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AN INVESTIGATION OF THE ISOTOPES OF AMERICIUM AND CURIUM

Gary Hoyt Higgins

(Thesis)

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AN INVESTIGATION OF THE ISOTOPES OF AMERICIUM AND CURIUM

Gary Hoyt Higgins
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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ABSTRACT

Isotopes of americium and curium with mass numbers less than 242 have been produced by cyclotron bombardment techniques, and several of their nuclear properties have been investigated. The partial alpha half-lives of Am^{239}, Cm^{241}, and Cm^{240} and the partial half-life for spontaneous fission of Cm^{240} were measured. The alpha decay daughter of Cm^{238} was found and evidence for the discovery of Am^{237} (an ~1 hour electron capture activity) and Cm^{239} (an ~10 hour electron capture activity) has been presented. An Appendix indicating the conditions for separation of the plus three actinides by selective elution from ion exchange resin with citric acid has been included.

AN INVESTIGATION OF THE ISOTOPES OF AMERICIUM AND CURIUM

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I. INTRODUCTION

The elements americium (Am) and curium (Cm) were discovered nearly simultaneously several years ago, and at that time their chemistry was indicated, and the properties of several of their isotopes were demonstrated.[1,2] Since that time there has been nearly continuous work pointed toward recognizing new isotopes of different mass number and refining the knowledge of properties of those isotopes previously known. A summary of the work previous to this publication is as follows.

Am^{243} emits 5.27 Mev alpha particles with an approximate half-life of 10^4 years.[3-5]

Am^{242m} decays by 0.628 Mev β^- transition, isomeric transition, and electron capture with a 15.7 hour half-life.[2,6-9]

Am^{242} emits alpha particles in 1 percent of the disintegrations and decays by 0.593 Mev β^- transition with a 100 year gross half-life.[6-9]

Am^{241} emits 5.47 Mev alpha particles with a 472 year half-life.[2,10,11]

Am^{240} undergoes electron capture with a 50 hour half-life.[2,3]

Am^{239} undergoes electron capture with a 12 hour half-life and emits 5.75 Mev alpha particles in ~0.1 percent of the transitions.[2,3]

Am^{238} undergoes electron capture with an approximate half-life of 1.5 hours.[3]

Cm^{244} emits alpha particles of 5.79 Mev with an approximate half-life of 20 years.[12-14]

Cm^{243} emits alpha particles of 5.79 and 5.89 Mev with a half-life of approximately 40 years.[12-15]

Cm^{242} emits 6.11 Mev alpha particles with a 162.5 day half-life.[1,16]

Cm^{241} undergoes electron capture with an approximate 35 day half-life.[1,17]

Cm^{240} emits 6.26 Mev alpha particles with a half-life of 26.8 days and less than 0.5 percent of the transitions are electron capture.[1,17]

Cm^{238} emits 6.5 Mev alpha particles in <10 percent of the transitions and decays with a gross half-life of ~2.5 hours.[4]

Chemically these elements represent the sixth and seventh members of the actinide series and experimentally their most stable oxidation state in aqueous solutions is the plus three. Americium may be oxidized to the (VI) state with the most powerful oxidizing agents such as ceric or argentic ion, while curium is probably not oxidized or reduced in aqueous solutions.

In order to work with the small quantities of materials usually produced in bombardments (in the order of 10^8 atoms), methods such as coprecipitation with a similar element, in this case a rare earth such as lanthanum, solvent extraction or ion exchange techniques may be used. A summary of the compounds with which americium and curium will coprecipitate and the approximate percentages of carrying is somewhat as follows: rare earth trifluoride, ~100 percent; rare earth oxalate, ~100 percent; any heavy metal hydroxide, ~100 percent;

zirconium phosphate, ~10-20 percent; rare earth iodate, 100 percent; and zirconium iodate, ~20 percent. The ions of these elements form more or less strong complexes with carbonate, high concentrations of chloride or hydrogen chloride, cyanide, thiocyanate, citrate and tartrate ions and organic chelating agents such as thenolytrifluoroacetone (TTA) and versine.

With these facts in mind the present work was begun with the object of investigating as many of the decay properties of the known neutron deficient isotopes as possible, and of finding any new one available to the present chemical and physical techniques in order to correlate the data with the existing concepts of nuclear decay systematics and general energetics of this region.

II. EXPERIMENTAL METHODS

A. Bombardment Techniques

The targets used in these bombardments were similar to those previously used; those used in the 60-inch cyclotron were the "pistol grip" type[15] and a double platinum foil envelope provided with a filtered air leak was constructed for bombardments in the 184-inch cyclotron. In the latter case the envelope was prepared by folding a piece of platinum foil and welding the edges. A copper tube was soft soldered into the side of the envelope which was to be away from the beam, and the tube led to a small filter which opened to the cyclotron vacuum during bombardment. After the material to be bombarded was placed inside the remaining edge was clamped between two copper blocks.

The target material was plutonium trifluoride which was prepared by precipitating plutonium(III) from solution with aqueous hydrofluoric acid. The plutonium trifluoride was placed in the target container in an acetone slurry and dried to a cake under a heat lamp.

The energy of the incident beam was determined by the radius at which the bombardments were made and corrections were made for degradation of beam energy in the platinum containers.[18]

B. Chemical Procedures

Dissolution of Target.-- In those bombardments during which the temperature of the target assembly did not exceed 500° - 800° C, the plutonium trifluoride remained unaltered and was simply dissolved in concentrated hydrochloric or nitric acid to which a little boric acid had been added to remove the fluoride ion by formation of

fluoboric acid. When higher temperatures were reached the plutonium trifluoride was partially converted to a form of oxide which tends to be more passive, and it was necessary to fume the targets with concentrated sulfuric acid and then dissolve the plutonium sulfate in dilute nitric acid.

Isolation of a Rare Earth Fraction.-- Lanthanum(III) carrier was added to the solution of plutonium and was separated along with the (III) actinides and the lanthanides by several methods. The basis for all of these was the fact that the most stable oxidation state of the lanthanum (and the actinides americium and curium) is (III) while plutonium is most stable in the (IV) and is easily oxidized to higher states which do not form insoluble fluoride precipitates.

One method is to oxidize the plutonium with MnO_4^- ion to a mixture of the (IV) and (VI) valences, precipitate zirconium phosphate to carry away the plutonium(IV), and then precipitate lanthanum trifluoride away from the remaining plutonyl ion. Several variations were used to prevent uniform errors from impurities.

Actinide-Rare Earth Separation.-- The actinide-rare earth separation was brought about by eluting their mixture on a Dowex-50 cation resin column with 13 M HCl. At this acid concentration the plus three actinides form fairly strong chloride complexes while the (III) lanthanides form either very weak complexes or none at all. Since the net positive charge on an ion complexed with Cl^- is less than one which remains hydrated, the actinides elute rapidly through a sulfonic acid type ion exchange resin while the lanthanides and other uncomplexed ions are strongly adsorbed.[19,20]

For those bombardments in which no curium was produced, the above described chemical procedures sufficed for the production of pure samples; in the instances where the bombarding particles were helium ions the americium and curium obtained from these procedures were separated from each other by selectively eluting them from Dowex-50 cation resin in a jacketed column maintained at 87° C with ethylene trichloride vapors. The elutriant was 0.25 M citric acid adjusted to pH 3.50 with ammonium hydroxide. In order to establish the best conditions for this separation it was studied to some extent and the data are included in the Appendix.

C. Counting Techniques and Equipment

Alpha active samples were volatilized onto platinum disks and examined with the 48-channel pulse analyzer first described by Ghiorso et al.[21] and since modified by A. Ghiorso in this laboratory. The beta particles and conversion electrons were counted in a windowless proportional counter whose threshold and counting efficiency were determined by counting samples of plutonium which had various amounts of Pu^{241} and $Pu^{239-240}$ in them as determined by mass spectrographic analysis. The counting efficiency for the electron capture process was assumed from these measurements, and others[22,23] to be 60 percent in this instrument. This value was used consistently for lack of any method for determining the efficiency of counting an electron capture transition. Experimentally under these conditions an alpha disintegration is counted with 100 percent efficiency since the recoil nucleus from the alpha disintegration has sufficient energy to cause an ionization pulse.

