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DECAY PROPERTIES OF Pu^{235}, Pu^{237}, AND A NEW ISOTOPE Pu^{233} *

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ABSTRACT

Electron capture and alpha decay properties of Pu^{237}, Pu^{235}, and the new isotope Pu^{233} have been measured. The overall half lives are 44±2 days for Pu^{237}, 26±2 minutes for Pu^{235}, and 20±2 minutes for Pu^{233}. Two alpha groups, one of 5.65±0.02 Mev and one of 5.36±0.02 Mev, were detected in the decay of Pu^{237}, one group of 5.85±0.02 Mev in the decay of Pu^{235}, and one of 6.30±0.02 Mev in the decay of Pu^{233}. The partial alpha half lives corresponding to these alpha groups are, for Pu^{237}, $(1.7\pm0.4) \times 10^4$ years and $(4.6\pm0.6) \times 10^3$ years, respectively; for Pu^{235}, (1.7±0.4) years; and for Pu^{233}, 11±4 days. On the basis of the experimental data it has been possible to calculate hindrance facotrs for the alpha decay and log ft values for the electron capture decay of the three isotopes and to correlate their properties with the alpha and electron capture systematics.

*This work was performed under the auspices of the Atomic Energy Commission. It is based in part on work done by T. Darrah Thomas and Robert Vandenbosch in partial fulfillment of the requirements for the Ph.D. degree at the University of California.

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I. INTRODUCTION

The odd-mass neutron-deficient isotopes of plutonium have received little investigation since the initial discovery experiments; in fact, Pu^{233} was heretofore undiscovered, although Pu^{232} and Pu^{234} have been known for some time. Plutonium-235 was first observed by Orth and Street,[1] who identified it as an isotope decaying primarily by electron capture with an overall half-life of 26 minutes. Orth[2] detected alpha particles of 5.85 Mev energy which he attributed to the alpha decay of Pu^{235} and estimated a partial alpha half life of about two years. James, Florin, Hopkins, and Ghiorso[3] first identified Pu^{237}, which was found to decay by electron capture with a half life of about 40 days. A study of the gamma spectrum and electron capture decay of Pu^{237} was made by Hoff,[4] who measured a ratio of L to K electron capture of 0.88. Kalkstein[5] has recently redetermined the L to K electron capture ratio to be 1.2.

In the present work, overall half lives, partial alpha half lives, and alpha decay energies have been redetermined or determined for the first time for these three isotopes. From these data it has been possible to calculate approximate values of the departure,[6] or hindrance,[7] factors for the alpha decay and log ft values for the electron capture decay of these nuclides and to correlate their properties with the alpha[6,7] and electron capture[8] systematics.

II. EXPERIMENTAL PROCEDURES

The three isotopes studied were produced with helium ions from the Crocker Laboratory 60-inch cyclotron by the reactions:

$$U^{233}\ (\alpha,4n)\ Pu^{233}$$

$$U^{233}\ (\alpha,2n)\ Pu^{235}$$

$$U^{235}\ (\alpha,4n)\ Pu^{235}$$

$$U^{235}\ (\alpha,2n)\ Pu^{237}.$$

The bombardments were carried out as part of a study of the excitation functions for reactions induced by helium ions in U^{233} and U^{235},[9] which in turn is part of a broad program of investigation of spallation-fission in the heaviest elements.[10]

For most of the experiments, uranium was electrodeposited[11] on a dish-shaped aluminum backing plate and bombarded in a microtarget assembly,[12] which served as a combined target holder and Faraday cup. For the production of Pu^{235} by the $(\alpha,4n)$ reaction on U^{235}, a 3-mil uranium metal foil was bombarded in a similar target assembly. The bombarded targets and aluminum cover and backing foils were dissolved in aqua regia and the plutonium was isolated by a precipitation and ion-exchange procedure.[12] The purified plutonium was electrodeposited[13] on a 2-mil platinum plate and the radiations were counted in a standard alpha-pulse-height analyzer. When the short-lived isotopes, Pu^{233} and Pu^{235}, were to be studied, the time for the chemical operations was kept to between an hour and an hour and a half.

