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Part A. An X-Ray Spectrometer for Use in Radioactivity Measurements, The L X-Rays of Neptunium and Plutonium

Part B. Some Lighter Isotopes of Astatine

G. W. Barton

May 3, 1950

Berkeley, California

PART A. AN X-RAY SPECTROMETER FOR USE IN RADIOACTIVITY MEASUREMENTS, THE L X-RAYS OF NEPTUNIUM AND PLUTONIUM

PART B. SOME LIGHTER ISOTOPES OF ASTATINE

G. W. Barton

Department of Chemistry, Radiation Laboratory
University of California, Berkeley, California

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A bent crystal focussing X-ray spectrometer has been designed and built for the observation of the X-and gamma-rays emitted in association with radioactive decay. The instrument uses proportional counter detection. The instrument and the associated electronic circuits are described. The plutonium L X-rays from Cm^{242} and the neptunium L X-rays from Am^{241} have been measured. Values of the energies and abundances are reported.

Part B. ---------- Page 31

Some isotopes of astatine hitherto unreported have been observed in bombardments of bismuth on the University of California 184" cyclotron. They are:

Z^A	Half-life	Decay Properties
At^{209}	5.5 hr.	5.65 Mev alpha particles, electron capture
At^{208}	6.3 hr.	Electron capture, no alphas observed
At^{207}	2.0 hr.	5.75 Mev alphas, presumably electron capture
At^{206}	2.6 hr.	Electron capture, no alphas observed
At^{205}	23 min	5.90 Mev alphas, electron capture
At^{204}	24 min	Electron capture, no alphas observed
At^{203}	7 min	6.10 Mev alphas
At^{201}	1.7 min	6.35 Mev alphas

PART A. AN X-RAY SPECTROMETER FOR USE IN RADIOACTIVITY MEASUREMENTS, THE L X-RAYS OF NEPTUNIUM AND PLUTONIUM

INTRODUCTION

In the observation of radioactive decay it is often of interest to determine the energies of the electromagnetic radiations more accurately than can be done by absorption measurements. For this purpose, the ten-inch X-ray spectrometer was designed and built.

Electromagnetic radiation of from 10 to 200 Kev energy, approximately the limits of this instrument, can arise from three sources: gamma-ray emission, X-rays from internal conversion of a gamma-ray, or X-rays following orbital electron capture. Precise measurement of the energy enables one to distinguish between these processes. The energy of the gamma ray is determined by the change in the nuclear configuration, that of the X-rays from the other two processes by the change in the atomic configuration. Internal conversion consists of the ejection from the atom of an orbital electron with a kinetic energy corresponding to the difference between the energy of the gamma transition and the binding energy of that electron as an orbital electron in the atom. The charge on the nucleus is not changed in this process. Consequently, the X-rays emitted are the same as those of the parent atom. Electron capture consists of the absorption and annihilation in the nucleus of one of the orbital electrons, thus decreasing the nuclear charge by one unit. The X-rays here emitted are those of the daughter atom, an atom of the element preceding the parent element in the periodic system.

One of the most important problems in the design of a spectrometer for the measurement of these radiations is the provision of the maximum possible sensitivity, since the intensities available from cyclotron bombardment are strictly limited. The possibility of making a focussing spectrometer was first examined by de Broglie[1] and by Dardord.[2] Du Mond and Kirkpatrick[3]

discussed the problems involved in the design of such an instrument. Cauchois[4] was the first to build a thoroughly satisfactory instrument. The essential feature of her design was the use of an elastically bent crystal, which enabled her to focus the radiations without structural complications in the instrument and at the same time to preserve a high resolution. Her design has since become generally adopted.

The first use of a spectrograph to identify X-rays associated with radioactive decay was by Abelson[5] at California, who observed the X-rays of molybdenum from the decay of element 43. Poole[6] at Ohio State has used this method in a number of experiments. Marmier, Blaser, Preiswerk, and Scherrer[7] at Zurich have built and used a Cauchois type instrument. Du Mond[8] at Cal Tech has described a gamma ray spectrometer using Geiger tube detection.

PRINCIPLE OF OPERATION

Consider a circle of radius R as in Figure 1. Define points C and C', and two conjugate foci F_R and F_V, for simplicity equidistant from and on opposite sides of C'. For focussing the two relations which have to be satisfied are the Bragg law

$$\sin \theta = \frac{n\lambda}{2d}$$

where θ is the angle of diffraction

λ is the wavelength of the radiation

d is the crystal spacing of the planes in use

n is an integer, the order of the diffraction; and the law of reflection, that the angle of incidence is equal to the angle of reflection.

It is seen that the angle θ subtended by F_R and C' and by F_V and C' from any point P on the circle are constant and equal, irrespective of the location of P on the circumference. This suggests that focussing would be

Figure 1

Geometrical Idealization of the Focussing Conditions

for a Cauchois Type X-ray Spectrometer

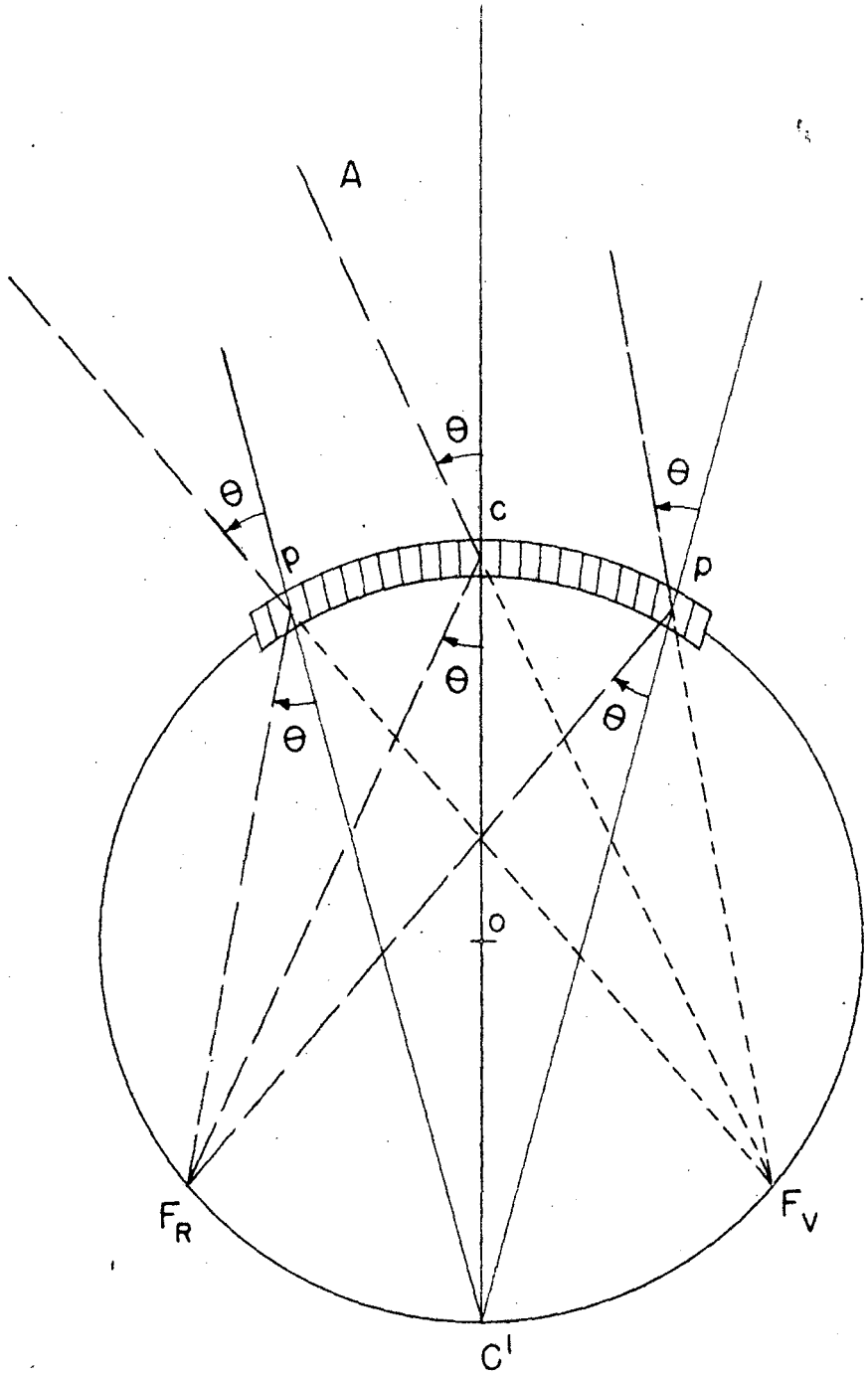


FIG. 1

obtained if a crystal were located on the circumference of the circle with all the diffracting planes pointing toward C'. In this case F_R, say, would be a real focus and F_V a virtual focus. In case the desired planes are not perpendicular to the crystal face but instead lie at an angle α from the perpendicular, it is only necessary to move both F_R and F_V an arc of 2 α around the focal circle in the same direction. Actually, to bend an originally plane crystal to one radius, and then to polish the surfaces to conform to another radius is a very difficult problem, which, it turns out, it is not necessary to solve. Cauchois[4] was the first to demonstrate that the focusing is perfectly satisfactory for a crystal which is merely bent to a radius 2R and the mounted tangent to the focal circle at C. This arrangement necessarily causes some aberration from perfect focussing. This is not serious, however, inasmuch as the crystal, for the practical reasons of procurement must be limited to a small aperture near C'. It might be thought that there would be some defocussing due to the finite thickness of the crystal. Cauchois,[4] though, showed that the compression on the inner face of the bent crystal and the stretching of the outer face are just sufficient to preserve the focus.

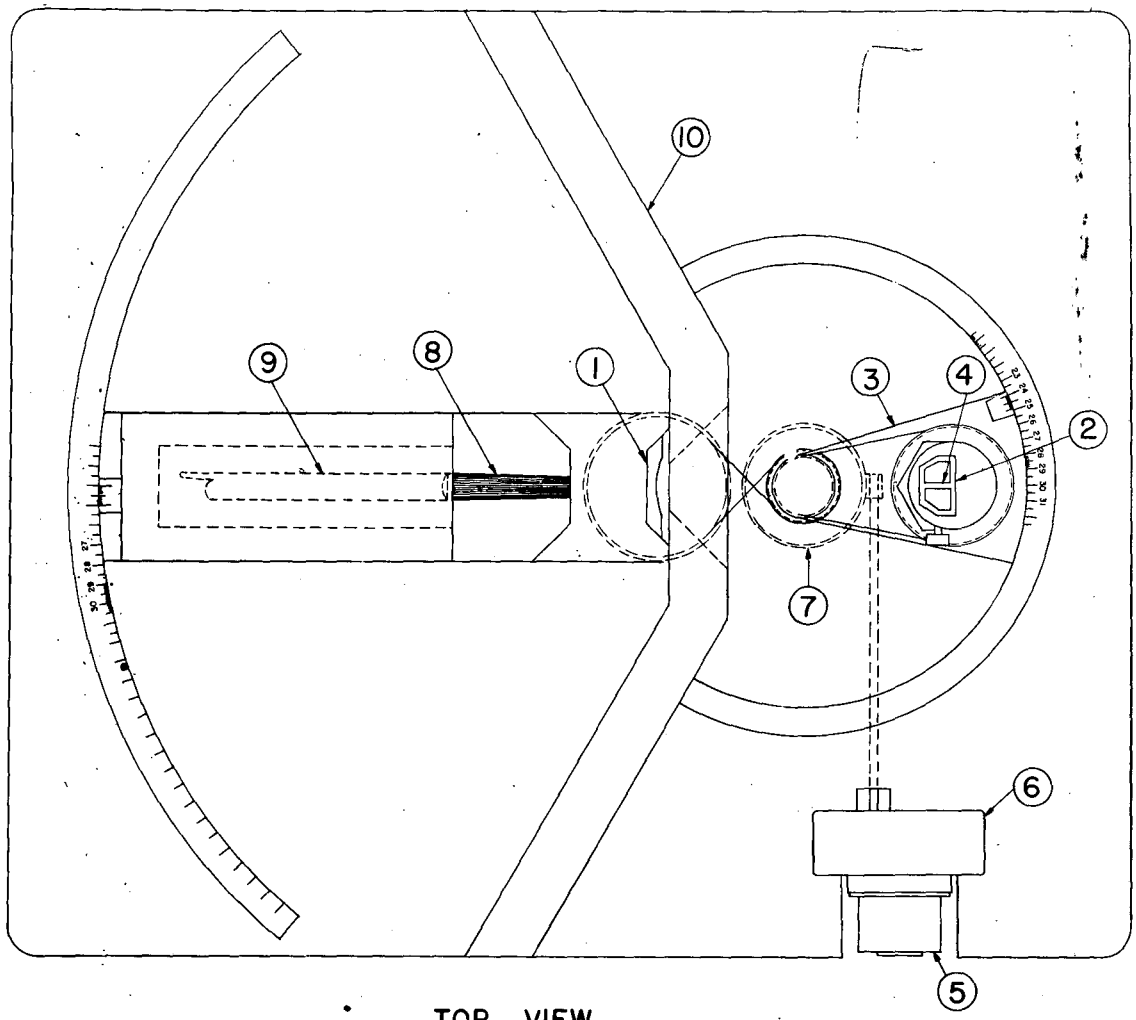
One sees that for a transmission spectrograph, the source may be placed in the region A and the detector, say a piece of photographic film, can be placed along the focal circle at F. On the other hand, the source can be placed at F and the detector, say a geiger tube, can be placed at A. Du Mond[8] has shown that there is a very significant increase in the sensitivity attainable if one uses the principle in this second manner. A naive calculation, assuming perfect focussing, gives a factor of 1000 in favor of the second arrangement for our particular crystal holder. This increase is due to all the X-rays from the source being incident upon the crystal at the appropriate angle for diffraction. The true increase in intensity is undoubtedly less than the calculated, but still quite appreciable.

CONSTRUCTION OF THE INSTRUMENT

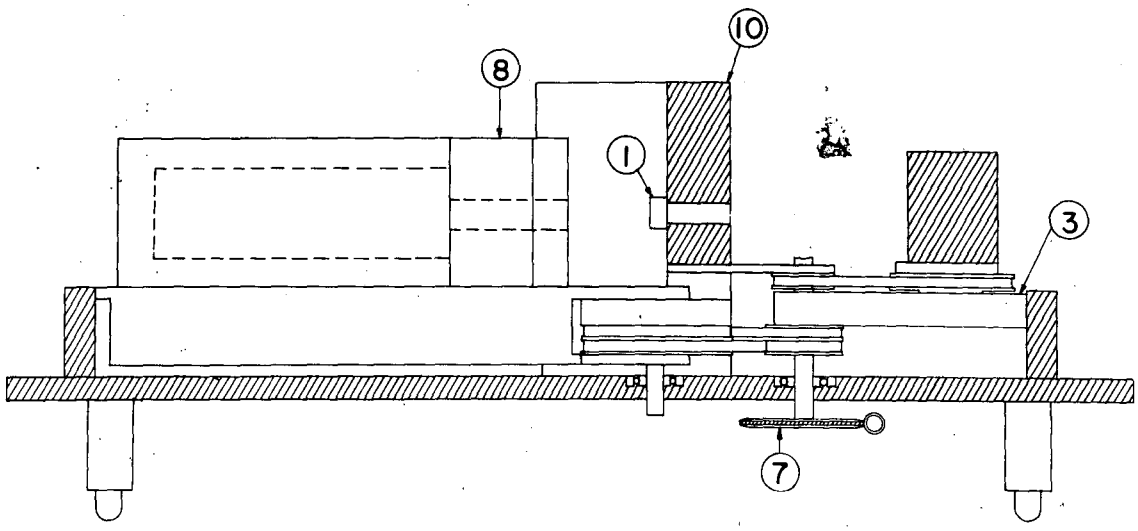
Following the reasoning of Du Mond,[8] and after some tests with a simpler spectrograph using photographic detection, it was decided to design the spectrometer as a monochromator using counting tube detection. Figure 2 is a scale drawing of the essential parts of the machine. Comparing Figure 1, the idealised geometry of the instrument, with Figure 2 one can see how it operates. The crystal is clamped in a tool steel holder (1) whose surfaces are machined to a radius of ten inches. The holder is mounted so that the center of the crystal is at point C. The sample (2) is placed on the arm (3) which rotates about the point O, tracing out the focal circle $CF_RC'F_V$ of five inch radius. A varible lead slit (4) defines the sample width, and a shield is placed around the sample to reduce the unwanted radiation. An arrangement of metal bands is provided so that the sample always faces the crystal aperture. The detector arm rotates about the point C with an angular velocity one half that of the sample arm. Metal bands are used to assure that the angles between the crystal planes and the sample, and between the crystal planes and the detector are always equal. The sample arm and the detector arm, when the instrument is on continuous sweep, are driven by the motor (5), through the gear changing box (6) and the worm and gear (7). The gear changing box together with the one rpm motor, furnish angular speeds of the sample arm of 1 degree per minute, 1/5 deg/min, 1/20 deg/min, or 1/100 deg/min. A lead collimator (8) prevents direct radiation from the sample from being seen by the counting tube (9) which is mounted inside a lead housing. An additional fixed shield (10) gives still more discrimination against direct radiation. A scale (11) permits the reading of the position of the sample. The scale is graduated in units of 4 degrees, the tenths of units marked and

Figure 2

Scale Drawing of the Essential Parts of the X-ray Spectrometer. Description will be found in text.