The ratio of electron capture to alpha particle emission of several isotopes was found by dividing the disintegration rate measured in this instrument by the disintegration rate of the alpha group or groups attributed to the activity in question. The latter rate was determined in the alpha particle pulse analyzer described above. The partial alpha half-life was found from the gross half-life and the ratio of electron capture to alpha particle emission. Since both calculated numbers are dependent on the counting efficiency of the flow counter, the results listed below are only as precise as that number. It is probable that the errors from this source are smaller than a factor of two.

The gamma and x-radiations of some of the samples were studied in a sodium iodide crystal scintillation counter which was attached to a 20 channel electronic pulse analyzer.[24] The counting efficiency was calculated from geometrical considerations only. One of the isotopes examined (Cm^{240}) was studied in an ionization type fission counter connected to a recorder. This instrument is described elsewhere.[25]

III. RESULTS AND CONCLUSIONS

A. Am^{240}

This isotope was observed in every bombardment of Pu^{239} and was produced by the (d,n) and (α,p2n) reactions. It decayed with a mean half-life of 53 hours and its decay was accompanied by electrons and x-rays, but the gamma ray previously reported[7] was not observed in any large abundance. The upper limit for any gamma ray of energy greater than 0.7 Mev may be set at 1×10^{-4} per disintegration using assumed counting efficiencies discussed above. Alpha pulse analysis of samples containing this isotope are not conclusive as regards the possibility of alpha branching since the alpha energy in question might well be nearly the same as that of Am^{241} and would be obscured by this alpha group. Am^{241} was present in all of the samples since it continually grows from the small amount of Pu^{241} present in the target material. From the absence of any decay of the alpha peaks including the 5.47 Mev energy region, the limit for the electron capture to alpha emission ratio is calculated to be 290 which corresponds to a lower limit of 1.5 years for the partial alpha half-life. This is in agreement with expectations for this odd-odd isotope on the basis of alpha systematics.[26]

The limits for the alpha energy region around 5.6 Mev are much lower: 5.6×10^{-5} alpha particles per disintegration or a minimum alpha half-life of 100 years. The disintegration energy expected from mass conservation is 5.6 Mev.

After one sample had decayed for a time it was examined carefully in the alpha pulse analyzer for alpha particles of Cm^{240} (6.26 Mev)

and since none were observed, an upper limit of the decay by β^- emission of 2×10^{-5} per disintegration was determined.

B. Am^{239}

Produced by the (d,2n) and (α,p3n) reactions on Pu^{239}, this isotope decays with a 12 hour half-life by orbital electron capture and alpha particle emission. Ten percent or more of the electron capture disintegrations are accompanied by a gamma ray of 0.3 Mev as determined by the scintillation counter described above.

Pulse analysis of the samples of americium show several alpha peaks as seen in Fig. 1 and the peak at 5.75 Mev was shown to decay with the 12 hour half-life and hence is assigned to this isotope. From simultaneous analysis of the decay curves plotted from the proportional counter data (see Fig. 2) and the alpha disintegration rate determined with the pulse analyzer, the ratio of electron capture to alpha particle emission is found to be $1.6 \pm 0.4 \times 10^4$. This leads to a partial alpha half-life of 22 ± 5 years. These figures represent the average of four determinations, and the spread is the maximum observed deviation.

C. Am^{238}

This isotope was previously assigned a 1.5 hour half-life, but in view of the discussion presented below this half-life probably represents a mixture of two isotopes (Am^{237} and Am^{238}) whose half-lives are very close to each other.

When Pu^{239} was bombarded with 18 Mev deuterons [an energy insufficient to cause an appreciable yield from the (d,4n) reaction

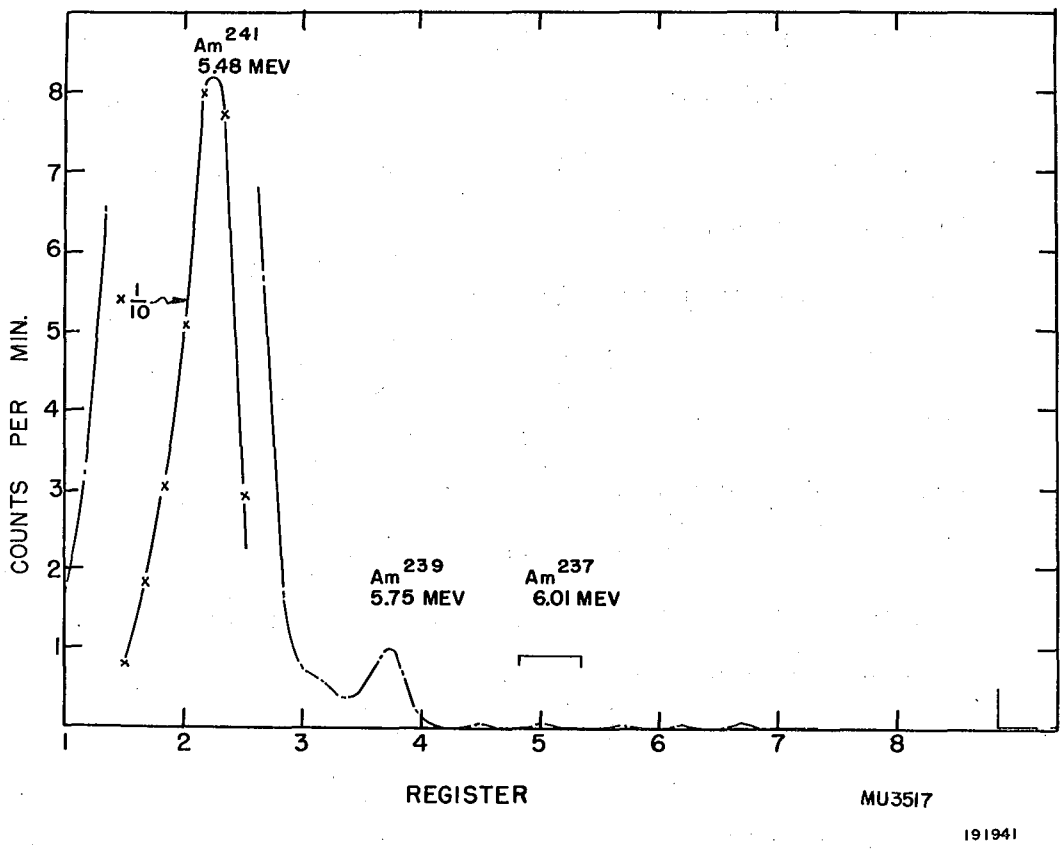

Fig. 1. Pulse analysis of alpha particles of americium produced by bombarding Pu^{239} with 19 Mev deuterons. Registers are numbered in groups of six.

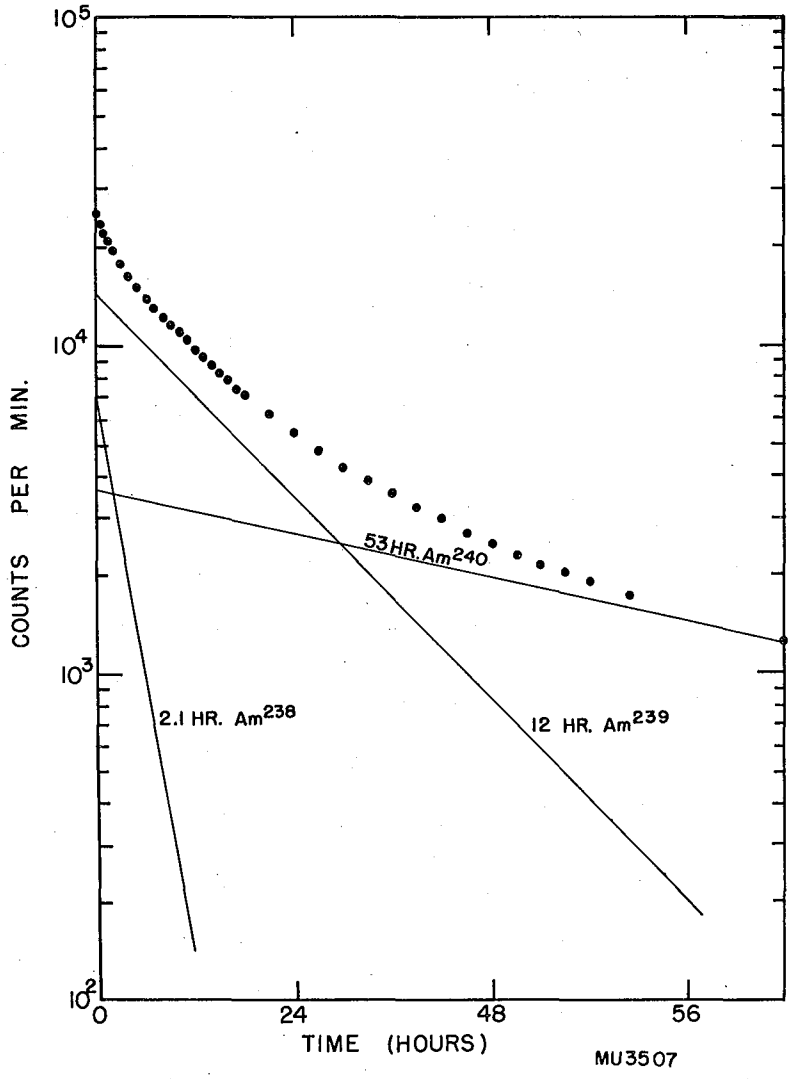

Fig. 2. Decay of americium produced by bombardment of Pu^{239} with 19 Mev deuterons. The measurements were made in a windowless methane flow counter.

