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ISOTOPES OF AMERICIUM AND CURIUM

Kenneth Street, Jr.

March 10, 1949

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Isotopes of Americium and Curium

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ABSTRACT

A new and rapid method for the separation of americium and curium from the rare earths has been developed. Utilizing this method, two new activities have been found; Am^{238}, decaying by orbital electron capture with a half-life of ca. 1.2 hours, and an alpha emitting (energy 6.5 Mev.) curium activity with a half-life of ca. 2.5 hours which is tentatively assigned to Cm^{238}.

The 50-hour americium activity previously reported has been reassigned to Am^{240}. The beta-particle decay of the long-lived Am^{242} has been demonstrated. A limit of less than 20% has been placed on the possible orbital electron capture branching of Cm^{240}.

The probable effect of fission competition on yields of alpha-particle and deuteron bombardments is discussed.

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Isotopes of Americium and Curium

Kenneth Street, Jr.
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Introduction

The search for the transuranium elements and the elucidation of their chemical and nuclear properties has had an interesting history. Fermi's[1] early attempt to produce elements of atomic number greater than 92 by neutron bombardment of uranium resulted not in the production of transuranium elements but in fission of the uranium nucleus, as was shown by Hahn and Strassmann[2] in 1939. The first successful attempt to produce a transuranium element was made by McMillan and Abelson[3] in 1940 when they discovered element 93, which they named neptunium. The number of transuranium elements was extended once more by the discovery of element 94 (plutonium) by Seaborg, McMillan, Kennedy, and Wahl in 1940[4].

With the development of chain reacting piles and the production of large quantities of plutonium a starting material became available for the attempt to produce the next higher elements. In order ot isolate and identify any transplutonium elements produced it was necessary to have some hypothesis as to the chemical nature of these elements. This was essential because of the large number of fission activities produced and the high level of alpha radioactivity of the target Pu^{239}. A consideration of the chemical properties of the elements actinium through plutonium led Seaborg[5] to the view that the heavy elements constitute a new "rare earth" series in which the 5f shell is being filled. On this basis it was expected that elements 95 and 96 would have very stable III oxidation states with higher states being formed with great difficulty if at all, and consequently these elements should coprecipitate with rare earth fluorides.

In 1944, using this chemical hypothesis, Seaborg, James, and Ghiorso[6] working

at the Metallurgical Laboratory of the University of Chicago identified isotopes of the element of atomic number 96 produced by the alpha-particle bombardment of Pu^{239} on the 60-inch cyclotron at the Crocker Radiation Laboratory of the University of California. Later the same year the element of atomic number 95 was found by Seaborg, James, and Morgan[7]. Element 95 was named americium after the Americas because of its analogy to the rare earth europium. The name curium was chosen for element 96 honoring the Curies in a manner analogous to the naming of gadolinium.

The status of the isotopes of americium and curium at the time the present work was begun may be summarized briefly as follows:

Seaborg, James, and Morgan[7] obtained Am^{241} by bombarding U^{238} with alpha-particles to make Pu^{241} and isolating the Am^{241} which grows from the beta decay of Pu^{241}. Am^{241} is an alpha emitter with an alpha energy of 5.45 Mev. and a half-life determined by Cunningham[8] to be 510 $\pm$ 10 years. Manning and Asprey[9] demonstrated the existence of a 17-hour beta emitting Am^{242} made by thermal neutron irradiation of Am^{241}. Morgan[10] showed that after the 17-hour Am^{242} had completely decayed from a neutron irradiated sample of Am^{241} the purified americium fraction still contained a long lived beta activity and that Np^{238} could be milked from this fraction six months after shutdown of the bombardment, thus proving the existence of a long lived isomer of Am^{242}. On the assumption that the beta-particle observed belonged to the long lived Am^{242} Morgan calculated the alpha branching as ca. 0.2%. Morgan also observed two orbital electron capturing activities in americium fractions produced by bombarding Pu^{239} with deuterons and Np^{237} with alpha-particles. The 12-hour activity which decays principally by electron capture but which also emits alphas to the extent of ca. 0.1% was tentatively assigned to Am^{239} and the 50-hour orbital electron capturing activity was tentatively assigned to Am^{238}.

Both these mass assignments were made on the basis of the relative yields of the two activities in a series of deuteron and alpha bombardments employing different bombarding energies. The reassignment of the 50-hour activity and a proposed explanation of Morgan's results are both discussed in later sections of this dissertation.

Seaborg, James, and Ghiorso[6] first produced Cm^{242} by the alpha bombardment of Pu^{239}. This same activity was later produced by neutron irradiation of Am^{241} to form Am^{242m} which decays by beta-particle emission to Cm^{242} [11]. The mass assignment of this activity was definitely established by milking its alpha decay daughter Pu^{238}. Cm^{242} is an alpha emitting activity with a half-life of 150-days and an alpha energy of 6.1 Mev. Cm^{240}, also produced by alpha-particle bombardment of Pu^{239}[11], emits alpha-particles of energy 6.3 Mev. and decays with a half-life of 26.8 days. The mass assignment of this activity was also definitely established by milking its alpha decay daughter Pu^{236}. In addition, James observed an orbital electron capturing activity of 55-day half-life, also formed in the alpha-particle bombardment of Pu^{239}, which he tentatively assigned to Cm^{241}.

The aim of the present investigation was to attempt to clarify any doubtful points in the previous work and to try to extend the known isotopes in the direction of higher masses and shorter half-lives.

EXPERIMENTAL METHODS

A. Bombardment Techniques. The bombardment techniques used in the bombardments made on the 60-inch cyclotron at the Crocker Radiation Laboratory were essentially the same as those employed by James[11]. The plutonium targets were prepared by slurrying approximately 10 mg. of Pu^{239} in the form of $(NH_4)_2PuF_6$ into a rectangular platinum boat of approximately 1 sq. cm. area and evaporating

to dryness. The boat was then covered with a cap made from 0.25 mil. tantalum foil and suspended by water-cooled finger in the deflected beam of the 60-inch cyclotron. As a further precaution to prevent contamination of the cyclotron, the target was separated from the cyclotron by another 0.25 mil. tantalum foil and a filtered air leak around this foil was provided to allow evacuation of the target.

In the higher energy bombardments on the 184-inch cyclotron at the University of California Radiation Laboratory, where it is necessary to introduce the target directly into the cyclotron chamber, this technique could not be used. In these bombardments the slurry of $(NH_4)_2PuF_6$ was placed in a welded platinum envelope, evaporated to dryness, and the open edge sealed by clamping between two copper blocks. This whole assembly could be attached to the probe in the cyclotron chamber.

B. Chemical Procedures. The chemical procedures used in isolating americium or curium fractions from the bombarded target material may be conveniently divided into three steps varying greatly in the degree of difficulty and the time required. These steps are, (1) the dissolution of the target material, (2) isolation of a rare earth fraction containing the fission product rare earths, americium, and curium, (3) separation of americium and curium from the rare earth fission products, and from each other.

(1) Dissolution of the Target. The difficulty encountered in dissolving the target material depends greatly on the temperature attained during the bombardment. In the bombardments on the 60-inch cyclotron, where the beam current and consequently the temperature attained is high, it is usually necessary to fume the target material to dryness with sulfuric acid and then dissolve the resulting plutonium sulfate in dilute nitric acid.

In the bombardments on the 184-inch cyclotron, where the beam current is lower by about a factor of ten, a very rapid dissolution of the target material can be

effected by simply heating with concentrated nitric acid with a little boric acid added.

(2) Isolation of a Rare Earth-Americium-Curium Fraction. The isolation of a rare earth-americium-curium fraction is easily attained by precipitating lanthanum fluoride from the solution after oxidizing the plutonium to the soluble VI oxidation state. Several different oxidizing agents may be used for this purpose with almost equal success: 0.1 N potassium dichromate and 0.1 N potassium permanganate are examples. The precipitate contained the beta-active rare earth fission products and the americium and curium. The precipitate is dissolved in a mixture of nitric and boric acids, the hydroxide precipitated with NH_4OH and the cycle repeated until the plutonium is eliminated.

(3) Separation of Am and Cm from the Rare Earths and From Each Other. The separation of americium and curium from the rare earth fission products, and from each other, presents by far the most difficult chemical problem encountered in the work and consequently a great deal of time was devoted to achieving a reasonably satisfactory separation procedure. Americium, curium, and the rare earths all have plus three oxidation states and the crystal radii of americium and curium overlap those of the light rare earths, being about the same as neodymium. Thus the solubilities of their compounds are very similar to those of the rare earths.

The only reasonably satisfactory group separation of americium and curium from the rare earths known when the present work was undertaken was the procedure known as "the fluosilicate precipitation"[11,12]. In this procedure LaF_3 carrier is partially precipitated by accurately controlling the fluoride ion concentration. The control of the fluoride ion concentration is attained by means of fluosilicic or fluoboric acid. These acids hydrolyze to produce a small fluoride ion concentration which can be accurately controlled by varying the hydrogen ion concentration.

Under suitable conditions 80-90% of all the rare earths are precipitated while only 30% of the americium and curium is found in the precipitate. The undesirable features of the method are the very carefully controlled conditions necessary, the large number of repetitions needed to achieve a high degree of decontamination, the relatively long time required (ca. 1 hr. per cycle), and the low yield of decontaminated americium and curium.

The very successful separation of the rare earths by selective elution from columns of cation exchange resins with ammonium citrate solution led Cunningham et. al.[13] to try this procedure for the separation of americium and curium. As is shown in Fig. 1, 0.25 M ammonium citrate solution adjusted to pH 3.0-3.1 with hydrochloric acid gives a satisfactory separation of americium and curium. It will be noted that americium is eluted with element 61 and presumably curium with samarium so that separation from these fission products is not obtainable by this method. The most unsatisfactory part of this separation is the long time (ca. 12 hours) required.

For many of the experiments contemplated a good yield and a very complete separation of americium and curium from rare earth fission products and from each other was necessary, but because of the relatively long half-lives involved, time was not a major consideration. In order to look for short half-lives a method for the very rapid and complete separation from the rare earths and some fractionation of americium and curium had to be found. Because a promising method of attack was at hand, a solution to the first problem was sought first, and very fortunately in the course of its solution the direction of a satisfactory solution to the second problem was indicated.

If a narrow band of cations is placed on the top of a cation exchange resin column, the volume of solution which must be passed through the column before the band is eluted depends on the distribution of the ions between the liquid and the

fixed resin phases. The larger the fraction of the cations in the liquid phase the more rapidly they will be eluted (i.e. smaller volume of solution needed). There are two major factors that can effect this distribution. There is the natural order of replacement of cations in the resin. In general +3 ions replace +2, +2 replace +1, etc., and with ions of the same charge those of larger crystal radii replace those of smaller radii. Then there is the effect of complexing agents in the liquid phase which reduce the activity of the cation and shift the distribution in favor of the liquid. Thus the separation of two species of cations by elution from an ion exchange column depends on the existence of a difference in their distribution coefficients. In the case of ions of similar charge this difference may be caused by differences in their size, or differences in the stability of their complexes with the eluting solution.

As was shown by Cunningham, et. al. the elution of americium and curium with ammonium citrate gives a satisfactory separation. However, due to the similarity in size and stability of their citrate complexes, some of the light rare earths, notably element 61, are eluted at the same time as americium and curium. Fig. 1 shows a typical elution of tracer amounts of americium, curium and several rare earths using 0.25 M ammonium citrate adjusted to pH 3.05 with ^{4}Cl.

