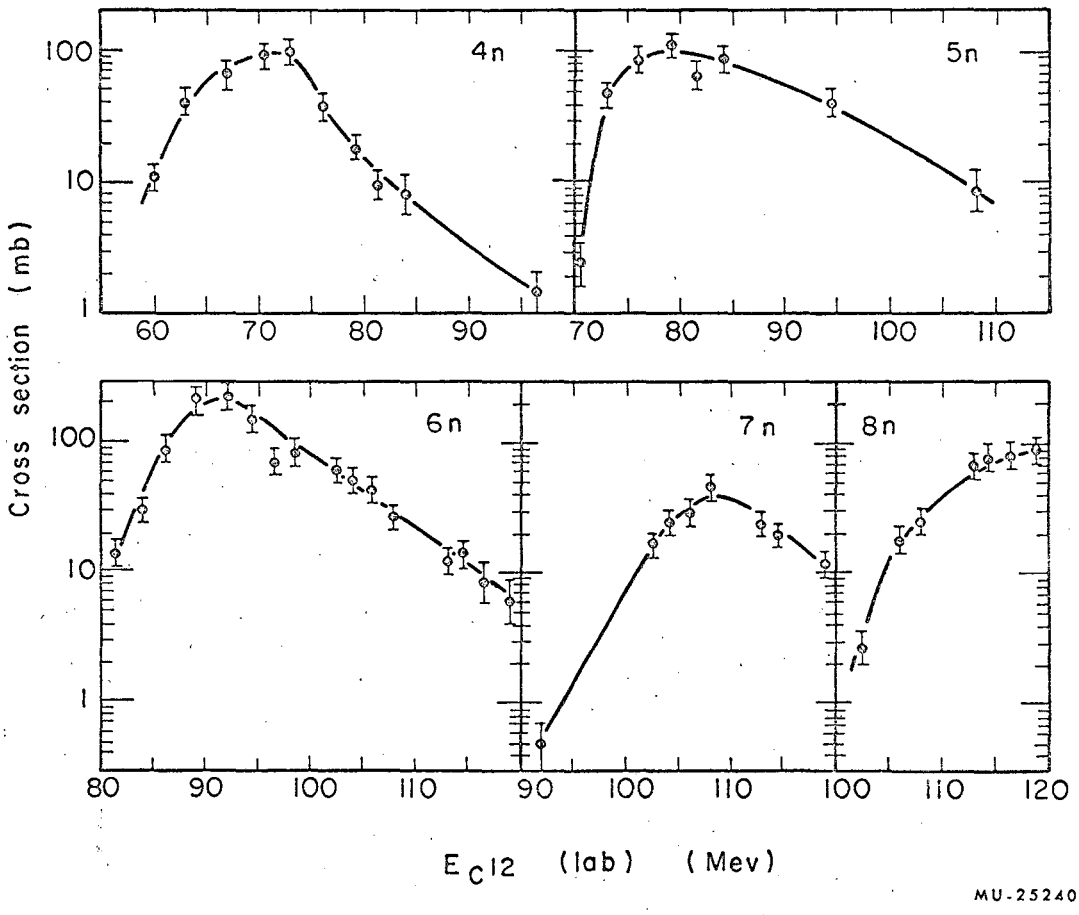


Fig. 1. Excitation functions for C^{12},xn reactions on Au^{197}.

a third platinum plate. The activity on this plate served as the source for alpha-spectra and gamma-spectra measurements. Many attempts were made by this stepwise vaporization method. It was found that this method worked well for astatine but not for polonium. The reason for this may be due to the fact that at 440° C the vaporization of polonium was quite incomplete and at higher temperatures bismuth would go with it. In all the attempts, the activities collected were too low and sometimes contaminated badly with bismuth activity.

2. Wet Chemistry Method

Since polonium forms a number of complex halides in solution, it was expected that some of these compounds could be extracted into organic solvents. Kimura and co-workers[7] found that polonium could not be extracted from HCl or HBr solutions with diethyl ether or isopropyl ether. However, polonium was extracted with isopropyl ether or methyl isobutyl ketone from HCl solutions containing KI. In the present work, the chemical procedures were as follows.

Following the bombardment (60 min), the target was removed from the Hilac and disassembled under a hood. The target gold foil was not melted until 5 hours after the bombardment, because most of the shorter lived polonium isotopes would decay away within the 5 hour period. Then the gold foil was melted as before in a glove box, and the activities were collected on platinum plate, mainly polonium, bismuth and lead. The platinum plate was removed from the glove box and washed with 1 cc of solution 0.25 M in KI and 3 N in HCl, 4 ml of isopropyl ether was then added to this solution and polonium was extracted into the organic phase with gentle swirling, while lead and bismuth remained in the aqueous phase. Polonium was scrubbed from the organic phase by shaking with 1 ml of 3 N HCl solution. The resulted HCl solution was free from bismuth and lead and contained polonium alone. This HCl solution was evaporated to dryness on a platinum plate and served as the source for the various measurements. The separation of polonium from bismuth and lead was very good. We

can see this by comparing the gamma ray spectrum of the polonium and bismuth fractions shown in Figs. 4 and 6. A yield of approximately 80 percent was obtained and time required for this separation was about 15 minutes.