TOP VIEW



SIDE VIEW

INCHES

FIG. 2

a vernier has been provided for the reading of hundredths of units.

The crystal in use at present in the spectrometer is quartz cut perpendicular to the 310 planes and 0.008 inch thick. The aperture in the crystal holder is 3/4 inch long by 1/4 inch high. As yet, only the 310 planes of quartz have been used in the instrument. Dr. D. A. Lind suggested the use of topaz. Such crystals are now in order. It is hoped that they will enable the sensitivity to be increased by a factor of six without seriously decreasing the resolution.

The standard for the neptunium and plutonium energies reported has been the grating spacing of the 310 planes of quartz as reported in the literature, 1.178 A. The conversion factor used has been

$$\lambda \times E = 12.395 \text{ A} \times \text{Kev}$$

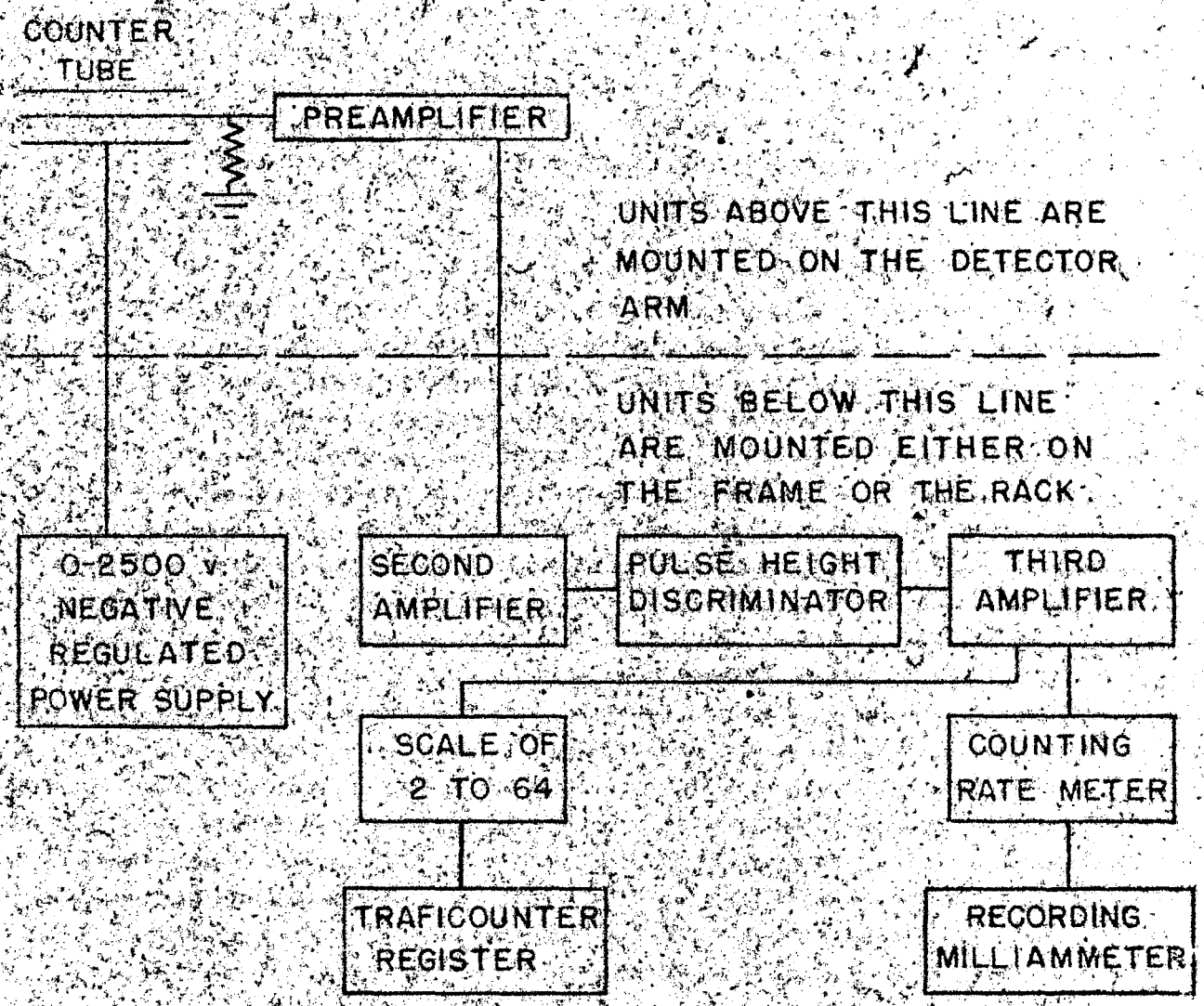
Measurements have been taken on both sides of the spectrometer zero point, in order to eliminate any error caused by inaccuracies in the location of the sample or the orientation of the crystal planes. The line width in the measurements reported has been limited by the finite extent of the sample.

The detector in use for these measurements is an end window Geiger-Müller type tube filled with 55 cm xenon and 15 cm methane and working in the proportional region. The tube has a tungsten wire anode and a copper cathode. The tube is operated in a pulse height selection circuit to discriminate against those counts occurring in the tube from particles not of the energy it is desired to detect. The efficiency of the tube has been measured for the L x-rays of neptunium using the decay scheme of W. W. T. Crane[9] for Am^{241} and found to be 27% in this energy region.

Figure 3 is a block diagram of the circuit. The tube is operated with the shell at a high negative voltage in order to reduce the problems of

Figure 3

Block Diagram of the Electronic Circuits Associated with the Proportional Counter Detector of the X-ray Spectrometer



BLOCK DIAGRAM OF THE ELECTRONIC COUNTING CIRCUITS.

FIG. 3

coupling the small pulses from the proportional counter to the preamplifier. The preamplifier is located as close as physically feasible to the tube, on top of the detector housing, in order to reduce the stray noise pickup at this low power level. The pulse is amplified by about a voltage factor of a hundred and delivered to the second amplifier, where it is amplified further. The pulse then passes to a pulse height discriminator which can be adjusted both in band width and in location of the pass band. Thus counts can be discriminated against which are not of the energy it is desired to detect. The gas amplification of the proportional counter is adjusted by changing the high voltage on the tube. The circuit is usually operated with the overall gain such as to make the pulses of interest about 70 volts at the input to the discriminator, and with a band width just sufficient to include most of the straggling, around 12 volts. This scheme has made it possible to reduce the background in this high efficiency tube to 3-10 c/m, varying with the amount of activity in the adjacent laboratory. From the pulse height discriminator the desired pulses are once more amplified and then fed to both a scaler and a counting rate meter. The counting rate meter indicates the amount of radiation entering the tube as a function of time on an Esterline-Angus graphic ammeter. This in turn can be related to the angle of diffraction since both the ammeter record chart and the sample holder are driven by synchronous electric motors. Ordinarily the counting rate meter is used only as a monitoring device, to detect large fluctuations in the counting rate, and the data are taken from the scaler. The register used on the scaler is a Streeter-Amet Traficounter with the variable stamping rate adjusted to a convenient interval. The angular position of the sample is recorded for selected stamps and the positions for other stamps are obtained by interpolation.

EXPERIMENTAL RESULTS

1. Plutonium X-rays from Cm^{242}

Cm^{242} decays by the emission of alpha particles of 6.08 Mev energy and has gamma rays of 49 Kev energy.[10] This gamma is partially converted in the L orbits of the plutonium atom. The X-rays arising from the refilling of these orbits have been observed and measured using a 24 μg sample of CmF_3 purified and mounted by Mr. W. W. T. Crane. The sample was mounted in a glass capillary making a line source about 1 cm long. This capillary was held vertically in the sample housing by a lucite holder. No slit was necessary for a source this well defined. The spectrometer was adjusted to sweep 1 unit every 80 minutes and the traficounter to stamp every 0.8 minutes. Thus a point was taken every 0.01 units. In presenting the data, a system of curve smoothing was employed which averaged over each five adjacent points and plotted the number so obtained at the center of the smoothing interval. Each number then representa a four minute count. These observations are presented in Figures 4 and 5 and in Table I. Figure 4 is the diffraction observed from the right of the spectrometer zero. Figure 5 is the diffraction observed from the left. Figure 6 compares the points directly observed with the line drawn through them after the smoothing operation. The top of the curve is depressed, due to the contributions from points not on the maximum.

In order to assign the transitions, an extrapolation of the known energies for thorium and uranium was attempted. The simplest of all extrapolation procedures, the use of the Moseley relation,[11] was chosen inasmuch as the extrapolation distance is small, and anyway, no exact theoretical calculation exists. Moseley's formula is

$$E^{1/2} = K(Z - \sigma)$$

where K and σ are almost constants for a given atomic level, E is the level energy,

Figure 4 and 5

The Observed Diffraction Pattern from a Sample of Cm^{242}. Figure 4 is the spectrum taken from the right of the spectrometer zero point. Figure 5 is from the left.

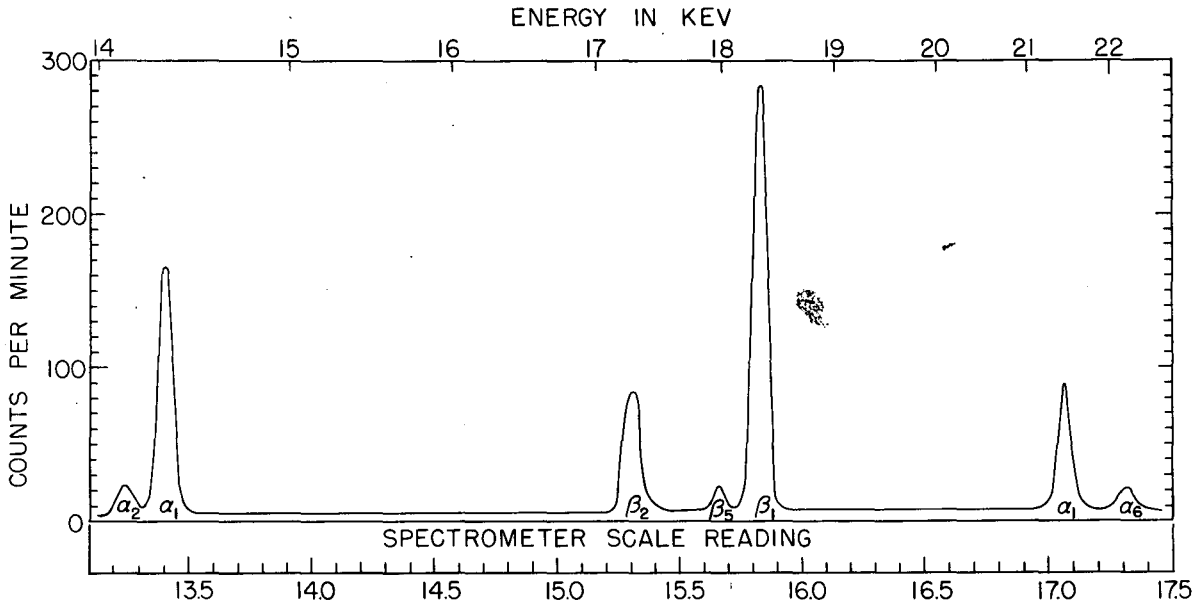


FIG. 4

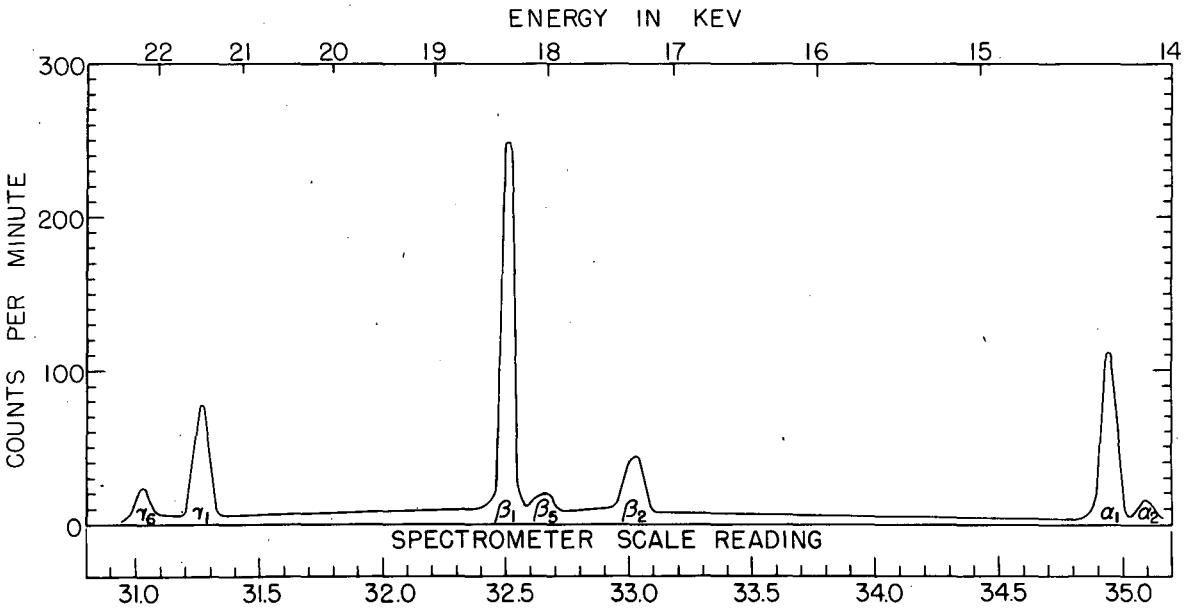
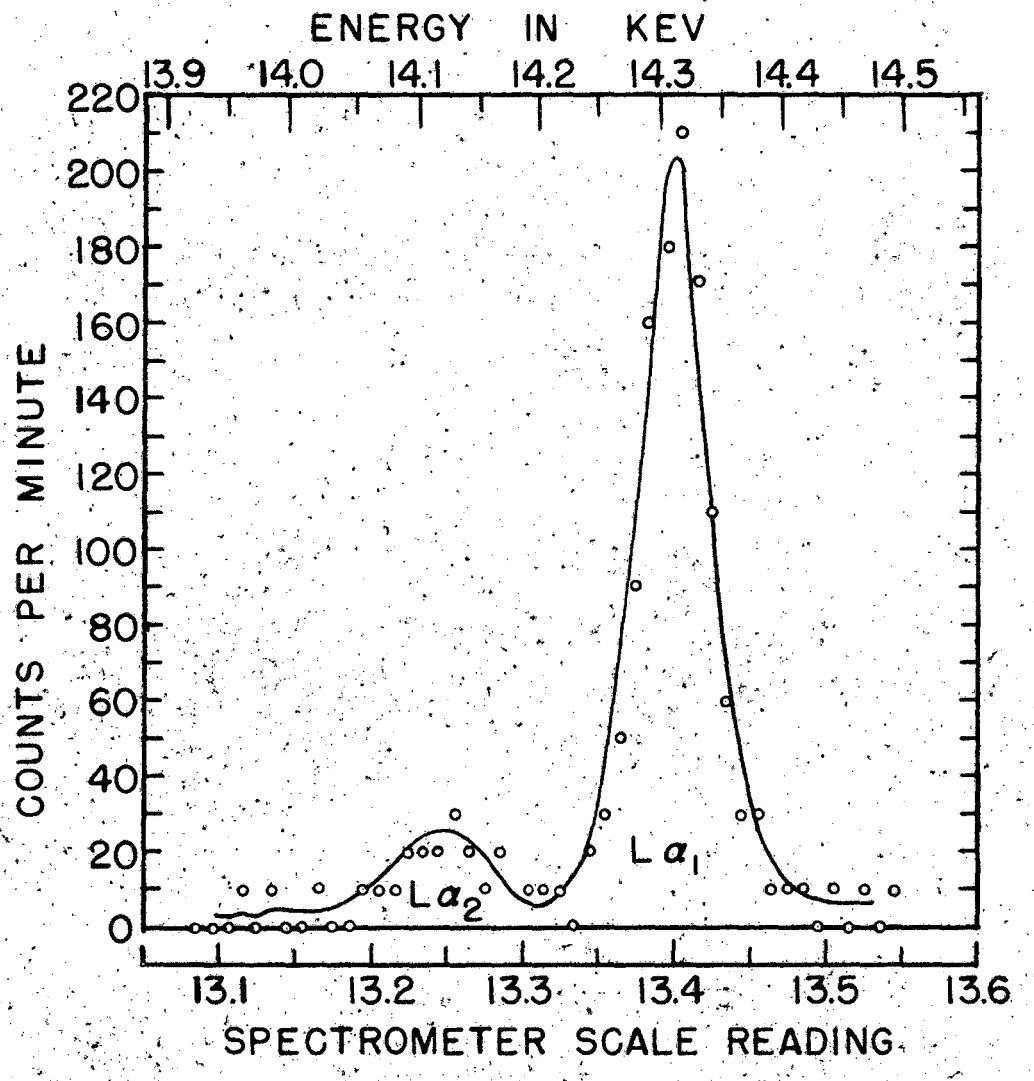


