Electron-capture-delayed fission properties of ^{232}Am

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Delayed fission from the electron-capture decay of ^{232}Am was studied. The ^{237}Np(α,9n)^{232}Am reaction with multiple ^{237}Np targets was used to produce ^{232}Am. The fission properties and half-life of ^{232}Am were measured using a rotating-wheel system. The half-life of ^{232}Am was found to be 1.31 ± 0.04 min from measurements of the fission activity. A highly asymmetric mass-yield distribution was observed for the fission activity, and the average total kinetic energy of the fission fragments was found to be 174 ± 5 MeV. Radiochemical separations confirmed the elemental assignment of the fissioning species to americium or fission following shortly after the decay of americium. The cross section for ^{232}Am produced by this reaction and decaying by electron capture was determined to be 1.3 ± 0.2 μb by measuring the intensities of the daughter plutonium K x rays in radiochemically separated americium samples. The delayed-fission probability was found to be $(6.9\pm1.0)\times10^{-4}$ from the measured ratio of fissions to plutonium K x rays. The observed fissions were unambiguously assigned to electron-capture-delayed fission of americium by measuring the time correlation between the plutonium K x rays resulting from americium K capture and the subsequent fission.

I. INTRODUCTION

Delayed fission (DF) is a nuclear decay process in which a decaying nucleus populates excited states in its daughter nucleus, which then fissions. The excited state can be above the fission barrier(s) of the daughter (yielding prompt fission), within the second well of the potential-energy surface (a fission shape isomer), or within the first well of the potential-energy surface (a spin isomer). This decay mode is believed to influence the production yields of heavy elements in multiple neutron-capture processes[1–5] followed by β decay, such as the astrophysical r process and nuclear weapons tests.[6,7] Delayed-fission processes may also provide a sensitive probe of fission barriers in the heavy-element region.[8]

The probability of electron-capture- (EC) delayed fission (εDF), P_{DF}, can be expressed experimentally as

$$P_{DF}=\frac{N_{\epsilon f}}{N_{\epsilon}}=\frac{\sigma_{\epsilon f}}{\sigma_{\epsilon}}\ , \qquad (1)$$

where N_ϵ is the number of electron captures, and $N_{\epsilon f}$ is the number of those decays leading to delayed fission. Similarly, $\sigma_{\epsilon f}$ and σ_ϵ are the corresponding partial cross sections. P_{DF} can also be derived[9,10] for EC as

$$P_{DR}=\frac{\int_0^{Q_\epsilon} W_\epsilon(Q_\epsilon-E)\frac{\Gamma_f}{\Gamma_f+\Gamma_\gamma}(E)dE}{\int_0^{Q_\epsilon} W_\epsilon(Q_\epsilon-E)dE}\ , \qquad (2)$$

where $W_\epsilon(Q_\epsilon-E)$ is the transition probability function for EC, $[\Gamma_f/(\Gamma_f+\Gamma_\gamma)](E)$ is the ratio of the fission width of excited levels within the daughter nucleus to the total depopulation width of these states, E is the excitation energy of the daughter nucleus, and Q_ϵ is the electron-capture Q value. The transition probability function is the product of the Fermi function for electron capture and the β strength function, which is governed by the structure of the daughter nucleus.

The fission width term $[\Gamma_f/(\Gamma_f+\Gamma_\gamma)](E)$ can be approximated[9,10] by an exponential dependence on the electron-capture Q value and the height of the fission barrier. As a result, ϵDF exhibits a large branching ratio for those nuclei in which the Q_ϵ is comparable to or larger than the fission barrier of the daughter nucleus. By this approximation, ^{232}Am is an excellent candidate for ϵDF since it has a Q_ϵ of 4.9 MeV (calculated using the predicted masses of Möller, Myers, Swiątecki, and Treiner[11]) and fission barriers in the midrange actinides are generally on the order of 5–7 MeV.