There was no a prioro reason to believe that equally good separation of americium and curium could not be effected with other eluting solutions. In addition the possibility existed that due to more subtle differences between the actinides and the rare earths the time of elution of americium and curium with respect to the rare earths might be shifted so that with a different eluting solution a rare earth other than element 61 would be eluted at the same time as americium and curium. Thus, although the time required might be long, a complete decontamination of americium and curium from the rare earths could be effected by eluting with one solution then placing the material on a second column and eluting with a different solution.

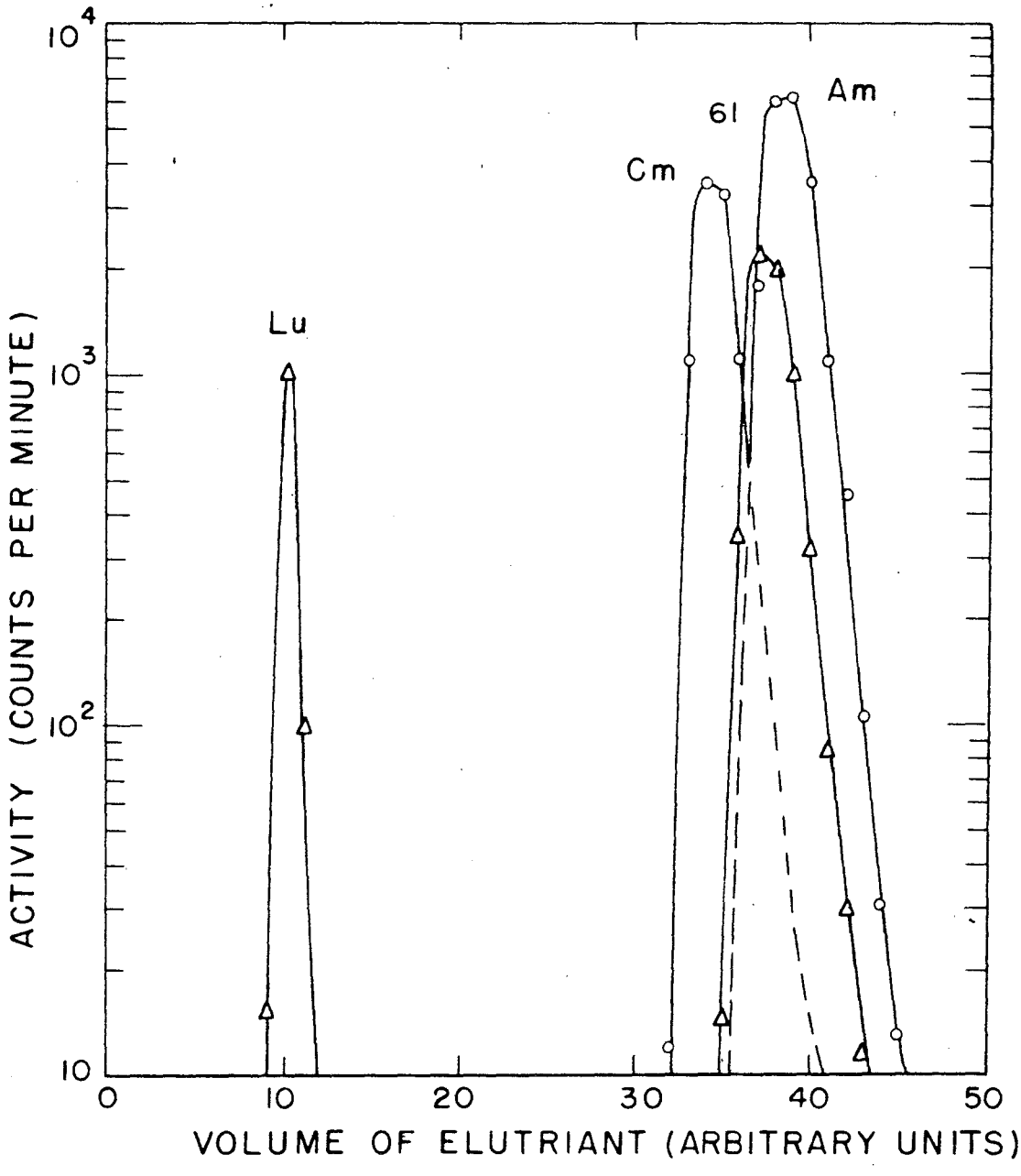
With this is mind a series of columns were run testing various eluting solutions such as 2 M NH_4NO_3, various concentrations of HCl, HBr, and $HClO_4$. With all of these eluting agents the separation of tracer amounts of americium and curium was essentially the same as with citrate. Fig. 2 shows an elution with 6 M HCl run with the same column as that used for the citrate run in Fig. 1. Thus 6 M HCl affords an equally effective and in some respects more convenient method of separating tracer quantities of americium and curium. The principal advantages are the reproducibility of results, and the convenience of the HCl solution for rerunning or doing additional chemistry.

In addition, increased concentrations of HCl showed a very marked effect on the time of elution of americium and curium with respect to the rare earths. This is illustrated in Figs. 3 and 4 which show elutions with 9M HCl and 13.3 M HCl respectively. It will be noted that with 9 M HCl americium and curium are eluted with the heavy rare earths while with 13.3 M HCl they have moved out well ahead of all the rare earths. The effects noted were of a much greater magnitude than anything anticipated when the experiments were undertaken. Thus a single fast elution with 13.3 M HCl separates americium and curium from all the rare earths.

Examination of Figs. 2, 3, and 4 will also reveal another striking fact. It will be noted that with 6 M HCl americium and curium are eluted in their normal order; that is, curium is eluted before americium in the order that would be predicted from their sizes and the order of elution of the rare earths. In 9 M HCl the order of elution is reversed, the americium coming off slightly ahead of the curium while the rare earths maintain their normal order throughout the whole range of HCl concentrations.

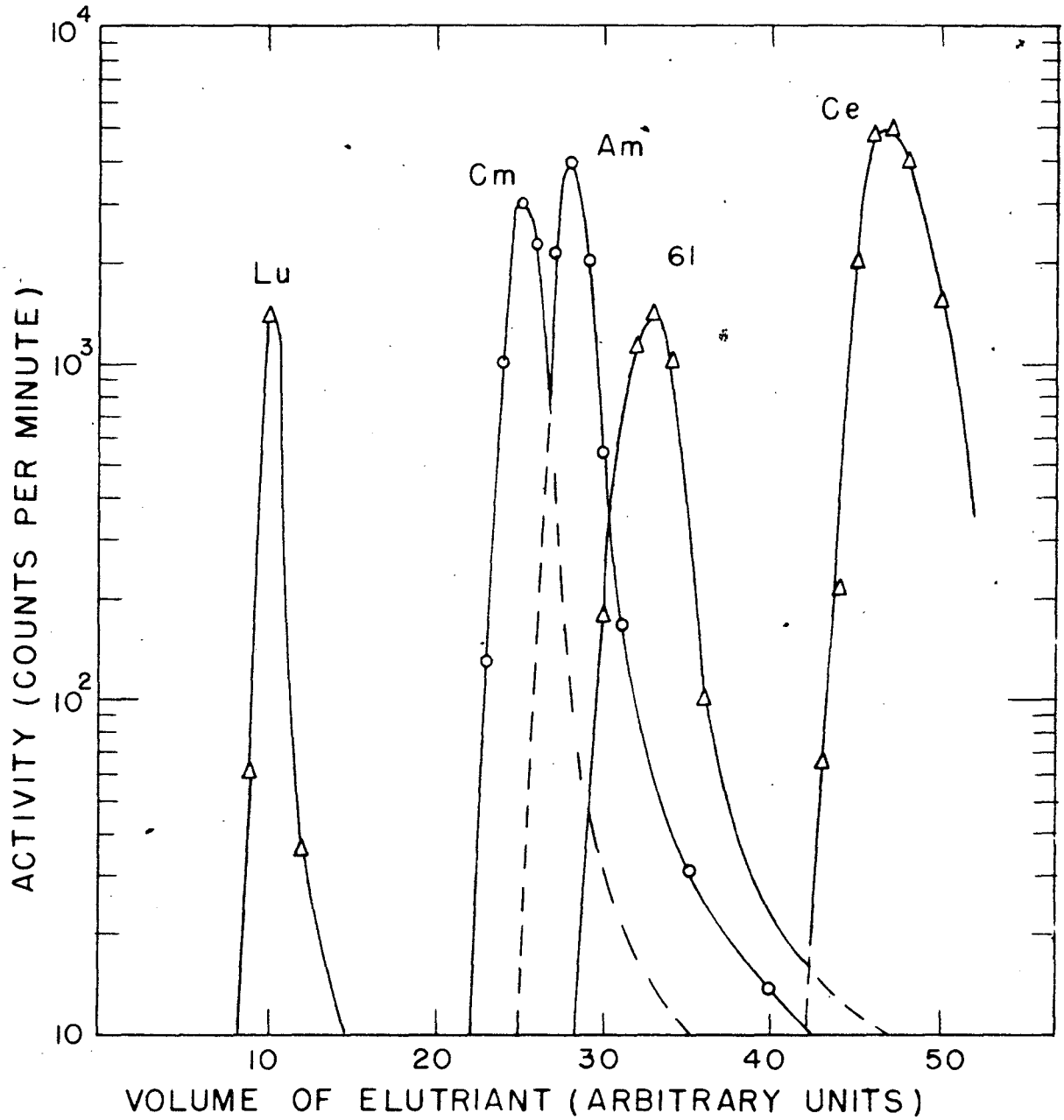
In addition to the very useful nature of the previous result in the bombardment work, this very anomolous behavior of americium and curium suggests some very interesting speculation. A more detailed investigation of the phenomena involved

Figure 1 Elution of Am, Cm, Lu, and element 61 tracers with 0.25 M ammonium citrate solution adjusted to pH 3.0-3.1 with hydrochloric acid. The column used was 1 mm. in diameter, 10 cm. long, and packed colloidal agglomerates of Dowex 50 cation exchange resin.


ACTIVITY (COUNTS PER MINUTE)
10^4
10^3
10^2
10
Lu
Cm
61
Am
0
10
20
30
40
50
VOLUME OF ELUTRIANT (ARBITRARY UNITS)