FIG. 5

OZ 901

Figure 6

A Comparison of the Observed Points With the Line Drawn Through Them After the Curve-smoothing Procedure


ENERGY IN KEV
13.9
14.0
14.1
14.2
14.3
14.4
14.5
220
200
180
160
140
120
100
80
60
40
20
0
COUNTS PER MINUTE
$L\alpha_1$
$L\alpha_2$
13.1
13.2
13.3
13.4
13.5
13.6
SPECTROMETER SCALE READING

14867-

Table I. Plutonium L X-Rays from Cm^{242}

L Designation	Spectrometer Scale Reading First	Second	Angle of Diffraction θ	Energy
$L\alpha_1$	13.404	34.932		
		34.947		
	13.404	34.940	21.536 ± 0.02	14.31 ± 0.01 Kev
$L\alpha_2$	13.244	35.090	21.846 ± 0.02	14.14 ± 0.01
$L\beta_1$	15.830	32.500		
		32.495		
	15.830	32.497	16.667 ± 0.02	18.35 ± 0.02
$L\beta_5$	15.665	32.650		
		32.660		
	15.665	32.655	16.990 ± 0.02	17.91 ± 0.02
$L\beta_2$	15.305	33.020	17.725 ± 0.02	17.28 ± 0.02
$L\alpha_1$	17.065	31.262		
	17.067	31.250		
	17.066	31.256	14.190 ± 0.02	21.46 ± 0.04
$L\alpha_6$	17.315	31.025	13.710 ± 0.02	22.20 ± 0.04
$L\beta_3$	16.05			18.53 ± 0.10

and Z is the atomic number. The data for this calculation were taken from Siegbahn.[12] Table II presents this extrapolation. Table III compares the measured energy values with the extrapolated values calculated from Table II. The transitions assumed are those listed in Compton and Allison.[13] Also listed in Tables II and III for comparison are the extrapolated values of Monk and Allison,[14] who used a formula of Sommerfeld derived from the old quantum theory. A term diagram including all the observed transitions is presented in Figure 7.

In addition in Table III, we have the observed and corrected intensities of the lines. The directly observed intensities in Figures 4 and 5 are subject to a number of corrections. The total absorption of the sample mount, crystal, and counter window, (about 150 mg/cm^2 of silica) and the self absorption of the sample have to be corrected for. The counting efficiency of the tube is a rapidly varying function of energy. The reflection coefficient of the quartz crystal is varying as $1/E^2$.[15] An attempt has been made to correct for all but the self-absorption, but too much trust must not be placed in these values. The absorption has been assumed to be equal to that of 150 mg/cm^2 of aluminum, the absorption coefficient for which has been taken from the compilation of S. J. M. Allen.[16] The calibrations of counting efficiencies for xenon tubes of Crane and Ghiorso[17] have been assumed. Table III contains the observed intensities, the corrected intensities, and the uranium values of Allison[18] for comparison.

Allison's values for the relative abundances of the L x-rays from uranium were made with an ionization chamber on x-rays produced by electron bombardment. A two-crystal spectrometer was used. His values were corrected for self absorption, reflection coefficient, and counting efficiency. It will be

Table II. Prediction of Energy Levels-Moseley Extrapolation Using Data from Siegbahn[12]

Level (Siegbahn)	$\sqrt{\nu/R}$ 92 Th (Siegbahn)	92 U (Siegbahn)	94 Pu (Extrap.)	ν/R Pu	Energy Corresponding (Kev)	Monk and Allison's Predicted Value[14]
K	89.93	92.05	94.17	8868.0	120.63	120.9
L_I	38.81	40.03	41.25	1701.6	23.15	22.9
L_{II}	38.07	39.28	40.49	1639.4	22.30	22.1
L_{III}	34.63	35.56	36.49	1331.5	18.11	18.0
M_I	19.50	20.21	20.92	437.6	5.95	5.91
M_{II}	18.82	19.53	20.24	409.7	5.77	
M_{III}	17.22	17.80	18.38	337.8	4.59	
M_{IV}	15.98	16.56	17.14	293.8	4.00	
M_V	15.61	16.16	16.71	279.2	3.80	3.75
N_I	9.81	10.30	10.79	116.4	1.58	
N_{II}	9.19	9.67	10.15	103.0	1.40	
N_{III}	8.34	8.75	9.16	83.9	1.14	
N_{IV}	7.14	7.58	8.02	64.3	0.87	
N_V	6.95	7.37	7.79	60.7	0.82	
N_{VI}	4.87	5.34	5.81	33.8	0.46	
N_{VII}	4.82	5.25	5.68	32.3	0.44	
O_I	4.43	4.87	5.31	28.2	0.38	
O_{II}	3.95	4.28	4.61	21.3	0.29	
O_{III}	3.39	3.73	4.07	16.6	0.23	
$O_{IV,V}$	2.26	2.65	3.04	9.3	0.13	
$P_{II,III}$		0.89				

Table III. Comparison of Predicted and

Observed Energies L Series X-rays of Plutonium

Line	Transition[13] Initial State	Transition[13] Final State	Energy: Monk & Allison (Kev)[14]	Energy: This Extrap. (Kev)[a]	Energy: Measured (Kev)[b]	Intensity: in Uranium[18]	Intensity: Observed Here[c]	Intensity: Corrected Here[d]
α_1	L_{III}	M_V	14.30	14.31	14.31	100	160	100
β_1	L_{II}	M_{IV}	18.27	18.30	18.35	49.4	276	91
β_2	L_{III}	N_V		17.29	17.28	28	78	28
γ_1	L_{II}	N_{IV}		21.43	21.46	12	80	23
α_2	L_{III}	M_{IV}	14.11	14.11	14.14	11	20	13
β_5	L_{III}	O_V		17.98	17.91	6.4	16	6
β_3	L_I	M_{III}	18.55	18.56	18.53	4.2		
β_4	L_I	M_{II}	17.60	17.58		4.1		
l	L_{III}	M_I	12.12	12.16		3.4		
γ_6	L_{II}	O_{IV}		22.17	22.20	2.2	14	4
β_6	L_{III}	N_I		16.53		1.6		
γ_2	L_I	N_{II}		21.75		1.5		
γ_3	L_I	N_{III}		22.01		1.4		
η	L_{II}	M_I	16.28	16.35		1.0		
β_7	L_{III}	O_I		17.73		0.4		

a) Predicted from energy levels in Table II.

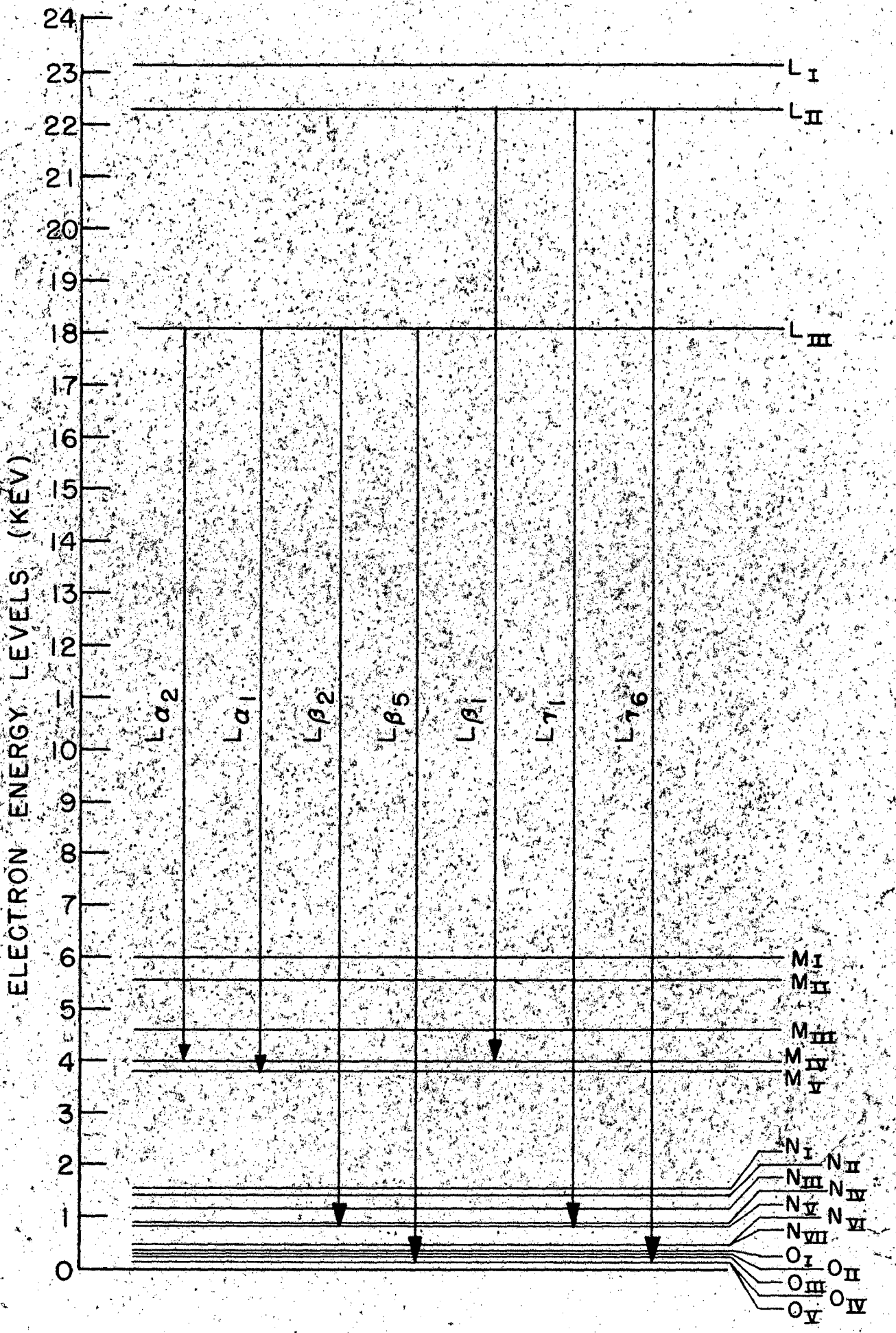
b) Measured in this work. See Table I.

c) See Figure 5.

d) Not corrected for self absorption.

Figure 7

Term Diagram for X-ray Energy Levels in the Plutonium Atom With All the Transitions Observed in This Study Plotted


ELECTRON ENERGY LEVELS (KEV)
24
23
22
21
20
19
18
17
16
15
14
13
12
11
10
9
8
7
6
5
4
3
2
1
0
L_I
L_{II}
L_{III}
$L\alpha_2$
$L\alpha_1$
$L\beta_2$
$L\beta_5$
$L\beta_1$
$L\gamma_1$
$L\gamma_6$
M_I
M_{II}
M_{III}
M_{IV}
M_V
N_I
N_{II}
N_{III}
N_{IV}
N_V
N_{VI}
N_{VII}
O_I
O_{II}
O_{III}
O_{IV}
O_V

14868-1

noted that the relative abundances of the plutonium transitions arising from the L_{III} initial state, the α_1, β_2, α_2 and β_5 groups, are in excellent agreement with Allison's measured values. The intensities of the lines arising from the L_{II} initial state, the β_1, γ_1 and γ_6 groups, are uniformly high by about a factor of two. Data on the interconversion between the L_I, L_{II} and L_{III} levels by Auger processes or other processes would enable the calculation of the relative excitations of these levels by the internal conversion. We can say, at least, that the internal conversion, together with atomic interconversion processes, excites the L_{II} level to twice the degree that it is excited in electron bombardment. The measurement of the relative excitations will in turn, indicate the spin change of the plutonium nucleus in undergoing this gamma emission.

2. Neptunium X-rays from Am^{241}

Am^{241} decays by the emission of an alpha particle of energy 5.45 Mev and a 62 Kev gamma ray.[19] These gammas are also highly converted in the L shell of the daughter atom, in this case Np^{237}. The sample, about 0.5 mg of AmF_3, was mounted in a lucite holder to reduce the absorption of the X-rays. This measurement was made in the same manner as described for curium. The traficounter was adjusted to stamp every 4 minutes and the spectrometer sweep set at 1 unit every 400 minutes. The smoothing interval was every tenth stamp (40 minutes). The results are shown in Table IV. No attempt is made to estimate the relative abundances since only about 10 c/m above a 10 c/m background was obtained. The $L\alpha_2$ line was also observed, but was not sufficiently well resolved from background to measure.

Table IV. Neptunium L X-rays from Am^{241}

Line Designation	Spectrometer Scale Reading First	Second	Angle of Diffraction	Energy (Kev)
$L\alpha_1$	35.15	13.05	22.10 ± 0.05	13.98 ± 0.03
$L\beta_2$	33.12 33.15 33.14	15.05	18.09 ± 0.06	16.94 ± 0.05
$L\beta_1$	32.69 32.71 32.70	15.50	17.20 ± 0.04	17.79 ± 0.03

SUMMARY

The data herein presented represent the first reported measurements of the X-ray energies of plutonium and neptunium. These energies are now measured to be

X-ray group	Neptunium	Plutonium
$L\alpha_2$		14.14 Kev
$L\alpha_1$	13.98 Kev	14.31
$L\beta_2$	16.94	17.28
$L\beta_5$		17.91
$L\beta_1$	17.79	18.35
$L\beta_3$		18.53
$L\gamma_1$		21.46
$L\gamma_6$		22.20

This data also proves that the gamma-ray is emitted after the alpha decay in these two rather than before, since if the emission occurred before, the X-rays would be those of the parent. Thus this instrument has proved itself

of use in elemental identification of activities. It can also be used to differentiate between electron capture and internal conversion.

Data on interconversion processes would enable one to use this method to calculate the spin changes undergone in the gamma transition. Intensity measurements give the relative level excitations.

