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ABSTRACT

Two new neutron deficient isotopes of polonium, mass 200 and mass 201 have been identified and improved measurements of the alpha particle energies of polonium isotopes mass 206 to 209, inclusive, are reported.

INTRODUCTION

Since 1946, eight isotopes of polonium from mass 202 to 209 have been reported.[1-3] In the course of further work, we have made improved measurements of the energies of the alpha particles emitted by several of these isotopes and by two others which have now been assigned to Po^{200} and Po^{201}.

Po^{201} AND Po^{200}

The new isotopes were first observed in pulse analysis[4] of polonium samples produced by bombardment of bismuth (as Bi_2O_3) with protons of energy 100 mev or more. Separation of the polonium was accomplished in about 30 minutes by dissolving the target in HCl, adjusting the acid concentration to

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*This work was performed under the auspices of the U. S. AEC.

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

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

3 D. G. Karraker and D. H. Templeton, Phys. Rev., in press.

4 Ghiorso, Jaffey, Robinson, and Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, "The Transuranium Elements: Research Papers," Paper No. 16.8 (McGraw-Hill Book Co., Inc., New York, N. Y., 1949).

approximately 6 molar, and extracting the polonium into 20 percent tributyl phosphate in dibutyl ether. The organic phase, containing the polonium, was purified by washing it several times with 6M HCl. Samples for alpha pulse analysis were prepared by evaporating a portion of the organic solvent on a platinum counting dish, then distilling a portion of the activity to a second disk. The chemical procedure serves to restrict the chemical identity of the product to either polonium or astatine. Since, after correction for decay, it was found that the ratio of the new activities to known polonium activities was not changed by partial volatilization, the new activities were positively identified as polonium.

The new alpha activities were shown by pulse analysis (Fig. 1) to have a 5.70 mev alpha particle group decaying with an 18 minute half-life, and a 5.84 mev group with a half-life of 11 minutes.

Mass assignments of these activities were made by successive separation of the electron capture daughter activities, where the yields of the end product of a decay chain serve to fix the half-life of the parent of the chain. A large quantity of polonium activity was prepared by bombardment of bismuth with 150 mev protons, and the polonium was separated and purified as previously mentioned. After a 10 minute interval for a growth of the bismuth electron capture daughters, the bismuth and lead were separated by washing the polonium (in tributyl phosphate solution) with 6M HCl. Five separations were done in this manner. The bismuth activities were allowed to decay for 24 hours, which should take Bi^{201} and Bi^{200} through their lead daughters to the Tl^{201} and Tl^{200} members of the electron capture chain (Fig. 2).

The thallium was separated by extraction with diethyl ether after oxidation with potassium permanganate. The decay of the thallium separated from each of the five bismuth fractions was followed. The yields of Tl^{200} and Tl^{201}

indicate that the polonium parent of the mass 200 chain has a half-life of 14 minutes, while the parent of the mass 201 chain is 21 minutes in half-life (Fig. 3). Thus, the 11 minute alpha emitter of 5.84 mev energy is identified as mass 200, and the 18 minute alpha emitter of 5.70 mev is placed at mass 201.

The timing of the experiments is such that it does not matter whether Po^{201} decays to one or both of the isomers of Bi^{201}. We have not attempted to determine this part of the decay scheme.

The discrepancy between the half-lives determined by chemical separation and the half-lives determined by alpha counting is to be expected, since the alpha decay of the Po^{205} and Po^{204} known to be present in the polonium fraction contributes to the yield of Tl^{200} and Tl^{201} and increases the half-lives determined by chemical separations. The half-lives determined by alpha counting are considered to be the correct values.

MEASUREMENT OF ALPHA PARTICLE ENERGIES

Previously published values[1,2] for the alpha particle energies of Po^{206}, Po^{207}, Po^{208}, and Po^{209} were measured on the 48-channel differential pulse analyzer[4] before improvement of techniques and the apparatus made possible the precision of the measurements reported here. Some of the increased resolution is due to care taken to produce thin, uniform samples, but improved design of the ionization chamber and the preamplifier are mainly responsible for the improvement. At the present time, the precision of a measurement is about 10 kev, with the accuracy estimated as about 20 kev.

Energies are obtained from the pulse analyzer by a standardization of the relation of the output pulse height to the energy of the alpha particle causing the pulse. This relation is very linear over an energy range of 2 mev or so, and the energy calibration was determined using as primary standards alpha emitters whose alpha energies have been previously determined by alpha ray spectroscopy. The primary standards used for these polonium isotopes were Po^{210} (alpha energy 5.298 mev),[5] Ra^{226} (alpha energies 4.795 mev (93.5 percent) and 4.611 mev (6.9 percent)),[6] Ra^{224} (alpha energy 5.681 mev),[7] and Th^{228} (alpha energy 5.418).[8] The pulse analysis curves for Ra^{226} do not agree with the presently accepted intensities of the two groups. The values determined in this work are 95.2 percent for the 4.795 mev group and 4.8 percent for the 4.611 mev group (Fig. 4).