but sufficient to yield a large (d,3n) cross section], the short half-life component obtained by subtracting all the longer lived activities decayed with a 2.1 hour period (as shown in Fig. 2). When the alpha pulse analyses (one of which is reproduced in Fig. 1) were examined, no group above 5.80 Mev was seen and the lower limit for the electron capture/alpha emission ratio was set at 1.6×10^5, corresponding to a minimum alpha half-life of 35 years. Because of the time limitation imposed by this short half-life the samples containing this activity were not examined with the gamma scintillation counter.

D. Am^{237}

When Pu^{239} was bombarded with 30-50 Mev deuterons the short half-life observed by analysis of the decay curves shown in Fig. 3 was about 1.5 hours. Pulse analyses of these samples (Fig. 4) indicated an alpha group of about 6.01 Mev which decayed with an ~1.3 hour half-life and since no alpha particles were seen from Am^{238}, this activity was assigned to Am^{237} on the basis of the alpha systematics.[26] Assuming that the (d,3n) and (d,4n) reactions had equal yield at these energies so that half the number of atoms decaying with the short half-life were Am^{237}, the electron capture to alpha emission ratio is calculated in the manner discussed above to be $1.1 \pm 0.4 \times 10^4$ which corresponds to a partial alpha half-life of 1.6 ± 0.5 years. These values are the average of two determinations and the limits represent the maximum deviation instead of the probable error.

E. Cm^{241}

This isotope was produced by the (α,2n) reaction on Pu^{239} with helium ions of from 22-38 Mev and by the (p,n) reaction on Am^{241}.

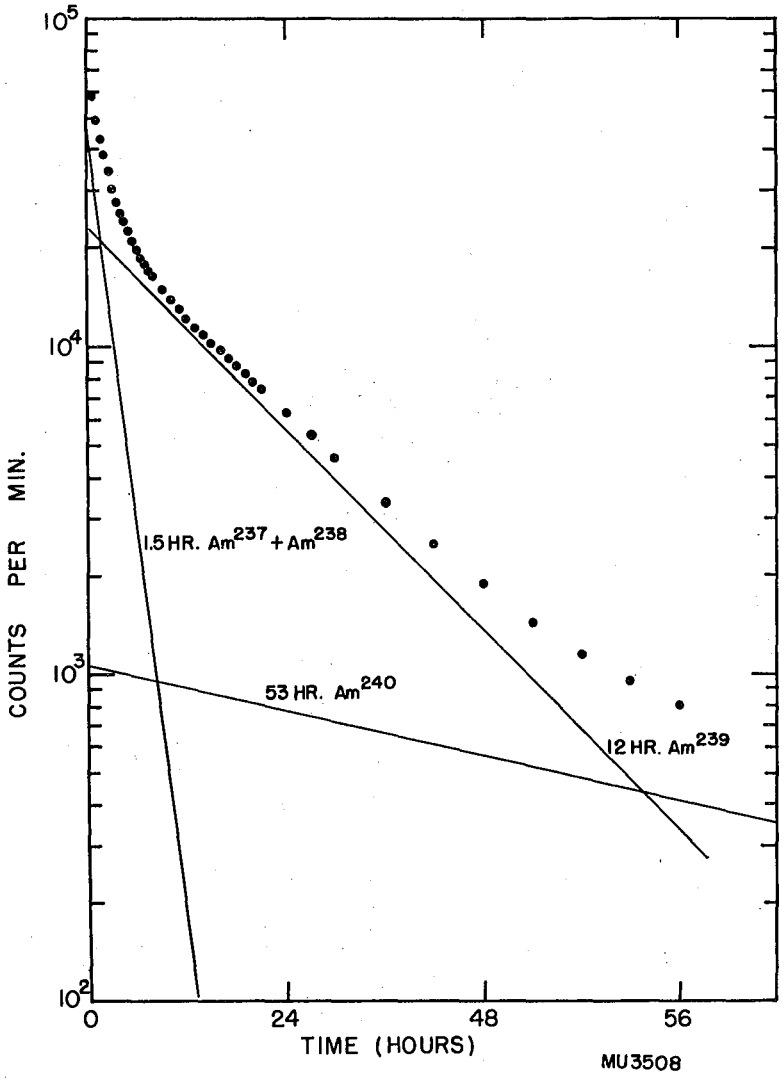

Fig. 3. Decay of americium produced by bombardment of Pu^{239} with 30-50 Mev deuterons. The measurements were made in a windowless methane flow counter.

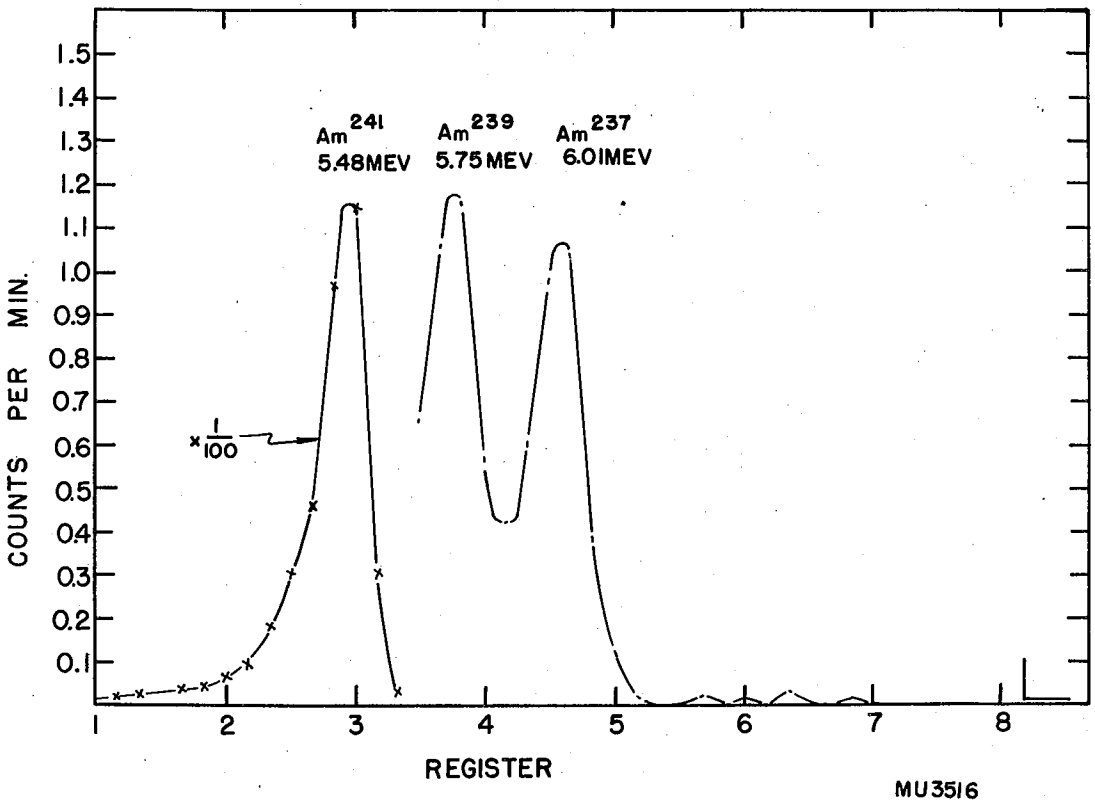

Fig. 4. Alpha pulse analysis of particles from americium produced by bombarding Pu^{239} with 30-50 Mev deuterons. The registers are numbered in groups of six.

