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RADIOACTIVE DECAY PROPERTIES OF Am^{238}, Am^{239}, Am^{240}, Cm^{240}, and Cm^{241}

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ABSTRACT

Some electron-capture and alpha-decay properties of neutron-deficient americium and curium isotopes have been investigated using 2π proportional counting, scintillation spectrometry, and alpha-pulse analysis. The results for Am^{238} include a new half-life of 1.86 hr and identification of gamma rays of ~370 kev (~ 12%), 580 kev (~ 29%), ~950 kev (~ 2%), 980 kev (~ 76%), and ~1350 kev (~ 17%). The data on Am^{239} (half-life 12.1 hrs) indicate gamma rays of 225 kev (~ 30%) and 275 kev (~ 20%) with a new alpha particle energy of 5.77 Mev and abundance of 5.0×10^{-3}%. For Am^{240} (half-life 51.0 hrs) gamma rays of 900 kev (~ 23%) and 1000 kev (~ 77%) and a negatron branching limit of $\leq 6 \times 10^{-6}$% were established. For Cm^{240} a new alpha particle energy of 6.26 Mev was established. For Cm^{241} a new alpha particle energy of 5.95 Mev and abundance of 0.96% and a gamma ray of 478 kev (97%) were established. Counting efficiencies for the 2π-geometry methane-flow proportional counter are 60, 90, and 82% for the electron capture decay of Am^{239}, Am^{240}, and Cm^{241}, respectively.

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These are in the range of values found for other beta-unstable isotopes of heavy elements.

The data on Am^{238} and Am^{240} are consistent with a rule that only levels ~1000 kev and higher in even-even plutonium isotopes are populated by relatively fast beta transitions (log ft 5 to 8) in the decay of odd-odd americium and neptunium isotopes.

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I. INTRODUCTION

The present investigation of decay properties of neutron-deficient americium and curium isotopes was undertaken during the course of a series of cross-section measurements[1] in order to obtain preliminary data on the decay schemes and counting efficiencies (on a 2π-geometry windowless proportional counter).

The gamma-ray and alpha-particle data obtained are of interest from the standpoint of correlation with the systematics of forbiddeness for beta decay and hindrance factors for alpha decay.[2-6] In most cases, however, more detailed studies with alpha and beta spectrographs and further coincidence studies are needed to complete the decay schemes.

The calculation of absolute cross sections or alpha-particle and gamma-ray intensities requires that absolute disintegration rates of the nuclides in question be determined. This has been a particularly troublesome problem for nuclides which decay by orbital electron capture. Calibration of a given type of counting system (preferably one of high geometry and low sensitivity to sample preparation method), however, allows one to estimate disintegration rates of other samples of the same nuclide with some confidence. In addition, repeated calibrations for as many different nuclides as possible form a basis for estimating disintegration rates of other nuclides. In the course of these studies counting efficiencies were determined for weightless samples of a number of nuclides on a 2π windowless methane-flow proportional counter.[7]

II. EXPERIMENTAL PROCEDURES

Targets of 1 to 100 mg of plutonium-239 oxide were bombarded with 27 and 35 Mev protons in the 184-inch synchrocyclotron to produce Am^{238}; with 15 to 20 Mev deuterons in the 60-inch cyclotron to produce Am^{239} and Am^{240}; and with 30 to 40 Mev helium ions in the 60-inch cyclotron to produce Am^{240}, Cm^{240} and Cm^{241}. Plutonium-239 targets of high isotopic purity were employed to avoid Am^{241} alpha-activity contamination from the decay of Pu^{241} present in ordinary plutonium. Chemical separations were performed to separate the curium and/or americium fractions from the target material, fission products, and unwanted spallation products. For the counting efficiency determinations the plutonium daughter activities were separated from the americium fraction after suitable growth periods.

Electron-capture isotopes were counted in a windowless 2π-geometry gas-flow proportional counter. Natural gas (mainly methane) was used as the counting gas. The sample, mounted on a 2.5 cm diameter platinum disc, was placed inside the counting chamber[8] at a distance of about 4 cm from a 1 mil center wire. The counter was operated on the proportional plateau at about 4,000 volts. Effect of sample preparation upon the counting efficiency was studied by comparing the counting rates of equivalent samples mounted by (1) evaporation of a carrier-free aqueous solution, (2) volatilization from a tantalum filament in vacuo at $\sim 1800^{\circ}C$ or (3) electrodeposition from neutral ammonium chloride solution.[9] Under proper conditions, the last two methods result in essentially weightless samples of $< 10\ \mu g/cm^2$ mass, whereas the first method may lead to 30 to 50 $\mu g/cm^2$ or greater. It was found that the sample thickness (up to $\sim 30\ \mu g/cm^2$) becomes critical only when a sizeable fraction of the counting rate for a nuclide depends on low-energy Auger electrons. For example successive volatilization of about 10 microgram amounts ot tantalum onto a weightless sample of Am^{239} (which is known[10] to have a high abundance of several hundred kilovolt conversion electrons) reduced the counting rate by 10% only after 50 $\mu g/cm^2$ had been transferred. Counting rates of Np^{234} are also reasonably independent of sample thickness.[11] On the other hand, Pu^{237}, which decays mostly (60%) to the ground state, counts most efficiently when electroplated, slightly less efficiently when vaporized, and considerably less efficiently when evaporated.[12]

A Frisch grid ionization chamber and a 50 channel pulse-height analyzer were used for alpha spectroscopy. Gamma spectra were determined with a scintillation counter employing a 1-1/2 (diam) x 1-inch thallium activated sodium-iodide crystal and a 50 channel pulse-height analyzer. Photopeak counting efficiencies and geometry factors tabulated by Kalkstein and Hollander were used.[13] In the gamma-gamma coincidence measurements the energy of the gating pulse from a second sodium-iodide detector was determined by analysis in a single channel pulse-height analyzer. The resolving time of the coincidence apparatus was 2 microseconds.