The polonium samples used for measurement were produced by deuteron and proton bombardment of bismuth. Polonium was separated as described above. Plates for pulse analysis measurements were prepared by distilling the polonium from one platinum plate to another platinum plate about one-half inch distant in air. A collimator restricted the activity on the second plate to an area 15 mm in diameter.

The energy calibration was accomplished by measuring the ratio of energy to channel width for the region of 5.418 to 5.681 mev, using Th^{228} and Ra^{224} as standards (Fig. 5). The same ratio was then measured in the region 4.795 to 4.611 mev, with Ra^{226} as the standard (Fig. 4). The values determined were 22.8 kev/channel at the high energy range, and 24.2 kev/channel in the low energy range. The mean value of 23.5 kev/channel was used in the determination of the alpha energies of Po^{208} and Po^{209}. A sample containing Po^{210}, Po^{208}, and Po^{209} was then run on the pulse analyzer. The Po^{210} was used as an absolute energy standard, and the number of channels separation of Po^{208} and Po^{209} from Po^{210} was measured. Using the value of 23.5 kev/channel, the energy of

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[5] M. G. Holloway and M. S. Livingston, Phys. Rev. 54, 18 (1938).

[6] Rosenblum, Guillot, and Bastin-Scoffier, Compt. rend. 229, 191 (1949).

[7] G. H. Briggs, Proc. Roy. Soc. A 157, 183 (1936).

[8] W. B. Lewis and B. V. Bowden, Proc. Roy. Soc. A 145, 235 (1934); recalculated according to reference 5.

Po^{208} was determined as 5.103 mev and the energy of Po^{209} as 4.865 mev (Fig. 6). The uncertainty in energy from the calibration amounts to 6 kev for Po^{208} and 13 kev for Po^{209}. With the energy of Po^{208} now standardized, it was used, with Po^{210}, as a standard for the determination of the 9 day Po^{206} as 5.21 ± 0.02 mev (Fig. 7). Po^{207}, which has a 5.7 hour half-life and extremely low alpha branching ratio ($\sim 10^{-4}$), was a more difficult problem, but samples were prepared by proton bombardment of thin bismuth foils which were about one-half Po^{207} by alpha activity. Pulse analysis (Fig. 8) of these samples showed that the Po^{207} alpha particles were identical in energy with those of Po^{208}, within experimental error. The results of these measurements, together with some values from other sources as indicated, are listed in the table below. Also tabulated are the decay energies, which include the recoil energy of the daughter.

Table I

Isotope	Half-Life	Alpha Particle Energy (mev)	Alpha Decay Energy (mev)
Po^{200}	11 min	5.84 ± 0.03	5.96
Po^{201}	18 min	5.70 ± 0.03	5.82
Po^{202}	52 min	5.59 ± 0.03[a]	5.70
Po^{203}	47 min	(not yet observed)	
Po^{204}	3.8 hr	5.37 ± 0.02[a]	5.48
Po^{205}	1.5 hr	5.21 ± 0.10[ab]	5.31
Po^{206}	9 days	5.21 ± 0.02	5.31
Po^{207}	5.7 hr	5.10 ± 0.02	5.20
Po^{208}	2.93 yr	5.10 ± 0.02	5.20
Po^{209}	~200 yr	4.86 ± 0.02	4.95
Po^{210}	138 days	5.298[c]	5.40

[a]Reference 3; [b]Value changed slightly because of new value for Po^{206}; [c]Reference 5.

The alpha decay energies of the polonium isotopes listed in Table I are plotted against mass number in Fig. 9. The points for even-even nuclei fall very well on a smooth curve with little curvature and small slope in the region below the abrupt change associated with the effect of 126 neutrons. Of the four even-odd isotopes, two and probably a third are below this curve. This greater regularity of the even-even nuclei and the negative deviations of even-odd isotopes with regard to alpha decay energy can be noted in most of the curves for even elements.[9] Of these polonium isotopes only Po^{209} deviates more than 0.1 mev from the curve, and the slope of the curve is relatively small; consequently, it is expected that estimates of the alpha decay energies of Po^{203}, Po^{199}, Po^{198}, etc., obtained by interpolation and extrapolation of this curve are rather reliable.

The polonium isotopes have abnormally small alpha decay rates compared to heavier elements, if their nuclear radii are assumed to be normal. Perlman and Ypsilantis[10] have calculated the deviations of radius necessary to account for the decay rates of some of the even-even polonium isotopes, and have shown that the deviations have some regularity, with especially large deviations at Po^{210} and Po^{208}. Our data indicate that the lighter isotopes continue to have these reduced radii, but the alpha decay rates are difficult to determine because the decay of these isotopes is primarily by electron capture. Our present values are based on uncertain estimates of electron capture counting efficiencies or on guesses of cross sections and are not of sufficient accuracy to warrant presentation at this time. In several of these cases the alpha branching fraction and thereby the alpha decay rate can be obtained more rigorously by calibrating the electron capture counting efficiency against another alpha decay

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[9] Cf. Fig. 1, Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).