It decays with a 35 day half-life (Figs. 5 and 7) emitting alpha particles of 5.90 Mev (Fig. 6). As seen from Fig. 6 it is produced nearly free from Cm^{240} when the bombarding helium ions have about 27 Mev of kinetic energy.

The ratio of the electron capture rate to the observed rate of emission of alpha particles in the 5.90 Mev group is $2.1 \pm 0.3 \times 10^2$ which corresponds to a partial alpha half-life of 20 ± 3 years. Since Cm^{242} was present in all of the samples, it is possible merely to set a lower limit of 30 years on the partial alpha half-life of any group in the energy range of 6.0 to 6.2 Mev which is the energy expected for the ground state transition by closed cycle considerations.

F. Cm^{240}

Produced by the $(\alpha,3n)$ reaction on Pu^{239}, and the $(C^{12},4n)$ reaction on Th^{232}, this isotope decays by the emission of 6.26 Mev alpha particles with a 26.8 day half-life. These decay characteristics are indicated in Figs. 7 and 8. The Pu^{236} indicated in Fig. 8 is the product of the alpha disintegration of Cm^{240}.

Several samples were allowed to stand for seven or eight days, and americium was then separated from them and examined in the proportional counter. Since no decay attributable to the 53 hour Am^{240} or any other activity of shorter half-life was observed, the maximum ratio of electron capture to alpha particle emission is 5×10^{-3}. The electron capture decay energy calculated from known decay data is 0.05 ± 0.20 Mev so a very long half-life is expected.

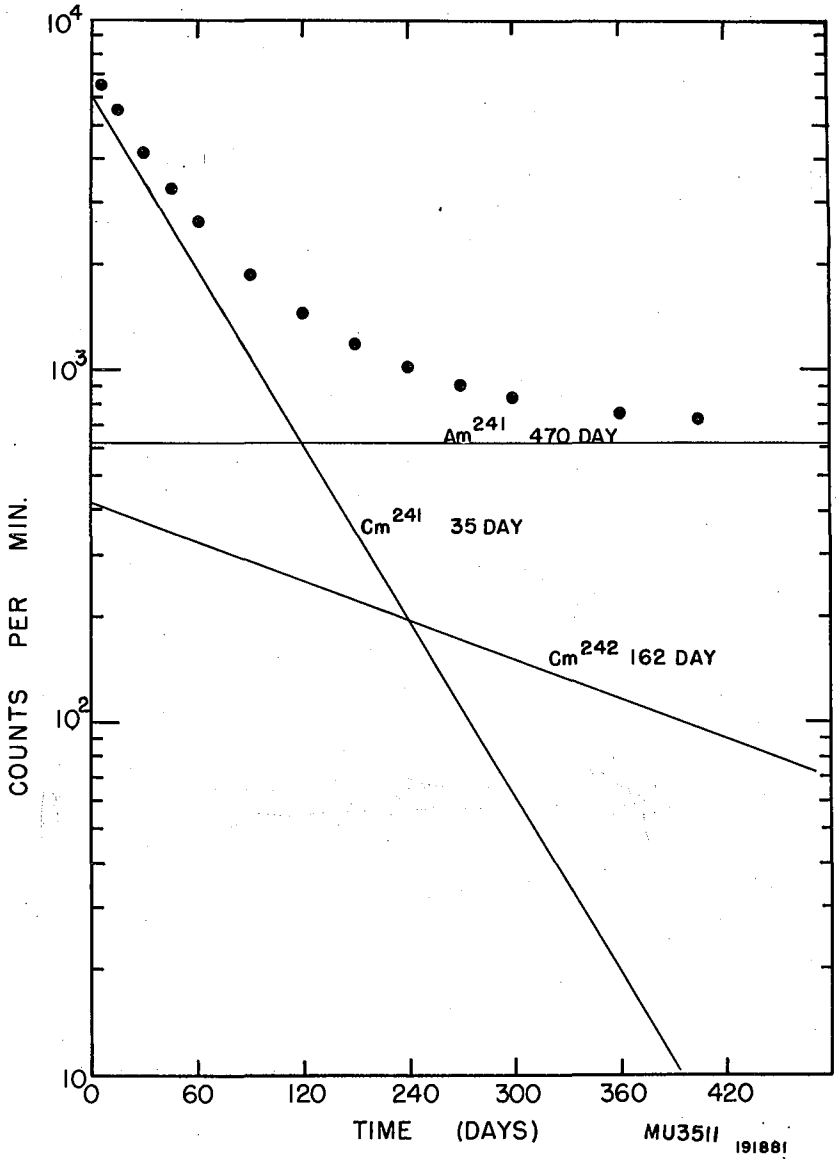

Fig. 5. Decay of curium produced by bombarding Pu^{239} with 27 Mev helium ions. The points were measured in a windowless methane flow counter.

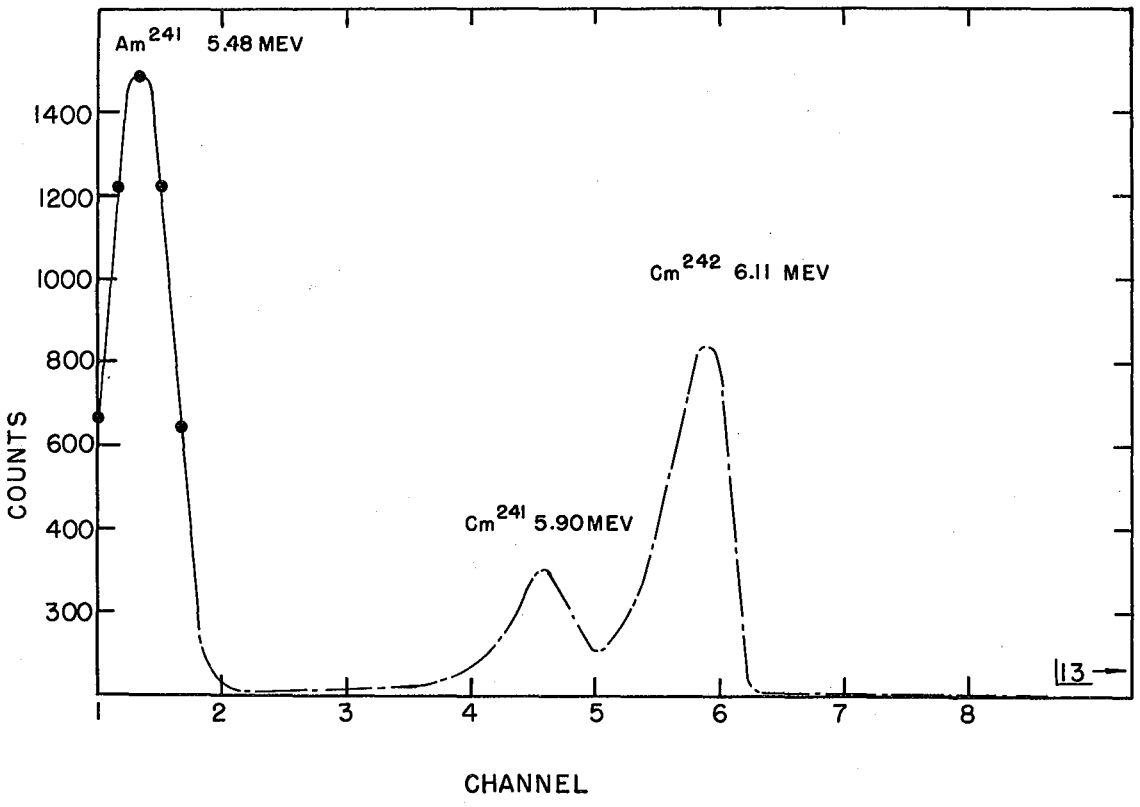

Fig. 6. Alpha pulse analysis of curium produced by bombarding Pu^{239} with 27 Mev helium ions. Each channel represents six registers.