III. MEASUREMENTS

A. Alpha-particle Spectra

The polonium fraction was measured in an alpha-particle ionization chamber. This ionization chamber utilized a grid to shield the collecting electrode from positive ions. A gas mixture of 93% argon and 7% methane was used. Through a preamplifier, discriminator, and amplifiers, the pulse was sent into a 400 channel pulse-height analyzer. Alpha particles have a counting efficiency of 42 percent when counted in this grid chamber. Alpha-particle spectra were obtained as shown in Fig. 2. Polonium-208 and polonium-210 standards were used for energy calibration. Two alpha-particle peaks, one with energy 5.375 MeV and the other with 5.581 MeV were observed in this measurement. The decay was followed for 24 hours. It was found that alpha-particles with energy of 5.375 MeV decayed with a half-life of 3.5 hours and the alpha-particles with energy of 5.581 MeV decayed with a half-life of 43 min. The decay curves are shown in Fig. 3. From the previously reported data (see Table I) the 5.375 MeV peak belongs to Po^{204} and the 5.581 MeV peak belongs to Po^{202}.

Karraker and Templeton observed Po^{204} in 1950. These early experiments were designed to establish the decay products and mass assignment. They separated its daughter activity at a sequence of equal time intervals, the intervals corresponding approximately to the half-life of the parents. A similar experiment was also done to identify the alpha decay daughter and fix the mass number. Their data together with other information are listed in Table I.

The assignment of Po^{202} was based on all the previous information given. The alpha branching ratio of Po^{202} has been reported by Stoner and Hyde[8] to be 2% from their experiments on the alpha-decay of Em^{206}. The alpha branching of Po^{203} has not been reported yet, but it would appear to be much smaller than Po^{202}. Therefore we would assign the 5.581 MeV to be Po^{202} in spite of the similar half-life of Po^{203} and Po^{202}.

Table I. Alpha spectra data.

Isotope	Energy (MeV)	Half-life	Reference number
Po^{204}	5.37	3.8 hr	(3)
	5.35 ± 0.05	3.5 ± 0.6 hr	(6)
	5.39	3.6 ± 0.2 hr	(10)
	5.375	3.5 ± 0.01 hr	This work
Po^{202}	5.580 ± 0.010	44 ± 7 min	(12)
	5.57	50 ± 10 min	(10)
	5.57 ± 0.05	51 ± 3 min	(8)
	5.61 ± 0.05	56 ± 5 min	(9)
	5.581	43 ± 4 min	This work

B. Gamma-spectra

Information on the gamma-rays data of Po^{204} has not yet been reported. The recently developed germanium crystal detector does provide a powerful tool in gamma-ray analysis. A germanium crystal detector with 3 cm by 2 cm by 0.9 cm dimension was used for the present measurements. A relative efficiency curve of the detector had been carefully made. A Na^{22} standard with known disintegration rate was used in determining the absolute efficiency. All the gamma-rays observed and their half-lives and possible assignments are listed in Table II. The gamma-spectrum is shown in Fig. 4.

1. Half-life Determination and Isotope Assignment

The decay of all the gamma-rays has been followed. It was found that there are eleven gamma rays in the polonium fraction which decayed with approximately the half-life of Po^{204} and are therefore assigned to this isotope. They are listed in Table II, and their decay curves are shown in Fig. 5. It should be noted that these gamma rays were not observed in any substantial intensity in the bismuth and lead fraction (Figs. 4 and 6). This latter fraction was obtained directly by vaporization from the melted gold target foil and hence contained other radioactive elements beside bismuth and lead.

In Fig. 4, (the gamma spectra of the polonium fraction) only two intense gamma peaks of Bi^{204} have been labeled (375 keV and 899 keV). No attempt has been made to label all Bi^{204} gamma rays which grow into the polonium fraction and decay with a half-life of about 12 hours.

Gamma-rays of energy 421 keV and 961 keV seem to be due to Bi^{202} which is made by the electron capture decay of Po^{202}. From the decay curve in Fig. 7, we see that these gamma rays grow into the polonium fraction and then decay with a half-life of about 98 minutes as would be expected. Gamma-rays of these energies had been previously reported by Herrlander and Bergström[15] who assigned them to Bi^{202}.