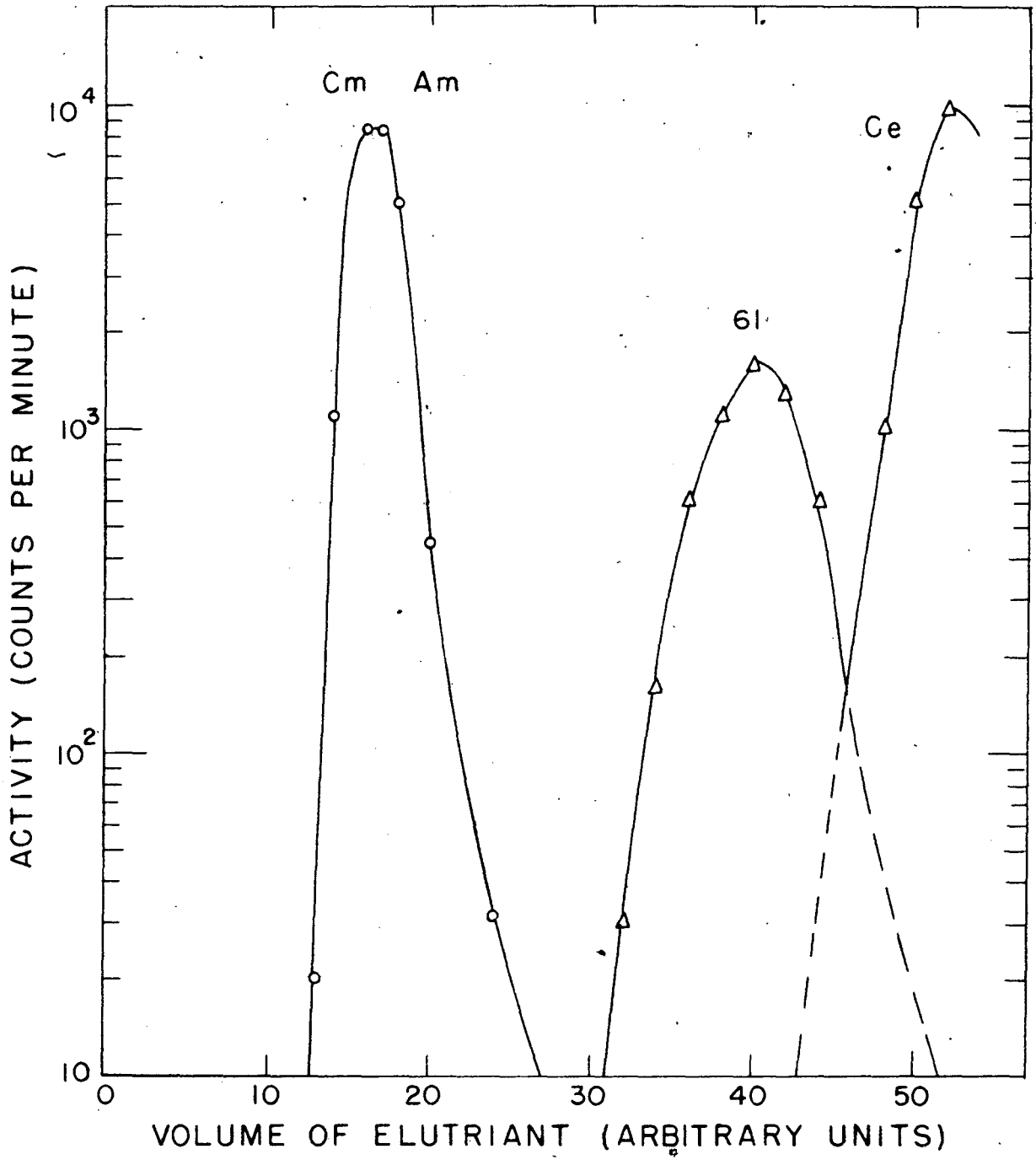
13477-1

Figure 2 Elution of Am, Cm, Lu, element 61, and Ce tracers with 6 M HCl. The column used was the same as in Fig. 1.


ACTIVITY (COUNTS PER MINUTE)
10^4
10^3
10^2
10
Lu
Cm
Am
61
Ce
10
20
30
40
50
VOLUME OF ELUTRIANT (ARBITRARY UNITS)

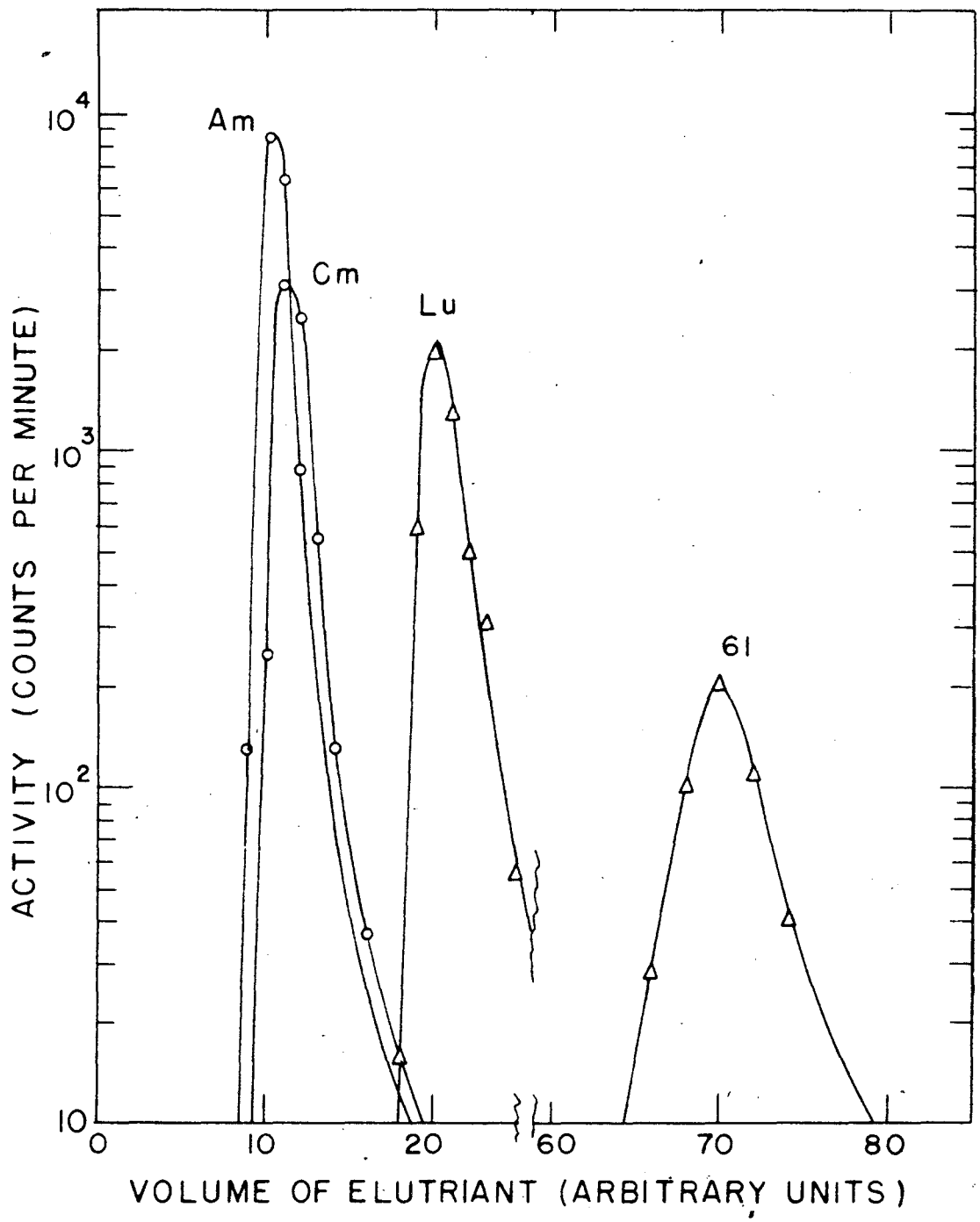
13478-1

Figure 3 Elution of Am, Cm, element 61, and Ce with 9 M HCl. The column used was the same as in Fig. 1.


Cm
Am
Ce
61
10^4
10^3
10^2
10
0
10
20
30
40
50
ACTIVITY (COUNTS PER MINUTE)
VOLUME OF ELUTRIANT (ARBITRARY UNITS)

13479-1

Figure 4 Elution of Am, Cm, Lu, and element 61 with 13.3 M HCl. The column used was the same as in Fig. 1


Am
Cm
Lu
61
10^4
10^3
10^2
10
ACTIVITY (COUNTS PER MINUTE)
0
10
20
60
70
80
VOLUME OF ELUTRIANT (ARBITRARY UNITS)

13480-1

is in progress so that only an outline of the working hypothesis will be given here.

On elution with solutions of HCl of increasing concentration americium and curium behave as though at high concentration there are on the average several more chloride ions involved in the complexes of americium and curium than in those of the rare earths. In view of the similarity in size and charge of the americium and curium and the rare earth ions this suggests that in addition to the interactions which the rare earths show with chloride ion perhaps americium and curium form weak covalent coordination complexes involving large numbers (say 6) of chloride ions. That is, covalent complexes in the sense of the transition series chloride complexes.

One of the most obvious differences between the actinide and the rare earth elements is the relative stability of the f electrons and orbitals. Thus in the actinide series the 5f orbitals have probably just become stable with respect to the 6d orbitals at uranium and are only a little more stable at americium and curium. While in the rare earths the 4f orbitals are already stable with respect to the 5d orbitals at cerium. This suggests that perhaps the 5f electrons and orbitals of the actinides can contribute to hybridized covalent bond orbitals, while in the rare earths the very stable and deeply buried 4f electrons and orbitals are not available. Thus if the covalent complex of americium were slightly more stable than that of curium, elution with increasing concentrations of HCl would result in both americium and curium moving up with respect to the rare earths, but americium moving slightly faster, as is observed to be the case.

As an example of the ion exchange column techniques used, a typical cyclotron bombardment will be outlined briefly. A rare earth fraction containing the americium and curium is isolated as described previously using from 0.2 to 0.5 mg. of lanthanum as a carrier. The $La(OH)_3$ is dissolved in ca. 100 ul. of 0.5 M $HClO_4$ and equilibrated for 5 minutes with ca. 25 ul of resin in three batches. The resin

fractions are combined and slurried on to the top of the column. The columns used are 2 mm. in diameter and 5 cm. long, and are packed with colloidal agglomerates of Dowex 50 cation exchange resin. On elution with 13.3 M HCl the americium and curium are eluted after ca. 0.7 ml. of solution have passed through the column. The time required is approximately 30 minutes. Separation from the rare earth fission products is complete, and in addition there is enough separation of the americium and curium so their radioactivities can be distinguished.

C. Alpha Energy Measurements. All of the alpha-particle energy determinations in these investigations were made by means of a multi-channel pulse analyzer[14]. With this instrument a thin sample is placed in an ionization chamber in which the total ionization of an alpha-particle can be measured as a voltage pulse. Individual pulses are sorted electronically and recorded on a number of fast mechanical registers in such a way as to separate the individual alpha-particle energies in a mixture of alpha emitters.

DISCUSSION OF RESULTS

A. Reassignment of the 50-hour Americium Activity. Morgan[10] assigned the 12-hour and the 50-hour orbital electron capturing activities which he observed in deuteron bombardments of Pu^{239}, to Am^{239} and Am^{238} respectively. His assignment was based largely on the very nearly equal yields of the 12 and 50-hour activities found in a series of bombardments in which the deuteron energy varied from 13 to 19 Mev., and on the fact that as the bombarding energy increased, the yield of the 50-hour activity increased very slightly relative to the 12-hour activity. The argument used was that in this range of bombarding energies the (d,2n) and the (d,3n) reactions should give comparable yields while the (d,n) yield should be much lower, and that the (d,3n) yield should increase with respect to the (d,2n) yield with increasing bombarding energy. The evidence that this situation probably does not hold in cases where fission is a possible competing process and a possible

explanation of it will be discussed in section G.