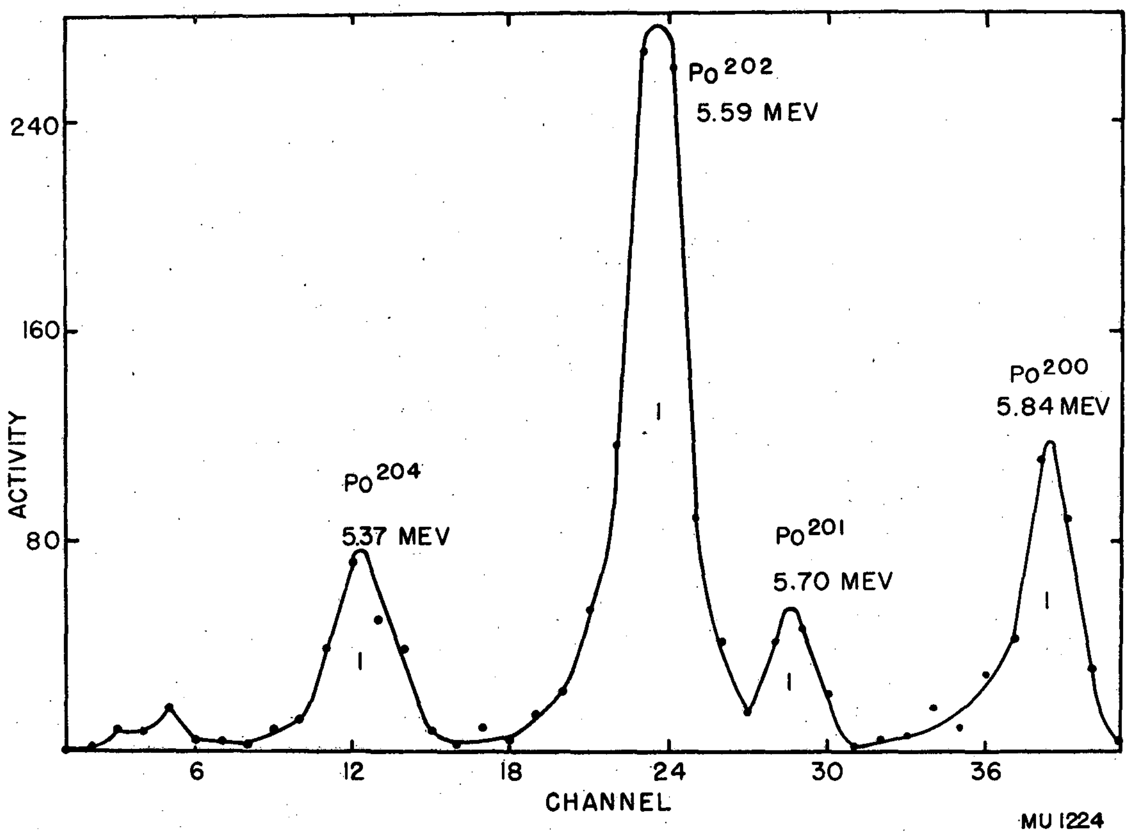
[10] I. Perlman and T. J. Ypsilantis, Phys. Rev. 79, 30 (1950).

process, such as of an astatine parent of a related bismuth radioactivity. Such experiments are in progress in this laboratory.

We are indebted to Mr. J. T. Vale, Mr. G. B. Rossi, and the cyclotron crews who carried out the irradiations on the 184-inch and 60-inch cyclotrons.

LIST OF ILLUSTRATIONS

Figure



ACTIVITY
240
160
80
6
12
18
24
30
36
CHANNEL
Po 202
5.59 MEV
Po 204
5.37 MEV
Po 201
5.70 MEV
Po 200
5.84 MEV
MU 1224

Fig. 1

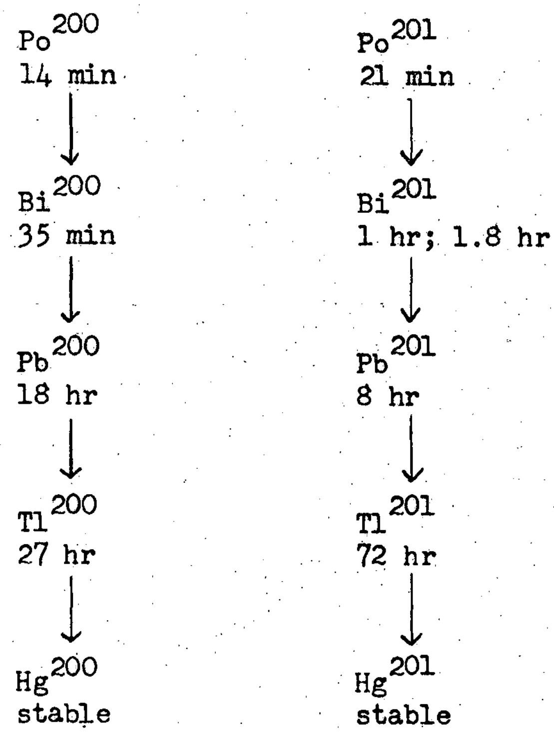

Po^{200}
14 min
Bi^{200}
35 min
Pb^{200}
18 hr
Tl^{200}
27 hr
Hg^{200}
stable
Po^{201}
21 min
Bi^{201}
1 hr; 1.8 hr
Pb^{201}
8 hr
Tl^{201}
72 hr
Hg^{201}
stable