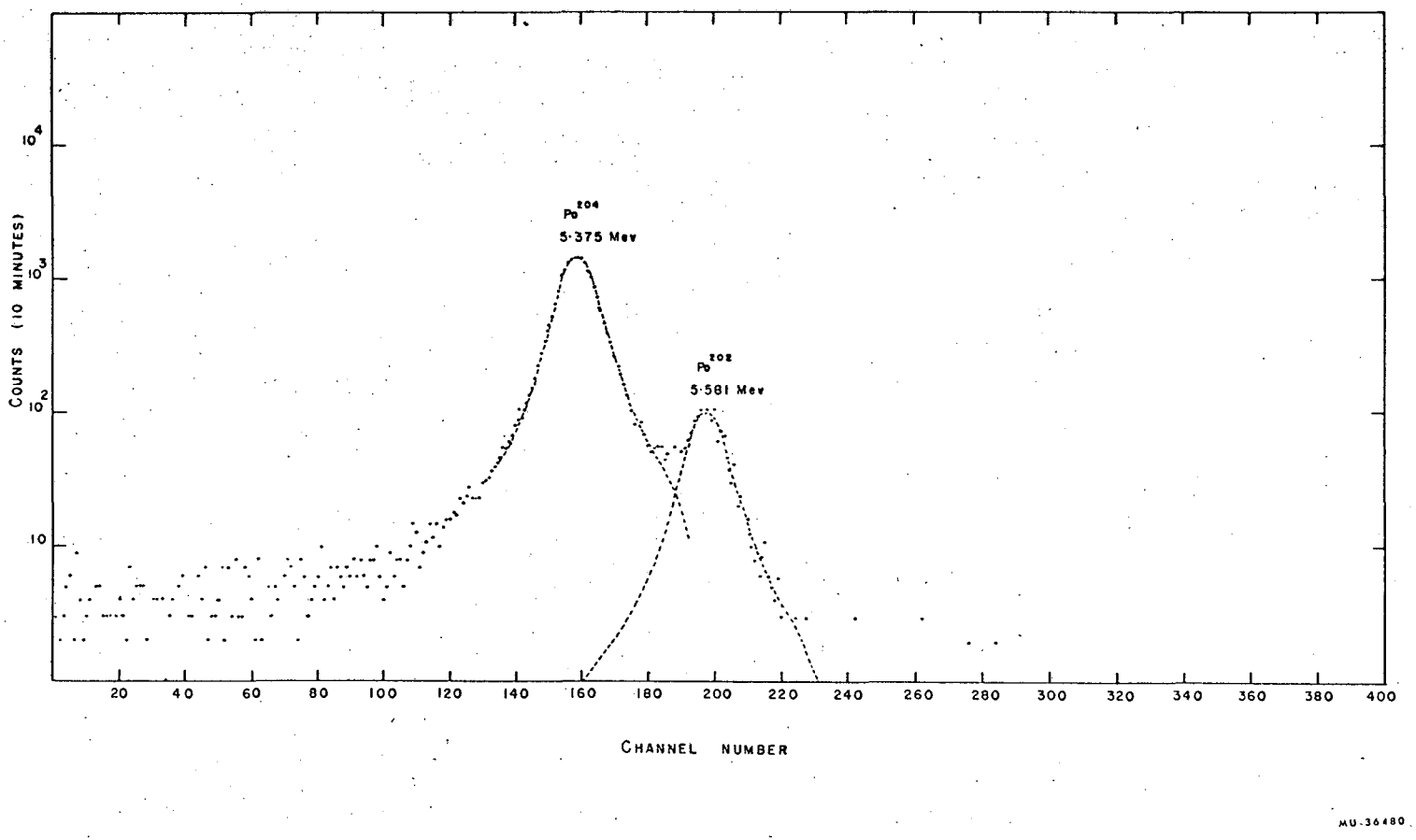


Fig. 2. Alpha-spectrum of chemically purified Po sample.