ACKNOWLEDGMENTS

Thanks are due to a number of persons who assisted in this work. Mr. Herman P. Robinson contributed many ideas to the design of both the instrument and of the associated electronic circuits. Mr. A. Ghiorso is largely responsible for the use of a proportional counter. Mr. Wallace Decker did the final design of the mechanical details and prepared the working drawings. Mr. Paul Lathrop built the electronic circuits. Finally, many thanks are due Prof. I. Perlman, who directed this work and Prof. G. T. Seaborg for their patient support during the painful months of design and de-bugging. This work was performed under the auspices of the United States Atomic Energy Commission at the Radiation Laboratory of the University of California.

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PART B. SOME LIGHTER ISOTOPES OF ASTATINE

INTRODUCTION

Astatine was first identified and reported by Corson, Mackenzie, and Segrè,[1] who observed the 7.5 hour alpha decay of the isotope of mass 211. Later Kelly and Segrè[2] reported the observation of the isotopes of masses 210 and 212. Meinke, Ghiorso and Seaborg[3] have reported isotopes of masses 214, 215, and 216 in the new collateral alpha decay series. At^{217} appears as a member of the artificially produced neptunium series.[4] A chart of isotopes in this region is shown in Figure 1.

All the isotopes yet found are radioactive, with fairly short half lives. All those with mass greater than 211 have half lives too short for chemical experiments. Only those of mass less than 212 are chemically identifiable. It has been suggested by Perlman, Ghiorso and Seaborg[13] that there seems to be a quite sudden decrease in the nuclear radius when the 127th neutron is removed from a nucleus. This change evidences itself in a decrease in alpha-energy in nuclei with 126 neutrons or less and an increase in the alpha half-life which is sufficient to once more give the nuclei observable half-lives. Figure 2 is a plot of alpha particle energy vs. mass number for astatine isotopes.

Chemically, these isotopes are of interest because of the light they may throw upon the chemical mechanisms of the halogens, of which astatine is the heaviest. From a nuclear point of view, these isotopes are of interest since they lie in the region above the closed nuclear shell of 82 protons and below the shell of 126 neutrons. It is hoped that a study of nuclides in this region may throw light on the nature of nuclear forces and the mechanisms of nuclear decay.

Figure 1

Chart of Nuclides in the Region Around Astatine

Proton Number	116	117	118	119	120	121	122	123	124	125	126	127
85	At^{201} 1.7 m α (1)		At^{203} 7 m α (1)	At^{204} 24 m K (1)	At^{205} 23 m α, K (1)		At^{207} 2.0 h α (1)	At^{208} 6.3 h K (1)	At^{209} 5.5 h 5% α (1)	At^{210} 8.3 h K (2)	At^{211} 7.5 h 40% α (2)	At^{212} 0.12 m α (2)
84				Po^{203} 40 m α, K (2)	Po^{204} 4 h α, K (4)	Po^{205} 1.5 h α, K (2)	Po^{206} 9 d 10% α (2)	Po^{207} 5.7 h K (2)	Po^{208} 3 y α (2)	Po^{209} ~200 y α (3)	Po^{210} 140 d α (2)	Po^{211} 10^{-3} s α (2)
83	Bi^{199} 25 m α, K (5)	Bi^{200} 35 m α, K (5)	Bi^{201} 90 m K (5)	Bi^{202} 1.5 h K (5)	Bi^{203} 13 h K (5)	Bi^{204} 12 h K (5)	Bi^{205} 14 d K (4)	Bi^{206} 6.4 d K (2)	Bi^{207} 10-100 y K (6)		Bi^{209} stable 100%	Bi^{210} 5 d β^-
82		Pb^{199} 80 m K (2)	Pb^{200} 18 h K (2)	Pb^{201} 8 h K (2)		Pb^{203} 52 h K (2)	Pb^{204} stable 1.4 %		Pb^{206} stable 26%	Pb^{207} stable 21%	Pb^{208} stable 52%	Pb^{209} 3.3 h β^- (2)
81		Tl^{198} 1.8 h K (2)	Tl^{199} 7 h K (2)	Tl^{200} 27 h K (2)	Tl^{201} 72 h K (2)	Tl^{202} 11.8 d K (2)	Tl^{203} stable 29.5%	Tl^{204} 3 y β^- (2)	Tl^{205} stable 70.5%	Tl^{206} 4.23 m β^- (2)	Tl^{207} 4.76 m β^- (2)	Tl^{208} 3.1 m β^- (2)

Neutron Number

References:

(1) This work
(2) G. T. Seaborg and I. Perlman, Table of Isotopes, Rev. Mod. Phys. 20, 585 (1948)
(3) E. L. Kelly
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(5) H. M. Neumann and I. Perlman
(6) H. M. Neumann and I. Perlman, Unpublished data

Figure 1

Figure 2

Plot of Alpha Particle Energy Vs. Mass Number for Astatine Isotopes

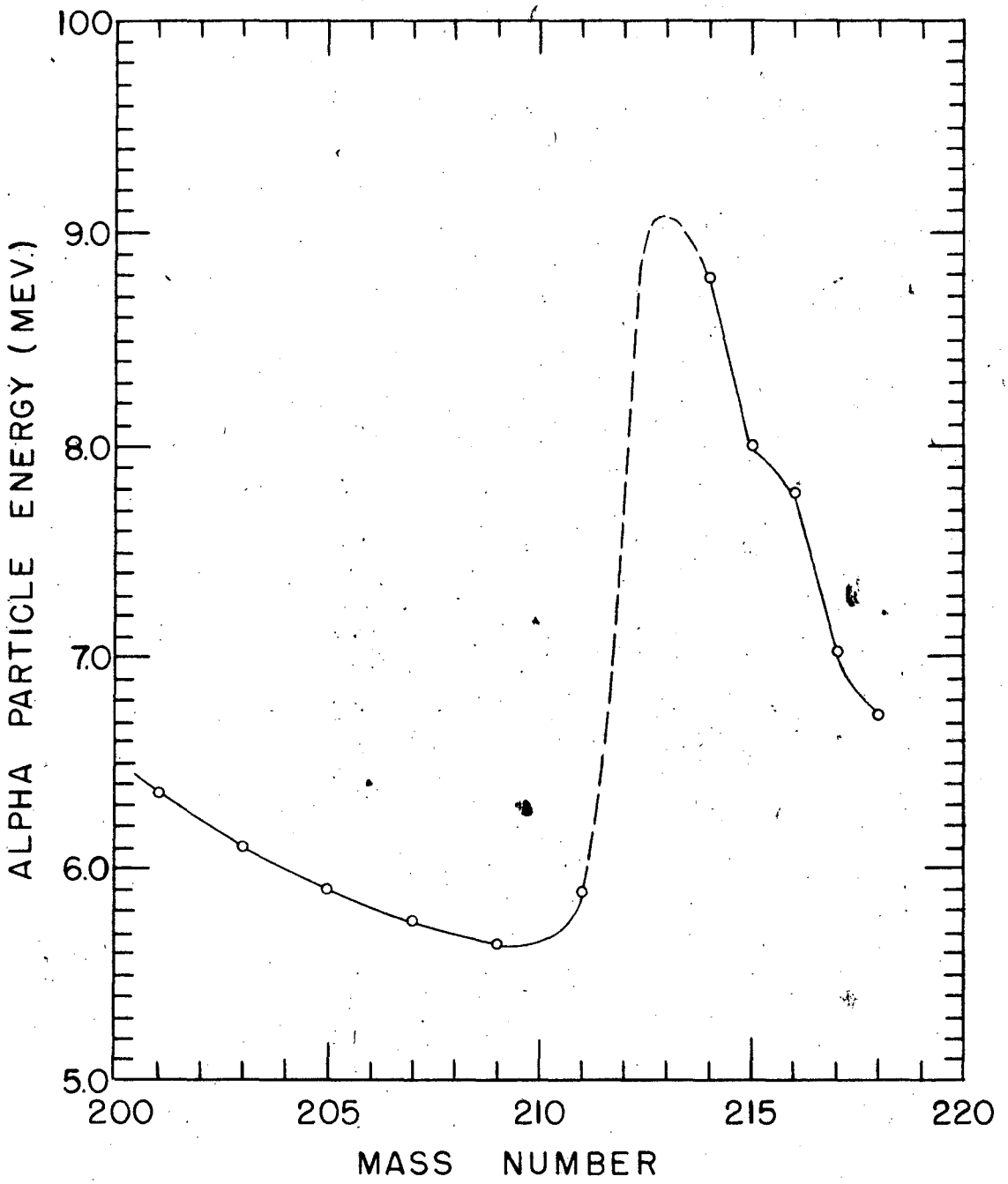

100
9.0
8.0
7.0
6.0
5.0
ALPHA PARTICLE ENERGY (MEV.)
200
205
210
215
220
MASS NUMBER

FIG. 2

EXPERIMENTAL METHODS

All bombardments herein reported were made by the University of California 184 inch cyclotron on a probe target mounted in the internal circulating beam of helium ions. The energy of bombardment was varied by changing the radial location of the target. Bombardments were made on strips of bismuth, on powdered bismuth, or on bismuth oxide. The bismuth strip was held in a watercooled copper clamp while being bombarded to prevent melting the strip. The bismuth oxide was clamped in an aluminum envelope for bombardment. For short bombardments either bismuth oxide or powdered bismuth was bombarded in the stainless steel target holders of the jiffy probe.

The techniques available allow one on the basis of nuclides previously identified to associate observed radiation characteristics with the mass number of the nuclide producing the radiations. The individual methods will be described later, but in general the tools available allow us to make the following observations. The alpha-particle energy and the individual decay of the alpha groups observed in a bombardment can be determined using the 48 channel pulse analyser.[7] The mass assignments are determined by what will be called "milking" experiments. These are experiments designed to identify the half-life of a parent activity by determining the yield of a daughter activity as a function of the time allowed for the parent to decay. Milking experiments were used to identify electron capture daughters by purely chemical methods, and alpha decay daughters by chemical methods or by recoil experiments (see below). Gamma-ray and conversion electron energies can also be determined by tools available, but complications which arise from the similarity of radiations of the various isotopes and confusion from the growth of daughter activities have not allowed these measurements to be useful.

In the course of this study, a number of different varieties of chemistry were tried, most of which turned out to be unsatisfactory. The principal reason for the unsuitability of the methods tried was the slowness of the reactions. The chemistry which was finally evolved consists in bombarding bismuth oxide, dissolving this in concentrated HCl, adding some $FeSO_4$ as a reducing agent, and extracting the astatine from this mixture with di-isopropyl ether (DIPE). In bombardments of the sort used this procedure, with one washing of the DIPE, gave undetectable amounts of any alpha activity other than astatine. If a plate was needed for alpha or geiger counting, an aliquot of this DIPE solution was plated out on platinum or stainless steel counting plates. If a milking experiment was to be done, some tri-butyl phosphate (TBP) was added to the ether solution after purifying the astatine. It has been found that polonium extracts quantitatively from TBP into a 2M HNO_3--4M HCl mixture.[5] At the same time under these conditions astatine has a partition coefficient of at least 200 in favor of the organic phase. This partition coefficient is measured after a period of contact between the aqueous and organic layers of only a minute. Difficulty was found in using the procedures described by Johnson, Leininger, and Segrè[6] in that when the chemistry was done rapidly, the astatine was always showing up in the wrong fraction. Since the partition coefficient measured at equilibrium for all the organic reagents they, or we, have tried is quite high, this must be due to some reaction rate effect. The extraction into TBP was the only one of the many we tried which gave a good separation of polonium from astatine. After removing the polonium, bismuth, and lead from the organic phase, polonium could be extracted into a new fraction of TBP by destroying the nitrate ion and making the solution 6M in HCl. This left the bismuth and lead in the aqueous phase. They could be separated by precipitating BiOCl and $PbSO_4$.

To perform a milking experiment, the astatine would first be purified and placed in TBP solution. After a known interval of time, the organic layer would be extracted, and the bismuth, lead, and polonium brought out. These could then be separated while the astatine was set aside for another growth period. This alternating procedure of growth, separation, growth, separation, was repeated 4 to 6 times.

For very rapid chemistry an "Astatine Boiler" was constructed. This consisted of a stainless steel crucible in which was placed a water-cooled metal finger with a platinum counting disk attached. The crucible had provision for the use of a thermocouple to measure the temperature. The bismuth was placed in the crucible, the metal finger placed above the sample and the bismuth melted. When the bismuth was just above its melting point, most of the astatine collected on the counting plate in 10 minutes and no other alpha activities could be detected. At a somewhat higher temperature, polonium began to distil over too. For the detection of the activities of a half hour or less, 10,000 alpha counts per minute could be collected using a two second bombardment and 10 seconds heating in the astatine boiler. This activity was produced on the jiffy probe in the cyclotron. Since the vacuum carrier system delivers the target in 18 seconds from the cyclotron to the chemistry laboratory, the total time from bombardment to the pulse analyser was less than 1.5 minutes.

Some experiments in which the long lived polonium were looked for were performed by extracting the astatine into benzene or DIPE, purifying the organic phase at intervals, and then plating out an aliquot of this pure astatine fraction. The astatine was allowed to decay completely and the yields of the poloniums were then determined by the use of the 48 channel pulse analyser.[7] (see below)

A method to obtain a physical separation of daughter atoms of alpha emitters consists of collecting the recoil atoms from alpha decay which in the present case are atoms of bismuth isotopes. In these experiments, the astatine was plated on silver to reduce the amount lost by evaporation. Now when an alpha particle is emitted into the plate, the energy of the recoiling daughter nucleus is great enough to cause it to leave the plate. In a vacuum this daughter nucleus can be caught on a second platinum plate placed close to the surface of the silver plate which contains the astatine. Some astatine does vaporize across the gap. This activity was removed by fuming the catcher plate with H_2SO_4 to convert the bismuth to the salt. The plate was then flamed in a Meker burner to remove the residual astatine. The bismuth could then be followed for decay.

Alpha energies and relative amounts of activities of the various astatine and polonium alpha emitters were determined using the 48 channel pulse analyser.[7] This instrument when used with an ionization chamber measures the alpha energy by measuring the amount of ionization produced by the alpha particle. It sorts out the ionization pulses and registers them on one of the 48 equal channels according to the pulse height. Since the amount of ionization produced by an alpha particle is directly proportional to the energy, within the limits of the measurements, a plot of the number of counts per channel against the channel number furnishes an intensity plot of the alpha spectrum versus the alpha energy. For the bombardments performed with the jiffy probe, a method of photographing the register dials was used in order to speed the time of taking points.

Alpha particles were counted in ionization chambers at 52% geometry from 0.001 inch thick platinum plates. Electrons and x-rays were counted from platinum or stainless steel plates in mica window argon or xenon filled geiger counters.