The light americium region has been the subject of the most intense experimental study of all regions in which delayed fission is postulated. Indeed, fission tracks attributed to the decay of nuclei in the light americium and neptunium regions were first observed[12,13] as early as 1966, the data of the first experimental report of anomalous fission activities which were later postulated[14,15] to arise from delayed-fission processes. Habs *et al.*[16] observed a fission activity which they attributed to the ϵDF of ^{232}Am in 1978, and the P_{DF} for this isotope was reported to be on the order of 1%. The first measurement of the correlation between the electron capture and the fission was recently reported[17] by Hall and co-workers for ^{234}Am. Hall *et al.* have also used the delayed-fission decay of ^{234}Am to measure[10] the fission properties of its daughter, ^{234}Pu. A summary of all reported delayed-fissioning species may be found in Ref. 18.

Using the Light Ion Multiple Target System[19] (LIM Target System) with multiple targets, it is possible to produce enough ^{232}Am to study the delayed-fission properties of this nucleus. Measurement of the total-kinetic-energy (TKE) and mass-yield distributions is important because the ϵDF mode allows the study of fission from nuclei with very low-excitation energy. This very low-energy mode is essentially inaccessible this far from stability with common techniques such as (n,f) and charged-particle reactions. Low-excitation-energy fission data also assist in understanding the dynamics of the fission process as the excitation energy of the fissioning nucleus goes to zero, approaching ground-state fission.[20–23] The ^{232}Am ϵDF system is also interesting because it is nearly isotonic with nuclei displaying the "radium anomaly," i.e., the onset of triple-humped fission mass-yield distributions.[24–27] Since ^{232}Am-^{232}Pu may be in the transition region between the Ra anomaly and "normal" double-humped mass-yield distributions, its fission properties may provide clues to understanding the Ra anomaly. An earlier study[10] of the TKE and mass-yield distributions of ^{234}Pu showed no evidence of the Ra anomaly, so it is important to study low-energy fission modes in as many nuclei in this region as possible so that the transition between "normal" and "anomalous" fission may be located and understood.

II. EXPERIMENTAL

A. Targets and irradiation

Approximately 10 mg of ^{237}Np was chemically purified and dissolved as the nitrate in isopropanol to form a stock solution. For the initial measurements of the fission properties of ^{232}Am, 12 targets were made by electrodepositing neptunium from aliquots of this solution containing 150–250 μg of ^{237}Np on 2.5-μm molybdenum foils in a 1.23-cm^2 area for each target. Measured target thicknesses ranged from 118 to 142 μg/cm^2. Each foil was mounted on a target holder frame, and ten of the target holder frames were mounted in the LIM Target System, with a spacing of approximately 1 cm between the targets. A 25-μm beryllium foil served as the volume-limiting foil (which confines the gas volume downstream of the last target to the same as that for the rest of the targets) for the LIM Target System, and another 25-μm beryllium foil upstream of the targets served as the vacuum window for the system.

Unfortunately, the interactions of the α-particle beam with the molybdenum target backings produced a very high β-γ background ($\sim 10^{7-8}$/sec after a 1-min irradiation) for these experiments. This did not affect the measurements made with the rotating wheel system, but with such a high-sample activity, fast radiochemical separations followed by γ measurements are difficult due to the background and hazardous to the experimenter in terms of radiation dosage. In order to minimize the background and reduce the risk associated with handling these samples, a new set of targets were made on thin beryllium backings. These targets were used for all subsequent studies on ^{232}Am.

For the second set of ^{237}Np targets, 25-μm beryllium foils were used as target backings. Neptunium was electrodeposited in a 1.27-cm^2 area from aliquots of the isopropanol solution containing 150–250 μg of ^{237}Np for each target. Fifteen targets were made, with measured thicknesses ranging from 124 to 197 μg/cm^2, and mounted on a target holder frames. Twelve of the target holder frames were mounted in the LIM Target System, with a spacing of approximately 1 cm between targets. Again, a 25-μm beryllium foil served as the volume-limiting foil for the LIM Target System, and another 25-μm beryllium foil served as the vacuum window for the system.

The 99-MeV α-particle beam was provided by the Lawrence Berkeley Laboratory 88-Inch cyclotron. The α-particle energy in the first target was 98 MeV, dropping to about 94 MeV after the last target in the stack. The beam intensity was 2–5 $p\,\mu$A for all irradiations. The recoiling reaction products were collected on KCl aerosols in helium, which swept out the volume behind each target continuously. The activity-laden aerosols were transported via a polyvinyl chloride capillary tube to either the Merry Go-'round-Realtime Acquisition Graphics System (MG-RAGS) rotating wheel (see Ref. 10) or to a chemistry laboratory.