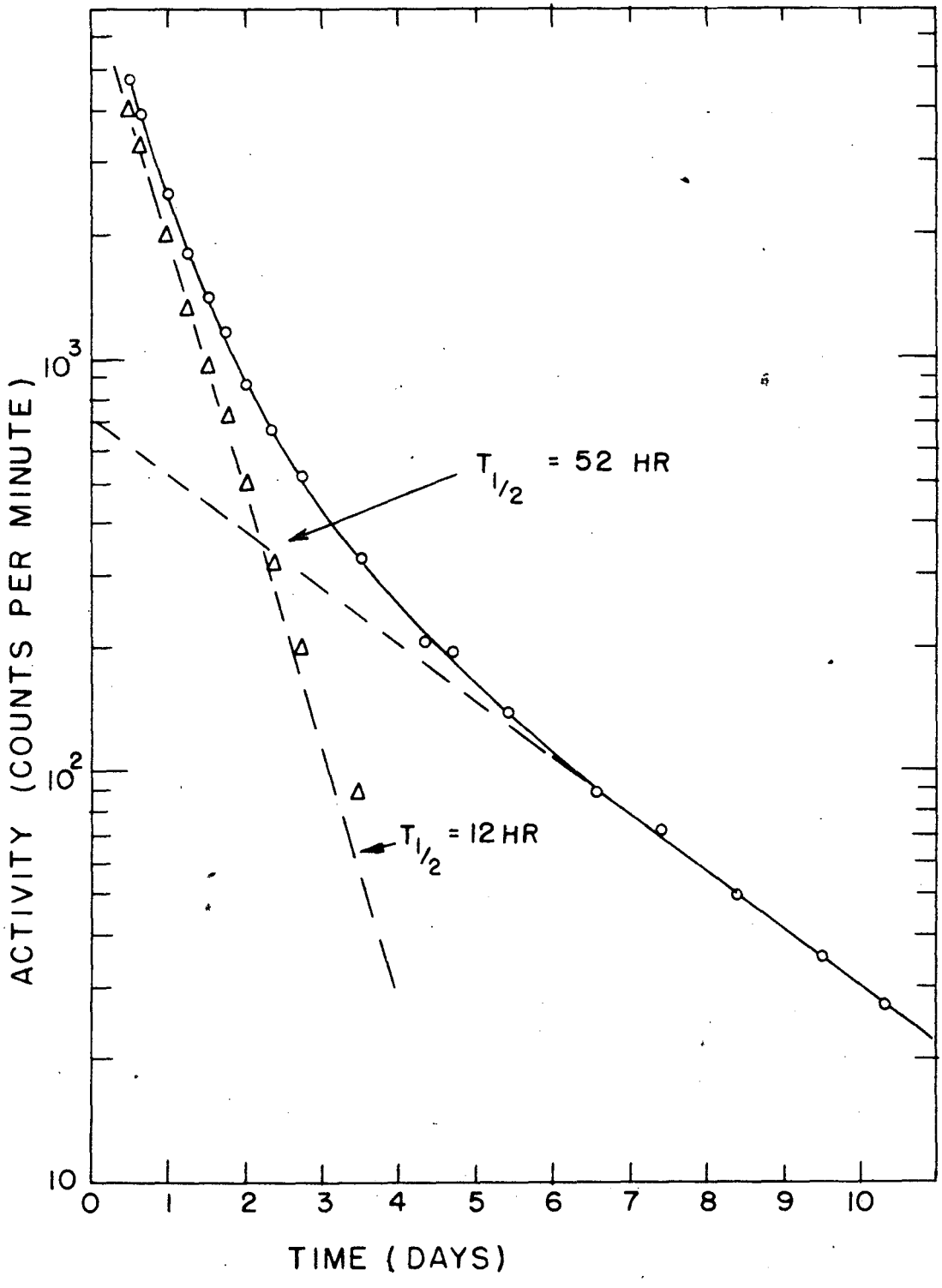
James and Orth[15] have shown that with 10 Mev. deuterons the yield of the (d,3n) reaction on U^{235} is very low if indeed the reaction goes at all. Thus in the same type of bombardment on Pu^{239} one would not expect to make an appreciable yield of Am^{238}. The Geiger counter decay of the americium fraction from a 10 Mev. deuteron bombardment of Pu^{239} is shown in Fig. 5. Both the 12-hour and the 50-hour activities were formed in good yield and thus are probably the products of the (d,n) and (d,2n) reactions rather than the (d,2n) and (d,3n) reactions. Therefore these activities should be assigned to Am^{239} and Am^{240}. This assignment was further substantiated by the discovery of a shorter lived activity which is best assigned to Am^{238} (section D).

Fig. 6 shows the Geiger counter decay of the americium fraction from a 9 Mev. proton bombardment of Pu^{239}. With 9 Mev, protons the yield of the $Pu^{239}(p,n)Am^{239}$ reaction should be very much larger than the yield of the $Pu^{239}(p,\gamma)Am^{240}$ reaction. In view of the very clean 12-hour decay observed this activity must be assigned to Am^{239} while the 50-hour activity must then be Am^{240}.

B. Alpha Energy of Am^{239}. Morgan[10] reported the fact that the 12-hour Am^{239} decays by alpha-particle emission to the extent of ca. 0.1%. However the chemical methods he used gave relatively thick samples which were not suitable for an alpha energy determination. The ion exchange procedures used in present work give carrier free samples. Fig. 7 shows the alpha decay of the americium fraction from the 10 Mev. deuteron bombardment of Pu^{239} mentioned in section A. Fig. 8 shows an alpha-particle pulse analysis of the same sample, giving an energy of 5.8 Mev. for the alpha-particle of Am^{239}.

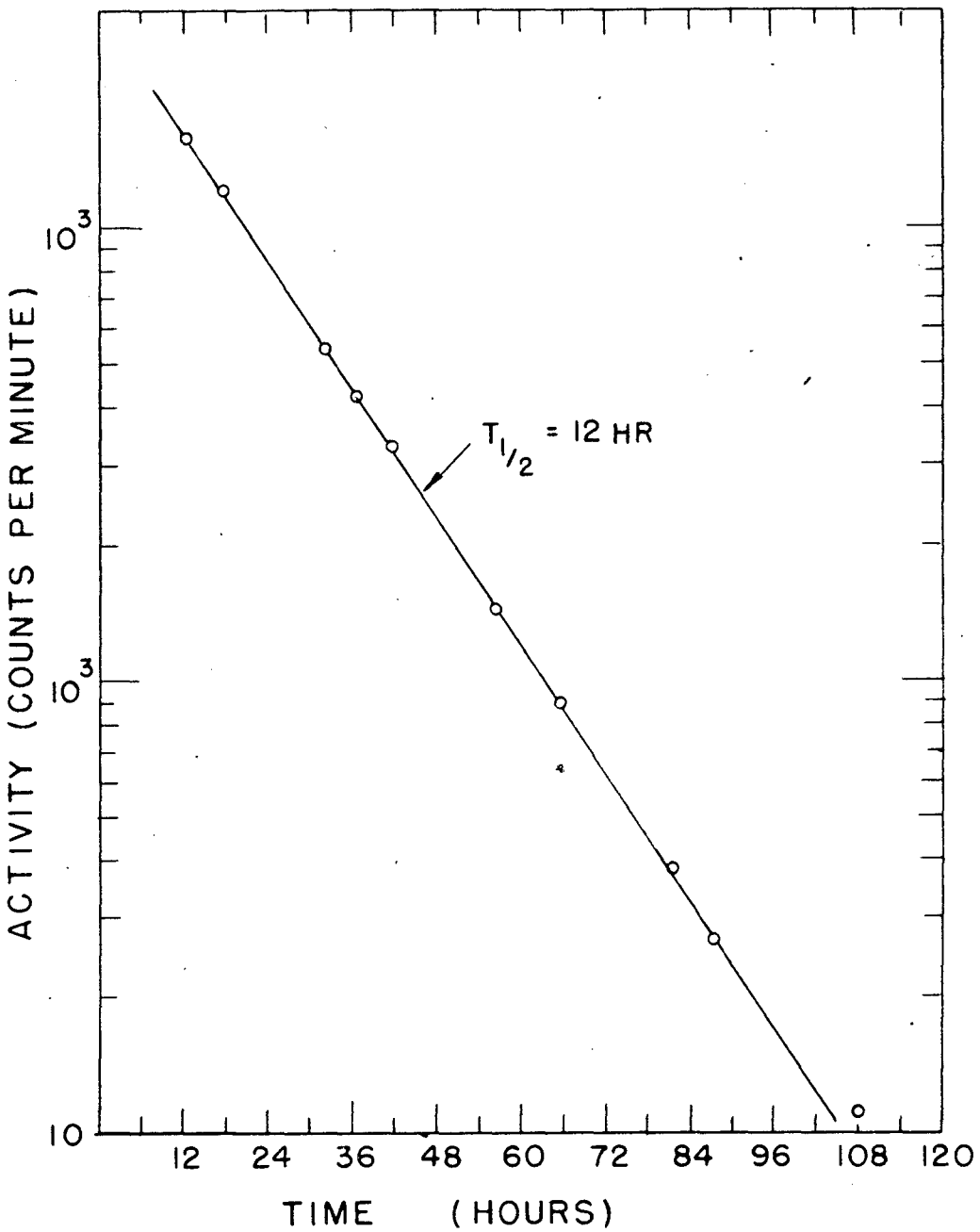
C. Growth of Cm^{242} from the Long-Lived Am^{242}. As mentioned in the introduction, Morgan[10] had demonstrated the existence of a long-lived Am^{242} decaying at

Figure 5 Geiger counter decay of the americium fraction from a bombardment of Pu^{239} with 10 Mev. deuterons. The counter used was of the cylindrical glass type with a window thickness of ca. 20 mg./cm.^{2}.


ACTIVITY (COUNTS PER MINUTE)
10^3
10^2
10
0
1
2
3
4
5
6
7
8
9
10
TIME (DAYS)
$T_{1/2}$ = 52 HR
$T_{1/2}$ = 12 HR

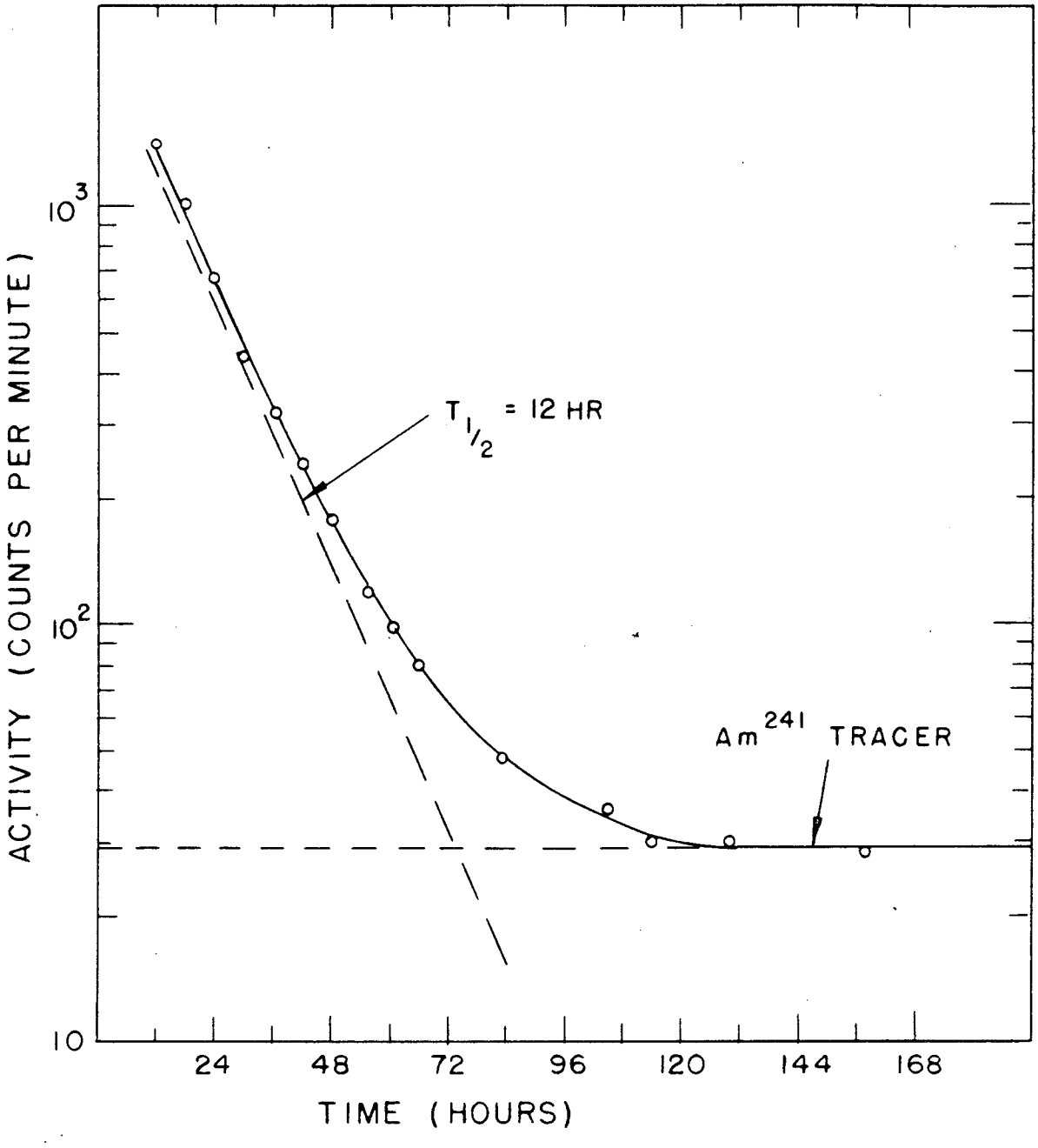
13481-1

Figure 6 Geiger counter decay of the americium fraction from a bombardment of Pu^{239} with 9 Mev. protons. The counter used was the same as in Fig. 5.