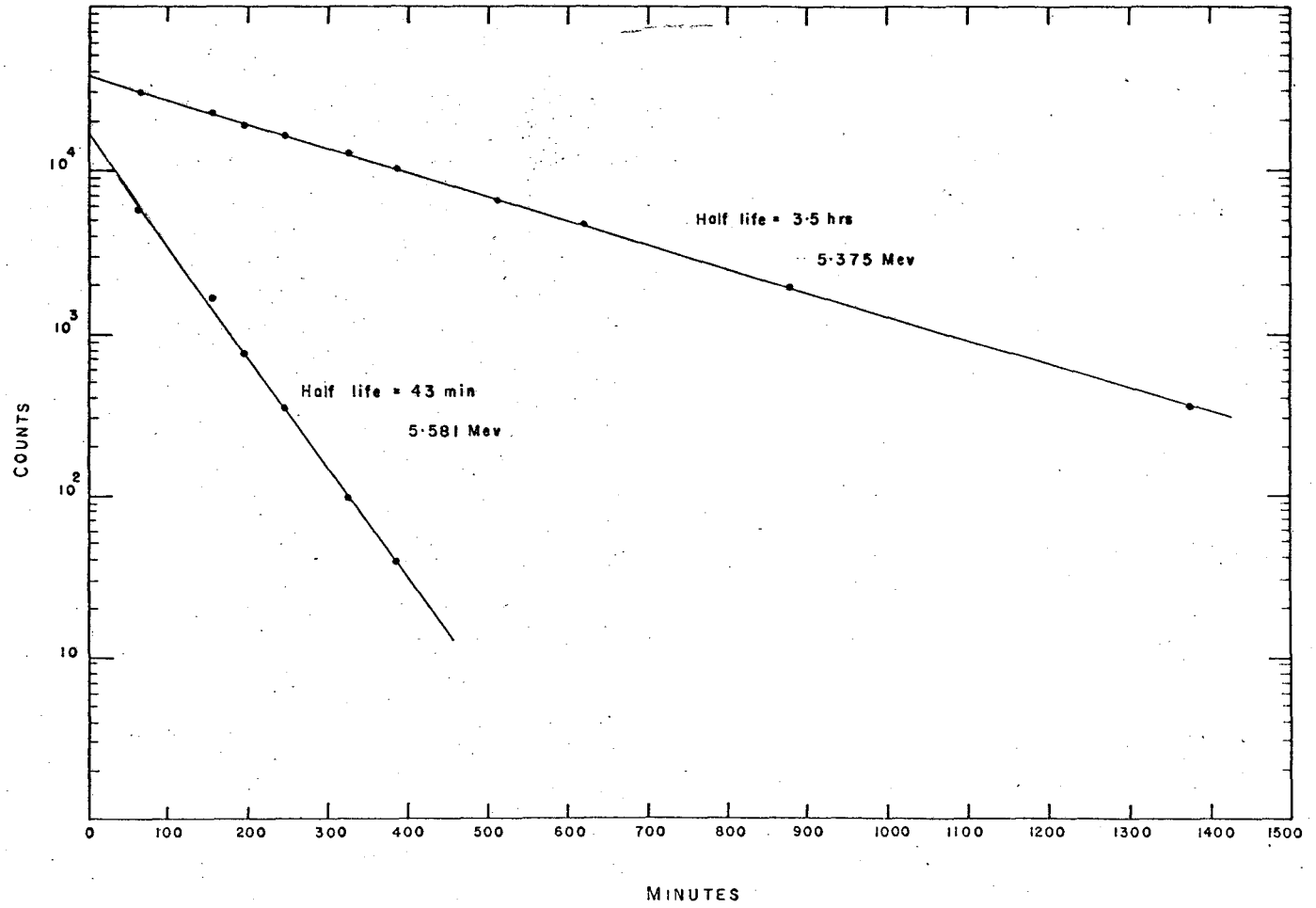


Fig. 3. Decay curves of alpha groups observed in the chemically purified Po sample.

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Two gamma rays of shorter half-life were observed. The gamma ray decay curves are shown in Fig. 8. The 707 keV gamma ray decayed with approximately 44 min half-life and the 1087 keV gamma ray decayed with approximately a 45 min half-life. These gamma rays might be due to Po^{203} or Po^{202}. This remains a question for further investigation.

2. Relative Intensities

In measuring the intensities of these gamma rays, a chemically purified polonium fraction sample was used. The gamma spectrum from this sample (as shown in Fig. 4) was observed with a germanium crystal detector connected to a 400 channel pulse height analyzer. The various peaks were integrated and the necessary corrections for counting efficiencies were made. The results are shown in Table III. Comparisons were made among those gamma rays which appear to be from the same isotope. The eleven gamma rays of Po^{204} were compared relative to the 268 keV gamma ray which was taken as one hundred.

C. Alpha-particle Branching Ratio

For the branching ratio measurement, the sample used was the chemically purified polonium fraction previously discussed. The measurement started about 50 minutes after the chemical separation.

A gamma spectrum is shown in Fig. 4. The 899 keV gamma ray of Bi^{204} was previously known to have an intensity of 100 percent. The 899 keV peak decayed with a half-life 11.9 ± 1.5 hours after most of the Po^{204} has gone (about 12 hours after the chemical separation). Since Bi^{204} comes from the electron capture decay of Po^{204} if we can find out the initial alpha intensity of Po^{204} (i.e. alpha intensity of Po^{204} at the time of chemical separation), and the initial Po^{204} electron capture intensity which would lead to the observed intensity of the 899 keV gamma ray at time t, we can calculate the branching ratio. Then we have:

Table II. Gamma rays observed in this work.

Possible assignment	Energy(keV)	Half-life measured
Po^{204}	140	205 ± 6 min
	202	210 ± 5 min
	268	208 ± 4 min
	305	210 ± 6 min
	316	218 ± 14 min
	537	215 ± 4 min
	680	212 ± 11 min
	764	210 ± 2 min
	883	215 ± 3 min
	1022	215 ± 4 min
	1037	220 ± 15 min
Bi^{202}	421	99 ± 4 min
	961	97 ± 5 min
Po^{203} or Po^{202} (?)	707	44 min
	1087	45 min