B. Chemical procedures

Two different chemical separations were performed on the reaction products of these irradiations. The first separation was designed to determine the elemental assignment of the fission activity, the second was used to produce an americium sample suitable for measurement of the plutonium K x-rays from the EC decay of ^{232}Am. Measurement of the EC-decay rate of ^{232}Am in conjunction with the ^{232}Am ϵDF rate allows a direct determination of P_{DF}. The separations performed for these measurements were the same as used in the earlier work[10] on ^{234}Am, with one exception. In experiments to measure the EC x-rays, the CeF_3 coprecipitation step was omitted to save time. As a result, this procedure took only about 90 sec instead of 4 min.

C. X-ray-fission correlation procedure

The time correlation between the K-capture x ray and the subsequent delayed fission was measured using aerosols collected directly without any chemical separation, as in the earlier studies[10,17] of ^{234}Am. The aerosols were collected on a fiberglass support and, after a suitable collection interval, the support was placed before a light-tight transmission-mounted 300-mm^2 silicon-surface-barrier (SSB) detector operated in air. The SSB detector and support were sandwiched between two planar germanium x-ray detectors to measure the plutonium K x rays following K capture in americium.

However, since low-energy fission is typically accompanied by about ten prompt γ rays[20,28] from the deexcitation of the fission fragments, the summing of the K x rays with these prompt γ rays must be considered when determining the x-ray-detector efficiencies for the K x rays emitted in ϵDF. By measuring the curium x rays in coincidence with the α particles emitted in the decay of ^{249}Cf, the efficiencies of the x-ray detectors were each determined to be 9% in the plutonium K x-ray region. By measuring the prompt γ rays in coincidence with fissions from a ^{252}Cf source, the prompt γ-ray summing rates were determined for each x-ray detector (assuming the prompt γ-ray multiplicities and energy distributions are similar for spontaneous fission of ^{252}Cf and ϵDF of ^{232}Am). Prompt γ-ray summing rates of 30% and 40% in the two detectors led to an overall efficiency for the detection of plutonium K x rays from ϵDF in americium of 12%. The timing resolution for the K x-ray-fission coincidences (determined with the prompt γ rays in coincidence with fission fragments from the ^{252}Cf source) was 8 nsec FWHM. The α-γ coincidences from the ^{249}Cf source were also used to determine the energy calibrations of the x-ray detectors. The background γ rate in the unseparated americium samples was approximately 10^3/sec. Signals from the detectors were digitized and recorded in the same manner as the earlier study[10] of ^{234}Am.

III. RESULTS AND DISCUSSION

A. Elemental assignment

Using the chemical procedure described in Ref. 10, 26 samples were processed and measured over about 3 h. In each case, the aerosols were collected for 3 min and then subjected to the chemical separation. Fissions from each sample were counted continuously for approximately 18 min. Eleven fissions were observed in the americium fractions, and none were observed in the Np-Pu fractions. The observed fissions decayed with a half-life consistent with the measured half-life of ^{232}Am.

Based on this distribution, the fission activity produced in the 99-MeV α irradiation of ^{237}Np was assigned to americium, delayed fission from an americium precursor, or from a short-lived daughter of americium.

B. On-line results

The ϵDF properties of ^{232}Am were measured over a 32 h irradiation using the MG-RAGS rotating-wheel system (described in Ref. 10). The MG wheel was stepped at 1.0-min intervals so that the samples would spend approximately six half-lives (based on the 55-sec half-life reported by Habs *et al.*[16] between the six detector pairs. After each full revolution of the wheel (80 positions), the wheel was replaced with a clean one so that any buildup of long-lived spontaneous fission activities was prevented. During a part of the experiment, the α activities were also recorded. However, a large number of α activities were produced in this irradiation (apparently from traces of lead and bismuth in the targets) which completely overwhelmed the region in which α particles from the decay of ^{232}Am and its daughters were expected to appear, making detection of the α-decay branch of ^{232}Am impossible.