The electron capture decay of Pu^{235} and Pu^{237} was detected by counting the Auger and conversion electrons accompanying the electron capture in a continuous-flow-methane proportional counter ("Nucleometer", Radiation Counter Laboratories, Inc., Skokie, Illinois), a windowless counter particularly sensitive to Auger electrons. The counting efficiency, or ratio of number of counts to number of disintegrations, of this instrument, which varies from nuclide to nuclide and appears to depend on the decay scheme and decay energy of the nuclide in question, has been a matter of considerable uncertainty. The problems involved and several determinations of the counting efficiency will be presented by Glass, Carr, Gibson and Cobble.[14] In the present experiments it was necessary to determine the counting efficiency of the counter for each isotope studied. In general, the most recently determined[14] values range from 60 to 95 percent, although lower values have been measured.[15] It was not feasible to make measurements on the electron capture decay of Pu^{233} because of the impossibility of resolving the activity due to the decay of Pu^{233} from that due to the decay of Pu^{235}, Pu^{232}, and Np^{233} (the daughter of Pu^{233}), all of which have similar half lives.

III. EXPERIMENTAL RESULTS

A. Plutonium-233. The alpha energy spectrum of the plutonium fraction isolated from U^{233} bombarded with 46-Mev helium ions showed a peak at the channels corresponding to 6.3 Mev, with an average on four experiments of 6.30 ± 0.02 Mev. One of the pulse analyses is shown in Fig. 1. The peak labelled

on the figure as Pu^{239} is due to Pu^{239} tracer which was added when the target was dissolved to determine the chemical yield. The other peaks are due either to plutonium activities produced by the U^{233} (α,xn) reactions or the U^{228} produced by the alpha decay of Pu^{232}. The peak corresponding to 6.3 Mev disappeared in successive pulse analyses with a half life of about 20 minutes. Individual values ranged from 17.5 to 24.1 minutes to give an overall half life of 20 ± 2 minutes. A typical decay curve is shown in Fig. 2.

The possibility was investigated that the observed activity might be due to contamination by Th^{226}, which has an alpha particle energy of 6.33 Mev and a half life of 31 minutes and which is a member of the Pu^{234} decay chain. However, a number of conclusive experiments showed that the amount of Th^{226} produced was not sufficient to account for the observed activity.

The assignment of this activity to the previously unobserved nuclide Pu^{233} is based primarily on three types of evidence: a rough excitation function, the appearance in the pulse analyses of alpha particles attributable to the U^{229} daughter of Pu^{233}, and the compatability of the alpha half life with the alpha decay systematics.[6,7]

Since it was not possible to measure the electron capture disintegration rate of Pu^{233} directly with the windowless proportional counter, Np^{233}, the electron capture daughter of Pu^{233}, was separated from an aliquot of the plutonium fraction by an extraction of neptunium-IV from aqueous solution into a benzene solution of thenoyltrifluoroacetone. The activity due to the electron capture of Np^{233} was measured in the proportional counter and from this measured activity and an assumed counting efficiency of 80% for the detection of electron capture disintegrations of Np^{233} the disintegration rate of Np^{233} was calculated. The electron capture disintegration rate of Pu^{233} was, in turn, calculated from the disintegration rate of Np^{233} and the extraction yield of Np^{237} tracer added to the plutonium before separation. The ratio of the alpha disintegration rate to the total disintegration rate, or alpha branching ratio, is $(1.2 \pm 0.5) \times 10^{-3}$, from which is calculated a partial alpha half life of 11 ± 4 days.