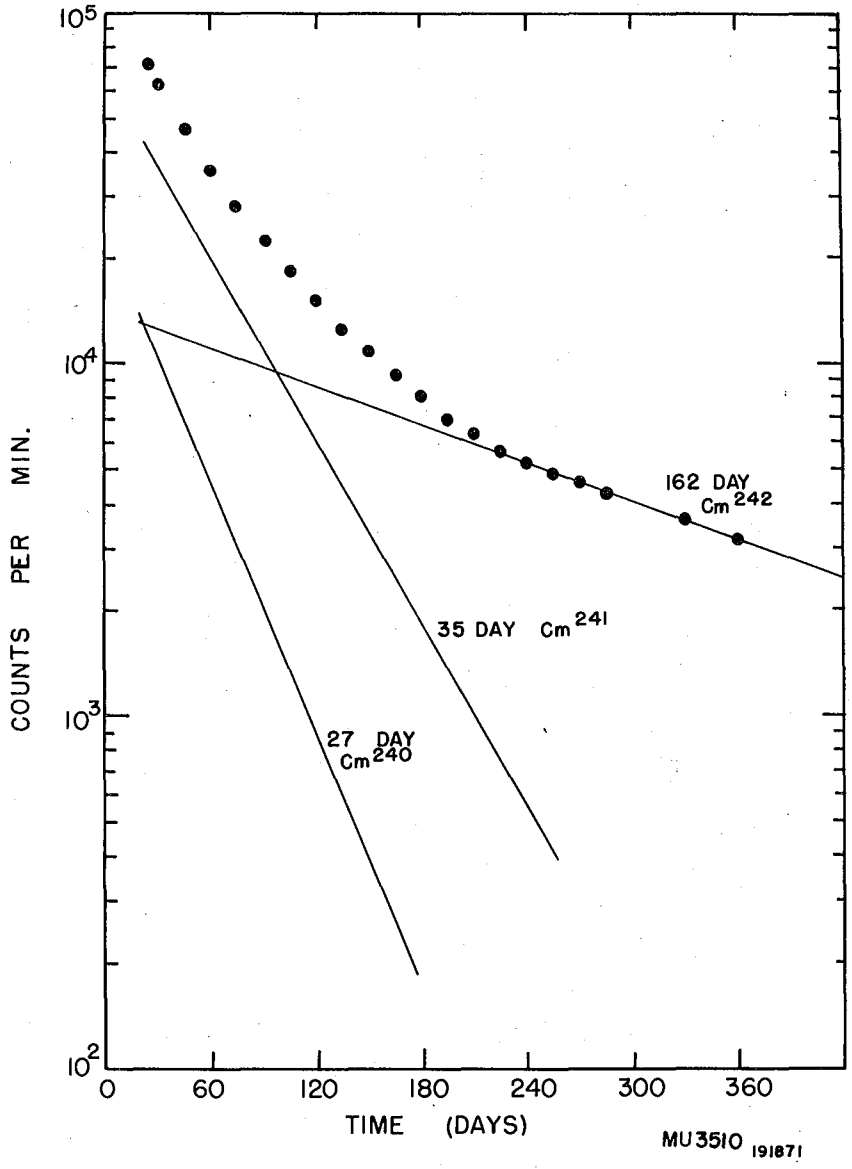

Fig. 7. Decay of curium produced by bombarding Pu^{239} with 38 Mev helium ions. Measurements were made in a windowless methane flow counter.

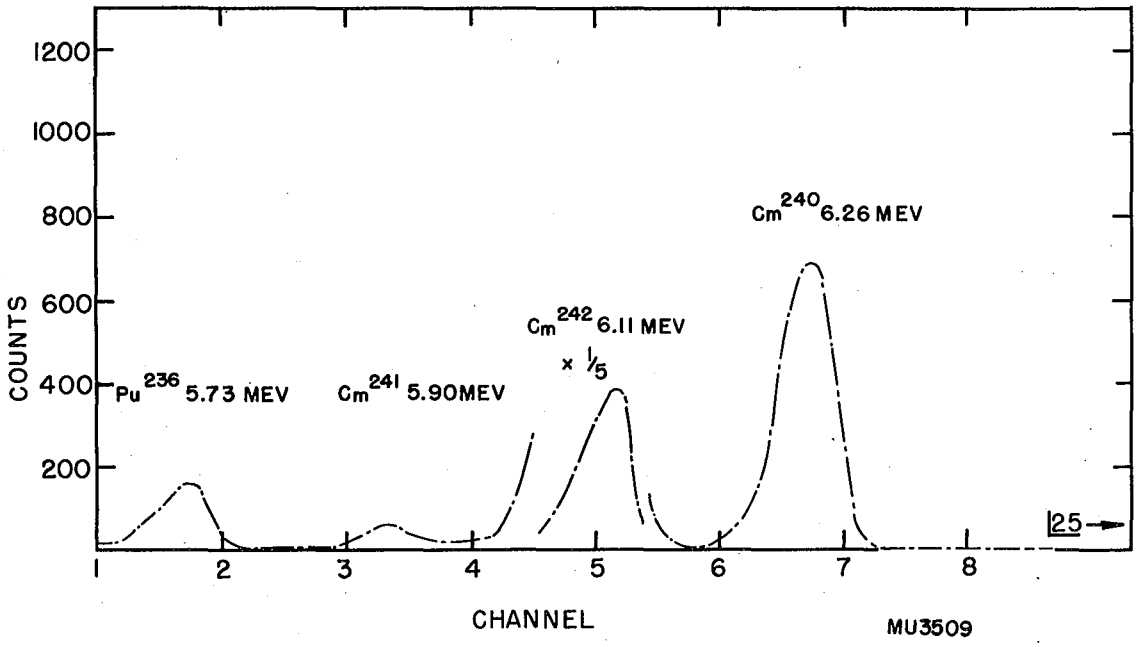

Fig. 8. Pulse analysis of the alpha particles of curium produced by bombarding Pu^{239} with 38 Mev helium ions. The Pu^{236} has grown from the alpha decay of the Cm^{240}. Registers are numbered in groups of six.

One sample was purified and examined in the fission counter described above. There were initially 15 events per day, and these were plotted with time as the ordinate (Fig. 9) and also analyzed statistically as a check of the apparatus. The sample was counted in a low geometry alpha counter and the alpha pulses of a small fraction were electronically analyzed. The spontaneous fission half-life calculated from these observations is 1.9×10^6 years.

The fact that the fission rate decreases with an approximate half-life of 1 month does not exclude the possibility of Cm^{241} contributing to the observations but since this isotope is of an even-odd type it seems highly unlikely that it could have contributed a measurable amount to the fission rate even though there were approximately equal numbers of atoms of Cm^{240} and Cm^{241} in the sample. From the considerations concerning spontaneous fission described by Seaborg[27] it may be concluded that Cm^{240} would undergo this sort of disintegration much more frequently than Cm^{241}.

G. Cm^{239}

Cm^{239} was produced from Pu^{239} by the $(\alpha,4n)$ reaction with 70-80 Mev helium ions. When the known activities of $Cm^{240-241}$ and Cm^{238} were subtracted from the counts measured in the windowless proportional counter of a sample of curium which had been twice purified from americium, there remained a growth and decay curve which was fitted by trial and error with a 10-12 hour parent producing a 12 hour daughter (Fig. 10). Since there was no indication of the 50 hour Am^{240} after the shorter lived activities had decayed, there was less than 5 percent contamination by americium and the 12 hour activity

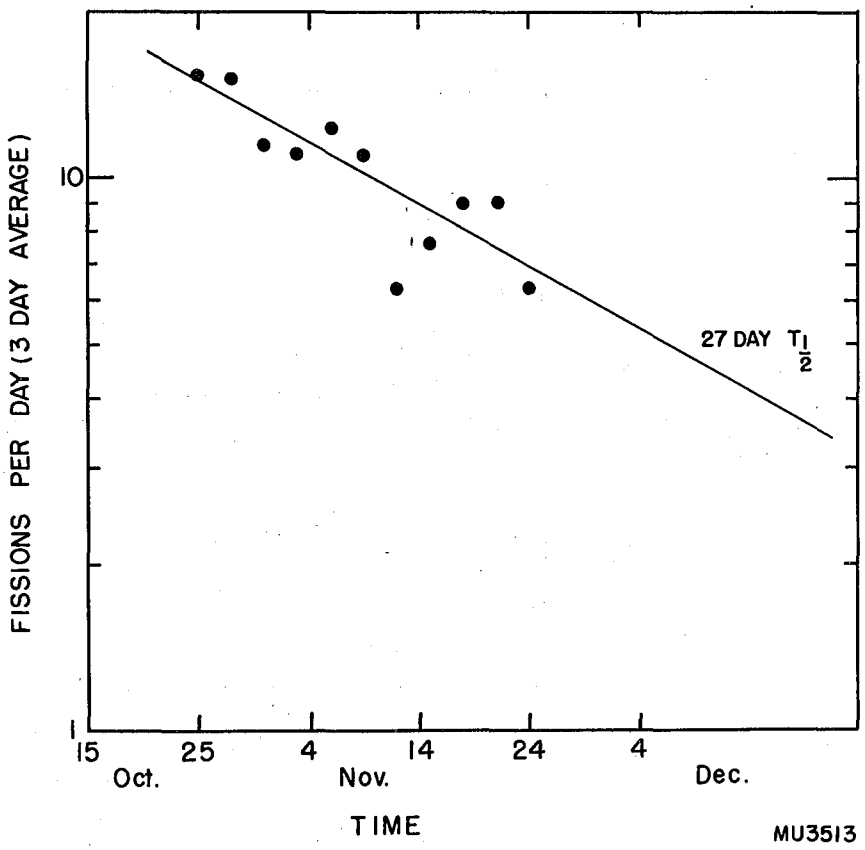

Fig. 9. Decay of the number of spontaneous fission pulses day observed in a sample containing Cm^{240}, Cm^{241}, and Cm^{242}.