Fig. 2

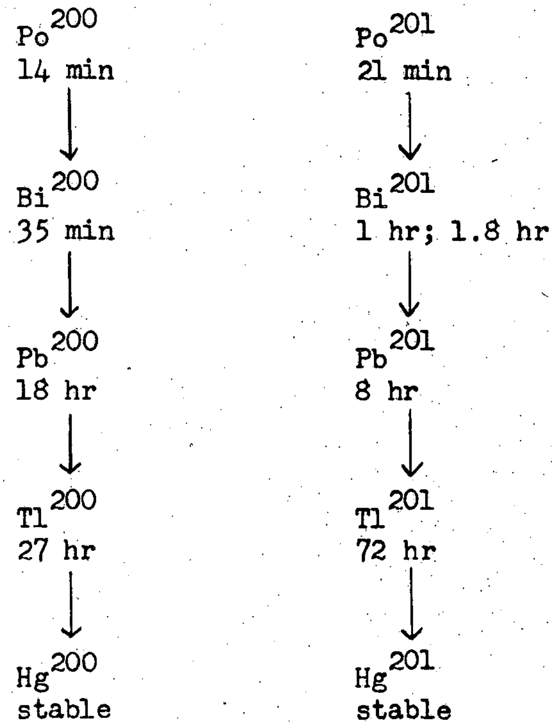

Po^{200}
14 min
Bi^{200}
35 min
Pb^{200}
18 hr
Tl^{200}
27 hr
Hg^{200}
stable
Po^{201}
21 min
Bi^{201}
1 hr; 1.8 hr
Pb^{201}
8 hr
Tl^{201}
72 hr
Hg^{201}
stable