ACTIVITY (COUNTS PER MINUTE)
10^3
10^3
10
$T_{1/2}$ = 12 HR
12
24
36
48
60
72
84
96
108
120
TIME (HOURS)

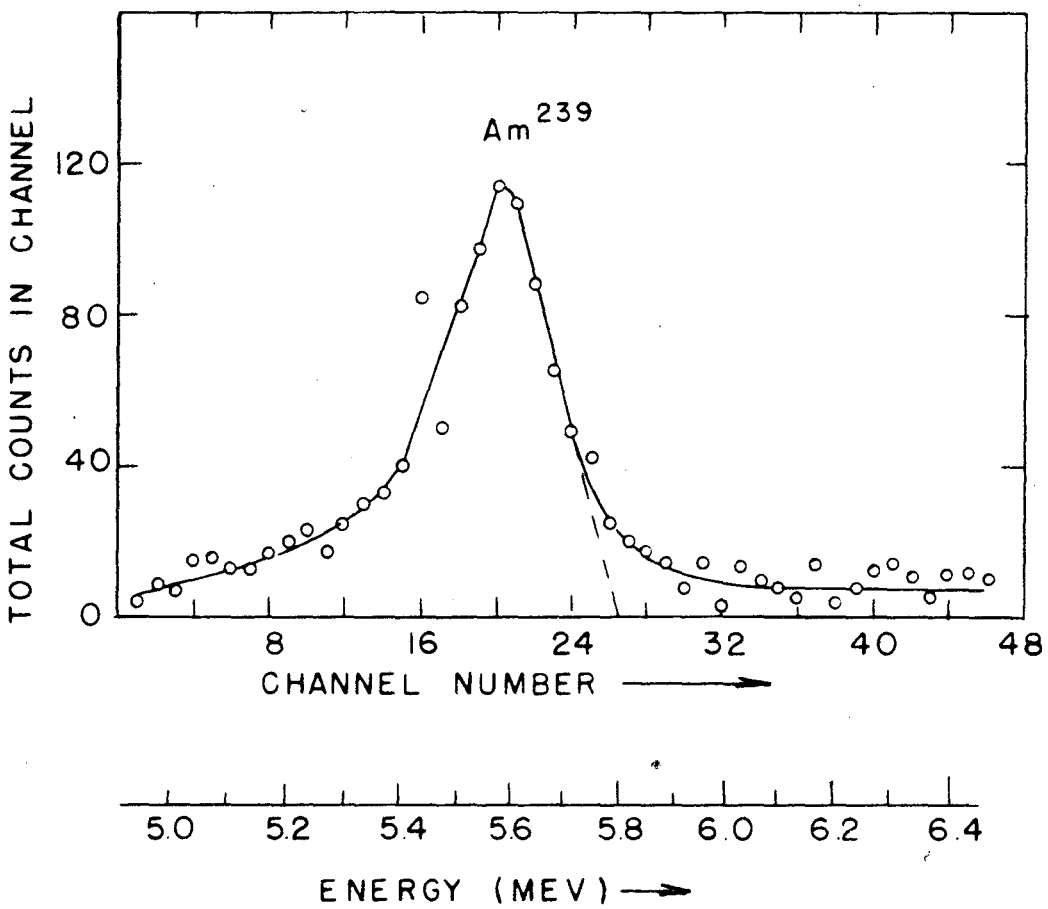
13482-1

Figure 7 Decay of the alpha activity in the americium fraction of a bombardment of Pu^{239} with 10 Mev. deuterons.


ACTIVITY (COUNTS PER MINUTE)
10^3
10^2
10
24
48
72
96
120
144
168
TIME (HOURS)
$T_{1/2}$ = 12 HR
Am^{241} TRACER

13483-1

Figure 8 Alpha-particle pulse analysis curve of the americium fraction the decay of which is shown in Fig. 7.


Am239
TOTAL COUNTS IN CHANNEL
120
80
40
0
8
16
24
32
40
48
CHANNEL NUMBER
5.0
5.2
5.4
5.6
5.8
6.0
6.2
6.4
ENERGY (MEV)

13484-1

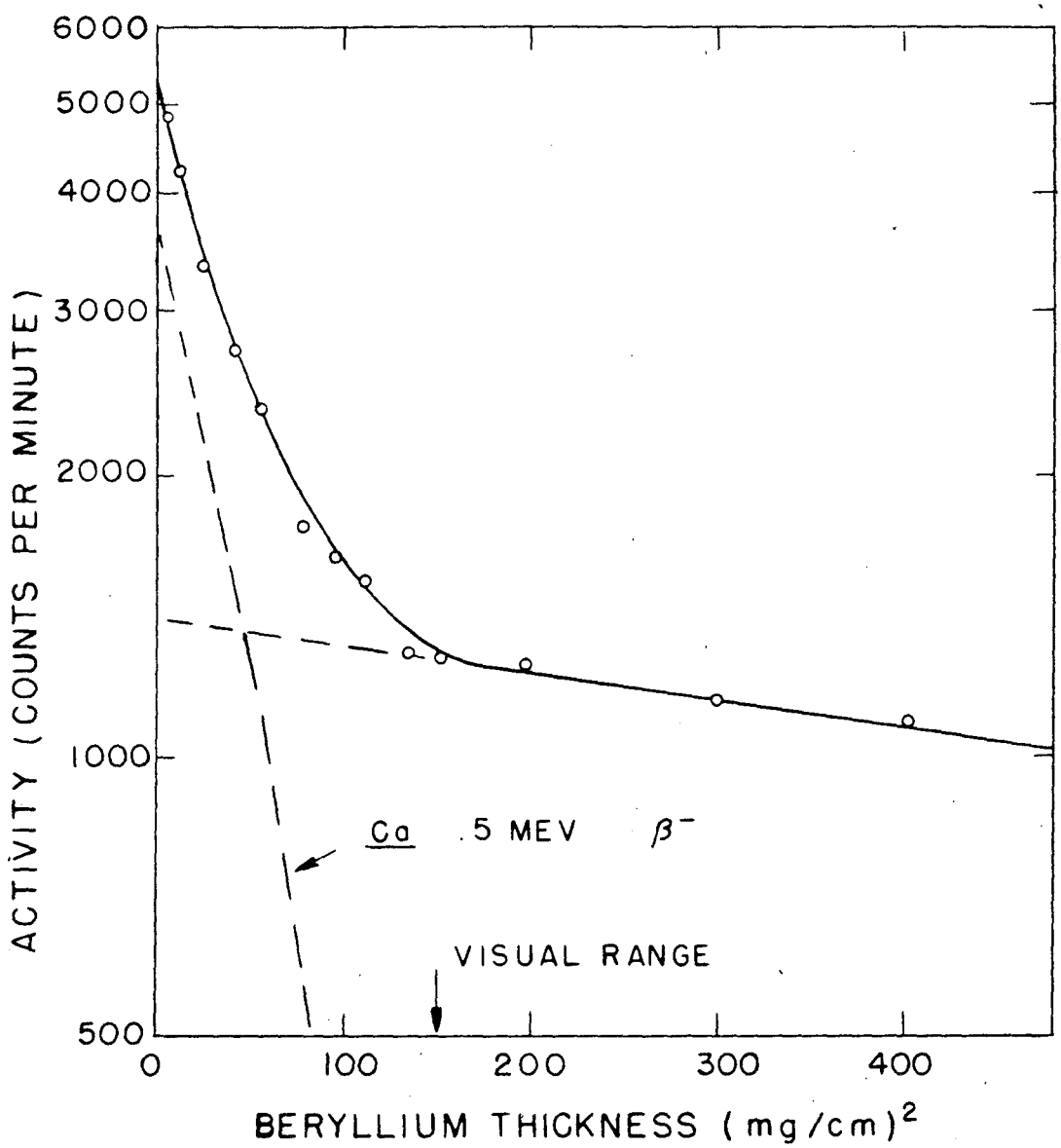
least partially by alpha emission. He did this by milking Np^{238} from Am^{241} irradiated with thermal neutrons long after the 17-hour Am^{242m} had completely decayed. He also observed a long-lived beta activity in the same americium fraction and concluded that Am^{242} probably decayed also by beta emission to Cm^{242}.

To prove the beta decay of the long-lived Am^{242} it is necessary to demonstrate the growth of Cm^{242} in a sample of the Am^{242} after the 17-hour isomer has completely decayed. This experiment was undertaken. Fig. 9 shows a beryllium absorption curve of the americium fraction from a thermal neutron irradiation of Am^{241}. The sample contained 1.30×10^6 alpha disintegrations per minute of Am^{241} and the absorption curve was taken at 10% geometry. Direct decay measurements on the beta-particles showed no detectable decay over a period of 18 months. Thus if all the beta-particles observed belong to Am^{242} one would expect to grow 2.9 alpha disintegrations per minute of Cm^{242} ($T_{1/2}$ = 150 days) at equilibrium for every 100 alpha disintegrations per minute of Am^{241}. To observe the growth in a satisfactory manner it was necessary to remove the Cm^{242} until the alpha activity due to it represented only about 0.1% of the alpha activity due to Am^{241}.

The neutron irradiated Am^{241} initially contained many times as much alpha activity due to Cm^{242} as that due to Am^{241}. Three successive ion exchange column separations of americium and curium reduced the Cm^{242} alpha activity to the desired 0.1% of the Am^{241} activity. Fig. 10 shows an alpha-particle pulse analysis of the americium fraction run 8 days after the final separation from curium.