B. Plutonium-235. The values determined by Orth[1,2] of 26 ± 2 minutes for the overall half life and 5.85 ± 0.03 Mev for the alpha energy have been confirmed by the present work. A revised alpha branching ratio of $(3.0 \pm 0.6) \times 10^{-5}$ has been calculated using an electron capture counting efficiency of

70 percent. The figure 70 percent is based on an absolute disintegration rate determined by counting the K x-rays accompanying the electron capture with a 50-channel pulse height analyzer, using a thallium-activated sodium iodide crystal coupled to a photomultiplier tube as a detector. The number of K-electron capture events per minute was determined from the K x-ray counting rate, the known geometry and counting efficiency of the counter, and a K fluorescence yield (i.e., the ratio of the number of vacancies in the K-shell to the number of K x-rays emitted) of 0.97, which is the value determined by Gray[16] for uranium. To calculate the total number of disintegrations per minute it was necessary to know what fraction of the electrons captured were from the L or higher shells. On the basis of the theoretical work of Brysk and Rose,[17] the L electron capture to K electron capture ratio was estimated to be 0.23. It was assumed that capture from the M and higher shells is negligible. This counting efficiency is consistent with one which can be estimated from the amount of Np^{235} activity[9] produced from the electron capture decay of Pu^{235}. (No reliable value was obtained by the second method because of the extremely low counting rates of the Np^{235}.) The partial alpha half life calculated from the above branching ratio is 1.7 ± 0.4 years.

C. Plutonium-237. In the early experiments on Pu^{237}, James, Florin, Hopkins, and Ghiorso,[3] Hyde, Studier, and Ghiorso,[18] and James, Thompson, and Hopkins[19] reported only an approximate half life of 40 days and detected no alpha particles which could be attributed to this isotope. The overall half life has been more accurately determined in the present experiments to be 44 ± 2 days. Furthermore, low intensity alpha activity decaying with this half life has been observed in pulse analyses, appearing at the channels corresponding to 5.65 ± 0.02 Mev and 5.36 ± 0.02 Mev (See Fig. 3). The peak attributed to Pu^{239} is due to Pu^{239} tracer and the peaks attributed to Pu^{238} and Pu^{236} are due to the products of the U^{235} (α,n) and U^{235} $(\alpha,3n)$ reactions, respectively. The 5.65 Mev group accounts for 21 ± 4 percent of the alpha activity and the 5.36 Mev group for 79 ± 8 percent. The fact that the energies of the two alpha groups are separated by 300 kev suggests that the lower energy group is populating a 300 kev level in the daughter, U^{233}. A level of this energy has also been observed in U^{233} from the beta decay of Pa^{233}.[20]

The counting efficiency of the proportional counter for Pu^{237} was determined by two methods: by x-ray counting, giving a value of 77% and by a mass spectrometric technique, giving 81%. The x-ray counting experiment was similar to that on Pu^{235}. The K x-rays following the electron capture were counted and the disintegration rate was calculated from the K x-ray counting rate, the known geometry and counting efficiency of the counter, a K fluorescence yield of 0.97, and the estimated contribution of electron capture from the L and higher shells. Kalkstein's[5] value of 1.2 for the L to K electron capture ratio was used and the contribution of electron capture from the M and higher shells was assumed to be negligible. In the mass analysis experiment, a portion of a sample containing both Pu^{236} and Pu^{237} was mass analyzed to determine the ratio of the number of Pu^{237} atoms to the number of Pu^{236} atoms. The activity from another portion of the same sample was measured in the proportional counter, in an ionization chamber with a known efficiency for the detection of alpha disintegrations, and in an alpha-pulse-height analyzer. The fraction of the alpha activity which was due to Pu^{236} was determined from the pulse analysis. From this fraction, the alpha disintegration rate, and the half life of Pu^{236}, the number of atoms of Pu^{236} was calculated, and, from this figure and the atom ratio of Pu^{237} to Pu^{236} in the sample, the number of atoms of Pu^{237}. The electron capture disintegration rate of Pu^{237} was, in turn, calculated on the basis of the number of atoms and half life of Pu^{237}. Finally, the counting efficiency is equal to the ratio of the proportional counter activity to the disintegration rate. Calculation of the alpha branching ratios for the two observed alpha groups, using 79 percent for the electron capture counting efficiency of the proportional counter, gives $(2.6 \pm 0.3) \times 10^{-5}$ for the 5.36 Mev group and $(7.1 \pm 1.5) \times 10^{-6}$ for the 5.65 Mev group. These values correspond to partial alpha half lives of $(4.6 \pm 0.6) \times 10^{3}$ years and $(1.7 \pm 0.4) \times 10^{4}$ years, respectively.