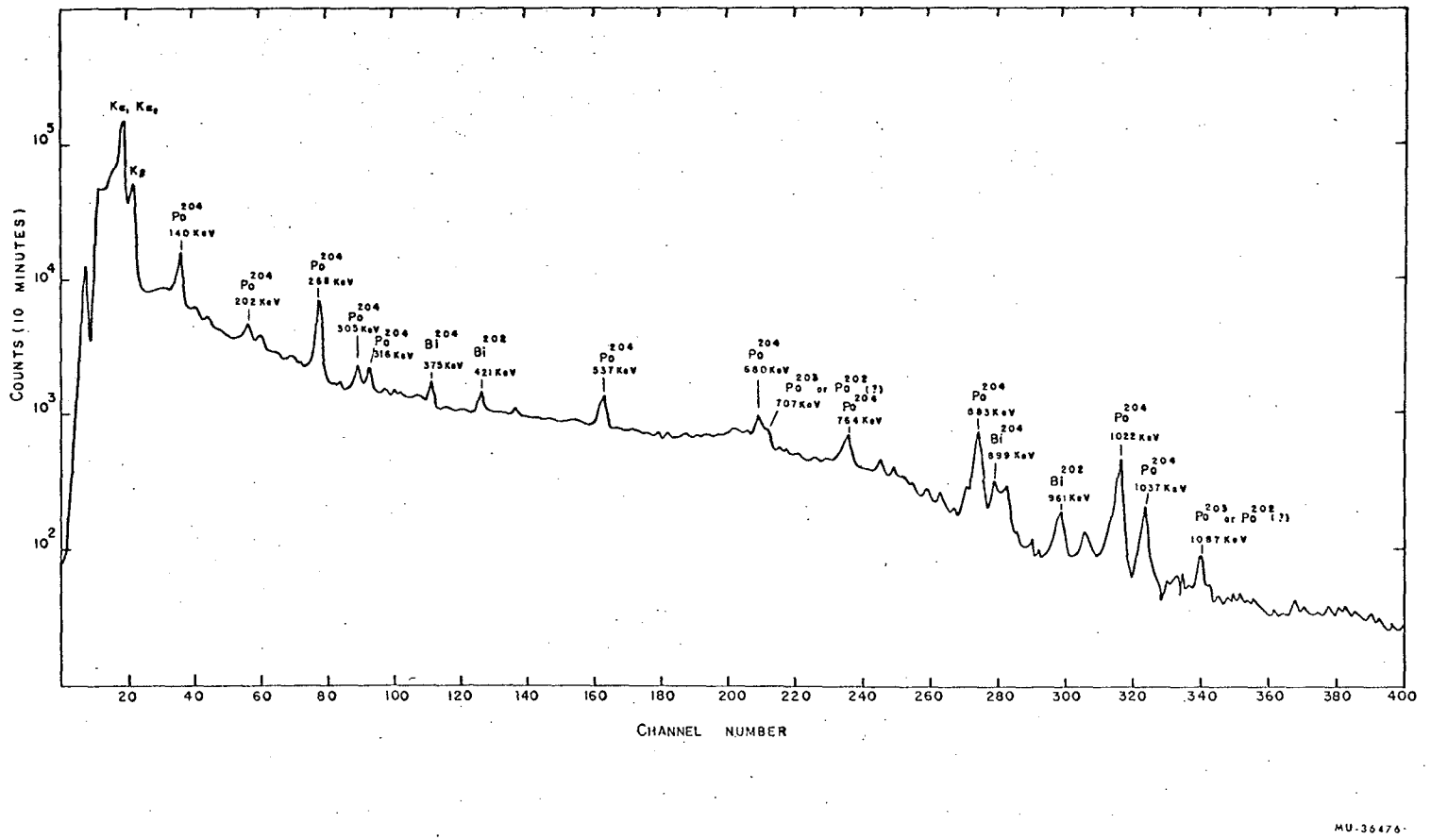

Fig. 4. Gamma spectrum of the chemically purified Po sample.