In all of the measurements, probable errors have been assigned on the basis of estimated systematic accuracy as well as the precision of the data.

III. RESULTS

A. Americium-238

The electron-capture decay of Am^{238} is of unusual interest among the heavy nuclides because the energy available for this process is almost uniquely high (2.26 Mev, estimated[14] from systematics) for known nuclides away from closed nucleon shells. The lower-level structure of the daughter nuclide has been studied through the alternate routes of alpha decay[9] of Cm^{242} and negatron decay[15] of Np^{238} with results which are of interest in terms of the collective model of the nucleus.[16]

Prominent gamma rays of 580 ± 25 kev and 980 ± 40 kev were observed in the americium fraction from bombardments with both 27 Mev and 35 Mev protons (see Fig. 1a). A half-life of 1.86 ± .09 hr was obtained by following the decay of the 580 kev and 980 kev photon peaks. (The previous value[17] (2.1 hr) was determined from resolution of a three component decay curve.) The assignment of these two high-energy gamma rays to Am^{238} rather than Am^{237} was made because their intensity relative to the 6.01 Mev alpha particles[17] of Am^{237} decreased appreciably when the proton bombarding energy was increased from 27 to 35 Mev. The gamma-ray spectrum in the region of 150 to 400 kev was completely obscured by the very prominent photopeaks of 225 kev and 275 kev belonging to Am^{239}. However, photopeaks having 1.9 hr components were observed

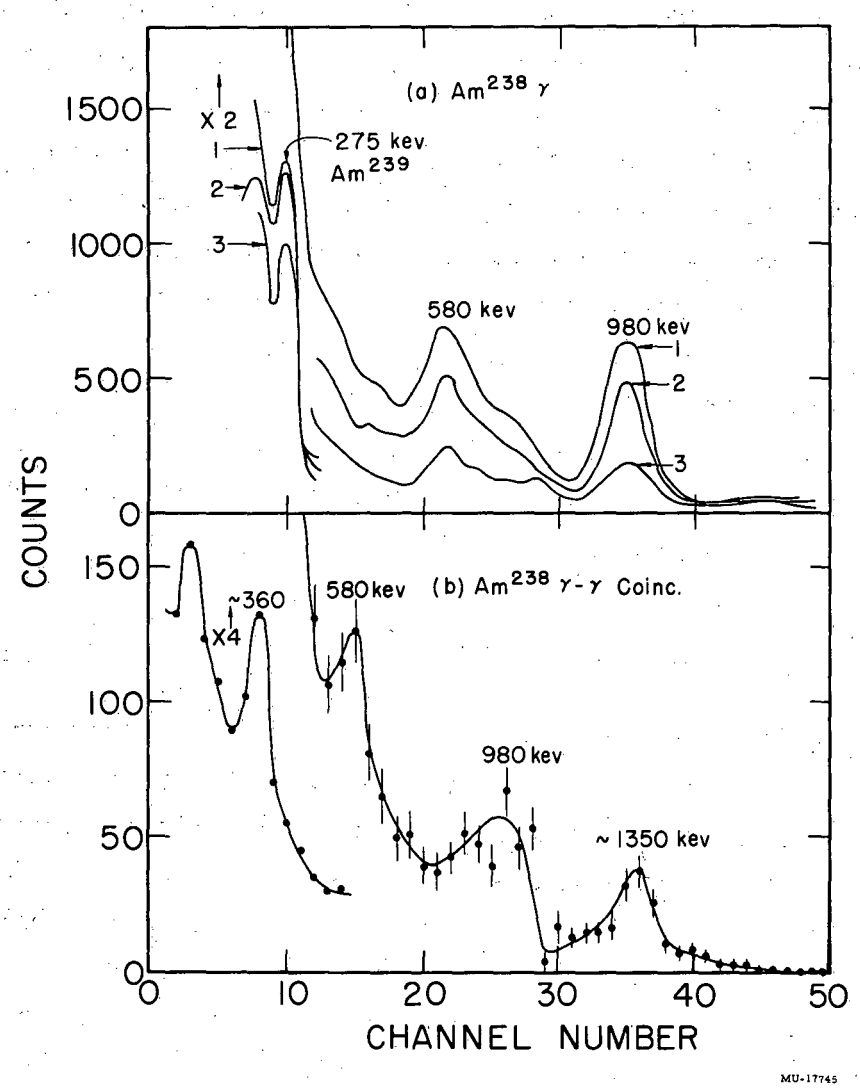

Fig. 1. Gamma-ray spectra of Am^{238}. (a) The direct spectrum measured at three different times, (b) spectrum of gamma rays in coincidence with gamma rays of 580-kev. The apparent self-coincidence peak at 580-kev is probably due to coincidences between 580-kev gamma rays and Compton-scattered electrons from initially 980-kev gamma rays.

with energies of 14 kev, 18 kev, and 102 kev. The first two of these, resolved with a xenon-filled proportional counter, correspond to the L_α and L_β x-rays of plutonium; the last is essentially all plutonium K x-rays, but also may contain a small contribution from the highly converted 104 kev gamma ray corresponding to a transition between levels of Pu^{238} known from Np^{238} and Cm^{242} decay studies.

In order to establish the relationships between the high-energy gamma rays in the decay scheme, gamma-gamma coincidence studies were made. In the course of these measurements new gamma rays of 1350 kev and 370 kev were found. The decay of all coincidence peaks was checked to confirm their assignment to Am^{238}. The observations are summarized in Table I, and the gamma spectrum coincident with the 580 kev gamma ray is shown in Fig. 1b.