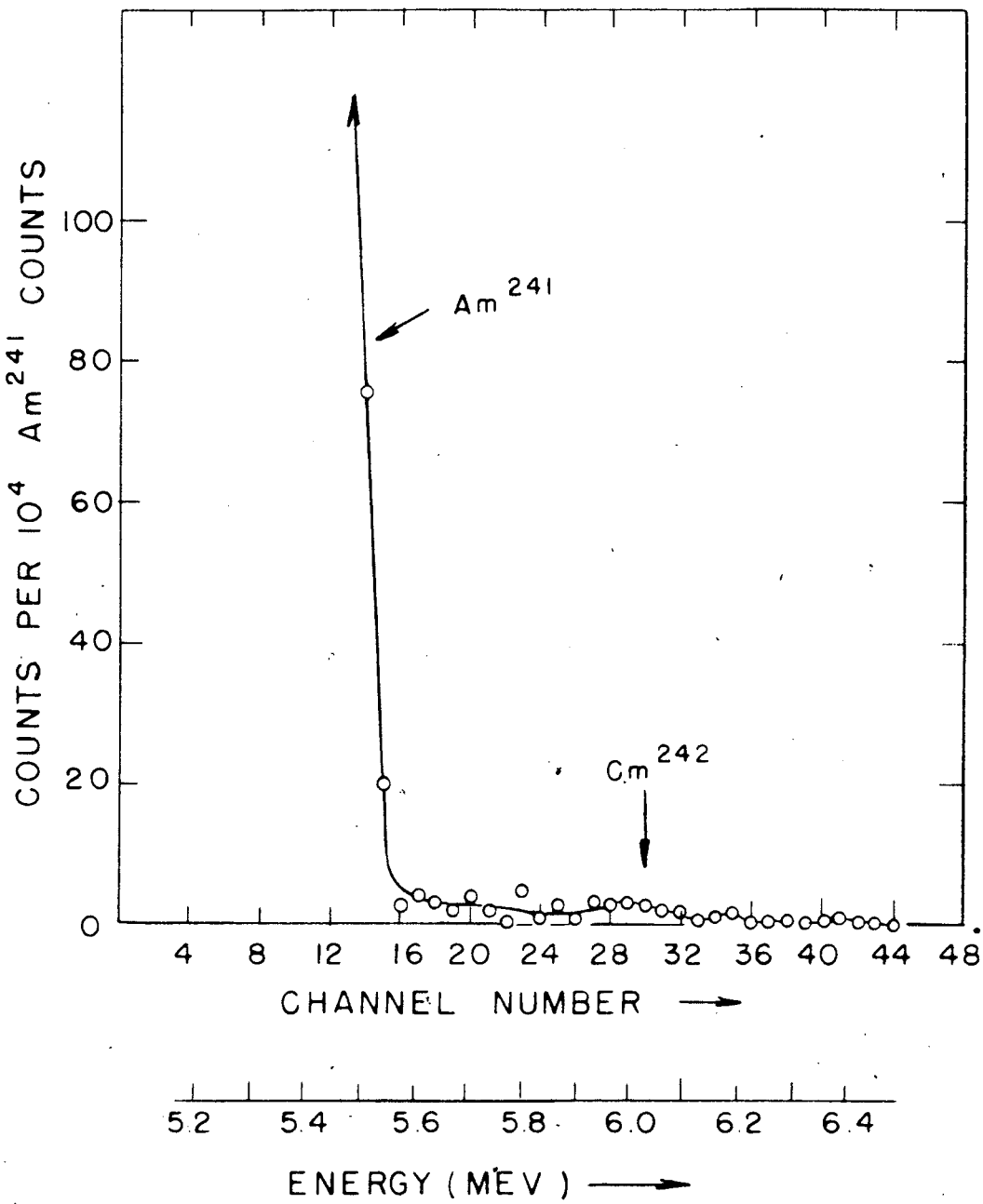
Additional pulse analyses were run at intervals over a period of 10 months in order to follow the growth of the Cm^{242}. Three different samples were followed to lessen the possibility of accidental contamination spoiling the experiment. Fig. 11 shows the pulse analyses taken 307 days after separation and demonstrating the definite growth of alpha-particle with the energy of Cm^{242} alphas. The growth of the Cm^{242} alphas is plotted in Fig. 12, the solid line indicating

Figure 9 Geiger counter beryllium absorption curve of the long-lived beta activity in the americium fraction from Am^{241} irradiated with thermal neutrons. The absorption curve was taken at 10% geometry on a bell type counter with a mica window of ca. 3 mg/cm^2 thickness. The sample contained 1.30×10^6 alpha disintegrations per minute of Am^{241}.


6000
5000
4000
3000
2000
1000
500
ACTIVITY (COUNTS PER MINUTE)
0
100
200
300
400
BERYLLIUM THICKNESS (mg/cm)^2
Ca .5 MEV β^-
VISUAL RANGE

13485-

Figure 10 Alpha-particle pulse analysis curve of the americium fraction from Am^{241} irradiated with thermal neutrons taken 8 days after the final separation from curium.


COUNTS PER 10^4 Am^{241} COUNTS
100
80
60
40
20
0
Am^{241}
Cm^{242}
4 8 12 16 20 24 28 32 36 40 44 48
CHANNEL NUMBER
5.2 5.4 5.6 5.8 6.0 6.2 6.4
ENERGY (MEV)

13486-1

Figure 11 Alpha-particle pulse analysis curve of the americium fraction of Fig. 10 taken 307 days after the final separation from curium.

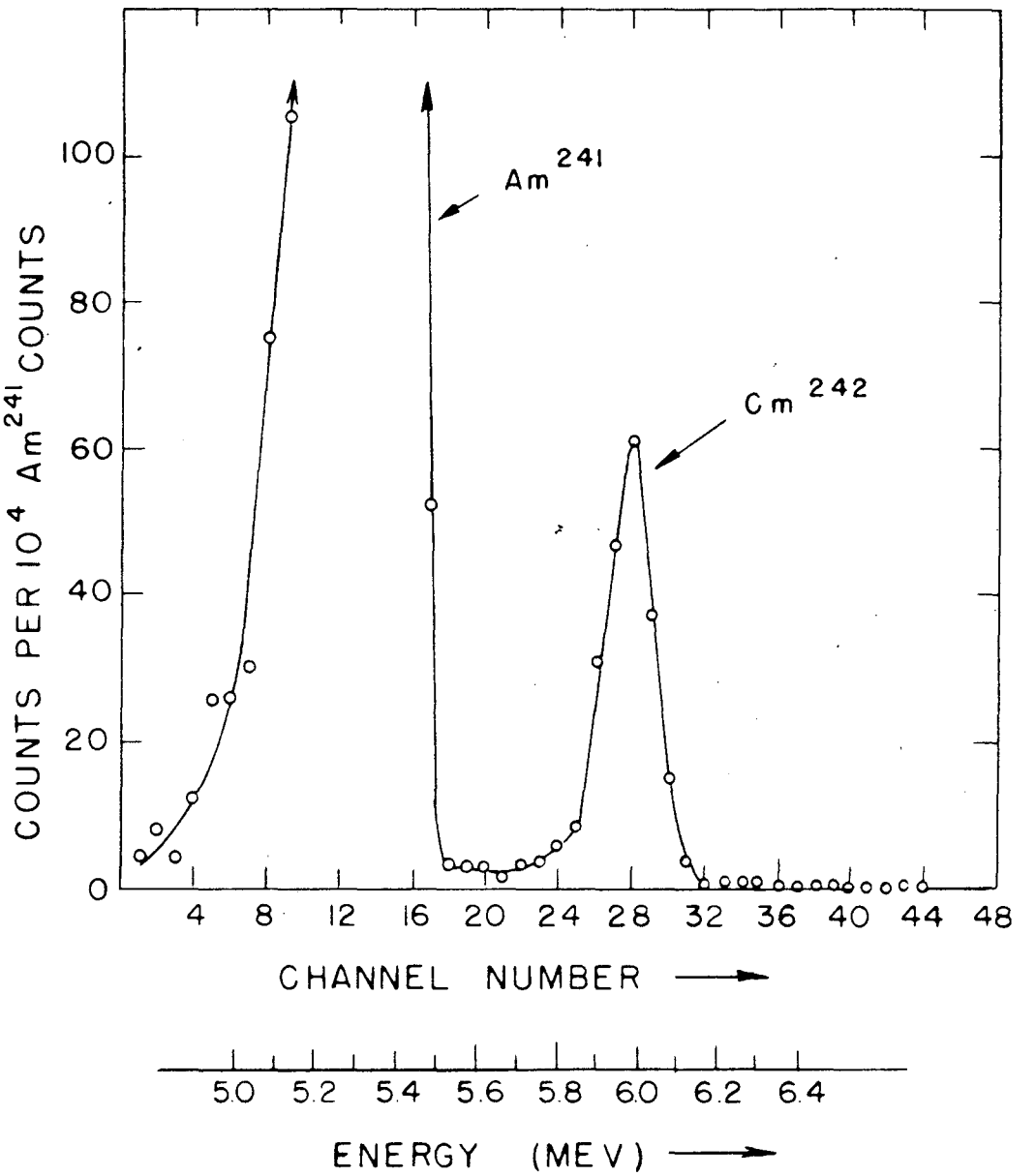

COUNTS PER 10^4 Am^{241} COUNTS
100
80
60
40
20
0
Am^{241}
Cm^{242}
4 8 12 16 20 24 28 32 36 40 44 48
CHANNEL NUMBER
5.0 5.2 5.4 5.6 5.8 6.0 6.2 6.4
ENERGY (MEV)

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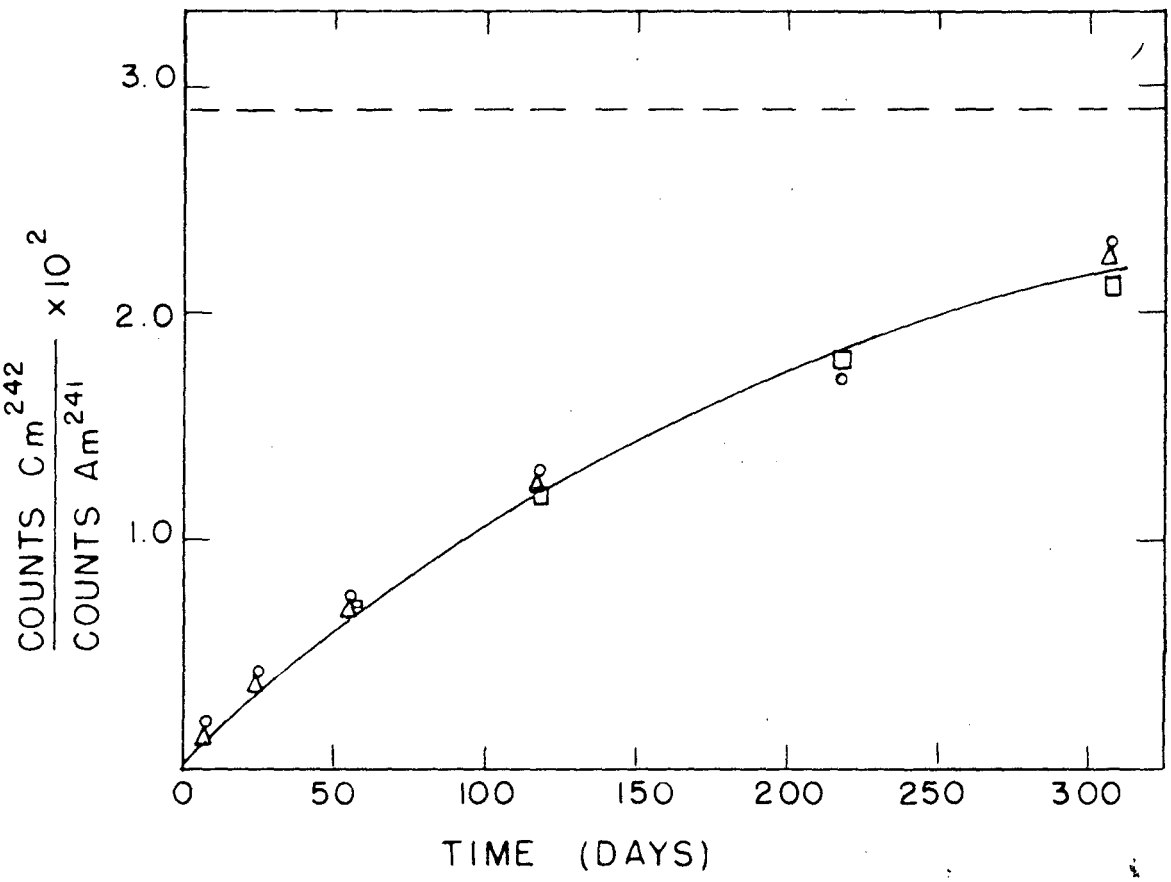
Figure 12 The growth of the Cm^{242} alpha-particles. The solid line indicating the growth expected on the assumption that all the betas observed in Fig. 9 are associated with the decay of Am^{242}. The experimental points are indicated for three different samples followed.