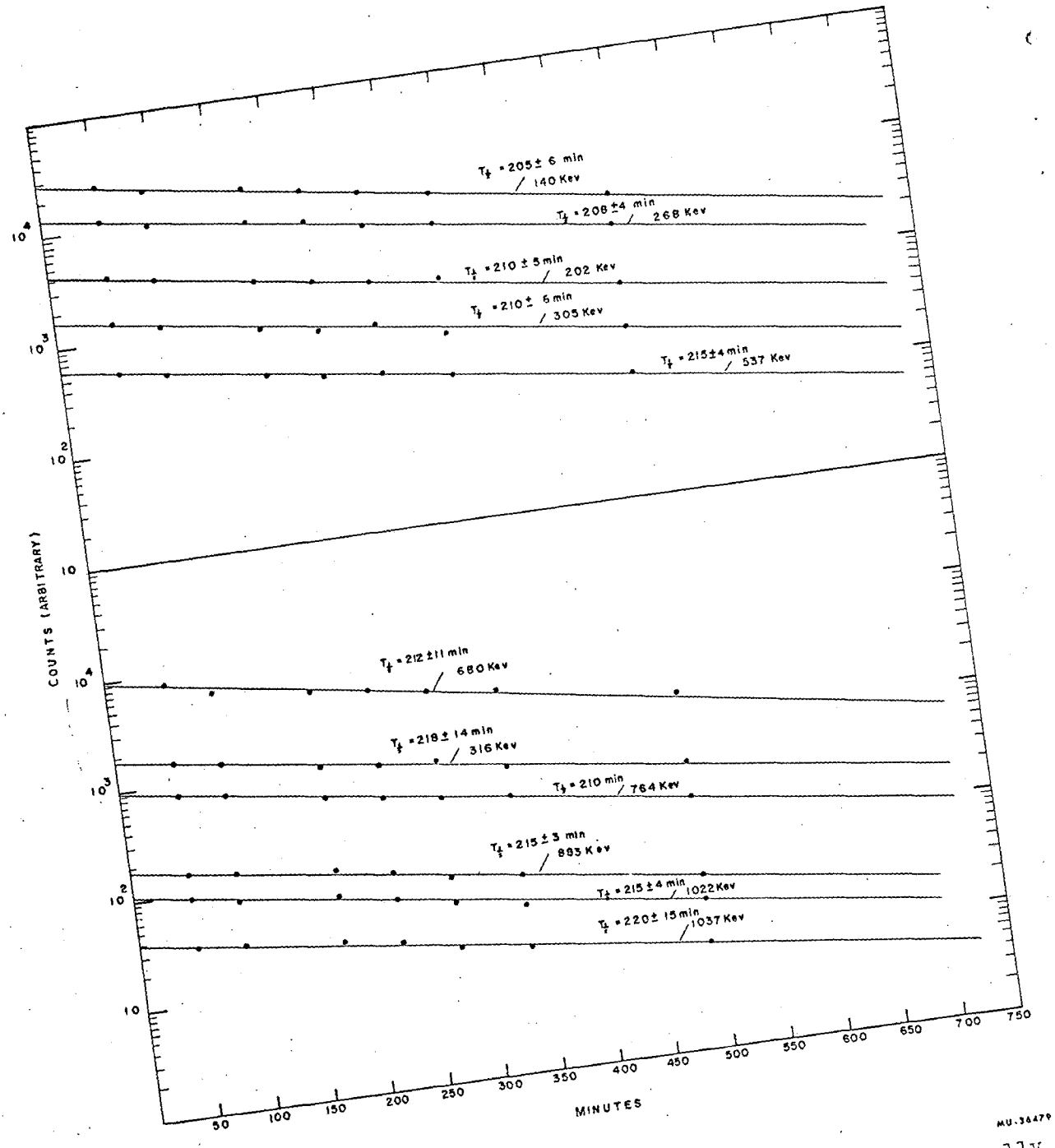

Fig. 5. Decay curves of the gamma rays of Po^{204} from the chemically purified Po sample.

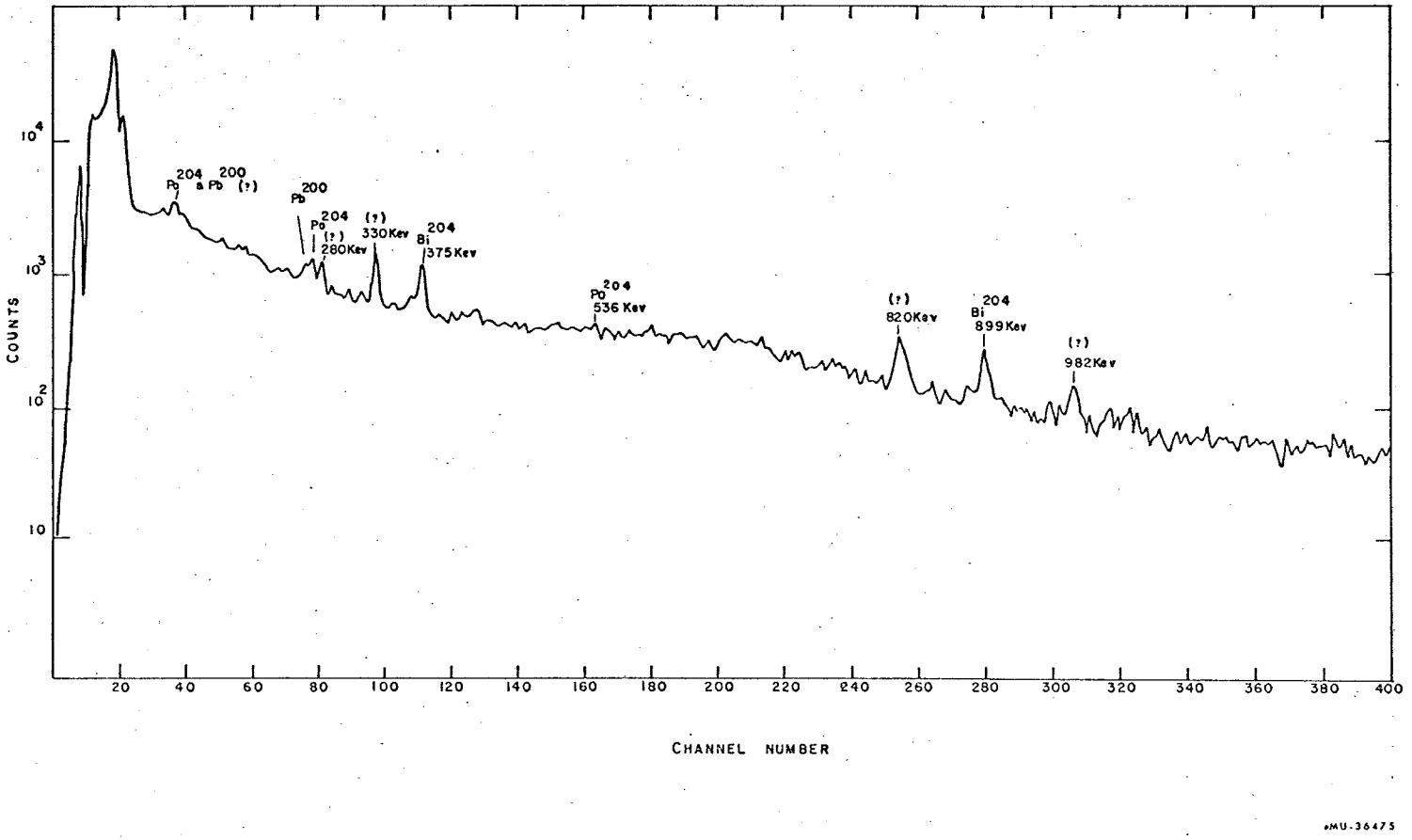

Fig. 6. Gamma spectrum of Bi and Pb fraction.