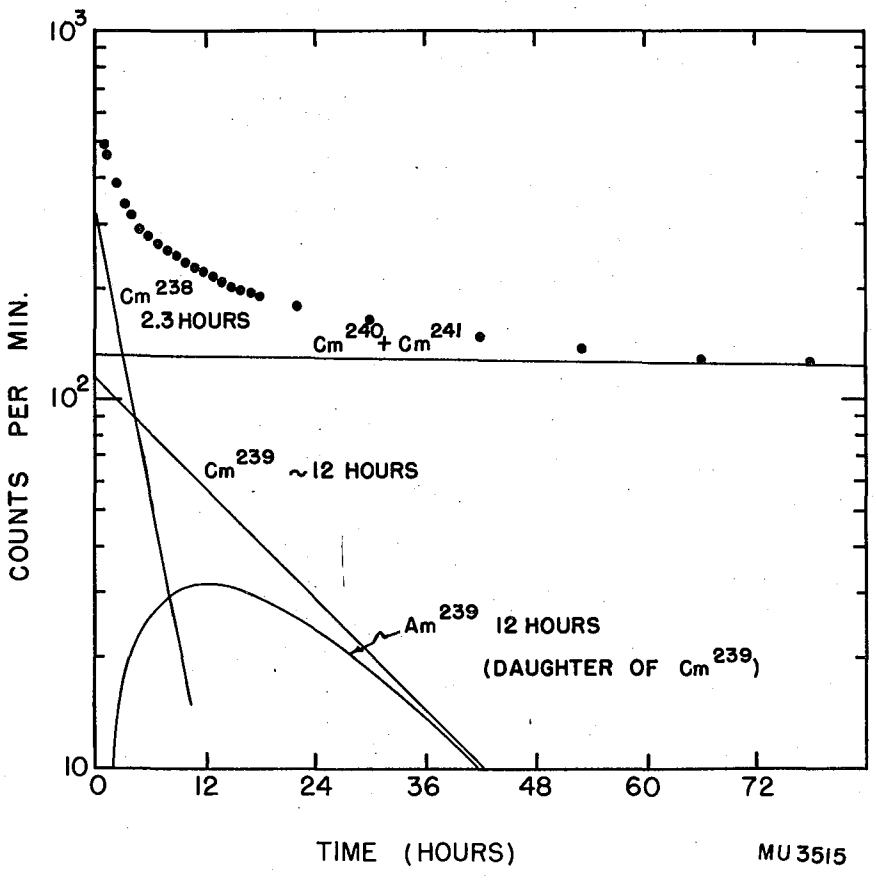

Fig. 10. Decay of curium produced by bombarding Pu^{239} with 70-80 Mev helium ions. Am^{239} is shown growing from Cm^{239}.

which grew in was Am^{239} produced by the decay of the 12 hour Cm^{239}.

H. Cm^{238}

This isotope was produced by the $(\alpha,5n)$ reaction on Pu^{239} and was found to have a 2.3 hour half-life brought about by decay by electron capture and alpha particle emission. The energy of the alpha particles measured with the alpha pulse analyzer was 6.52 ± 0.05 Mev and the electron capture to alpha particle emission ratio was calculated from data from the pulse analyzer and windowless proportional counter to be 240 ± 50 based on counting assumptions mentioned above. The partial alpha half-life calculated from this ratio is 23 ± 5 days based on four determinations. The decay and pulse analysis are shown in Figs. 10 and 11. One sample was allowed to stand until more than 90 percent of the Cm^{238} had decayed at which time a sample of plutonium was chemically isolated and examined in the alpha pulse analyzer.

The amount of Pu^{234} observed was consistent with the number of Cm^{238} alpha emission events and a branching ratio for Pu^{234} of approximately 20.

A fraction of the curium isolated from another bombardment was deposited in a photographic emulsion and allowed to decay for two days before the emulsion was developed. Five events consisting of two coincident tracks were observed. This number was consistent with the amount of activity of Cm^{238} and branching ratio of Pu^{234}.

Since all curium isotopes of mass number 239 to 243 decay by alpha emission to daughters which have long partial alpha half-lives, it is highly improbable that they would produce even one double track

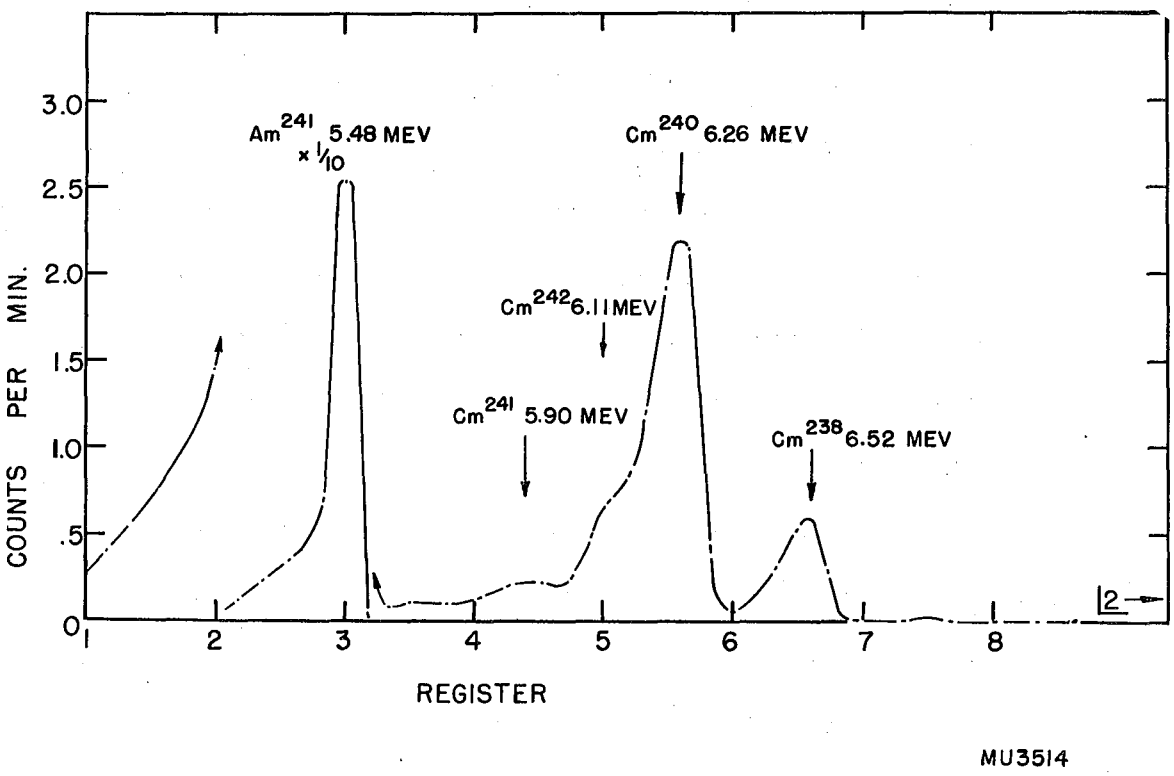

Fig. 11. Pulse analysis of the alpha particles of curium produced by bombarding Pu^{239} with 70-80 Mev helium ions. Registers are numbered in groups of six.

in the short exposure time so the multiple events observed must have been due to the

$$Cm^{238} \xrightarrow{\alpha} Pu^{234} \xrightarrow{\alpha} U^{230} \xrightarrow{\alpha}$$

decay sequence. From the track lengths it is possible to calculate the energies and these were 6.5 ± 0.2 Mev and 6.1 ± 0.2 Mev corresponding to Cm^{238} and Pu^{234}. One photograph of these tracks is shown in Fig. 12. This track was chosen because it was parallel to the plane of focus. With the optical system used the magnification was such that there were 0.80 divisions per micron and the relation between track length and energy in the emulsion used is

$$E_\alpha = 1.95 + 0.15\ S$$

where E_α is the energy in Mev and S is the track length in microns.