Table I

Americium-238 Gamma-Gamma Coincidences[a]

Gate	Coincident gamma rays (signal)
980 kev	~370 kev, 580 kev, ~950 kev
580 kev	~360 kev, 580 kev, 980 kev, ~1350 kev
~1350 kev	~580 kev

[a]The energy measurements of the coincident gamma rays are approximate since the inherent accuracy of the instrument used is only about 4% and the counting rates were very low (of the order of 3 counts per minute for some of the peaks). Further, the short half life of the nuclide drastically limits the amount of time available for the measurements.

The apparent self coincidence of the 580 kev gamma ray is probably not real. Radiations in the 580 kev region in coincidence with the 580 kev photopeak are believed to be due to the Compton edge of the 980 kev photopeak. Thus the apparent self-coincidence is probably a result of the 980 kev - 580 kev cascade. The apparent 580 kev - 360 kev coincidences may also include come contribution from electrons and photons resulting from Compton scattering of the 980 kev gamma ray.

From the gross spectra the relative intensities of the L x-rays, K x-rays, 580-kev gamma rays, and 980-kev gamma rays can be calculated. The L x-ray counting efficiencies and geometry factors of Hoff,[18] for the xenon-filled proportional counter were used. Corrections were made for escape peaks in the cases of the K x-rays, and Compton scattered electrons from the 980 kev gamma ray in the case of the 580 kev gamma ray. The relative intensities are: L x-rays: K x-rays: 580 kev: 980 kev = 63: 100: 29: 78.

It is reasonable to assume that about one K x-ray is emitted per disintegration in order to establish rough absolute intensities. This is based on: (1) the expectation that the 102 kev gamma radiation that has been observed in Np^{238} and Cm^{242} decay, and which has the same energy as K x-rays is negligible; (2) the assumption that K-shell conversion of the more energetic gamma rays is small; (3) the assumption that L-electron capture is probably small (near the theoretical limit[19] of 14%) since the observed L x-rays can be accounted for by the L-shell vacancies arising from electron transitions to fill K-shell vacancies and L-shell conversion of gamma rays; and (4) the fact that the fluorescent yield[20] of K-shell vacancies is almost 100%. It will be noted later that measured K x-ray intensities (~ 85% for Am^{239}, ~ 75% for Am^{240}, and ~ 90% for Cm^{241}) are somewhat less than 100%, although most probable errors would tend to increase these experimental values. From the assumption of one K x-ray per disintegration the intensity of the 580 kev gamma ray is determined as ~ 29%. The 580 kev gamma ray intensity along with the relative intensities obtained from the coincidence data allows the absolute intensities of the other gamma rays to be computed.[21]

The decay scheme indicated by these data is given in Fig. 2. An alternative possibility would have the 580-kev and the ~370-kev gamma rays depopulate the 980-kev level; that is, in a path parallel to the lower rather than the upper ~950-kev gamma transition (a 605-kev level has been observed in Pu^{238} from the alpha decay of Cm^{242} [9]). However, the observed 580 kev - ~1350 kev coincidences as well as the fact that the 605-kev level (or any nearby level) is not observed in the negatron decay of Np^{238}, either from direct population or from cascade from the levels near 1 Mev, have led us to place the 580 kev gamma transition solely in the high-energy branch. Also shown for comparison purposes, is the level diagram of Pu^{238} as indicated by the