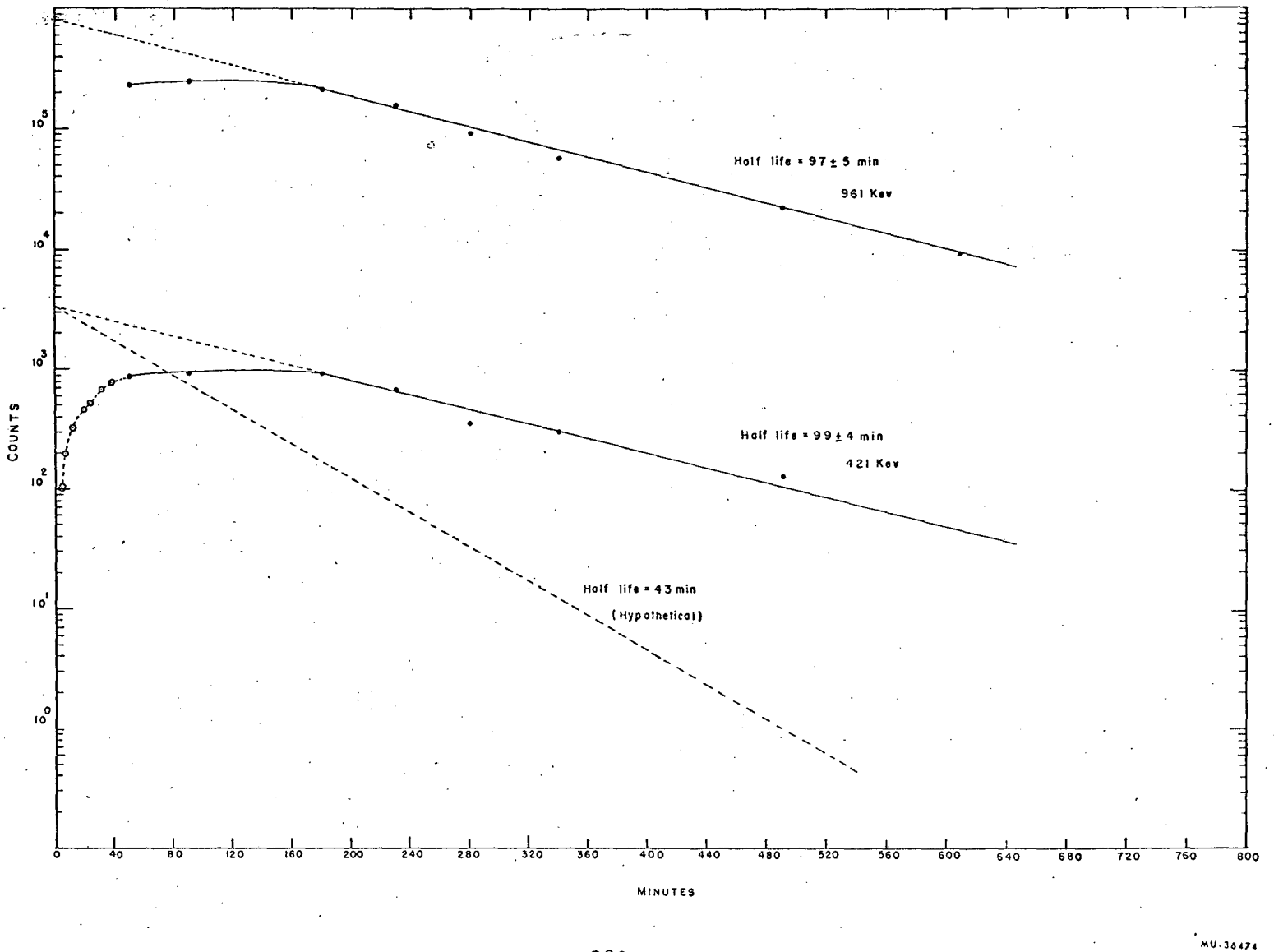

Fig. 7. Decay curves of gamma rays of Bi^{202} in the chemically purified Po sample. (The dashed lines and circles are hypothetical and based on a 43 min half life parent and the growing of a 99 min daughter.