3.0
2.0
1.0
COUNTS Cm242 / COUNTS Am241 × 10^2
0
50
100
150
200
250
300
TIME (DAYS)

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the growth expected on the assumption that all the betas observed are associated with the decay of Am^{242}. These results prove that Am^{242} decays by beta emission to Cm^{242} and indicate very strongly that the 0.5 Mev. beta-particles observed in the americium fraction are associated with this decay.

D. The Isotope Am^{238}. With the development of the more rapid chemical separation discussed in the section on chemistry it became feasible to look for americium isotopes of shorter half-life. The Geiger decay of an americium fraction isolated from the bombardment of Pu^{239} with 50 Mev. deuterons is shown in Fig. 13. The decay was followed down and showed the presence of the 12 and 50-hour americiums. Subtraction of these activities shows very clearly an americium activity of ca. 1.2 hours half-life.

A subsequent bombardment with 17 Mev. deuterons showed the same half-life in good yield. Differential counting with beryllium and lead absorbers indicate that the decay is accompanied by the emission of conversion electrons and x-rays. The production of this activity with 17 Mev. deuterons indicates that its mass number must be greater than 237, since there is good evidence[16,17] that even with the heavy elements the (d,4n) yield is very small at 17 Mev, if indeed it goes at all. In view of its half-life and method of formation this activity is best assigned to Am^{238}.

E. The Isotope Cm^{238}. The curium fraction of a bombardment of Pu^{238} with 80 Mev. alpha-particles showed the presence of about 30 alpha counts/minute with an energy of 6.5 Mev. Fig. 14 shows an alpha pulse analysis of this curium fraction. The decay of the 6.5 Mev. alphas was followed by taking successive pulse analyses and as is shown in Fig. 15 gives a half-life of ca. 2.5 hours.

Another bombardment was made the object of which was to make enough of the activity to milk its alpha decay daughter Pu^{234} and thus definitely establish the mass assignment. However the results of the bombardment were nearly identical

Figure 13 Geiger counter decay of the americium fraction from a bombardment of Pu^{239} with 50 Mev deuterons. The counter used was the same as in Fig. 5.

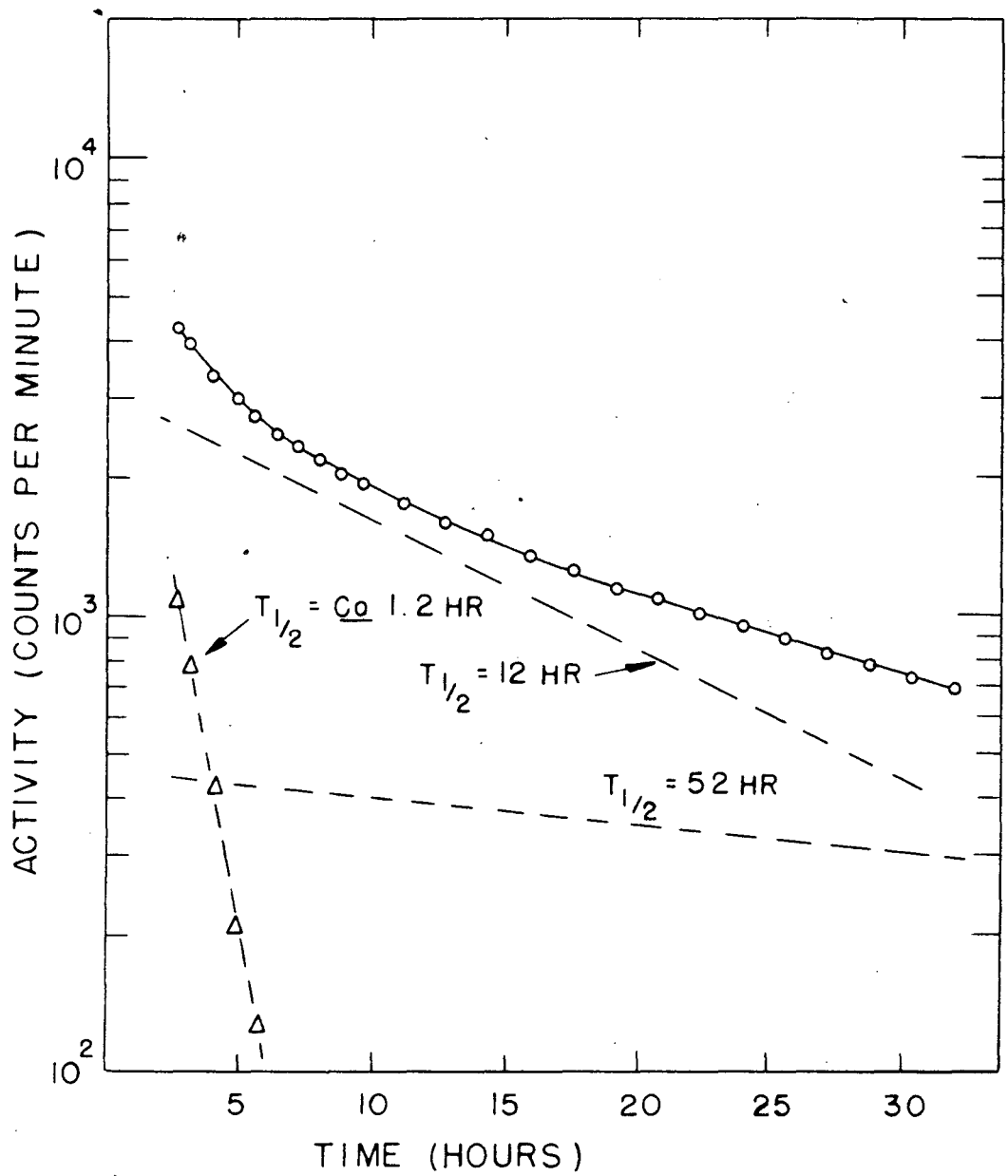
ACTIVITY (COUNTS PER MINUTE)
10^4
10^3
10^2
5
10
15
20
25
30
TIME (HOURS)
$T_{1/2}$ = Co 1.2 HR
$T_{1/2}$ = 12 HR
$T_{1/2}$ = 52 HR

Figure 14 Alpha-particle pulse analysis curve of the curium fraction of a bombardment of Pu239 with 80 Mev. alpha-particles.

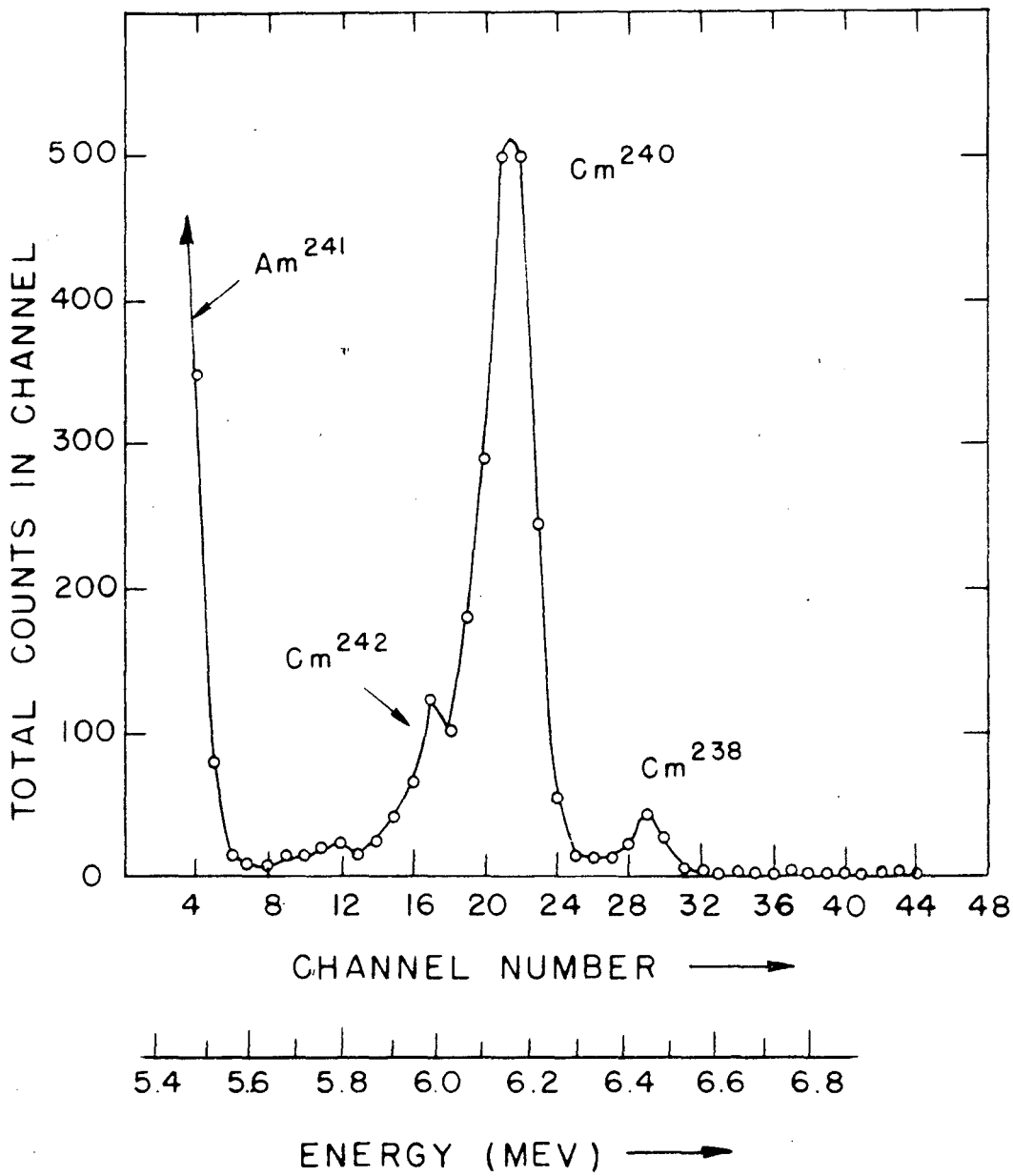
TOTAL COUNTS IN CHANNEL
500
400
300
200
100
0
Am241
Cm242
Cm240
Cm238
4
8
12
16
20
24
28
32
36
40
44
48
CHANNEL NUMBER
5.4
5.6
5.8
6.0
6.2
6.4
6.6
6.8
ENERGY (MEV)