Fig. 2

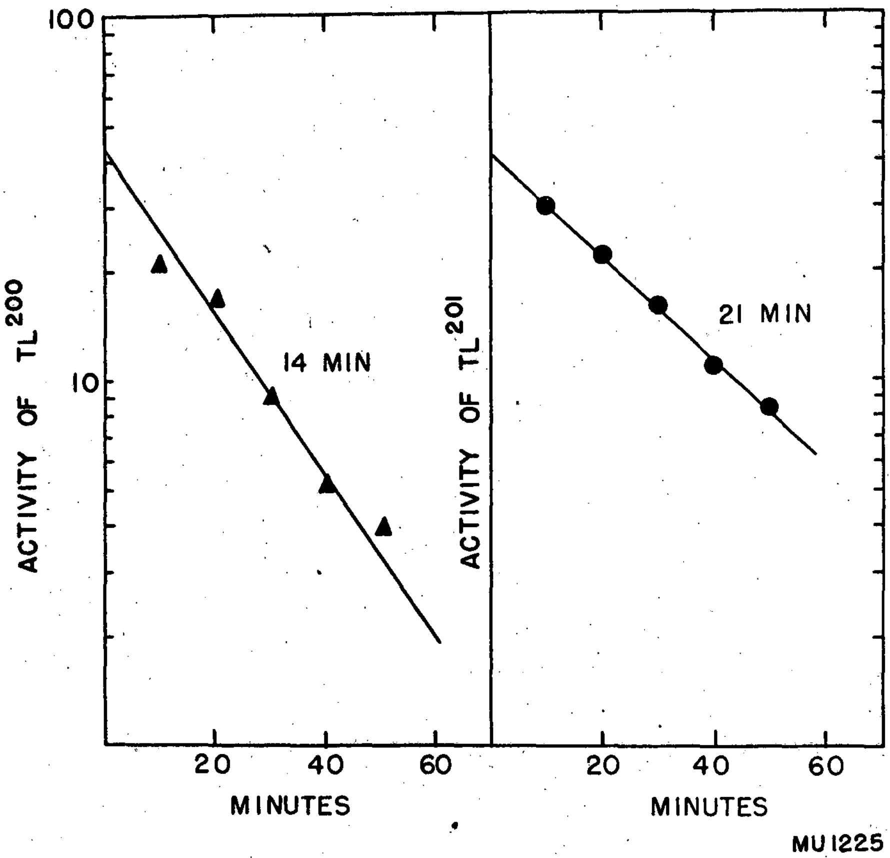

100
10
ACTIVITY OF TL200
14 MIN
20
40
60
MINUTES
ACTIVITY OF TL201