I. General

From examining the aforementioned alpha systematics[26] in comparison with the partial alpha half-lives and energies of alpha emission listed above several things are apparent.

Since americium has an odd number of protons (95) and the last proton is presumed to be in the highest nuclear level, the odd-even alpha emitters, $Am^{243,241,239,237}$, must emit a particle which comes from either splitting the last pair of protons or taking the last pair and leaving the odd one in an excited state, or in some other way forming the alpha particle from unpaired or deep lying nucleons. Under these conditions the most abundant alpha group is usually hindered in the transition in that the available disintegration energy

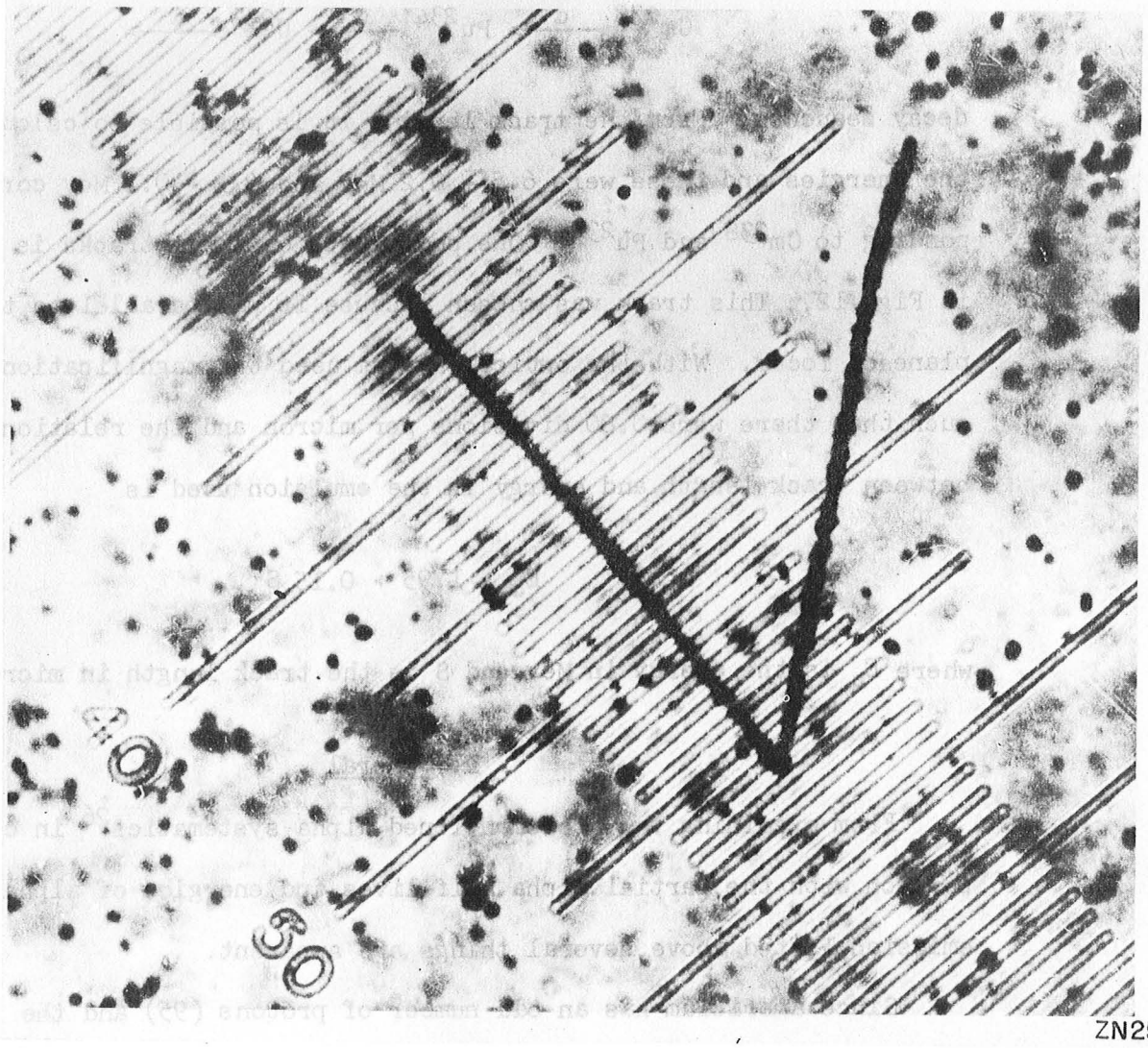

Fig. 12. Enlargement of a section of photographic emulsion in which Cm^{238} had decayed. The tracks are attributed to Cm^{238} (6.5 Mev) and Pu^{234} (6.1 Mev).

is not fully utilized or the particles must be rearranged, and the result is a lengthening of half-life for a given disintegration energy. The half-lives mentioned above are very nearly those which would be expected for unhindered transitions of the same energies so it may be presumed that the most abundant group represents the case in which the alpha particle is formed from paired nucleons and that the measured energies are lower than the disintegration energies by several tens of kilovolts. In the case of Am^{241} the alpha particle fine structure has been investigated and these conclusions seem justified.[28] In addition, the groups must be represented in roughly the same abundances in the various isotopes since they all follow a linear log half-life-energy relation, and any large change in abundances would result in a corresponding deviation in half-life, except if the change is regular.

The fact that no alpha particles of Am^{238} or Am^{240} are seen is easily understood since they have odd numbers of both protons and neutrons and decay by alpha emission would be hindered by an even greater amount while decay by electron capture is enhanced because the daughters have even numbers of both neutrons and protons and the pairing energy is gained by decay.

The even-even isotopes of curium, except Cm^{238}, all follow the half-life energy relation within experimental error, and as expected, the even-odd isotopes of $Cm^{239,241,243}$ are hindered, having half-lives which are longer for a given energy than they would have if they had even numbers of neutrons. Cm^{238} is peculiar since the measured alpha half-life is one-half as long as that of Cm^{240} which

has ~200 kev less energy. A decrease in half-life of about a factor of ten would be expected for this energy increase, and it seems reasonable that the measured values are in error and that the partial alpha half-life is between 3 and 5 days. Possible systematic errors in counting efficiency could easily reduce the measured value of alpha half-life by a factor of three or four, especially since separations of americium and curium are at present too slow to observe the growth of the americium daughter. Another possibility is that the isotope Cm^{237} has a half-life which is very nearly the same as Cm^{238}, in which case radiation from it would have been included in those electron capture events attributed to Cm^{238} in the branching ratio calculation.

The slope of the curve obtained by plotting alpha energy versus mass number of the even-even curium isotopes is less than for other elements in this region.

ACKNOWLEDGMENTS

I wish to thank Professor Glenn T. Seaborg for his advice concerning all phases of this work, and Dr. Kenneth Street, Jr. for suggesting several experiments and helping with the early experimental procedures. The Messrs. A. Ghiorso, A. E. Larsh, D. L. Martin, R. Glass, and R. Carr assisted in many phases of the experimental work. The crews of the 60-inch Crocker Laboratory and 184-inch cyclotrons were very helpful in performing the bombardments and, along with L. F. Deckard of the Health Chemistry group, assisted in designing the targets. This work was performed under the auspices of the Atomic Energy Commission.