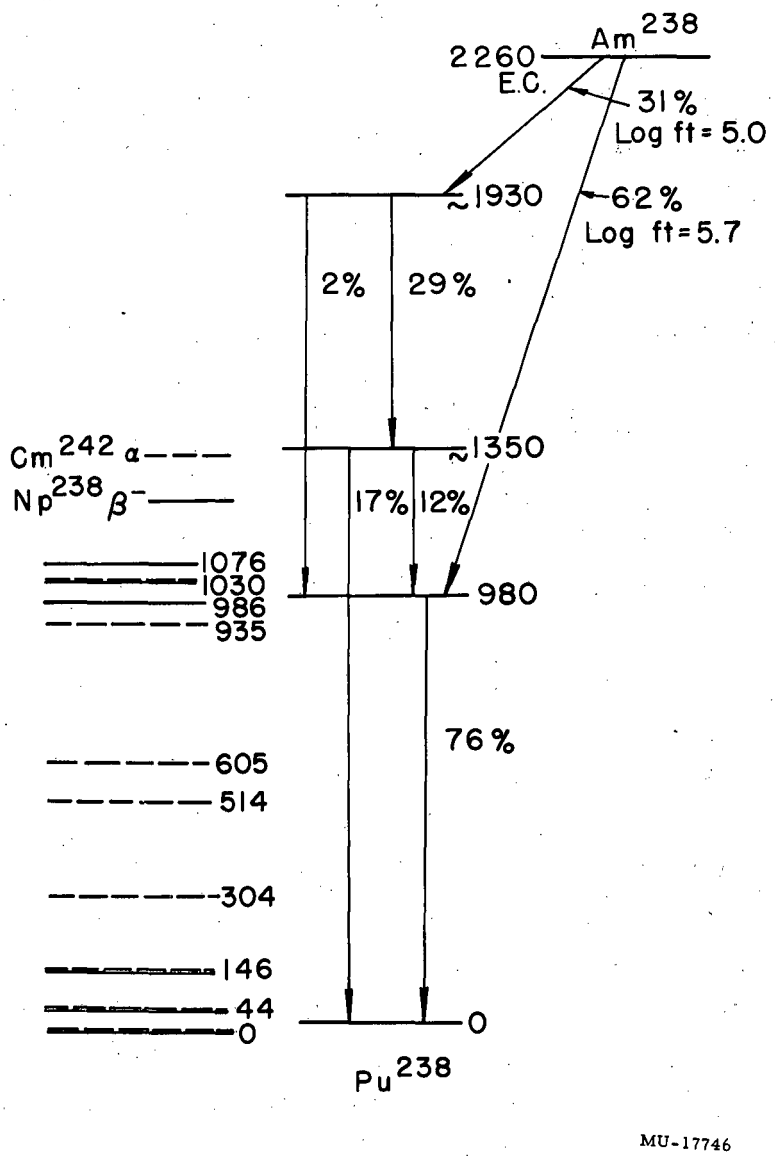

Fig. 2. Preliminary decay scheme for Am^{238} showing gamma ray intensities (and therefore electron-capture-branching intensities since conversion of these high energy gamma rays should be very low) based on the assumption that one K x-ray is emitted per disintegration. The unaccounted-for 7% of decay may be due to an error in the above assumption but could also be due to branching to the ground state or to a low energy state of Pu^{238}. The Pu^{238} levels on the left are those populated from negatron decay to Np^{238} and alpha decay of Cm^{242}, as listed by Perlman and Rasmussen (Ref. 4). The exact alignment of Am^{238} associated levels is not certain due to uncertainties in energies and in the precise levels near the ground state populated by the high energy gamma rays.

decay of Np^{238} and Cm^{242}. It should be noted that in these studies many transitions were observed between the band of levels at ground and that near 1 Mev. There are at least four and possibly five gamma transitions between 927 kev and 1029 kev from Np^{238}. These would appear as a single photopeak in the equipment used in this study. It is highly probable, therefore, that the 980 kev gamma ray of Am^{238} is also complex. Indeed, it may be that all the observed gamma rays are complex and that the levels indicated are really bands of levels.

Approximate log ft values[22] of 5.0 and 5.7 were estimated to the ~1930 kev level and the 980 kev level, respectively, corresponding to allowed transitions. It is perhaps worth pointing out a few observations about the energy level of ~1930 kev (which is one of the highest ever observed in decay studies in the heavy element region). If the band of levels around 1 Mev corresponds to a first vibrational excitation, according to the collective model a second vibrational band is expected [16] at about twice this energy.

B. Americium-239

The level scheme of Pu^{239} has been characterized in extensive studies of the beta decay[23] of Np^{239} and the alpha decay[24] of Cm^{243}. In addition, the conversion-electron spectrum associated with the electron-capture decay of Am^{239} has been determined,[10] with results of value for correlation with the present gamma-ray studies. Gamma rays in coincidence with the alpha particles of Am^{239} have also been reported.[25]

Intense samples containing Am^{239} and Am^{240} were counted repeatedly over many half lives of both isotopes in the proportional counter giving a value of 12.1 ± 0.4 hours for the half life of Am^{239}.

The presence of Am^{240} in the samples limited the accuracy of counting efficiency determinations for Am^{239}. Since the daughters of both Am^{239} and Am^{240} (Pu^{239} and Pu^{240} respectively) have almost the same alpha-particle energy, a growth vs time analysis of the alpha radiations of this energy was necessary. The proportional counting efficiency for Am^{239} (volatilized onto platinum from a tungsten filament) obtained from a single experiment was 60 ± 20%.

Gamma rays of 225 ± 10 kev and 275 ± 33 kev with the 12 hr half life were observed in the gross gamma spectra. Abundances obtained (based on a 60% proportional counter efficiency) were: K x-rays, 85 ± 30%; 225 kev gamma rays, 30 ± 15%; 275 kev gamma rays, 20 ± 10%. These abundances are somewhat at variance with the conversion electron data;[10] principally in that the K x-ray intensity is low. Considering K x-ray counting difficulties in this experiment and the proportional counting efficiency uncertainties, however, it is believed that the difference is not significant.

Results of gamma-gamma coincidence studies are given in Table II.

Table II

Americium-239 Gamma-Gamma Coincidences

Gate	Coincident gamma rays (signal)
K x-rays	K x-rays, ~225 kev, ~275 kev
~225 kev	K x-rays, ~225 kev, ~275 kev
~275 kev	K x-rays, (~180 kev?), ~225 kev

These measurements confirm the cascade relationship that a 225 kev gamma ray leads to a level depopulated by 225 kev and 275 kev gamma rays.

These data have not been analyzed further because of the uncertainty in the intensity measurements and because log ft values and level assignments have been treated elsewhere.[10]

Well resolved alpha particles from Am^{239} were observed in samples from two bombardments. The alpha energy was redetermined as 5.77 ± .02 Mev.

The percent of Am^{239} decays proceeding by alpha particle emission was evaluated in duplicate experiments with results of 5.0×10^{-3} and 4.9×10^{-3} (based on a proportional counting efficiency of 60% for electron capture decay). The best value is taken as $(5.0 \pm 1.0) \times 10^{-3}$, corresponding to a partial alpha half life of 28 ± 5 years. The hindrance factor is 2.6 for this alpha group, to be compared with values[4] of 1.2 and 1.1 for the alpha-particle groups in highest abundance of Am^{241} and Am^{243}, respectively.

C. Americium-240

The electron capture decay characteristics of Am^{240} are of interest for comparison with the decay of Am^{238}, since both isotopes are odd-odd nuclei and decay to even-even plutonium isotopes. For the levels of Pu^{240}, information has been obtained through study of the alpha decay of Cm^{244} [26] and negatron decay of the 7.3 minute[27] and 60 minute[28] isomers of Np^{240}.