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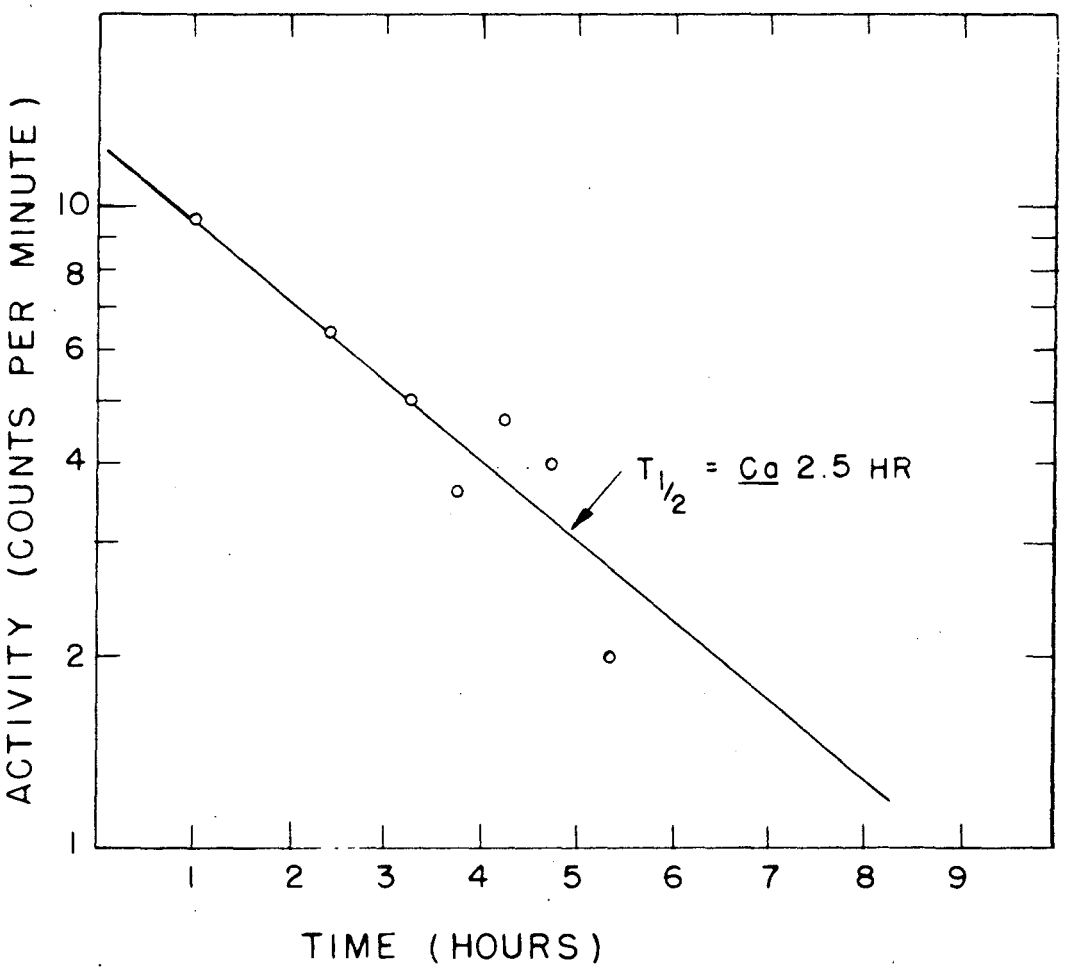
Figure 15 Decay of the 6.5 Mev. alpha-particles followed by taking successive pulse analyses.


ACTIVITY (COUNTS PER MINUTE)
10
8
6
4
2
1
1
2
3
4
5
6
7
8
9
TIME (HOURS)
$T_{1/2}$ = Ca 2.5 HR

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with those obtained in the first case and the low yield precluded the identification of its daughter.

In view of the method of formation, the half-life, and the alpha energy, this activity is best assigned to Cm^{238} although this assignment must be regarded as very tentative.

F. Limit on K-Capture Branching of Cm^{240}. The alpha decay of Cm^{240} to Pu^{236} was definitely established by James[11]. James also found that the decay was accompanied by an unusually large amount of electromagnetic radiation. This suggested the possibility that Cm^{240} decays also by orbital electron capture to Am^{240}.

To check this possibility a sample of Cm^{240} was completely separated from americium and allowed to stand for two weeks. Subsequent milking of an americium fraction from 3000 alpha disintegrations/minute of Cm^{240} gave less than 10 counts/minute of Geiger activity counted at 30% geometry. If one takes Morgan's[10] value of 0.06 hard electrons per disintegration of Am^{240} then the branching decay of Cm^{240} by orbital electron capture to the 50-hour Am^{240} must be less than 20%.

G. Effect of Fission Competition in Alpha and Deuteron Bombardments. One of the most striking facts noted in the bombardment of Pu^{239} with 36 Mev. alpha-particles is t e very nearly equal yields (ca. 10^{-3} barns) of the $Pu^{239}(\alpha,n)Cm^{242}$ reaction and the $Pu^{239}(\alpha,3n)Cm^{240}$. The threshold of the $Pu^{239}(\alpha,3n)Cm^{240}$ reaction is somewhere between 25 and 30 Mev.[18] and thus the yield of this reaction is expected to be at a maximum at about 36 Mev.

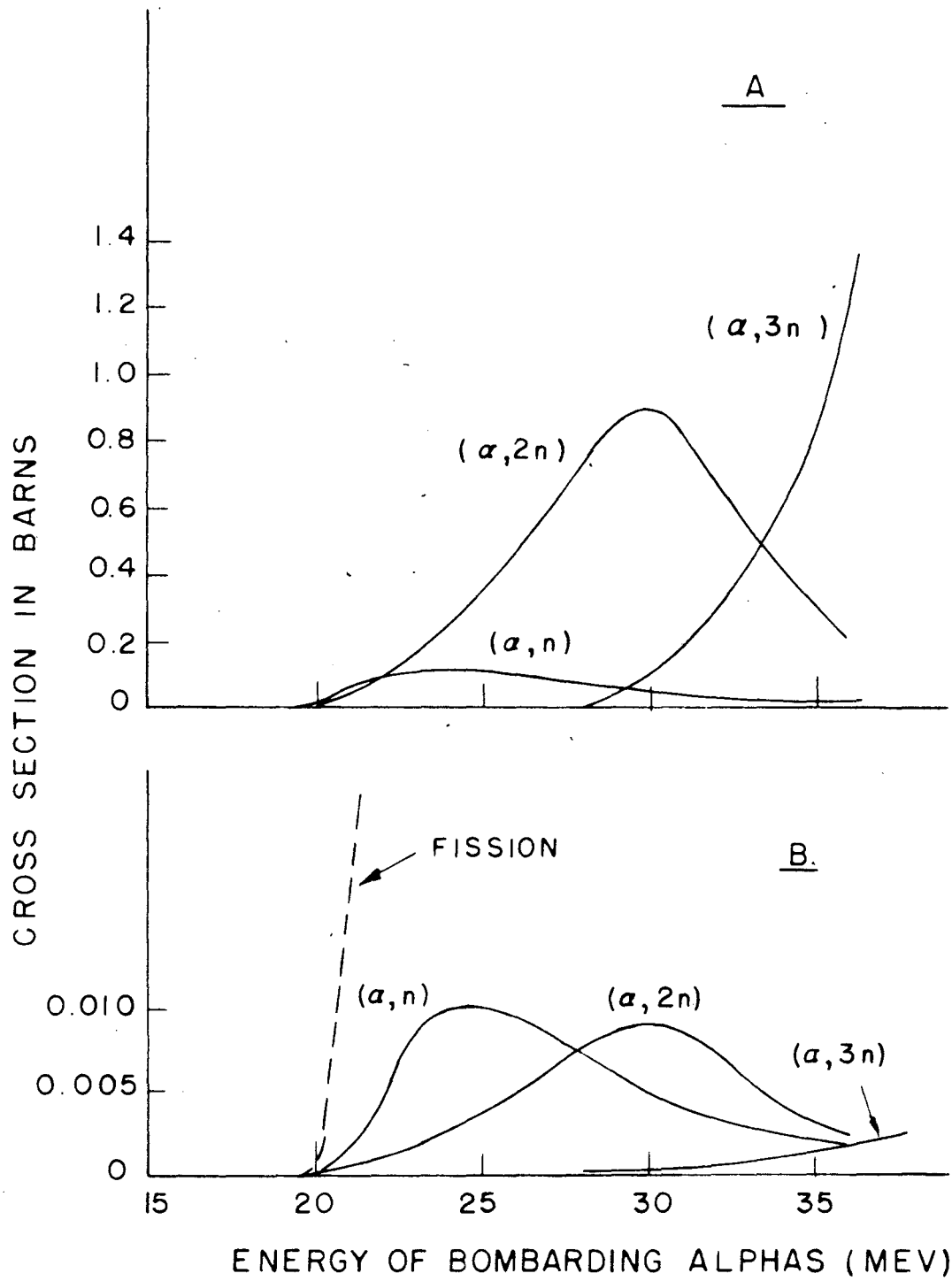
This situation is in direct contrast to that which prevails in bombardments in which fission is not a competing process. Thus, for example, in the rare earth region when the bombarding energy is such that the yield of the $(\alpha,3n)$ reaction is somewhere near its maximum the yield of the (α,n) reaction has already dropped to from 1% to 0.1% of the yield of the $(\alpha,3n)$ reaction[19].

If one neglected the effects of fission one would expect to have an excitation function for the bombardment of Pu^{239} with alpha-particles about like the one shown in Fig. 16A. But if we make the very likely assumption that fission competition takes a much larger fraction of those nuclei which ordinarily go to the (α,2n) product than of those which ordinarily go to the (α,n) product, and a still larger fraction of those which usually give the (α,3n) product, we obtain an excitation function more like the one shown in Fig. 16B. These curves were constructed from the corresponding ones in Fig. 16A by assuming that 90% of the (α,n), 99% of the (α,2n), and 99.9% of the (α,3n) products have been taken by the fission competition. This type of excitation function fits the observed facts that the (α,xn) yields in the Pu^{239} bombardments are all low (ca. 10^{-3} barns) and that the relative yields of the (α,n) and (α,3n) reactions change only slowly with a change in bombarding energy[18].

That fission should take a much larger fraction of those nuclei which ordinarily give the (α,3n) product is very reasonable and probably arises from a combination of two effects. First, those nuclei which usually give the (α,3n) product are ones that remain excited even after the emission of two neutrons (i.e. they emit slow neutrons) and since they remain excited longer the fission competition should be greater. Secondly, with the emission of each successive neutron the Bohr-Wheeler[20] parameter Z^2/A increases slightly making the probability of fission at a given excitation energy somewhat greater.

Deuteron excitation functions for bombardments of Pu^{239} undoubtedly show similar effects since fission is a competing process. Fig. 17A shows the type of excitation functions expected if fission is neglected and Fig. 17B the probable effect of fission. Morgan observed nearly equal yields of the (d,n) and (d,2n) products, with a slight increase in the (d,n) relative to the (d,2n) on increasing the bombarding energy from 13 Mev. to 19 Mev. Fig. 17B accounts for Morgan's

Figure 16 Hypothetical excitation function for the bombardment of plutonium with alpha-particles; A neglecting fission, B including the probable effect of fission competition.


A
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
(α,3n)
(α,2n)
(α,n)
CROSS SECTION IN BARNS
FISSION
B.
0.010
0.005
0
(α,n)
(α,2n)
(α,3n)
15
20
25
30
35
ENERGY OF BOMBARDING ALPHAS (MEV)

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results quite satisfactorily.

ACKNOWLEDGMENT

The work described in this dissertation was performed under the direction of Professor G. T. Seaborg, Mr. A. Ghiorso, Mr. R. A. James, and Mr. D. A. Orth aided in many of the experiments. We wish to thank Professor J. G. Hamilton, Mr. T. M. Putnam, Mr. B. Rossi and their associates in the Crocker Radiation Laboratory for their cooperation in operating the 60-inch cyclotron; and Mr. D. C. Sewell, Mr. J. T. Vale, and the operating crew of the 184-inch cyclotron for their aid in the bombardments performed at the Radiation Laboratory of the University of California. We also wish to thank the operating crews of the uranium-graphite chain reacting units at the Hanford Engineering Works for their aid in the neutron irradiation of the americium samples. This research was carried out under the auspices of the Atomic Energy Commission.

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