EXPERIMENTAL RESULTS

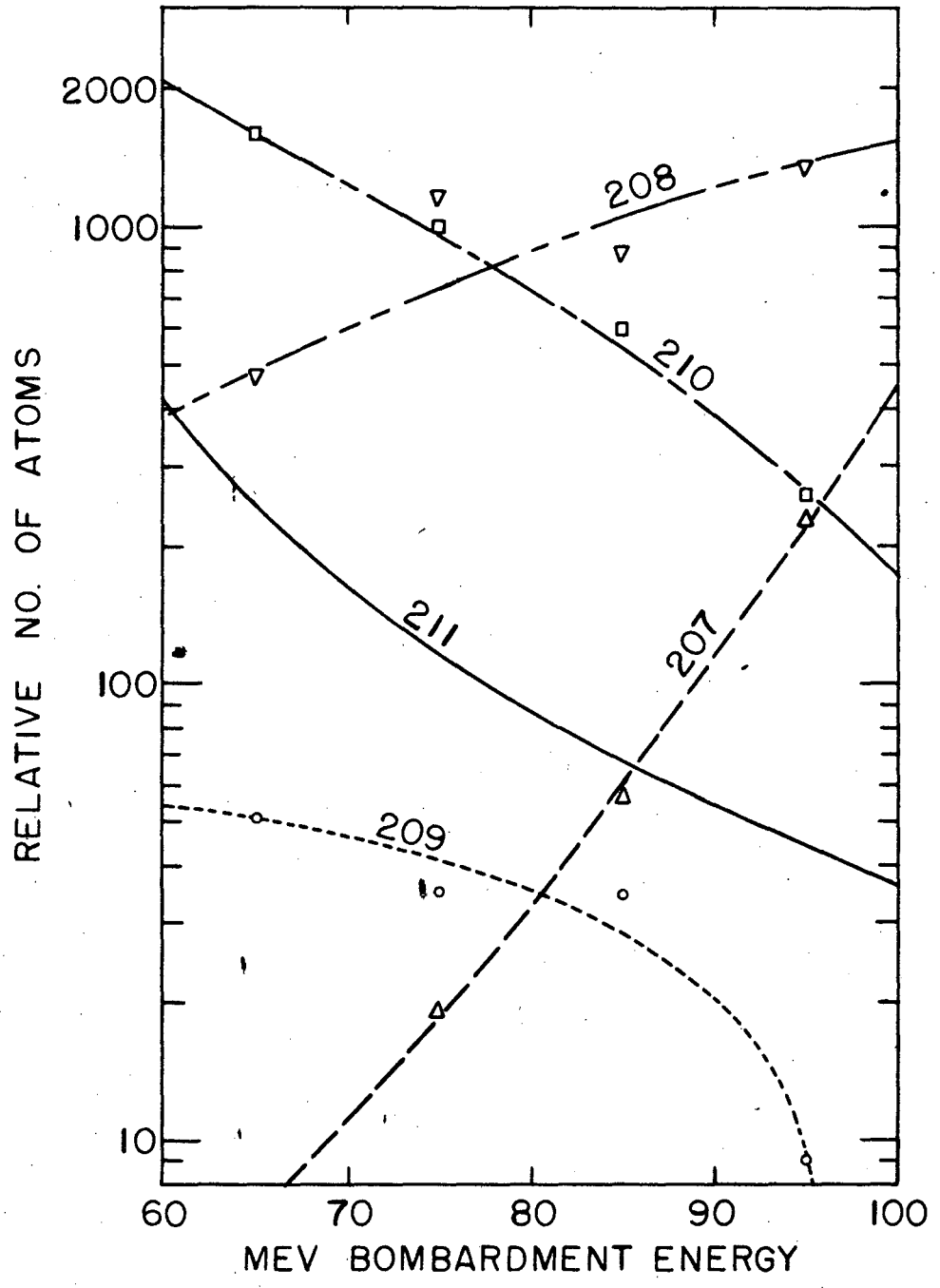
1. At^{211}. This is the isotope first discovered by Segrè and co-workers. It has a half life of 7.5 hours and an alpha particle of 5.89 Mev. It decays 60% by electron capture to the known very short lived Po^{211}(AcC') which has a 7.434 Mev alpha particle. This activity was seen in all bombardments.

2. At^{210}. This isotope was also reported by Segrè.[2] It is identified by its electron capture daughter, the 140 day Po^{210}, which decays by the emission of an alpha particle of 5.298 Mev. Also reported is an 1 Mev gamma ray associated with its decay. The half life is 8.3 hours. No alpha particle has yet been observed, but Neumann and Perlman,[8] by counting the alpha decay daughter activity, 6.4 day Bi^{206}, have measured a branching ratio of ca. 1% for alpha decay. At^{210} was observed whenever looked for in these bombardments.

3. At^{209}. This isotope has not previously been reported. An alpha group of 5.65 Mev, 5.5 hours half life is first observed in pulse analyser curves at a bombarding energy of 60 Mev. At 70 Mev bombardment energy, this 5.65 Mev group was observed to decay with a half life of 5.5 ± 0.3 hours. Figure 3 is a rough excitation function of the activities observed below 100 Mev. The amounts of activity observed at different bombardment energies were normalized to the excitation function of At^{211} determined by E. L. Kelly.[9] This 5.65 Mev activity seems to drop off before the At^{208} as indicated by Po^{208} has reached its peak yield. Figure 4 is a reproduction of the alpha spectrum from astatine prepared at 70 Mev showing alpha-groups of only At^{211} and the new activity assigned to At^{209}. Figure 5

Figure 3

A Rough Excitation Function of the Astatine Activities Observed Between 60 and 100 Mev Bombarding Energies. The points observed have been normalized to fit the excitation function of Kelly[2] for At^{211}.



At^{211} BY E.L. KELLY (ASSUMED)

At^{210} FROM YIELD OF Po^{210}

At^{209} FROM ALPHA YIELD

At^{208} FROM YIELD OF Po^{208}

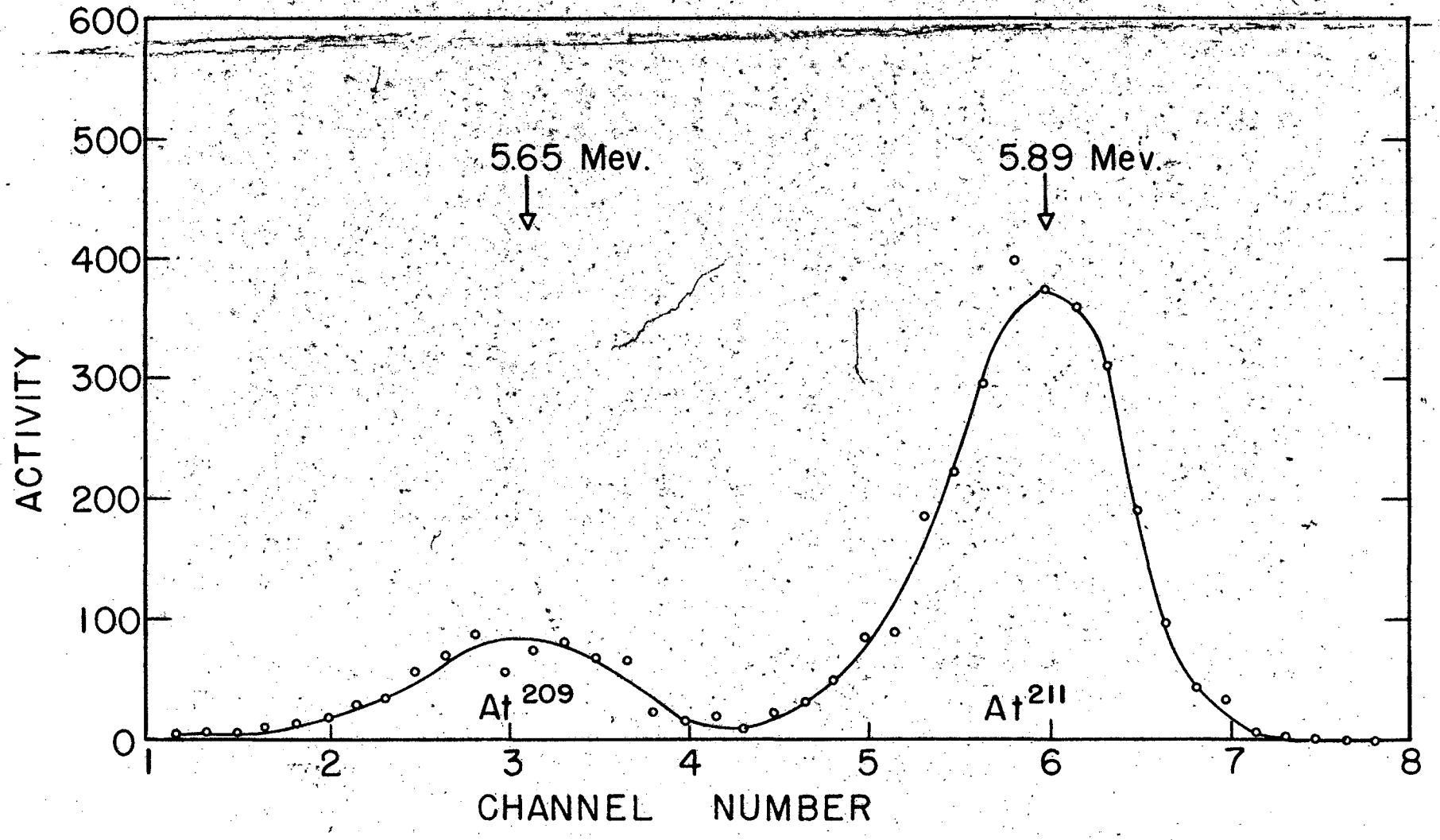
At^{207} FROM ALPHA YIELD

FIG. 3

14861-1

Figure 4

Alpha Spectrum from Astatine Made at 70 Mev as Observed on the Pulse Analyser



ALPHA SPECTRUM FROM At FRACTION OF Bi + 70 MEV He^{++}

FIG. 4

14863-1

shows the decay as determined on the pulse analyser of the two alpha groups observed.

Attempts have been made to identify the daughter activities by direct chemical milking and by recoil experiments. Pure astatine was prepared at 120 Mev purified aliquots of this were plated out at intervals, allowed to decay away, and the yields of the various polonium isotopes measured by use of the pulse analyser. The method did not permit an accurate standardization of the amount of astatine plated out so the yields are normalized to an assumed 8.3 hour decay of At^{210}. These results are plotted in Figure 6 and for illustration the alpha spectrum of one of the P_o milkings is plotted in Figure 7. These results indicate a half-life for the parent of 5.7 hours.

The 14 day Bi^{205} [10] has been observed in recoil experiments from astatine made by 70 Mev bombardment. The yields of activities observed in this experiment are plotted in Figure 8. In this experiment the chemical identification of the activities was not possible. Another experiment, particularly designed to chemically identify the 14 day activity, was a direct milking of the bismuth activity from a sample of astatine prepared at 120 Mev. The yields of the 14 day activity in the bismuth fraction is presented in Figure 9. The first of these experiments indicates a half-life of At^{209} of 5.4 hours. The second indicates a half-life of 6.5 hours. From yield considerations, we estimate the alpha branching to be 5%.

4. <u>At^{208}</u>. This isotope appears, according to Figure 3, at an energy less than 60 Mev. The Po^{208} daughter from the astatine originally on the plate persists at a higher bombardment energy than does the At^{209} alpha activity, and appears at lower energy than the activity which we have assigned to At^{207}. The experiment plotted in Figure 6 indicates a half life of 5.9 hours for this isotope. Another similar experiment indicated a half life of 6.7 hours. No

Figure 5

Decay of Sample Shown in Figure 4

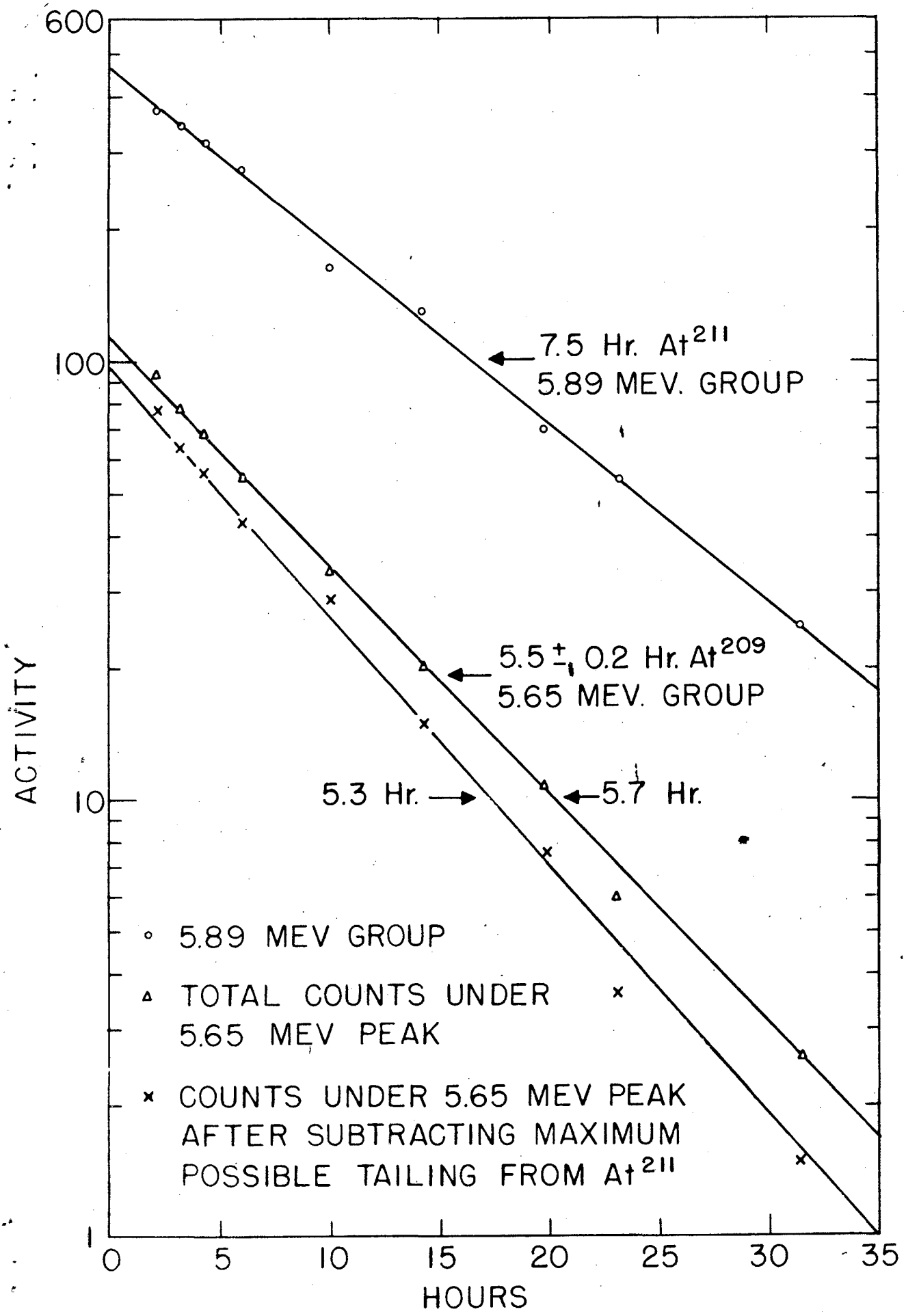

600
100
10
1
ACTIVITY
0
5
10
15
20
25
30
35
HOURS
7.5 Hr. At211
5.89 MEV. GROUP
5.5 ± 0.2 Hr. At209
5.65 MEV. GROUP
5.3 Hr.
5.7 Hr.
5.89 MEV GROUP
TOTAL COUNTS UNDER
5.65 MEV PEAK
COUNTS UNDER 5.65 MEV PEAK
AFTER SUBTRACTING MAXIMUM
POSSIBLE TAILING FROM At211

FIG. 5

14865-1

Figure 6

Decay of At Activities as Shown by Yields of Polonium Activities Obtained in a Milking Experiment