The half life of Am^{240} was determined to be 51.0 ± 0.5 hr by proportional counting.

The electron-capture proportional counting efficiency was determined for an evaporated sample for one plutonium daughter "growth" period (following one bombardment) and for vaporized samples for two "growth" periods (following a second bombardment). The values derived were 91%, 109%, and 67%, respectively, leading to an average value of 90 ± 15%.

Photopeaks of 900 ± 20 kev, 1000 ± 30 kev, and 1400 ± 50 kev, decaying with a 51 hr half life were observed in the gross gamma spectra. The abundances obtained from a series of independent measurements (all based on a 90% proportional counting efficiency) were: L x-rays, 55 ± 10%; K x-rays, 75 ± 15%, 900 kev, 23 ± 15%; 1000 kev, 77 ± 20%; and 1400 kev, < 10%. The electron-capture counting efficiency uncertainty is not included in the estimated probable errors.

The partial decay scheme suggested by the gamma ray measurements is shown in Fig. 3, together with Pu^{240} levels populated by alpha particle and negatron decay. Values of log ft[22] for decay to the 900-kev and 1000-kev levels based on this tentative decay scheme are 7.2 and 6.4, respectively, corresponding to allowed hindered or first forbidden unhindered transitions. Both electron capture decay branches agree with correlations for first forbidden electron capture transitions of Hoff and Thompson.[3]

It is interesting to note that levels around 1000 kev (possibly corresponding to a vibrational band) are populated in both electron capture and negatron decay to both Pu^{238} and Pu^{240}. A level at about 1400 kev is also common to Am^{238} and Am^{240} decay.

A search was also made for negatron branching in Am^{240}, since the results of closed cycle calculations[14] of heavy-element decay energies are

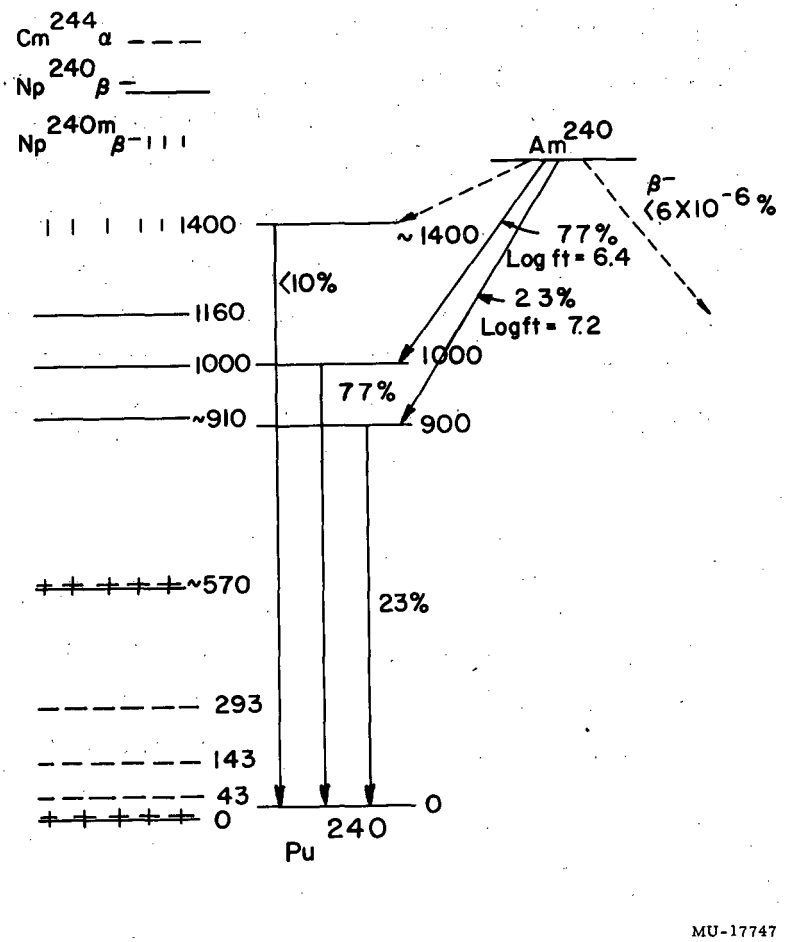

Fig. 3. Preliminary decay scheme for Am^{240} showing gamma ray intensities (and therefore electron capture branching intensities since conversion of the high energy gamma rays is expected to be negligible) based on a 2π-proportional counting efficiency of 90%. The 1000 kev and 900 kev gamma rays could possibly proceed from the same level and populate the 43 and 143 kev levels, respectively. The Pu^{240} levels on the left are those populated from negatron decay of Np^{240} (Ref. 28) and Np^{240m} (Ref. 23), and alpha decay of Cm^{244} (Ref. 26). The alignment of Am^{240} associated levels is uncertain due to uncertainties in energy measurement and in the precise levels near the ground state populated by the high energy gamma rays.

ambiguous concerning the relative beta-instability of Am^{240} and Cm^{240}. The calculations indicate that electron capture of Cm^{240} is exoergic by 60 ± 100 kev (neglecting the electron binding energy). Experimentally, Higgins[17] has searched for both negatron emission of Am^{240} and orbital electron capture of Cm^{240} and on the basis of his negative results, has set upper limits of 2×10^{-3}% negatron emission for Am^{240} and 0.5% electron capture for Cm^{240}. In the present work, a sample of Am^{240} decaying at the rate of 4.6×10^{6} disintegrations per minute was prepared. After the Am^{240} had decayed, an upper limit of 0.02 disintegrations per minute was set for alpha particle emission at 6.26 Mev, the energy of Cm^{240} alpha particles. This limit corresponds to an upper limit of 6×10^{-6}% for the negatron-decay branching of Am^{240} and a lower limit of 1×10^{5} years for the partial half life for this process.