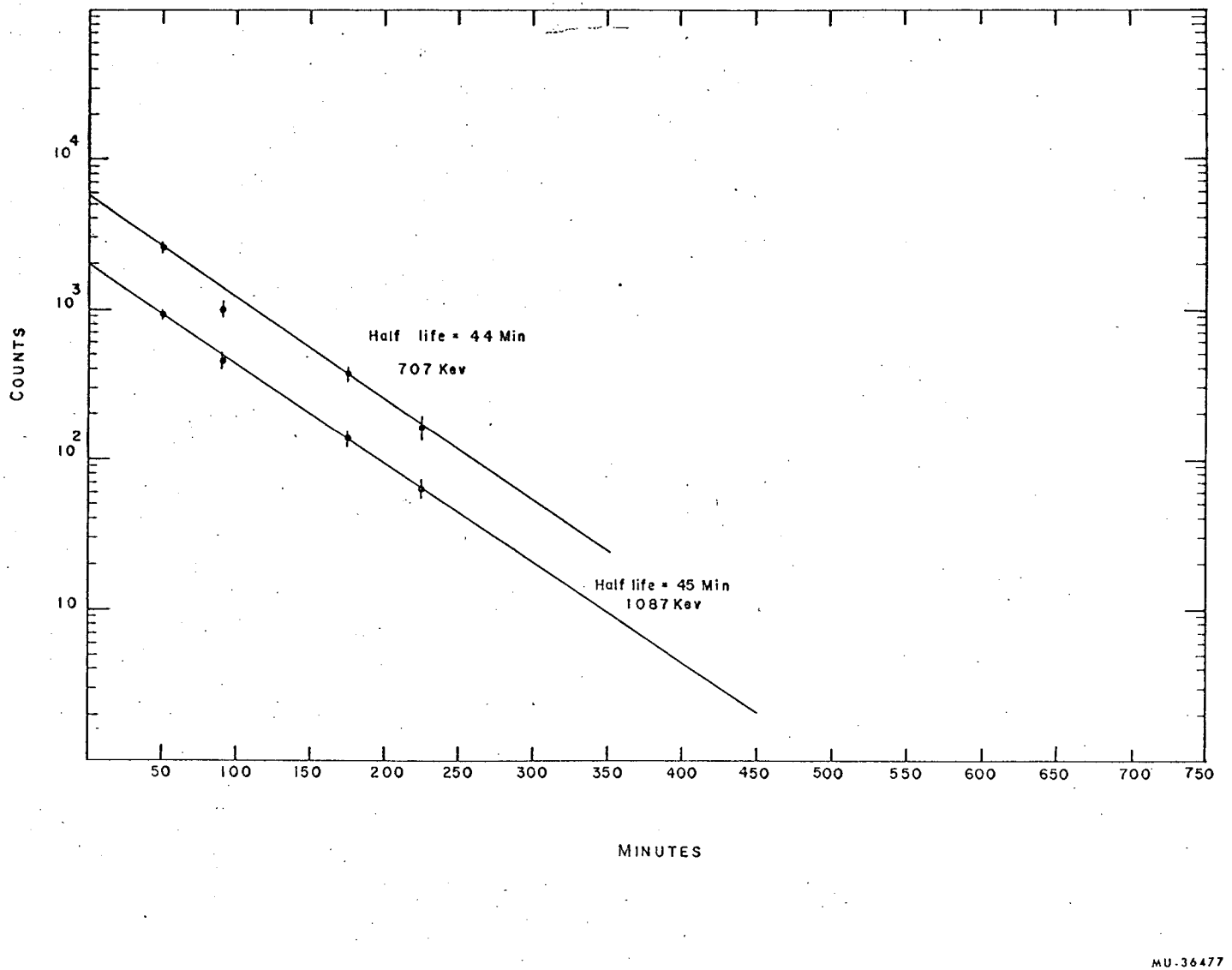

Fig. 8. Decay curves of shorter half-life gamma rays in the chemically purified Po sample.

Table III. Relative intensities.

Energy (keV)	Efficiency	Relative counting rate	Relative intensity
Gamma-rays of Po^{204}			
140	2.6×10^{-3}	23764	66
202	1.42×10^{-3}	2620	13
268	8.6×10^{-4}	11827	100
305	6.9×10^{-4}	1600	17
316	6.5×10^{-4}	1733	19
537	2.6×10^{-4}	1857	52
680	1.71×10^{-4}	917	40
764	1.38×10^{-4}	810	43
883	1.1×10^{-4}	1523	101
1022	8.9×10^{-5}	937	77
1037	8.8×10^{-5}	339	28
Gamma-rays of Bi^{202}			
421	3.9×10^{-4}	600	100
961	9.6×10^{-5}	233	158
Gamma-rays of Po^{203} or Po^{202} (?)			
707	1.6×10^{-4}	159	100
1087	8.4×10^{-5}	93	111

$$\text{Alpha branching ratio} = \frac{Po^{204}\ (\text{alpha intensity})_{t=0}}{\dfrac{Bi^{204}(899\ \text{keV gamma total intensity})_{t=t}}{e^{-k_{Bi}t} - e^{-k_{Po}t}} \times \dfrac{k_{po} - k_{Bi}}{k_{Bi}}}$$

$$= (0.66 \pm 0.007)\ \%$$

$$k = \frac{0.693}{T_{1/2}}$$

A grid chamber was used for the alpha measurement with counting efficiency of 42%. A known source of Na^{22} containing $(1.06 \pm 0.6) \times 10^{6}$ dpm was used for the absolute efficiency calibration. A conversion coefficient correction (0.85%) for the 899 keV gamma ray was also made.

IV. DISCUSSION

The alpha particle branching ratio of Po^{204} has been estimated by Karraker and Templeton[3] to be 1% based on their experiment of comparison of expected reaction cross sections and actual yields. Latimer, Gordon and Thomas[16] have plotted the log partial alpha half-life of even-even polonium isotope versus the inverse square root of the effective alpha-decay energy. They have been able to predict a value of 0.0063 ± 0.0016. Our result of 0.0066 ± 0.0007 in the present work is in good agreement with their calculated value.

There is a vast amount of work on the energy levels remaining to be done. High resolution γ-γ and e^--γ coincidence experiments in particular would be worth further investigation.

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I would like to express my gratitude to Dr. Frank Asaro and Professor Isadore Perlman, under whose continued help and guidance this work was conducted. The discussions with Dr. William C. McHarris are appreciated as well as the opportunities to share bombarding time with Mrs. Janis M. Dairiki.

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