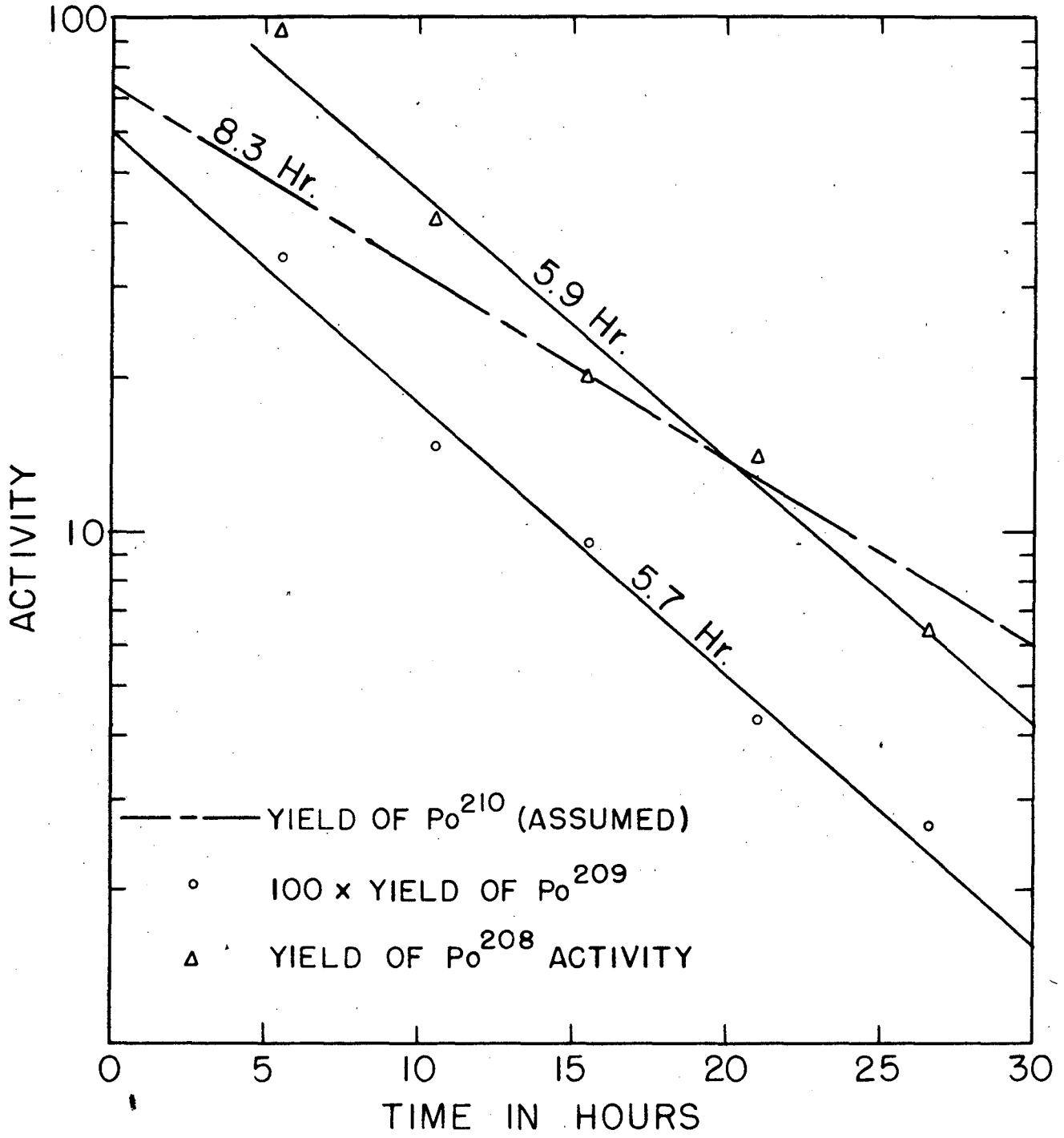

100
10
ACTIVITY
8.3 Hr.
5.9 Hr.
5.7 Hr.
YIELD OF Po210 (ASSUMED)
100 x YIELD OF Po209
YIELD OF Po208 ACTIVITY
0
5
10
15
20
25
30
TIME IN HOURS

FIG. 6

14866-1

Figure 7

Alpha Spectrum of One of the Polonium Milkings of Figure 6

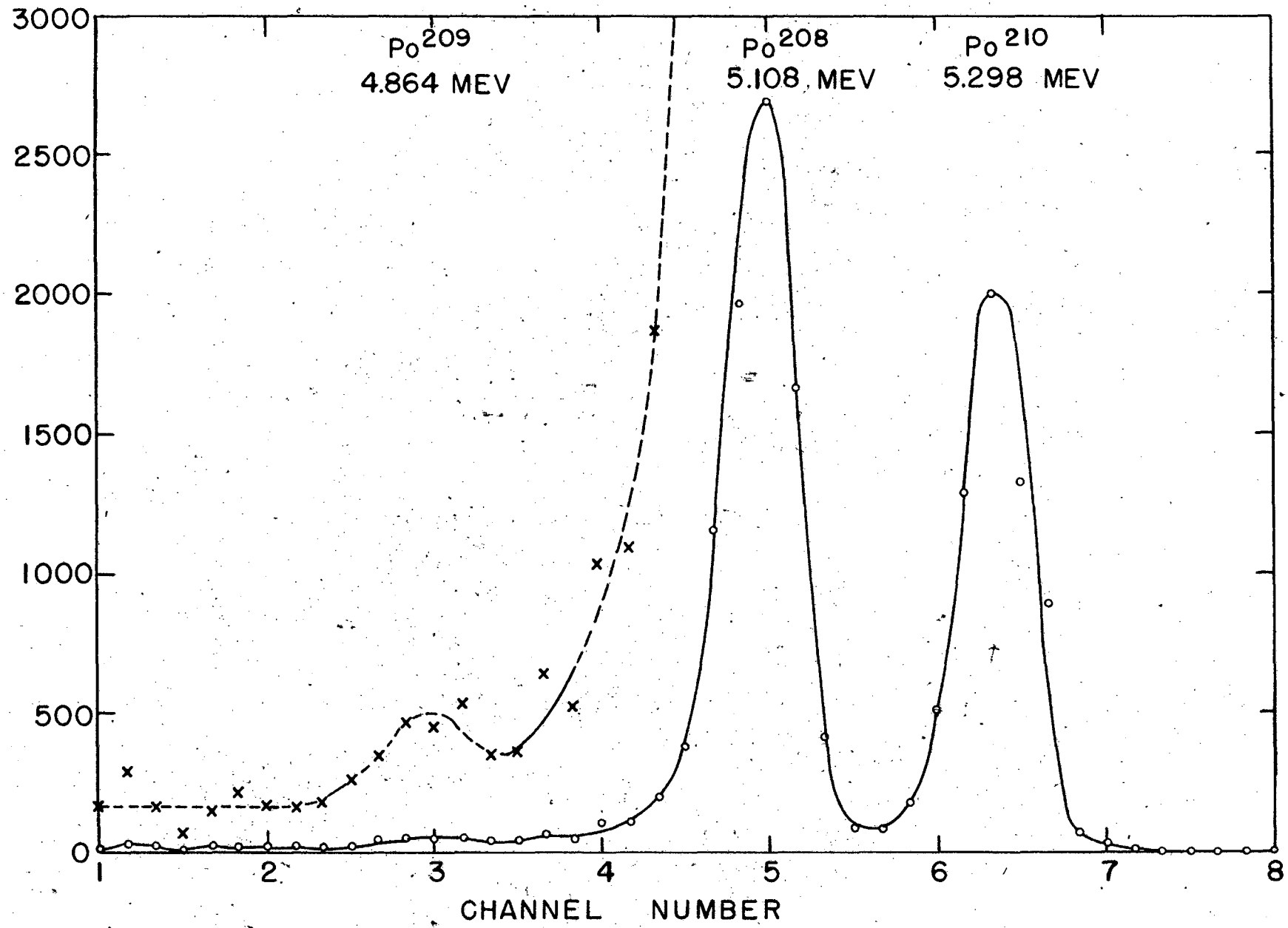

3000
2500
2000
1500
1000
500
0
1
2
3
4
5
6
7
8
CHANNEL NUMBER
Po209
4.864 MEV
Po208
5.108 MEV
Po210
5.298 MEV

FIG. 7

14869-1

Figure 8

Yield of Recoil Activities of 14-day Bismuth and 52-hr. Pb^{203} As a Function of Time from Astatine Prepared at 70 Mev

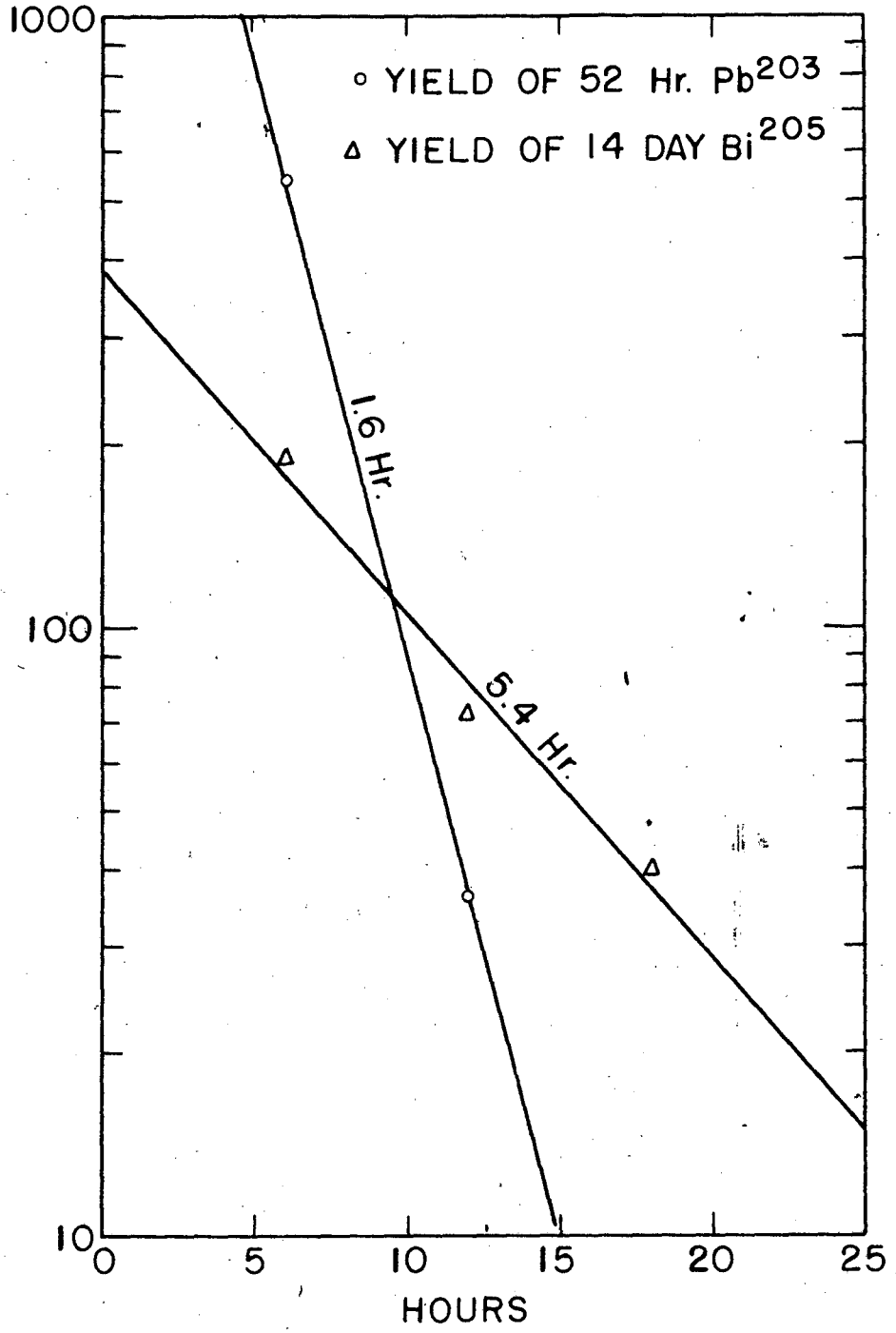

1000
100
10
0
5
10
15
20
25
HOURS
○ YIELD OF 52 Hr. Pb203
△ YIELD OF 14 DAY Bi205
1.6 Hr.
5.4 Hr.

FIG. 8

Figure 9

Yield of Chemically Identified 14-day Bismuth as a Function of Time From Astatine Prepared at 120 Mev

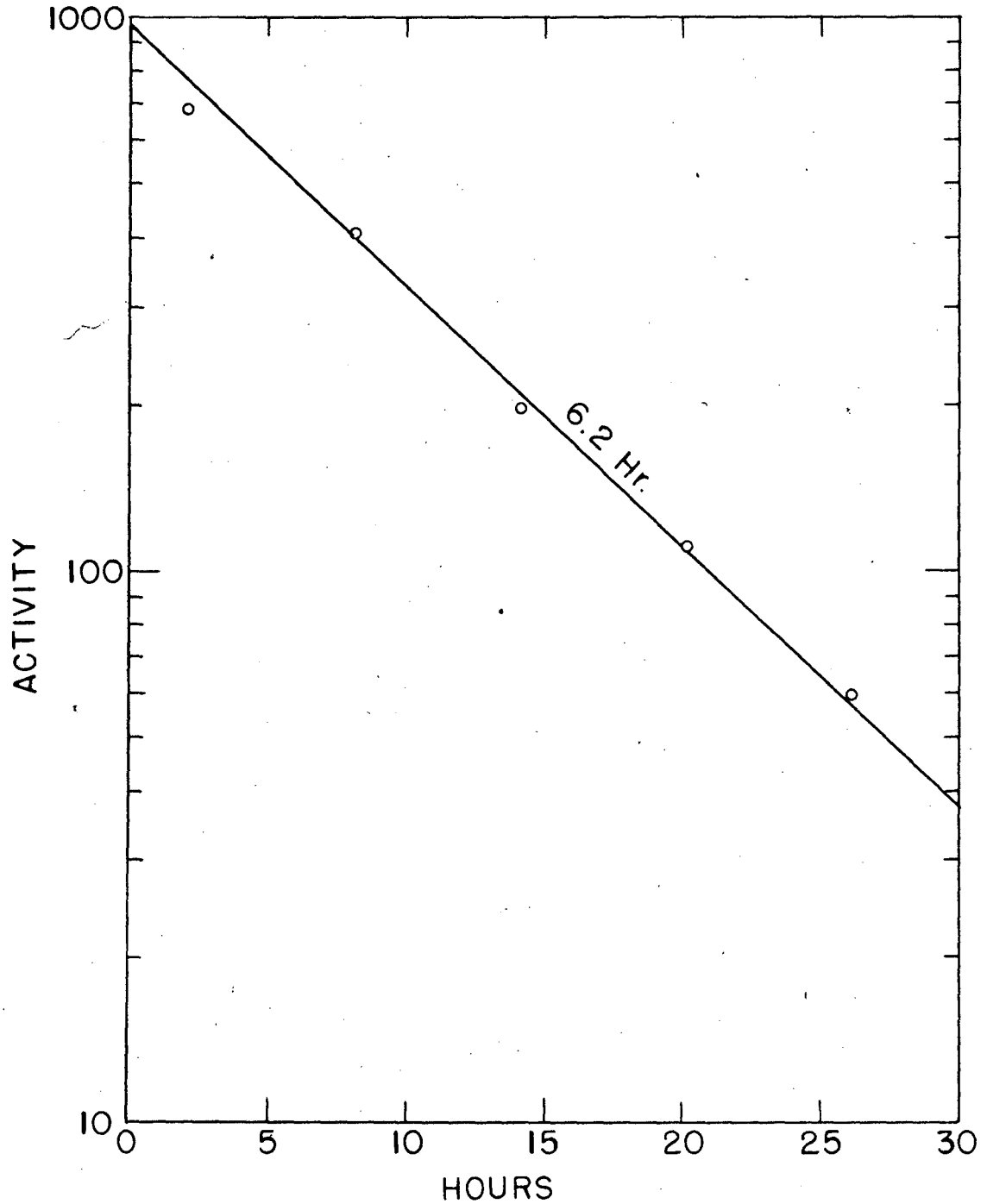


FIG. 9

alpha particles corresponding to this activity have been observed. Thus the branching ratio must be 1% or less. Absorption curves were taken of all these activities. Nothing which was distinctive was found.

Hyde[14] has reported a 1.7 hour activity with a 5.65 Mev alpha particle formed from the alpha decay of Fr^{212} at this mass number. We have been unable to definitely identify this activity in astatine formed from direct bombardment of bismuth since the sensitivity is poor because of tailing of alpha peak from 2.0 hr. At^{207} and the presence of 5.65 Mev At^{209}.

5. $\underline{At^{207}}$. Starting at 80 Mev, this activity appears in the astatine fraction. An alpha spectrum of astatine prepared at 90 Mev is presented in Figure 10. The decay of this activity as measured on the pulse analyser is shown in Figure 11. These results indicate a half-life of 2.0 hours and an alpha energy of 5.75 Mev.

On Figure 8 are also plotted the yield of 52 hr. activity, presumably Pb^{203}, [11] from the decay of Bi^{203} caught as recoils. This result, although only two points had a perceptible amount of Pb^{203}, indicates that this activity is probably correctly assigned to At^{207}. A chemical experiment was performed in which a mixed fraction of lead, bismuth and polonium was milked at two hour intervals from astatine prepared at 110 Mev. The fractions were allowed to decay for two days before final separations were made so that the 12 hr. Bi^{203} could decay to 52 hr Pb^{203}. Polonium and lead fractions were removed at this time and followed for decay. The yields of Pb^{203} were corrected for the varying times of growth allowed. These results are plotted in Figure 12. The yield of 52-hr. Pb^{203} is seen to drop off with a 1.9 hr. half-life. The 2.0 hour alpha group is thus established as At^{207}.