D. Curium-240

This isotope is observed to decay solely by alpha-particle emission. The present work consisted of an attempt to improve the alpha-particle energy with numerous alpha pulse height analyses. The result was an energy of 6.26 ± .02 Mev, as compared with an earlier preliminary value of 6.2_5 ± 0.1 Mev.[29]

E. Curium-241

This isotope decays predominantly by electron capture to excited levels of Am^{241},[30] which are also populated by alpha decay[9,31] of Bk^{245}.

The electron-capture proportional-counting efficiency was determined by observing the growth of Am^{241} alpha activity in a vaporized sample of Cm^{241} (see Fig. 4). The isotope Cm^{241} is a particularly favorable case for the "alpha-daughter" method of measuring the counting efficiency. Chemical separations are unnecessary, since both the parent and daughter radiations can be counted in the same sample. The resultant electron-capture counting efficiency was 82 ± 8%.

A prominent gamma ray of 478 ± 10 kev was observed. A number of determinations established the following intensities: L x-rays, 65 ± 10%; K x-rays, 90 ± 10%, 478-kev gamma ray, 97 ± 10%.

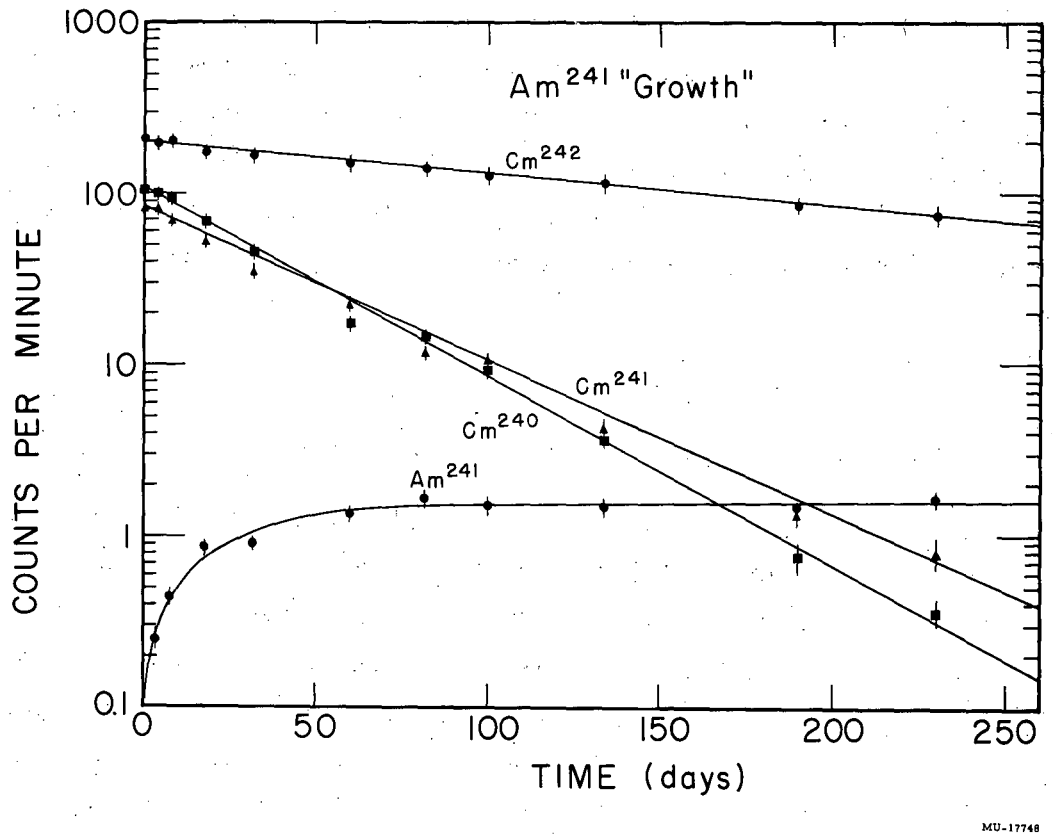

Fig. 4. Growth of Am^{241} alpha activity into a chemically purified curium fraction. The curium was volatilized onto a platinum disk for alpha pulse-height analysis. The alpha activities of Cm^{240}, Cm^{242}, and Am^{241} represent 100% of the decay of these isotopes, whereas the Cm^{241} activity represents only 0.96% of its decay.

A log ft value of 7.4 was calculated[14,22] for electron capture decay to the 478 kev level indicating an allowed hindered or first forbidden unhindered transition. This is in close agreement with the value given by Mottelson and Nilsson,[6] who also show a decay scheme indicating decay to an additional level at 640 kev.

The numerous alpha-pulse analyses of Cm^{241} gave an alpha energy of 5.95 ± 0.02 Mev. This is slightly higher than the previous figure of 5.90 Mev.[17] A total alpha branching of 0.96 ± 0.09%, corresponding to a partial alpha half life of 10 ± 1 years, was calculated from these data. The hindrance factor is 2.5 for this alpha group, to be compared with values[4] of 1.4 and 2.5 for the alpha-particle groups in highest abundance of Cm^{243} and Cm^{245} respectively.

IV. DISCUSSION

Electron-capture counting efficiencies for the 2π-geometry windowless methane-flow proportional counter (for thin samples mounted on platinum) from the present work and other sources[32] are summarized in Table III.