1. Fission properties

A total of 2201 coincident fission-fragment pairs were observed in these measurements using the wheel-stepping interval of 1.0 minute. From these events, the half-life was found to be 1.31±0.04 min, closer to the early half-life of 1.4±0.25 min reported by Skobelev[15] than the more recent value of 0.92±0.12 min reported by Habs *et al.*[16] The decay curve for this fission activity is shown in Fig. 1. Each point on the decay curve has been normalized to represent the same number of samples per detector station. This is necessary since, for each wheel, the first station sees 80 foils before the acquisition is stopped while the second station sees 79, and the third 78, and so on. The correction is fairly small (0% for the first station, rising to 12% for the last), but can significantly affect the measured half-life.

From the decay curve, an apparent fission cross section (a partial cross section for ^{232}Am nuclei produced in this reaction and decaying by ϵDF) was estimated for the ^{232}Am ϵDF mode from this reaction. The effective target thickness was estimated by extrapolating low-energy recoil ranges[29] for the compound nucleus linearly to zero energy.[30] This gave an estimate of the effective target thickness of 100 μg/cm^2 per target. The efficiency of the aerosol-transport system was taken as 100%. These assumptions result in an apparent fission cross section of about 2.5 nb. (It should be noted that this is, in fact, a lower limit due to the 100% efficiency assumed for the gas jet; however, other experiments have indicated normal operating efficiencies of the LIM system at about 80–100% so the 100% estimate is not grossly wrong. In these experiments, unfortunately, there was no way to directly measure the efficiency.)

The fission-fragment distributions were analyzed using the method originated by Schmitt, Kiker, and Williams

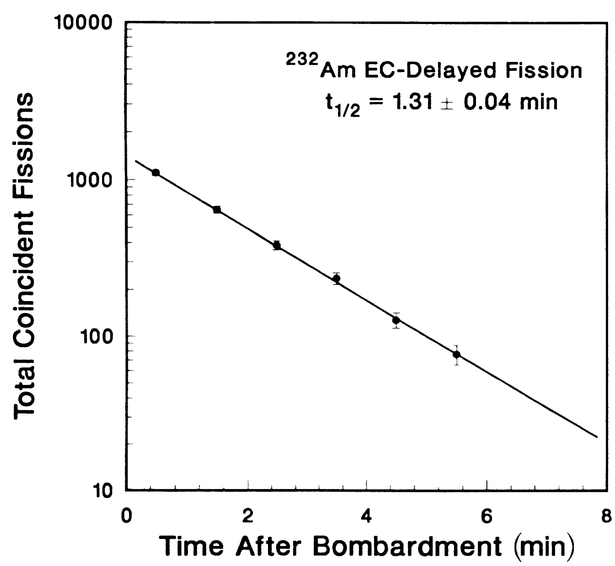

FIG. 1. Decay curve of the ^{232}Am EC-delayed fission activity as measured on MG-RAGS. The wheel-stepping times was 1.0 min per station.

(SKW).[31] The ^{252}Cf calibration constants were taken from Weissenberger and co-workers.[32] The measured post-neutron-emission distributions were corrected for neutron emission using a neutron-emission function $\overline{\nu}(A)$ similar to that of ^{252}Cf normalized to $\overline{\nu}_T = 2.40$ (estimated from systematics[20]).

Fission from ^{232}Am was observed to have a highly asymmetric mass distribution, with no trace of the triple-peaked mass distribution characteristic of the radium anomaly. The preneutron-emission mass-yield distribution is clearly two humped, with a well-defined valley and no evidence shown of a symmetric component. The preneutron-emission TKE distribution is symmetric about 174 MeV with no evidence of multiple components. The preneutron-emission TKE and mass-yield distributions are presented graphically in Fig. 2. The $\overline{\text{TKE}}$ value of 174 MeV for the ^{232}Am ϵDF is comparable to the predicted $\overline{\text{TKE}}$ (Refs. 33 and 34) for ground-state fission from ^{232}Pu, as shown in Fig. 3, and very similar to the measured[10] $\overline{\text{TKE}}$ of ^{234}Pu, 175 MeV.