6. $\underline{At^{206}}$. The polonium fraction from the experiment of Figure 12 showed Po^{210}, Po^{208}, Po^{206} and perhaps Po^{209}. The yield of Po^{206} was measured by

Figure 10

Alpha Spectrum of Astatine Prepared at 90 Mev

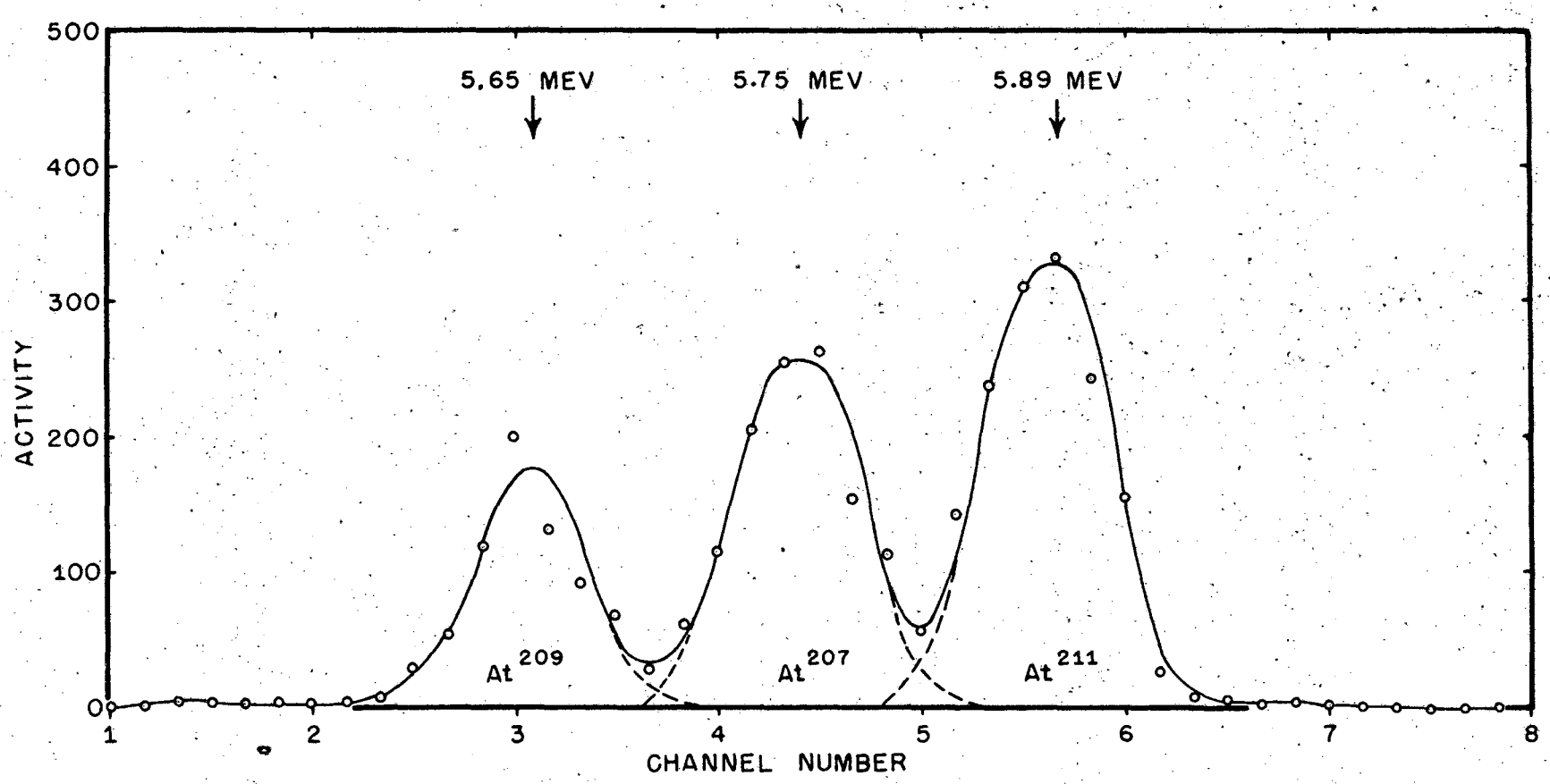


FIGURE 10

14872-1

Figure 11

Decay of the Sample of Figure 10 As Observed on the Pulse Analyser

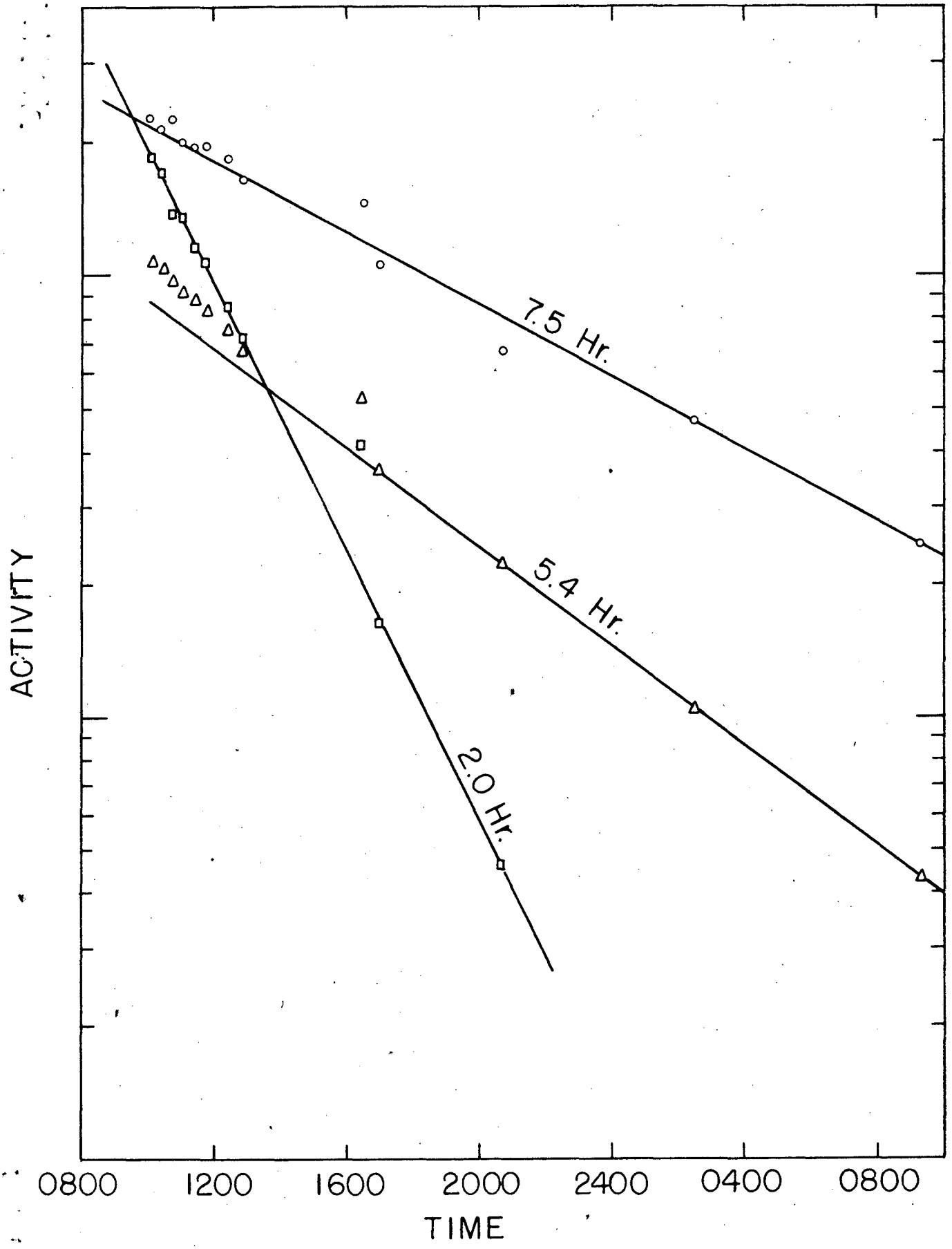


FIG. 11

14873-1

Figure 12

Yields of Pb^{203} and Po^{206} Observed in Milkings from Astatine Made by a 110 Mev Bombardment

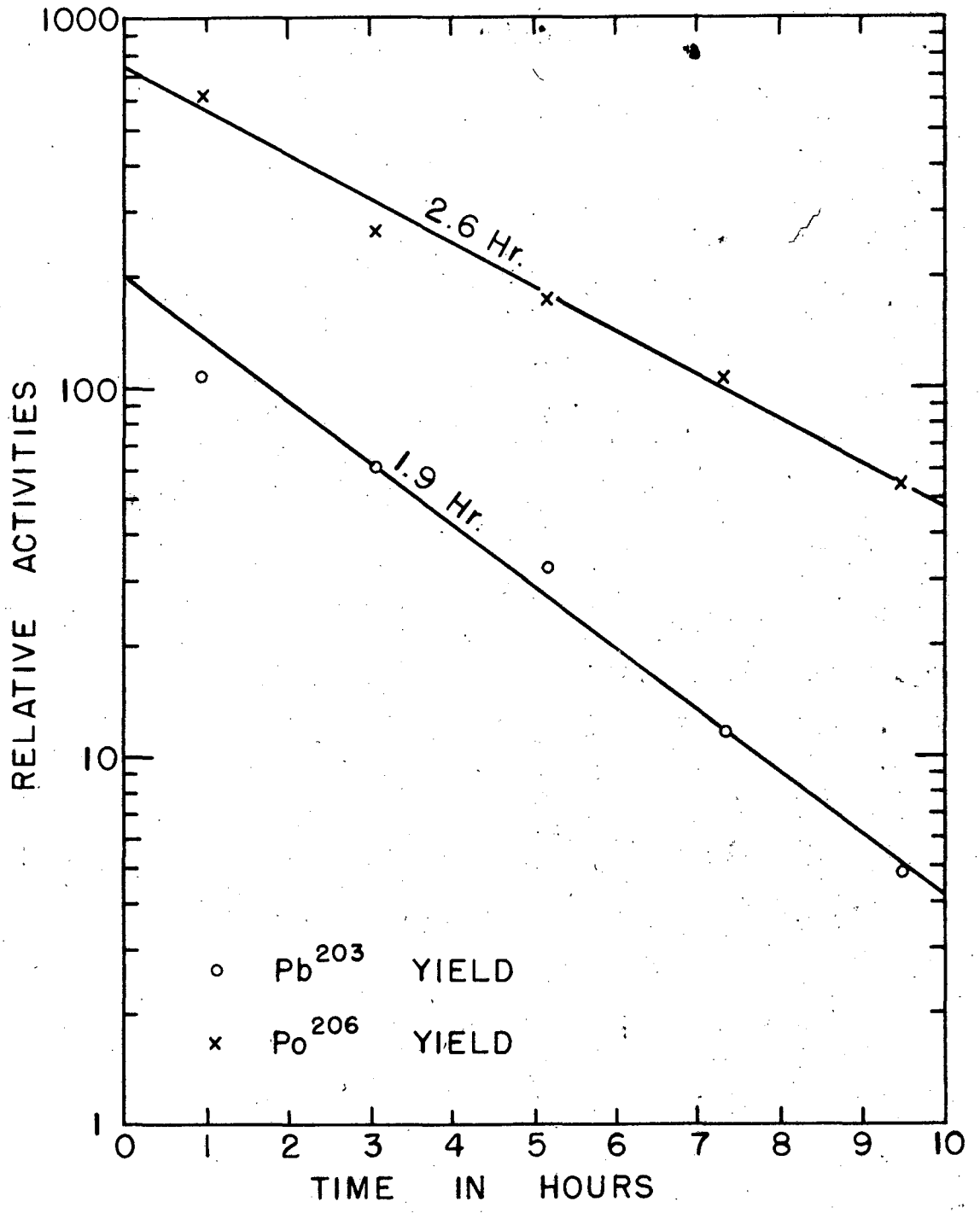


FIG. 12

14874-1

following the gross alpha decay of the milked polonium. The At^{206} polonium half-life indicated is 2.6 hr. No radiations have been identified as belonging to this isotope.

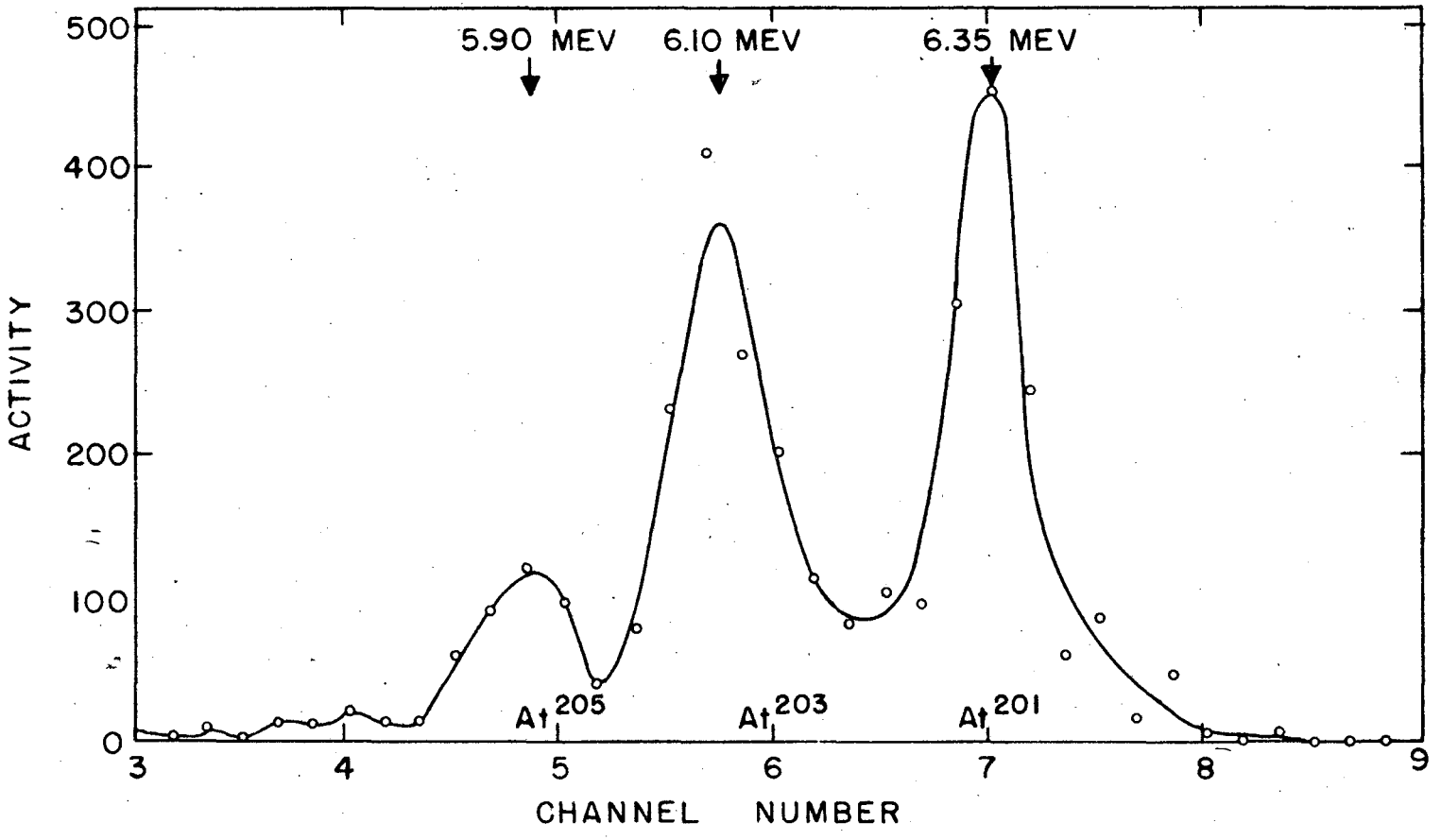
7. At^{205}. Figure 13 is the pulse analysis of astatine made from bismuth bombarded in the jiffy probe with 275 Mev helium ions. The decay of these activities is shown in Figure 14. Values are 5.90 Mev -- 23 min., 6.10 Mev -- 7 min, 6.35 Mev -- 1.7 min.