The results for the three isotopes have been summarized in Table I.

IV. ALPHA AND ELECTRON CAPTURE SYSTEMATICS

In comparing partial alpha half lives, it is informative to calculate hindrance factors,[7] F, for the alpha decay. The results of such calculations for Pu^{233}, Pu^{235}, and Pu^{237} are given in Table I. A comparison of these

hindrance factors with those of other odd mass plutonium isotopes[7] shows that the hindrance factors for the most abundant alpha transition in these isotopes are all of the same order of magnitude. For Pu^{241}, F is 3.2, and for Pu^{239}, F is 2.9.

On a plot of alpha particle energy versus mass number, the alpha energies[7] of the most abundant transitions of the odd mass plutonium isotopes fall low relative to the line joining the points corresponding to the energies of their even mass neighbors. (See Fig. 4). If one plots, instead of the energy of the most abundant transition, the energy of what is presumed to be the ground state transition for Pu^{237} and Pu^{241}, the resulting points lie on the line joining the energies of the even mass isotopes. On the possibility that those isotopes whose highest known energy transition falls on the lower line have a higher energy transition, an experiment was performed to look for a possible higher energy alpha group in Pu^{235}. No such group was seen and it was possible to set an upper limit of 5 percent for the abundance of any alpha group in the energy range 5.5 to 6.5 Mev.

The electron capture half lives of Pu^{235} and Pu^{237} have previously been correlated with other electron capture half lives by Hoff and Thompson,[8] who calculated log ft values of 5.1 for Pu^{235} and 6.8 for Pu^{237}, using the assumption that the electron capture transitions were ground state transitions. From closed energy cycles[21,22] and the measured alpha decay energy for Pu^{233} one can calculate an electron capture decay energy of 2.08 Mev for Pu^{233}. If the assumption that most of the electron capture events in Pu^{233} proceed to the ground state is valid, the 2.08 Mev decay energy and 20 minute half life correspond to a log ft value of 5.6[17,23] indicating that the transition is allowed. The half life and decay energy correlate well with the half lives and decay energies of similar transitions[8] in other heavy elements. Using the values for the electron capture decay energies of Pu^{235} and Pu^{237}, calculated from their alpha decay energies by the method of closed cycles, the above values for the half lives, and the assumption of ground state transitions, log ft values of 5.2 for Pu^{235} and 6.5 for Pu^{237} have been calculated.

It is interesting to note that the electron capture decay of Pu^{237} is first forbidden and that the alpha-decay hindrance factor for what is presumed to be the ground state transition is relatively very high. On the other hand, in the case of both Pu^{233} and Pu^{235} the electron capture decay is allowed and the hindrance factors are low.

V. ACKNOWLEDGMENTS

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Table I

Isotope	Overall Half life	Alpha Decay: Partial Alpha Half life	Alpha Decay: Alpha Energy (Mev)	Alpha Decay: Hindrance Factor F	Electron Capture Decay: Calculated[a] Electron Capture Decay Energy (Mev)	Electron Capture Decay: Log ft
Pu^{233}	20 ± 2 m	11 ± 4 d	6.30 ± 0.02	3.1	2.08	5.6
Pu^{235}	26 ± 2 m	1.7 ± 0.4 y	5.85 ± 0.02	1.2	1.17	5.2
Pu^{237}	44 ± 2 d	$(1.7\pm0.4)\times10^{4}$ y	5.65 ± 0.02	1.1×10^{3}	0.22	6.5
		$(4.6\pm0.6)\times10^{3}$ y	5.36 ± 0.02	7.0		

[a] The methods of reference 22 together with data given in reference 21 and the energies given in the present work were used to calculate the electron capture decay energies.