The behavior of the TKE and $\overline{\text{TKE}}$ as a function of mass fraction is shown in the TKE contour[35] plot in Fig. 4. From this figure, it is noteworthy that the $\overline{\text{TKE}}$ for near-symmetric mass division is about the same as the TKE's for asymmetric mass division. The statistical significance of this point is poor (only 46 events were observed at this mass division), but an elevated $\overline{\text{TKE}}$ at symmetry is unusual for light actinides (other plutonium isotopes display $\overline{\text{TKE}}$'s at symmetries which are lower by 20 MeV or more than the $\overline{\text{TKE}}$'s for the most-probable asymmetric mass division[10,36–39]). Such an effect was not observed in the ϵ(DF) (Ref. 10) of ^{234}Am-^{234}Pu. If the symmetric fragments were approaching a spherical shell, this behavior might be expected, since elevated $\overline{\text{TKE}}$'s at symmetry has been observed in the heavy-fermium region[28] where the fission fragments can approach two doubly magic ^{132}Sn nuclei and thus have higher $\overline{\text{TKE}}$'s at symmetry. For ^{232}Pu, the symmetric fragments would be $^{116}_{47}Ag_{69}$. This is probably too far from the $Z=50$ shell to

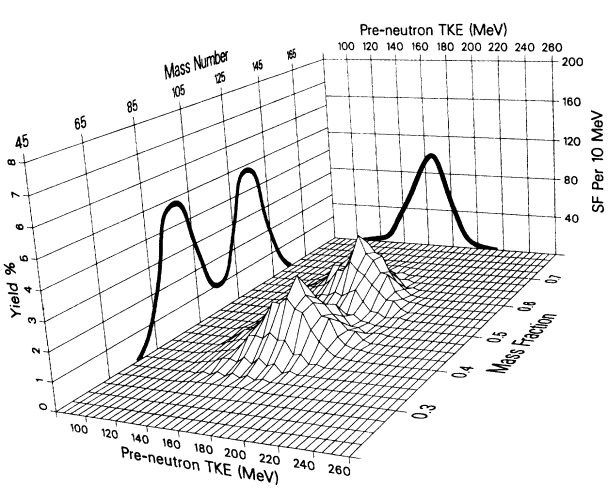

FIG. 2. Preneutron emission total-kinetic-energy (TKE) distribution of the ^{232}Am ϵDF mode and preneutron emission mass-yield distribution.

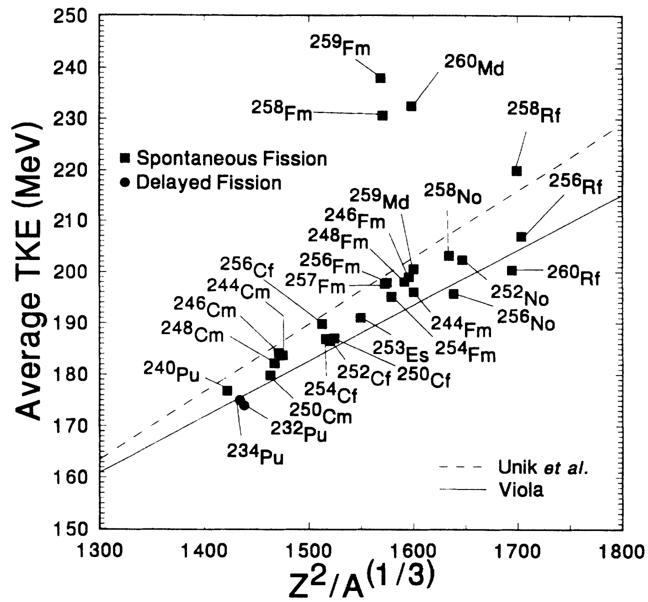

FIG. 3. Average total kinetic energy as a function of $Z^2/A^{1/3}$. The solid line is a linear fit of Viola (Ref. 33), and the dashed line is from Unik *et al.* (Ref. 34). Ground-state (spontaneous) fission data for the trans-berkelium actinides are taken from Hoffman and Somerville (Ref. 28), and data for the lighter actinides are from Hoffman and Hoffman (Ref. 20). $Z^2/A^{1/3}$ for delayed fission is calculated for the daughter since that is the fissioning nucleus. Solid square data points (■) are for spontaneous fission, solid circles (●) are for delayed fission.