21 MIN
20
40
60
MINUTES
MU 1225

Fig. 3

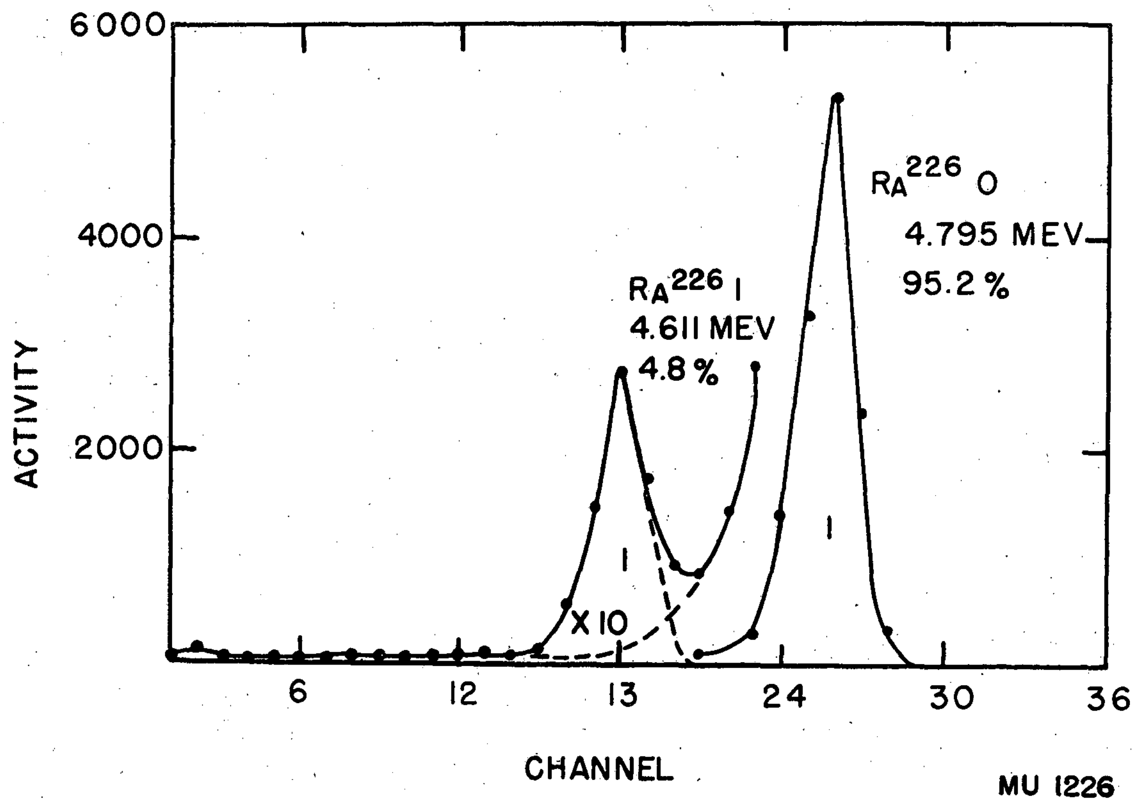
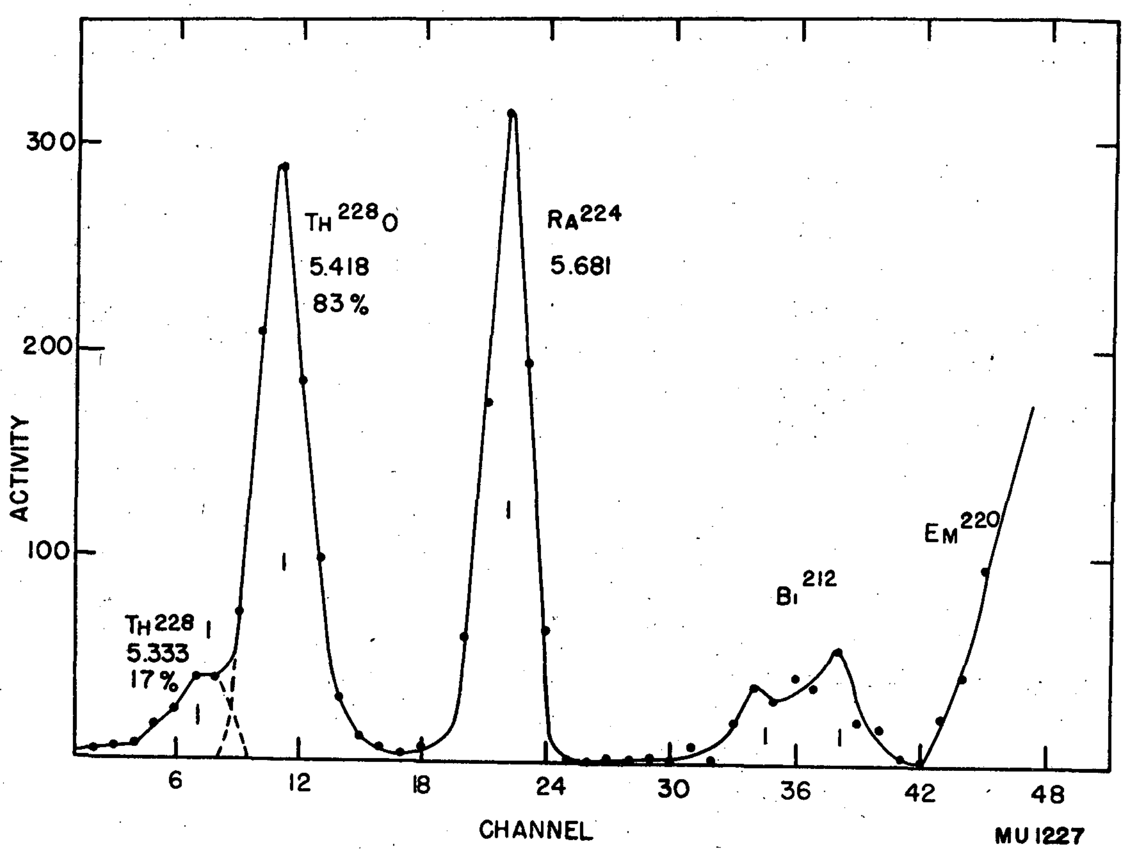