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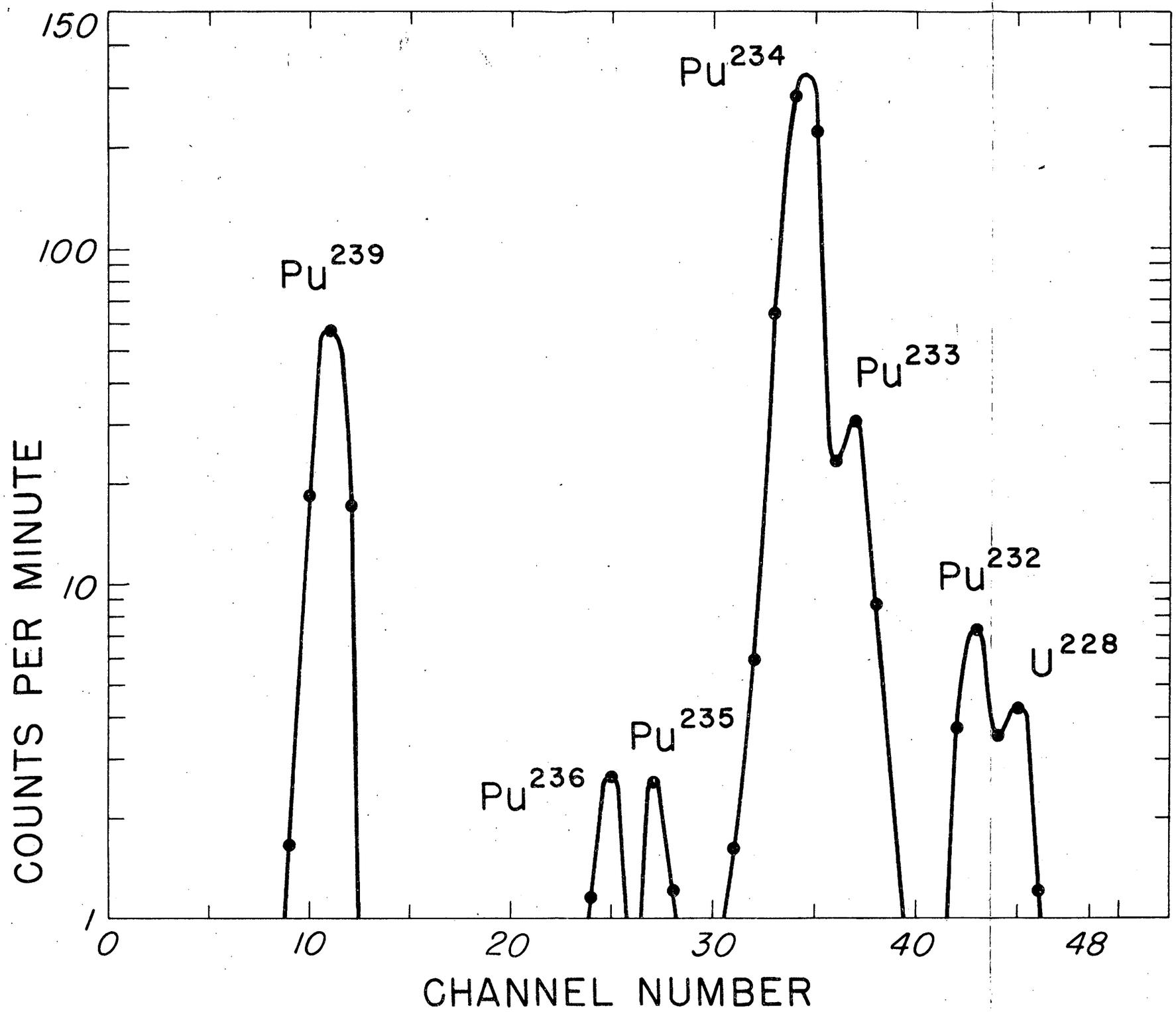
FIGURE 1. Pulse analysis showing alpha activity due to the decay of Pu^{239} and of the products from the U^{233} (α,xn) reactions.

FIGURE 2. Decay curve showing the decay of the pulse analysis peak corresponding to 6.3 Mev.