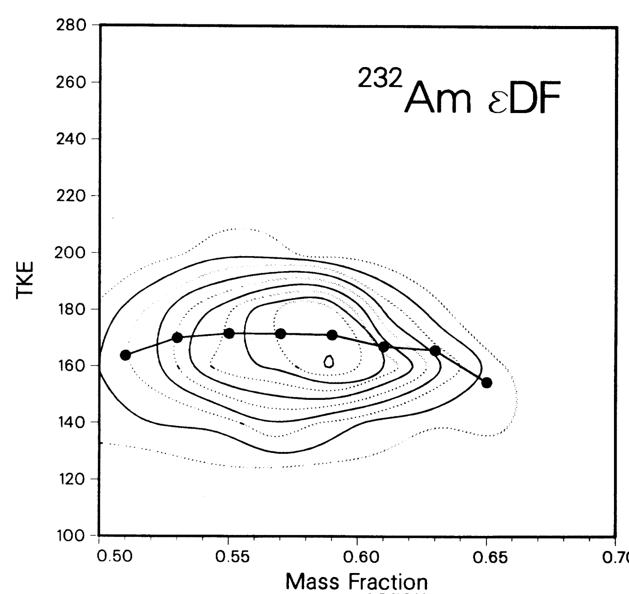

FIG. 4. Total kinetic energy and average total kinetic energy of ^{232}Am as a function of mass fraction. The solid points are average TKE values at each mass fraction. The contours represent the mass yield (normalized to 200%) as a function of TKE and mass fraction. The transition between a solid and dashed contour represents a change of 0.5% in the mass yield.

TABLE I. Summary of the fission characteristics of ^{232}Am ϵDF.

Post-neutron $\overline{TKE}$ [a]	173 ±5 MeV
Preneutron $\overline{TKE}$	174 ±5 MeV
Post-neutron $\overline{KE}$ [b] of high-energy fragment	99.4±1.9 MeV
Post-neutron $\overline{KE}$ of low-energy fragment	73.6±2.0 MeV
Preneutron $\overline{KE}$ of high-energy fragment	100.2±1.9 MeV
preneutron $\overline{KE}$ of low-energy fragment	74.2±2.0 MeV
Average mass of the light-fission fragment	98.7±0.3
Average mass of the heavy-fission fragment	133.3±0.3

[a]Average total kinetic energy.
[b]Average kinetic energy.

expect an effect, and is quite far from the $N=50$ and 82 shells. However, it may be that fission of ^{232}Pu is being affected by the half-filled shell at $N=66$. It has been observed that there is a transition from spherical to deformed nuclei at about $N=60$ for elements of lower Z than silver,[40] so the high $\overline{\mathrm{TKE}}$ observed near symmetric mass division for ^{232}Pu may signal the gradual onset of a similar transition in the silver isotopes. The fission properties of the ^{232}Am ϵ mode are summarized in Table I.

The highly asymmetric mass division and symmetric TKE distribution show no trace of the radium anomaly, although the data in the contour plot hint at a possible effect. Therefore, the transition region between "normal" double-humped mass distributions and the triple-humped distribution of the radium anomaly must begin with lighter elements for this neutron number.

C. P_{DF} and σ_ϵ results

Americium fractions were repeatedly isolated chemically from 4-min collections taken over a irradiation period of about 24 h in order to measure the x rays from americium K capture. Fission measurements were made on an alternating basis with the chemical separations. The chemically purified americium samples were repeatedly γ counted for 20 min each, and the fission samples were each counted for 4 min in a windowless gas-flow proportional counter, and the integrated fissions were recorded. The γ- and x-ray peaks were fitted and integrated using the SAMPO (Ref. 41) computer code, and a half-life analysis was performed with the CLSQ (Ref. 42) code.

The initial activities determined for the americium electron-capture-decay mode were corrected for detector efficiency, individually measured chemical yields, branching ratio, and K-fluorescence yield (taken as 97.7% (Ref. 43)]. The resulting initial disintegration rates were used for the calculation of σ_ϵ and P_{DF}.

The partial cross section for ^{232}Am nuclei produced and decayed by electron capture σ_ϵ was calculated based on the following assumptions. First, the effective target thicknesses were estimated the same way as for the determination of the apparent fission cross section, yielding an effective total target thickness of 100 μg/cm^2 per target for ^{232}Am. Second, the gas-jet yield was assumed to be 100%. Third, because of the lack of discernible γ lines in the spectrum with half-lives consistent with the decay of ^{232}Am, it was assumed that the level density of the plutonium daughter was high enough that deexcitation proceeded through a series of high-energy (500–1000 keV), low-multipolarity transitions. Based on this assumption, the K x-ray production from internal conversion was taken to be negligible. Of course, the last few transitions should be more highly converted, but without detailed information about the daughter-level scheme, any estimates on K conversion would be nearly meaningless. Finally, it was assumed that K capture was by far the dominant mode of electron capture for ^{232}Am; L and M capture were neglected.