6000
4000
2000
ACTIVITY
RA226 1
4.611 MEV
4.8 %
RA226 0
4.795 MEV
95.2 %
X10
6
12
13
24
30
36
CHANNEL
MU 1226

Fig. 4



ACTIVITY
300
200
100
TH 228 O
5.418
83 %
RA 224
5.681
TH 228
5.333
17 %
BI 212
EM 220
6
12
18
24
30
36
42
48
CHANNEL
MU 1227

Fig. 5

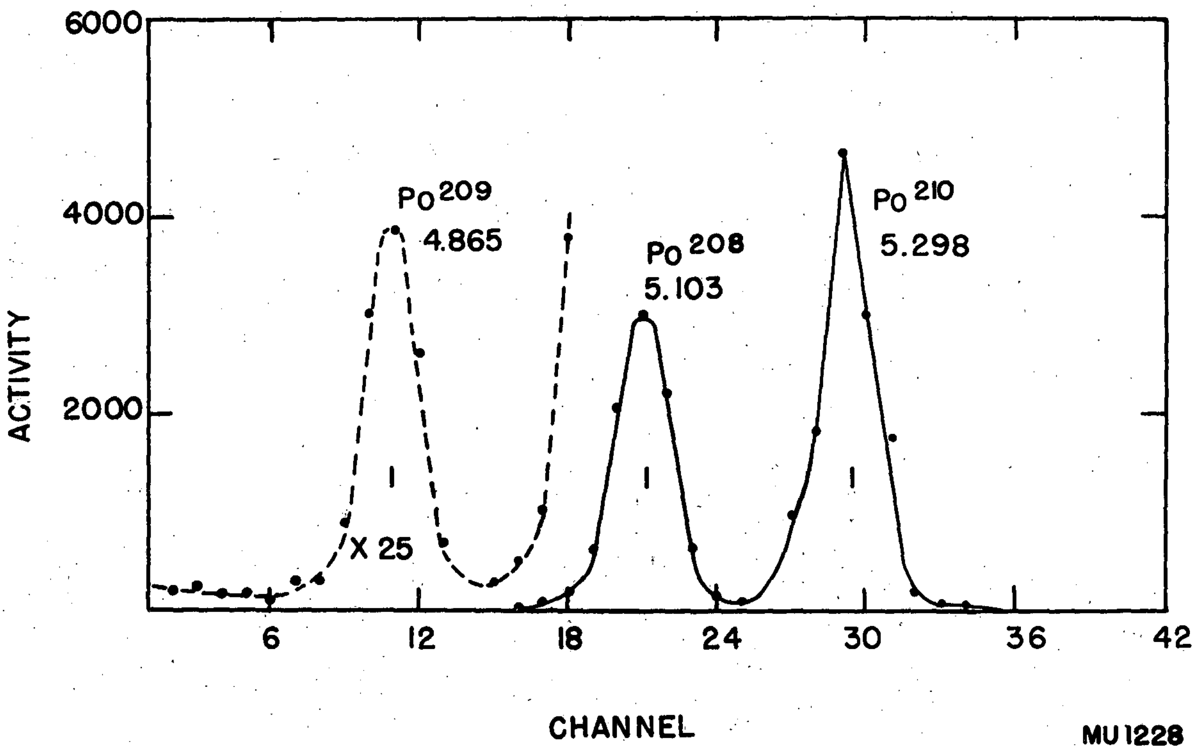
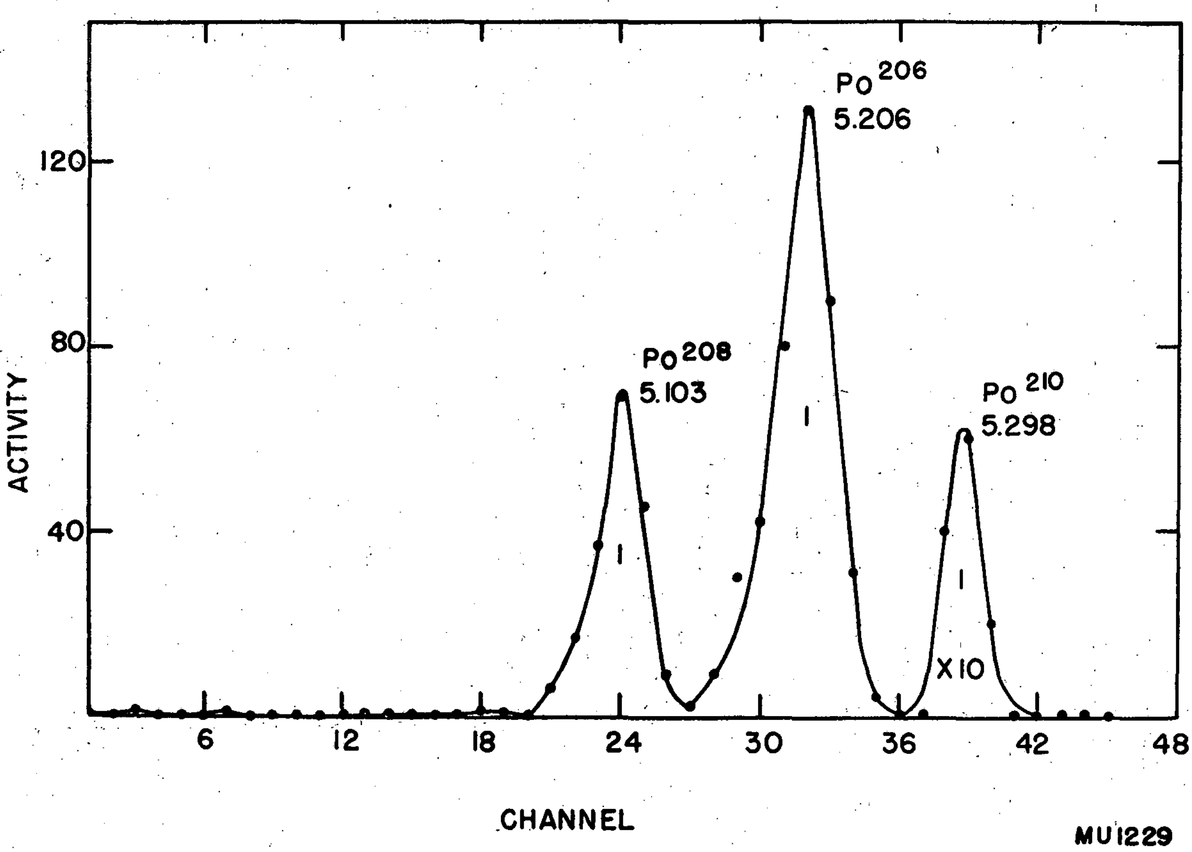