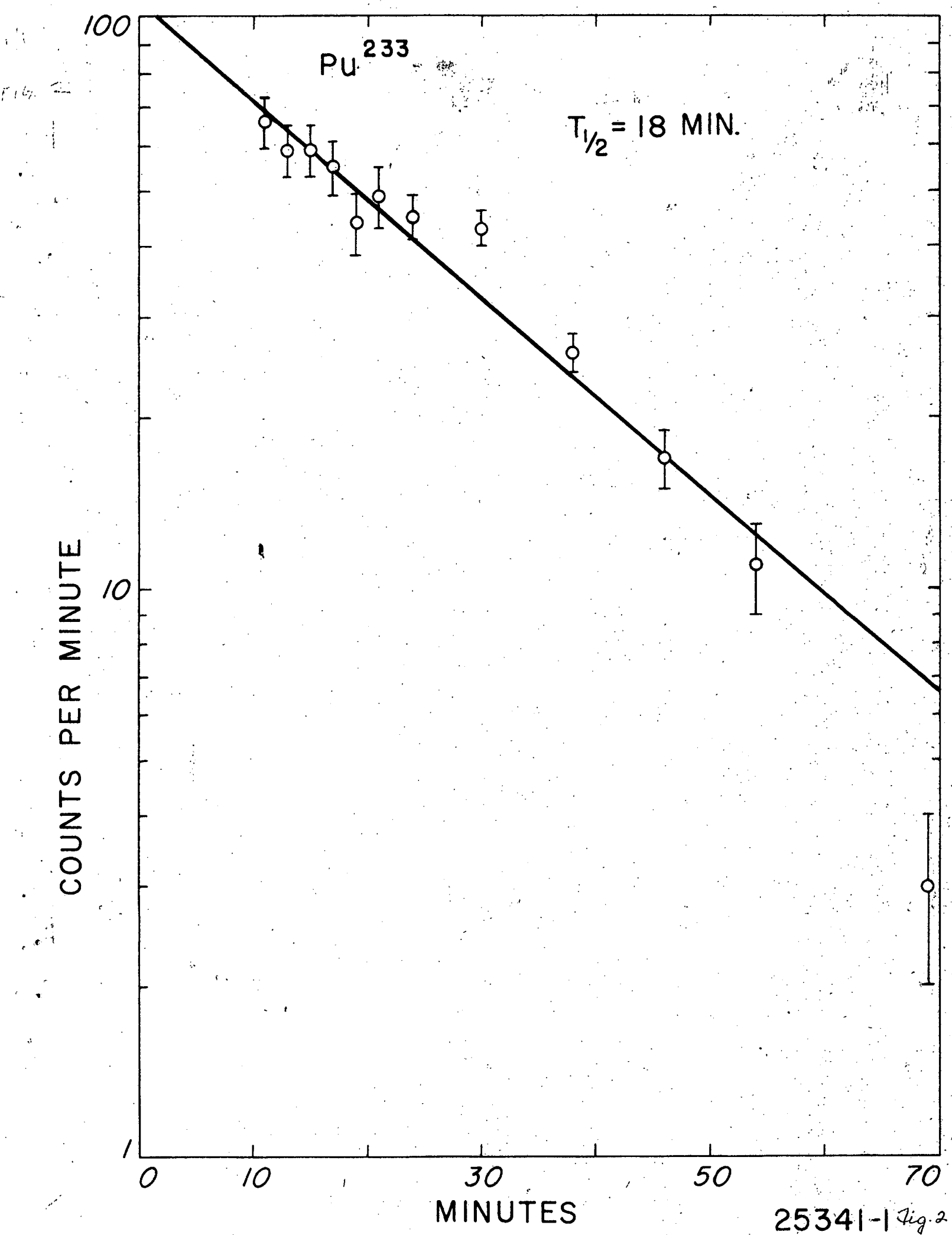
FIGURE 3. Pulse analysis showing alpha activity due to the decay of Pu^{239} and of the products from the U^{235} (α,xn) reactions.

FIGURE 4. Alpha decay energy vs mass number for plutonium isotopes.
● - most abundant transition ▲ - highest energy transition (for Pu^{237} and Pu^{241}).