APPENDIX

Separation of Americium and Curium by Means of Ion Exchange

Since the best way to separate elements as chemically similar as the actinides and lanthanides has been by their selective elution with various complexing agents from sulfonic acid ion exchange resins, a more thorough study of this separation was begun. The choice of citric acid as eluting agent was somewhat arbitrary but it was chosen since most data had been collected with that elutriant and successful separations had been brought about.[29]

The resin chosen was Dowex-50 cation exchanger, spherical form. It was chosen because of its high capacity and apparently good kinetic properties.[30]

The first experiments were performed at room temperature with a column 10 cm long and 2 mm in diameter with ammonium form resin which had been graded by allowing it to settle through water. The fraction of spheres which settled about 1 cm/min was used. It was found that the fastest successful separation was performed with a flow rate of elutriant of about 1λ/min under these conditions and even then there were evidences of nonequilibrium such as "tailing" of the peaks in great deviation from the logarithmic form expected.[31]

Since some workers had veen very successful using columns run at around 100° C,[25,26] it was decided to investigate the distribution of a typical actinide [Am(III)] between citric acid and this resin at various temperatures and hydrogen ion concentrations. These experiments were performed by placing a weighed amount of oven dried resin in a small vial along with a measured volume of 0.24 M citric

acid which had been adjusted to a predetermined pH at room temperature. A quantity of Am(III) tracer was then added and the vials were placed in a water bath. After agitation for several minutes a known volume of the supernatant liquid was pipetted out, dried and counted for alpha particles. A second sample was taken after several more minutes of shaking and the two samples compared to make sure equilibrium had been attained.

The results of the experiments are indicated in Fig. 13 in which K_d, an arbitrary distribution constant calculated as the ratio of total americium in the resin divided by the total americium in solution times the ratio of the volume of solution in ml to weight of resin in grams, is plotted against temperature.

Since very little change in K_d was noted between 80° and 100° C and since the high vapor pressure of water at 100° C makes column operation difficult, it was decided to attempt operation near 90° C. Trichloroethylene boils at 87.5° C and was chosen as a heat transfer material. A boiler was prepared from a round bottom flask; the vapors were led to an ion exchange column jacketed in a manner similar to a jacketed condenser and then to an air condenser from which the liquid ran back to the boiling pot.[15]

A K_d of approximately 10 was desired to facilitate rapid separations, so pH 3.50 citrate solution was used. The optimum rate under these conditions was 10λ per minute and there was little evidence of any nonequilibrium effect. A typical separation on a 20 cm x 2 mm column is shown in Fig. 14.

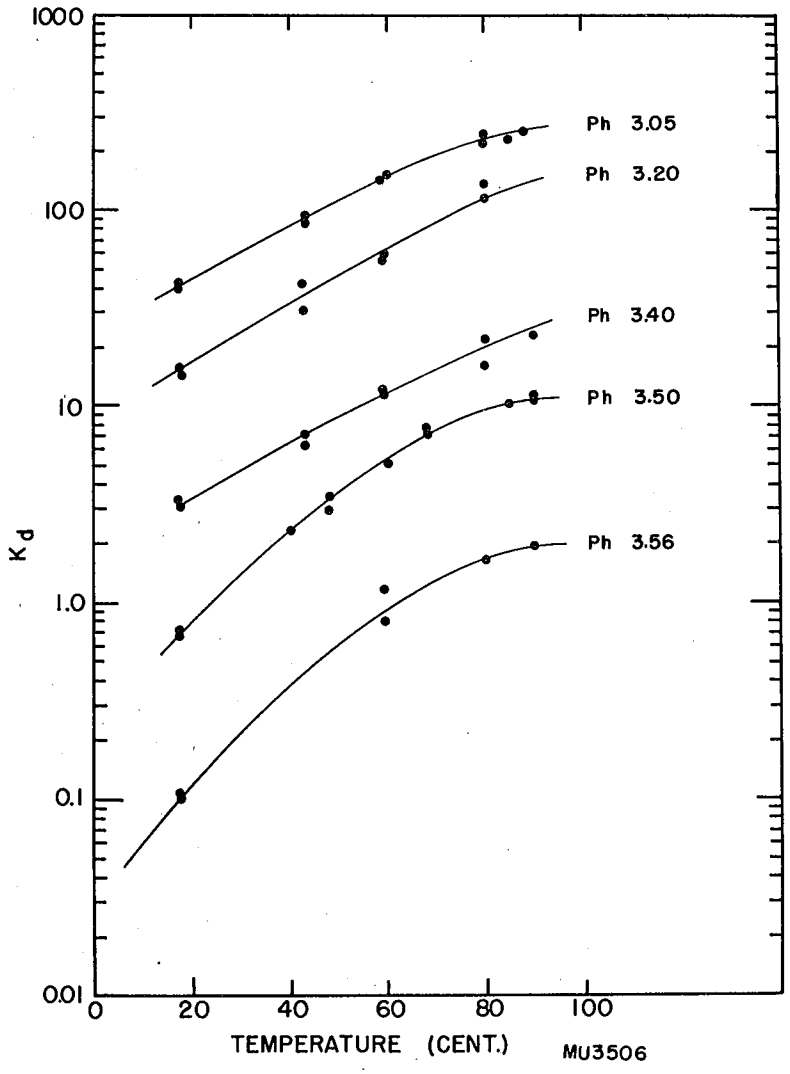

Fig. 13. A plot showing the distribution of americium(III) between 0.24 M citric acid and Dowex-50 cation exchange resin at various temperatures and hydrogen ion concentrations. K_d is defined as

$$\frac{\text{counts/min on resin x vol. of soln.}}{\text{counts/min in soln. x wt. of resin}}$$

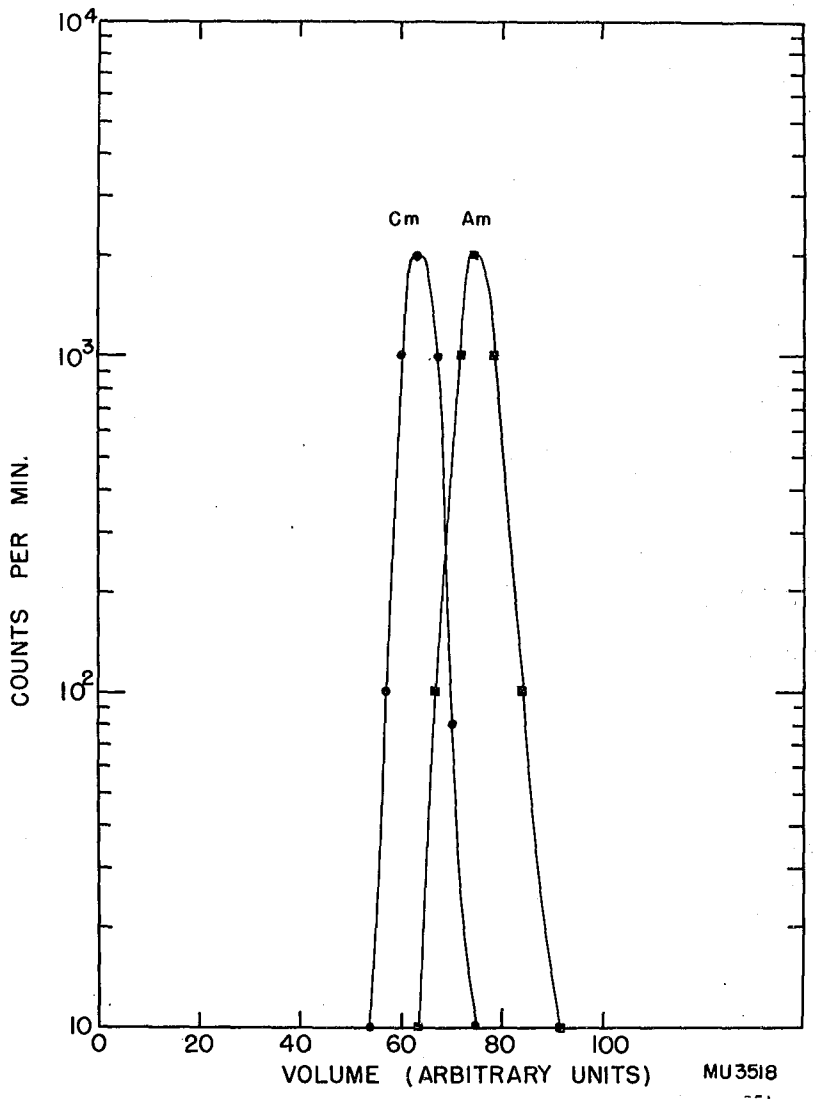

Fig. 14. Typical elution of americium(III) and curium(III) from Dowex-50 cation resin with 0.24 M citric acid adjusted to pH 3.50. The temperature was maintained at 87° C with trichloroethylene vapors.

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