6000
4000
2000
ACTIVITY
Po 209
4.865
Po 208
5.103
Po 210
5.298
X 25
6
12
18
24
30
36
42
CHANNEL
MU 1228

Fig. 6



Po 206
5.206
Po 208
5.103
Po 210
5.298
X10
120
80
40
ACTIVITY
6
12
18
24
30
36
42
48
CHANNEL
MU1229

Fig. 7

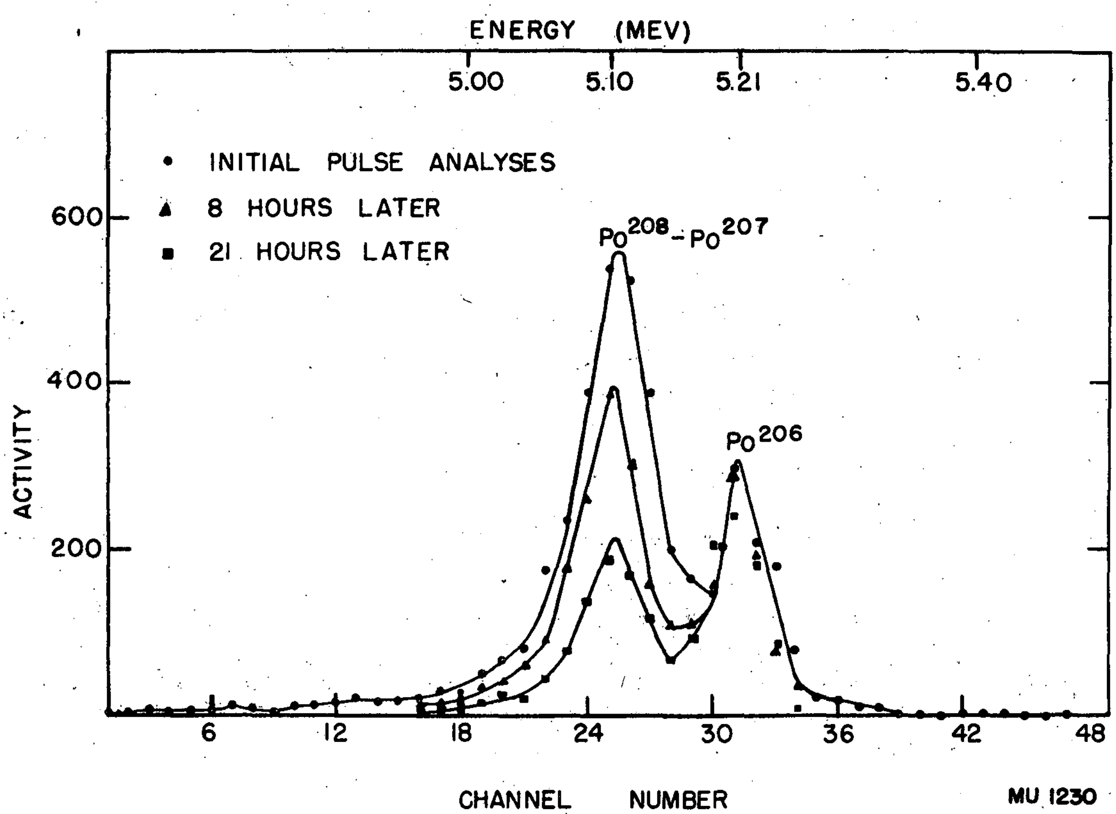

ENERGY (MEV)
5.00
5.10
5.21
5.40
INITIAL PULSE ANALYSES
8 HOURS LATER
21 HOURS LATER
Po208 - Po207
Po206
600
400
200
ACTIVITY
6
12
18
24
30
36
42
48
CHANNEL NUMBER
MU 1230

Fig. 8

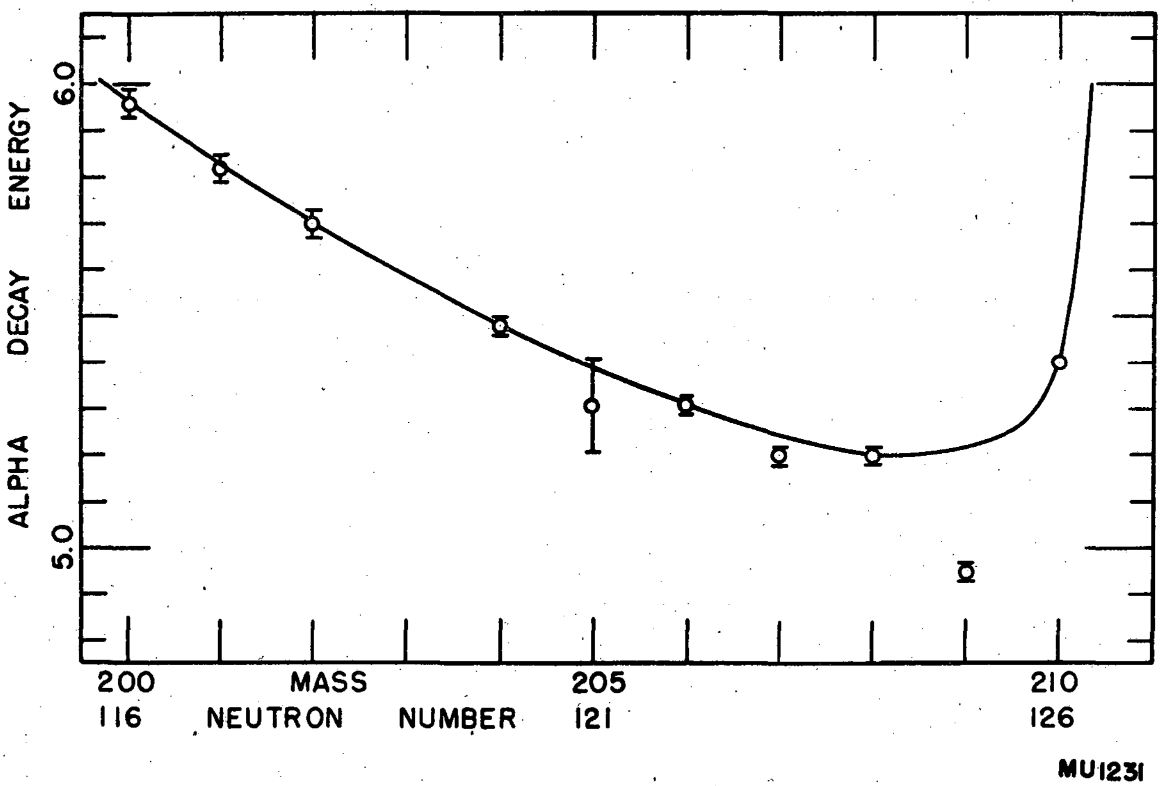

6.0
5.0
ALPHA DECAY ENERGY
200
116
MASS
NEUTRON NUMBER
205
121
210
126
MU1231

Fig. 9