150
100
10
1
COUNTS PER MINUTE
0
10
20
30
40
48
CHANNEL NUMBER
Pu239
Pu234
Pu233
Pu232
U228
Pu235
Pu236

25342-2

Fig 1


100
10
1
Pu 233
T1/2 = 18 MIN.
COUNTS PER MINUTE
0
10
30
50
70
MINUTES
25341-1 Fig. 2

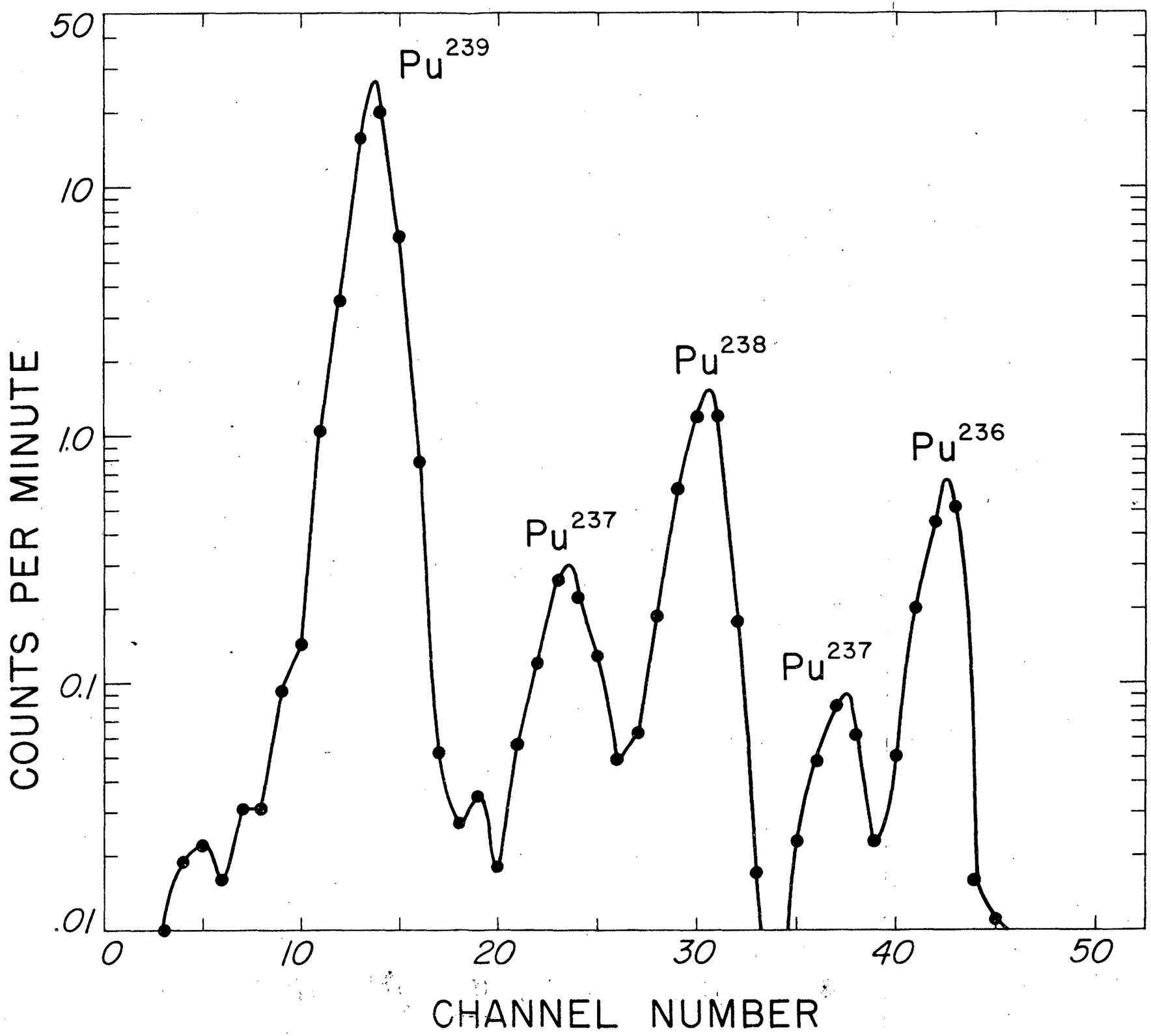