The average counting efficiency for electron capture nuclides included in Table III is about 80%, and it is this value that is recommended for use for those nuclides for which no direct determination has been made, although caution should be exercised since the counting efficiency depends to some extent on the decay scheme. The high (greater than 50%) counting efficiencies measured for this type of decay in a 2π-geometry windowless proportional counter, and the similarity to the efficiency of the same counter for nuclides which decay by negatron emission should be noted. These high efficiencies can be partly attributed to the backscattering factor[34] on platinum of 1.6 to 1.8 for beta particles (and presumably conversion electrons) of moderate and high energies, but a large contribution to the high counting efficiency must come from Auger electrons and low energy x-rays are usually present in high abundance. An efficiency of 77% has been observed for Pu^{237} which decays 60%[6] to the ground state of Np^{237}.

Table III

Electron-Capture and Negatron Proportional-Counting Efficiencies

Isotope	Counting Efficiency (on platinum)	Sample Mounting Method[a]
Electron Capture		
Np^{234}	63[b]	Ev.
"	68[b]	V.
"	65[b]	Ep.
Np^{236}	89[c]	V.
"	95[c]	Ep.
Pu^{237}	77[c]	Ep.
Am^{239}	60[d]	V.
Am^{240}	90[d]	Ev., V.
Cm^{241}	82[d]	V.
Negatron		
Rare Earths	78[c,e]	
Bi^{210}	85[f]	
Np^{238}	70[c]	
Pu^{241}	66[f]	

a. Ev. refers to samples mounted by evaporation of a carrier-free aqueous solution onto a 2 mil platinum disc followed by heating in a burner to a dull red glow; V. refers to samples volatilized onto a platinum disc from a tantalum filament at ~1800°C in vacuo; Ep. refers to samples electroplated onto a platinum disc from neutral ammonium chloride solution (see reference 33 for further details).

b. W. M. Gibson, Ref. (33).

c. R. Vandenbosch, unpublished data.

d. This work.

e. This value is the average for Y^{91}, Zr^{95}-Nb^{95}, Nd^{147}, Pm^{149}, Sm^{153}, Gd^{159}, Tb^{164}, and Er^{169}.

f. S. G. Thompson, A. Ghiorso and B. G. Harvey, unpublished data.

Most of the decay data presented need little further amplification. The preliminary decay schemes of the two odd-odd isotopes, Am^{238} and Am^{240}, however, have an unusual and revealing characteristic: essentially all of the electron capture decay leads to levels of about 1000 kev or higher energy. From this observation and inspection of the decay schemes of other odd-odd americium isotopes and odd-odd neptunium isotopes one concludes that it is a general rule that levels > 1000 kev must be populated for relatively fast beta decay to even-even plutonium isotopes. The decay of the ~ 100 yr isomer of Am^{242} (believed to be the ground state[30]) is one case. The decay energy for electron capture is only about 650 kev and the log ft value of 12 for decay to the lowest-lying states of Pu^{242} implies highly forbidden decay.[35,36] On the other hand, the decay of the 16 hr isomer is characterized by allowed or first forbidden log ft values, so the forbiddenness rule may be associated only with decay from ground states. The negatron decay of Np^{236}, also to an even-even plutonium isotope may be another example of the rule. If the long-lived isomer[32] of Np^{236} is assumed to be the ground state and the negatron and electron-capture decay energies are assumed to be similar to those noted for the 22 hr isomer of Np^{236} (which is reasonable from decay-energy systematics[14]) then the log ft value is > 12 for negatron decay and higher still for electron capture decay. Again the decay from the excited state (22 hr isomer of Np^{236}) is relatively unforbidden. Among the other beta-minus unstable neptunium isotopes, Np^{238} exhibits forbidden decay to low-lying levels of Pu^{238} (log ft = 8.5 to the 2+ level) and more allowed decay to levels above 1000 kev.[16] The decay of Np^{240} is apparently similar to that of Am^{242} and Np^{236}. The recently assigned 60-minute isomer[28] of Np^{240} (probably the ground state) again decays mainly to excited levels, whereas the 7-minute excited state of Np^{240} undergoes beta transitions[27] to the ground and 560 kev levels of Pu^{240} with log ft values of about 6.5

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REFERENCES

1. For a study of helium-ion reactions in Pu^{238}, Pu^{239}, and Pu^{242}, see Glass, Carr, Cobble, and Seaborg, Phys. Rev. 104, 434 (1956). For a study of helium-ion reactions in Np^{237} and deuteron reactions in Pu^{239} see Ph.D. thesis of W. M. Gibson, University of California, June 1957, also reproduced as University of California Radiation Laboratory Report UCRL-3493, November 1956 (unpublished).

2. For various correlations of heavy element decay properties see, G. T. Seaborg, The Transuranium Elements, (Yale University Press, New Haven, 1958).

3. R. W. Hoff and S. G. Thompson, Phys. Rev. 96, 1350 (1954).

4. I. Perlman and J. O. Rasmussen, Handbuch der Physik (Springer-Verlag 1957) Vol. 42, p. 109 also reproduced as University of California Radiation Laboratory Unclassified Report UCRL-3424, June 1956.

5. Stephens, Asaro, and Perlman, Phys. Rev. 113, 212 (1959).

6. B. R. Mottelson and S. G. Nilsson, Mat. Fys. Skr. Dan. Vid. Selsk. 1, No. 8 (1959).

7. Previous techniques used include measuring K x-ray counting rates (which necessitates detailed knowledge of the decay-scheme), measuring counting rates in a 4π-geometry proportional counter, and measuring disintegration rates of alpha particle emitting daughters produced from decay of the electron-capture parents. The last of these methods is the most accurate and was employed in this investigation.

8. The detector used was manufactured by the Radiation Counter Laboratories of Chicago Illinois and carried the trade mane "Nucleometer".