A milking experiment in which polonium from astatine prepared at 150 Mev was first separated at timed intervals, the polonium then allowed to grow bismuth for about 5 hours and the bismuth then separated out and plated is shown in Figure 15. Both the 12 hour Bi^{204} [12] and the 14 day Bi^{205} [10] were observed in this experiment. Both seem to have come from about a half-hour parent. The half lives are not different enough to allow us to decide which of the two is the 23-min alpha emitter.

As has been pointed out by Perlman, Ghiorso & Seaborg,[13] the observed lengthening in half-life from what is expected on the basis of the Gamow formula [15] using a smoothly varying function for the nuclear radius may be due to the existence of unpaired nucleons in the nucleus. These unpaired nucleons probably lie in the highest energy levels. As the highest energy levels are the one from which the alpha emission takes place, the odd nucleon must be paired with a nucleon from a lower level, which in turn must be removed from its partner. This mechanism decreases the probability of having nucleons in the appropriate configuration, two paired neutrons and two paired protons, for the emission of an alpha particle. If only one nucleon is unpaired, then the half life is lengthened by an amount, if two nucleons are unpaired the half life would be expected to be lengthened by a much greater amount.

Figure 13

Pulse Analysis of Astatine Prepared at 275 Mev on the Jiffy Probe of the Cyclotron



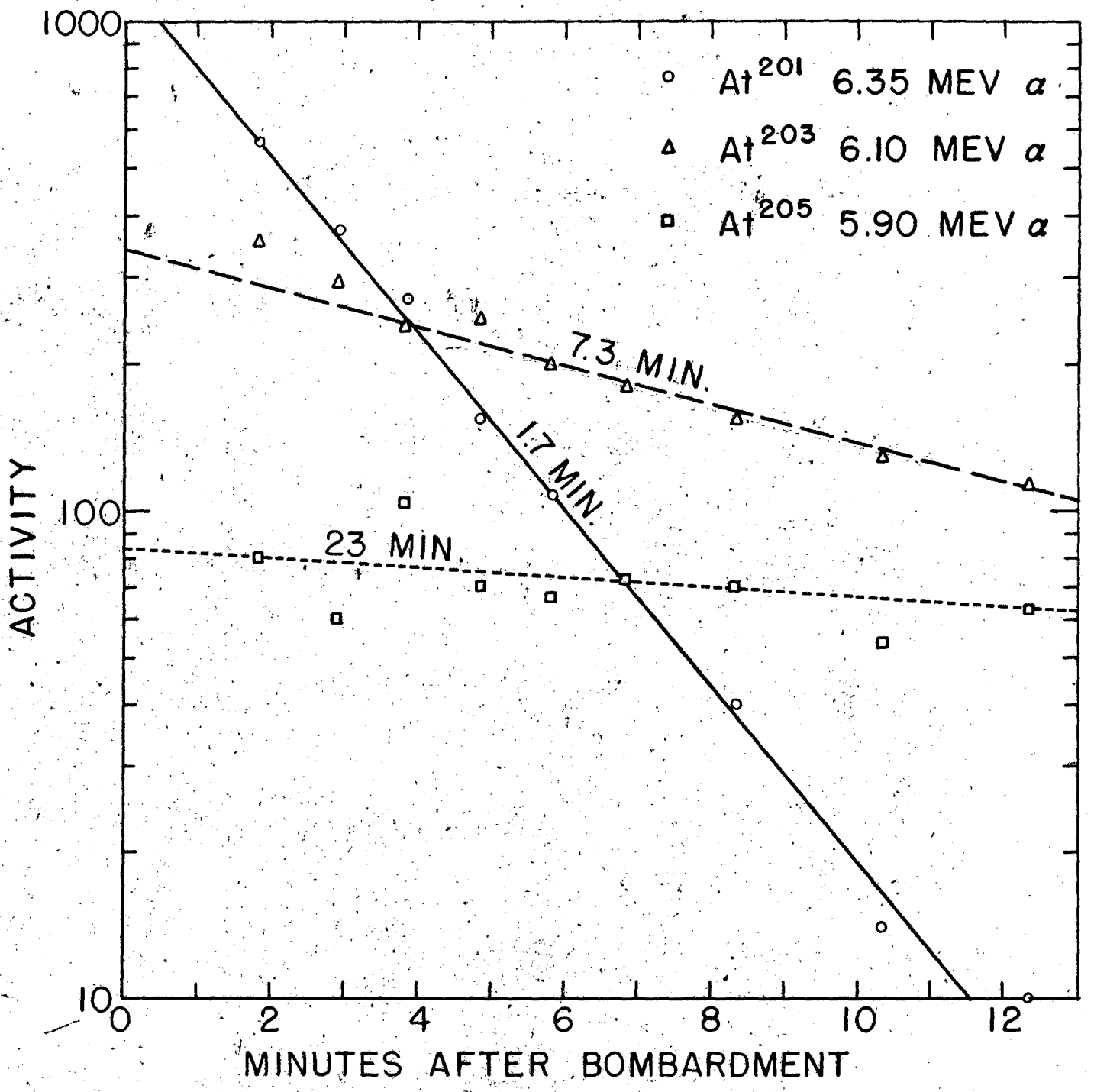
At FROM Bi + 250 MEV He^{++}

FIG. 13

14875-1

Figure 14

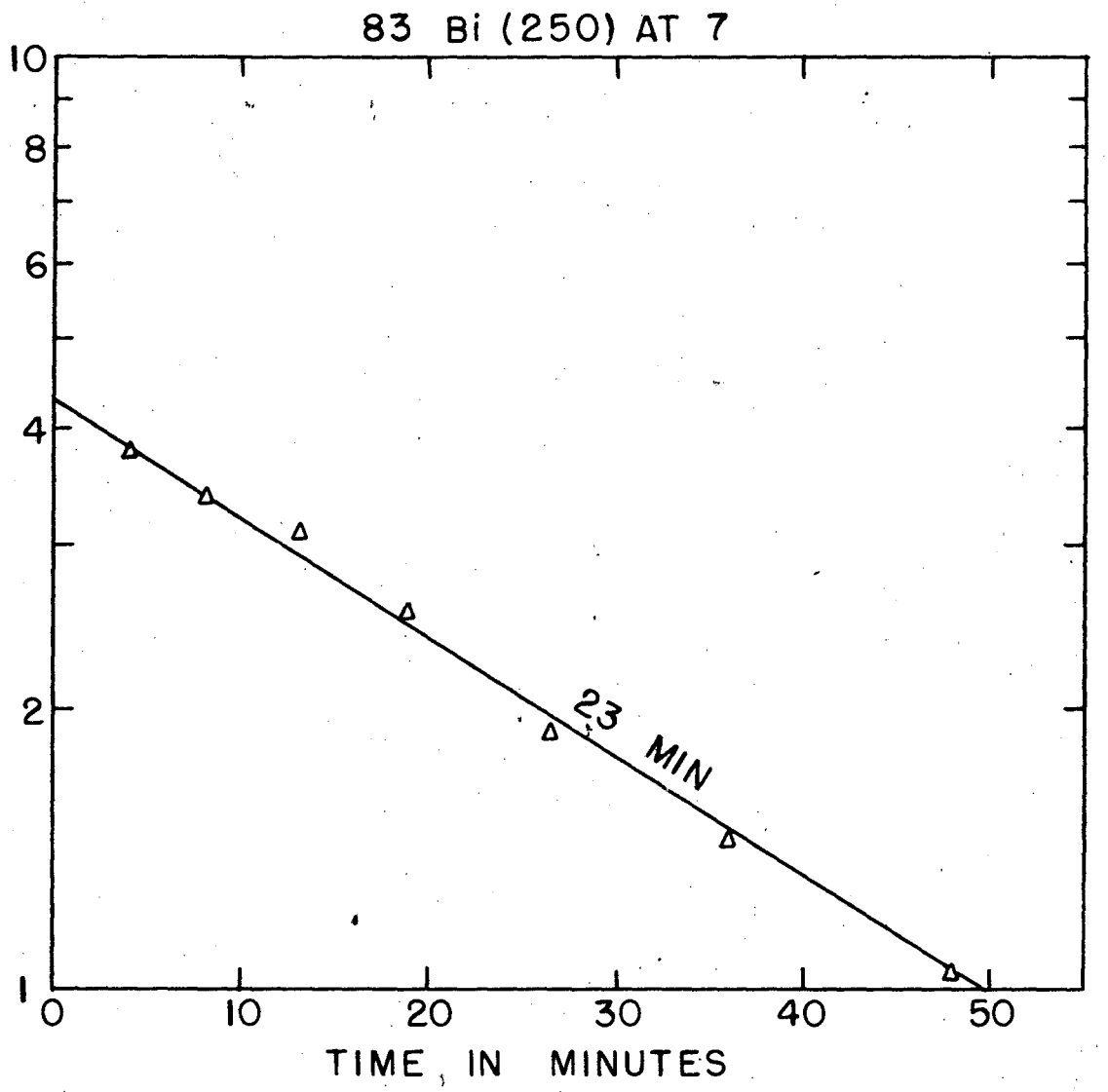
Decay of the Alpha Groups Shown in Figure 13



DECAYS FOR At FROM Bi + 275 MEV He++

FIG. 14

14876-1

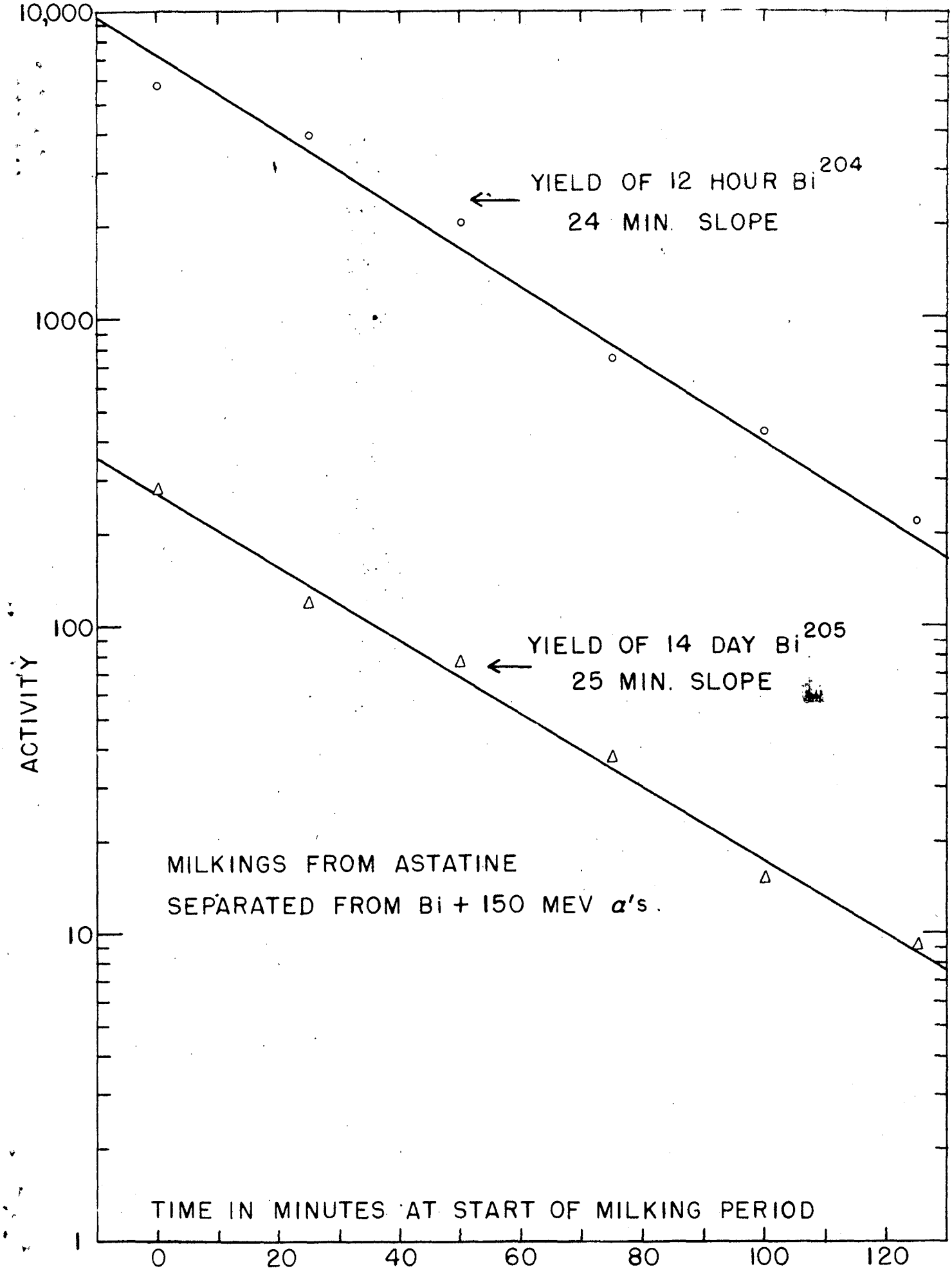


DECAY OF 5.90 MEV At^{205}

FIG. 14 a

Figure 15

Milkings of Polonium, and in Turn Bismuth, from Astatine Made at 150 Mev. Yields of Bismuth Activities Are Plotted


10,000
1000
100
10
1
ACTIVITY
YIELD OF 12 HOUR Bi204
24 MIN. SLOPE
YIELD OF 14 DAY Bi205
25 MIN. SLOPE
MILKINGS FROM ASTATINE
SEPARATED FROM Bi + 150 MEV α's.
TIME IN MINUTES AT START OF MILKING PERIOD
0
20
40
60
80
100
120

Inasmuch as the electron capture half lives are not affected by this phenomenon, alpha emission might be seen only from the odd-even isotopes, the ones which have the shorter alpha half lives. In addition, the results reported herein show that we have seen the alphas from only At^{211}, At^{209}, and At^{207}, all odd-even isotopes. Therefore we assign the observed 23 min alpha-group to At^{205}.

8. $\underline{At^{204}}$. From the reasoning set forth above, with regard to At^{205}, we must say that this isotope has a half life of 24 minutes, no measurable decay by alpha emission.

9. $\underline{At^{203}}$. There has been no chemical identification of the daughters from this isotope. As in Figure 14, there is an astatine activity of 7 minutes half-life. It is the next higher energy alpha group observed after exciting the 23 minute group, and it is observed at higher bombardment energy. The alpha energy is 6.10 Mev, as shown in Figure 13.

10. $\underline{At^{202}}$. We will assign none of the observed activities to this mass number.

11. $\underline{At^{201}}$. Also in the bombardment shown in Figure 13 is seen a 6.35 Mev alpha group. This decays (Fig. 14) with a 1.7 minute half life.

12. $\underline{At^{201}}$. One other astatine activity has been observed. This is a 6.50 Mev alpha particle, having a half-life of the order of 12 sec.

SUMMARY

In view of the above results we can make the following statements. At^{209} is a 5.5 hr. activity exhibiting alpha particles of 5.65 Mev energy. The branching ratio is not well known, but is of the order of 5%. Both the alpha decay and electron-capture daughter activities have been observed. At^{208} is a 6.3 ± 0.4 hr. activity having very small alpha branching. The

half life is known to at least 10%. At^{207} is a 2.0 hr. alpha emitter having 5.75 Mev alphas. This activity can only be confused with At^{206} which by milking Po^{206} has been shown to have a 2.6 hr. half life. At^{205} and At^{204} are both 23 minute activities as shown by milking experiments. A 5.90 Mev alpha is observed with this half life, and we somewhat arbitrarily assign it to At^{205}. At^{203} and At^{201} are both assigned on the basis that the observation of the alphas from only the odd mass numbered At^{207}, At^{209}, At^{211} implies that only the odd mass numbers are observed to alpha decay.

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