50
10
1.0
0.1
.01
COUNTS PER MINUTE
0
10
20
30
40
50
CHANNEL NUMBER
Pu239
Pu237
Pu238
Pu236
Pu237

Fig. 3

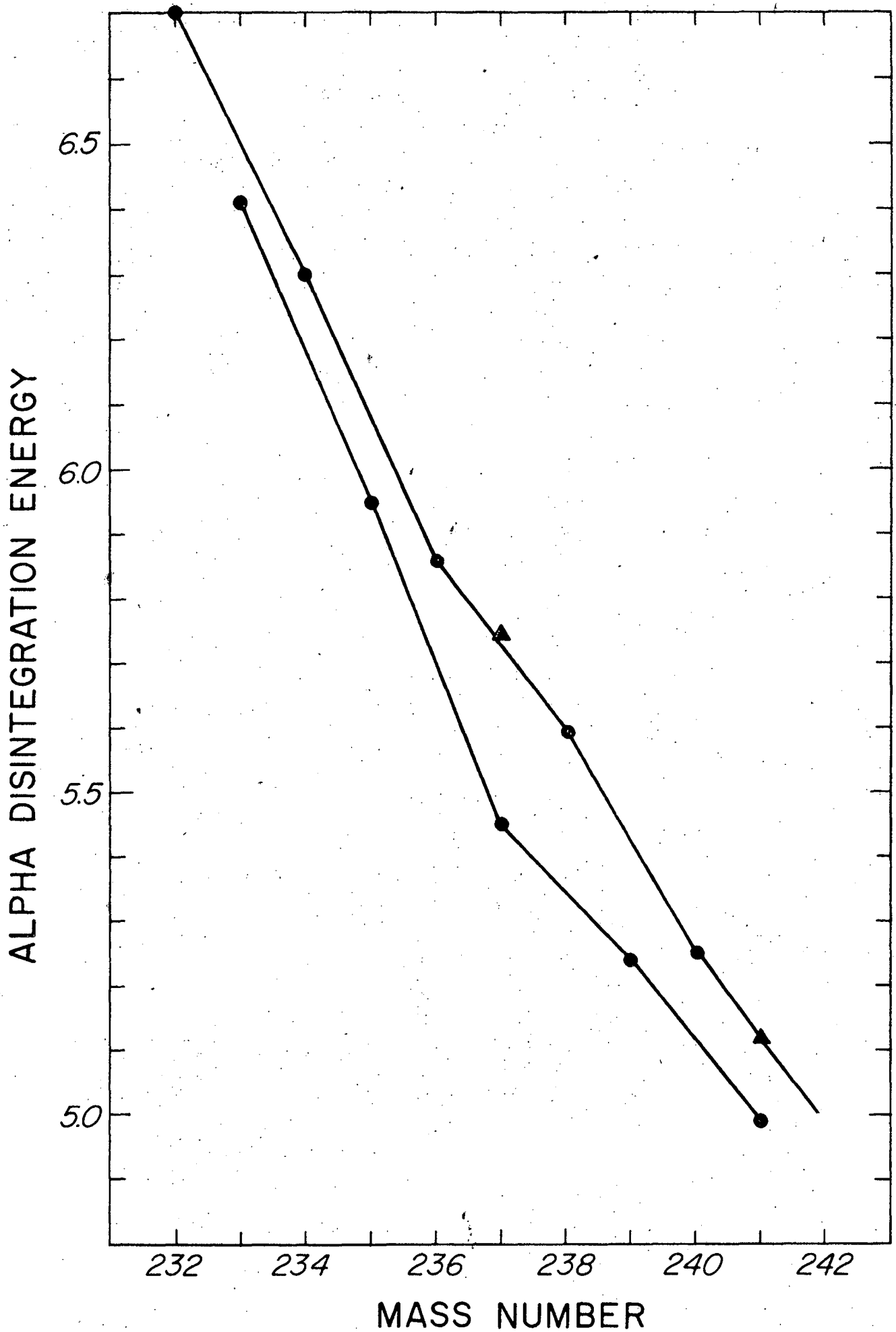

ALPHA DISINTEGRATION ENERGY
6.5
6.0
5.5
5.0
232
234
236
238
240
242
MASS NUMBER

Fig. 4

24805-1