9. Ph.D. thesis of A. Chetham-Strode, Jr., University of California, September 1956, also available as University of California Radiation Laboratory Report UCRL-3322, June 1956 (unpublished).

10. W. G. Smith, W. M. Gibson, and J. M. Hollander, Phys. Rev. 105, 1514 (1957).

11. Ph.D. thesis of W. M. Gibson, University of California, June 1957, also available as University of California Radiation Laboratory Report UCRL-3493, November 1956 (unpublished).

12. R. Vandenbosch, unpublished data.

13. M. I. Kalkstein and J. M. Hollander, University of California Radiation Laboratory Report UCRL-2647, October 1954 (unpublished).

14. Glass, Thompson, and Seaborg, J. Inorg. Nuclear Chem. 1, 3 (1955).

15. Rasmussen, Slätis, and Passell, Phys. Rev. 99, 42 (1955); Rasmussen, Stephens, Strominger, and Åström, Phys. Rev. 99, 47 (1955).

16. A. Bohr and B. R. Mottelson, Kgl. Danske Viden Skal. Selskab. Mat.-fys. Medd. 27, 16 (1953).

17. Ph.D. thesis of G. H. Higgins, University of California 1952; also available as University of California Radiation Laboratory Report UCRL-1729, June 1952 (unpublished).

18. R. W. Hoff, unpublished data (1954).

19. H. Brysk and M. E. Rose, Oak Ridge National Laboratory Report 1830, 1955 (unpublished).

20. P. R. Gray, Phys. Rev. 101, 1306 (1956).

21. Only the intensity of the 580-kev gamma ray is determined uniquely from the gross gamma spectra and the assumption of one K x-ray per disintegration. The intensities of the other transitions, however, can be derived on the basis of the proposed decay scheme from the gross spectral intensities and the relative coincidence intensities. The relative intensities of the 370-kev and the 580-kev gamma rays in coincidence with

the 980-kev photopeak are the same as the relative intensities obtained using the 580-kev gamma rays as gating pulses. This indicates that there is little direct population by electron capture to the 1350-kev level. The intensity of the 580-kev radiations (determined to be 29% from the gross spectra) must then equal the sum of the intensities of the 370-kev and the 1350-kev gamma rays. Since the relative intensities of the 370- and the 1350-kev gamma rays are established from the coincidence measurements, the absolute intensities of these transitions can be calculated. To assign intensities to the ~950-kev and 980-kev transitions, one may use the relative intensities of the 580-kev and ~950-kev gamma rays in coincidence with the 980-kev photopeak (taking into account that either the 980-kev or ~950-kev gamma ray may be acting as a gating pulse and either appearing in the coincidence spectrum). Since the 580-kev – 980-kev cascade has been fixed at 12%, the ~950-kev gamma ray intensity is calculable. Finally, the 980-kev gamma ray intensity is equal to the difference between the gross 980-kev gamma ray intensity and the ~950-kev gamma ray intensity.

22. The log ft calculations for Am^{238} and other isotopes were made using the graphs of H. Brysk and M. E. Rose (Ref. 19) for K, L_I, and L_{II} electron capture. These lead to log ft values about 0.1 to 0.2 lower than those calculated from the nomogram of S. A. Moszkowski, Phys. Rev. 82, 35 (1951) for K electron capture.

23. Hollander, Smith, and Mihelich, Phys. Rev. 102, 740 (1956); D. Engelkemeir and L. B. Magnusson, Phys. Rev. 99, 135 (1955).

24. I. Perlman and F. Asaro, Ann. Rev. Nuc. Sci. 4, 157 (1954).

25. F. Asaro, F. S. Stephens, Jr., W. M. Gibson, R. A. Glass, and I. Perlman, Phys. Rev. 100, 1541 (1955).

26. Ph.D. thesis of J. P. Hummel, University of California 1956; also available as University of California Radiation Laboratory Report UCRL-3456, July, 1956 (unpublished).

27. Knight, Bunker, Warren, and Starner, Phys. Rev. 91, 889 (1953).

28. Ph.D. thesis of R. M. Lessler, University of California, February, 1959; also available as University of California Radiation Laboratory Report UCRL-8439, October 1958 (unpublished).

29. G. H. Higgins and K. Street, Jr., Phys. Rev. 86, 252 (1952).

30. Gamma rays of 480 kev and 600 kev were found in preliminary experiments by Amiel, Albridge, Stephens, and Asaro (unpublished data, 1957).

31. Magnusson, Friedman, Engelkemier, Fields, and Wagner, Phys. Rev. 102, 1097 (1956).

32. Less certain values have been reported for At^{211} (39%), U^{231} (33%), Bk^{243} (26%), and Bk^{245} (33%) by E. K. Hulet, (Ph.D. thesis, University of California, 1954, and University of California Radiation Laboratory Report UCRL-2283, July 1953); and for At^{211} (60%) by Higgins (Ref. 14); and for Bk^{245} (~100%) and Cf^{247}(80-90%) by A. Chetham-Strode and B. G. Harvey (unpublished results). The first set of values, probably much too low, have lead to the widespread, but probably erroneous, belief (in connection with later results) that there is a very wide range of counting efficiencies.

33. Ph.D. thesis of W. M. Gibson, University of California, 1957; also available as University of California Radiation Laboratory Report UCRL-3493, 1956 (unpublished).

34. B. P. Burtt, Nucleonics, 5, No. 2, 28 (1949).

35. Hoff, Jaffe, Passell, Stephens, Hulet, Thompson, Phys. Rev. 100, 1403 (1955).

36. Studier, Gindler, and Stephens, Phys. Rev. 97, 88 (1955).

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