The K x-ray region from a representative γ spectrum is shown in Fig. 5. The plutonium x rays resulting from the electron capture of americium are weak, but visible. Half-life analysis of the Pu K x rays revealed a two-component decay curve, with one component being consistent with 1.31 min and the other on the order of an hour. The long component is attributed to a mixture of

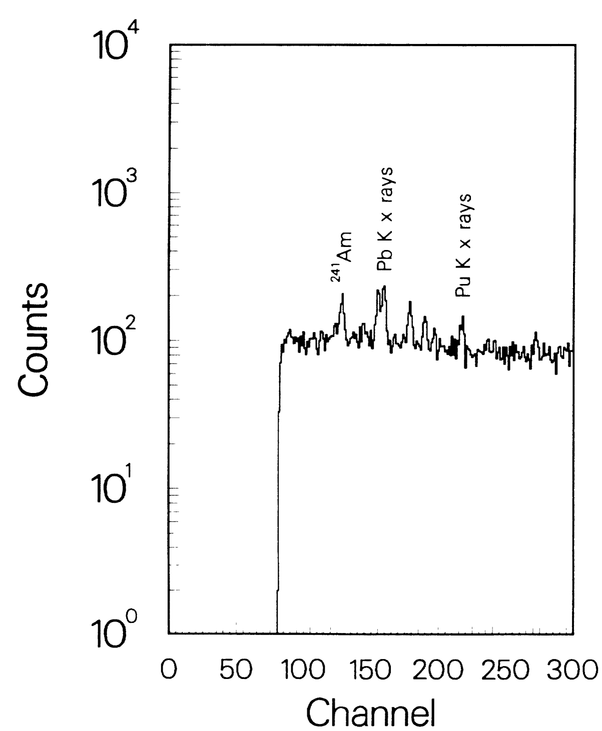

FIG. 5. The K x-ray region of the gamma spectrum of a chemically purified ^{232}Am sample.

^{237}Am ($t_{1/2}$=73 min) and ^{238}Am ($t_{1/2}$=1.63 h), and the short to ^{232}Am. The K x rays were fitted with two components using CLSQ, with the short component being set at 1.31 min and the long component allowed to vary to produce the best fit. An example of such a fit is shown in Fig. 6.

The delayed-fission probability was calculated from the electron-capture initial activities and the average number of fissions observed in the bracketing fission samples. By measuring each quantity nearly simultaneously, experimental variables such as the target thickness, the beam flux (since the flux was held at a constant 7 $e\,\mu$A throughout the experiment, with less than 5% deviation), and the gas-jet yield should all cancel out. This allows us to calculate P_{DF} with a variant of Eq. (1),

$$P_{DF}=\frac{\lambda I_f/[e^{-\lambda t_1}-e^{-\lambda(t_1+t_c)}]}{D_{0,\epsilon}}, \quad (3)$$

where I_f is the number of fission observed in a counting time t_C, t_1 is the time from the end of bombardment to the start of the fission counting, and $D_{0,\epsilon}$ is the initial activity for electron capture. Employing this relationship and performing an error-weighted average over all of the separate determinations yielded a value of P_{DF} of $(6.9\pm1.0)\times10^{-4}$ at the 1σ (68%) confidence level. From these D_0 values, σ_ϵ was also estimated to be 1.3 ± 0.2 μb at the 1σ confidence level. Individual measurements are given in Table II.

This value for P_{DF} is approximately a factor of 20 smaller than the value reported by Habs *et al.*,[16] and nearly a factor of 100 smaller than the estimate of Kuznetsov.[44] However, Kuznetsov's P_{DF} estimates rely on evaporation codes to estimate σ_ϵ, and Hab's report uses a σ_ϵ estimated from α data. Either of these estimates could easily be wrong. Our measurement, however, uses nearly 30 separate experimental determinations

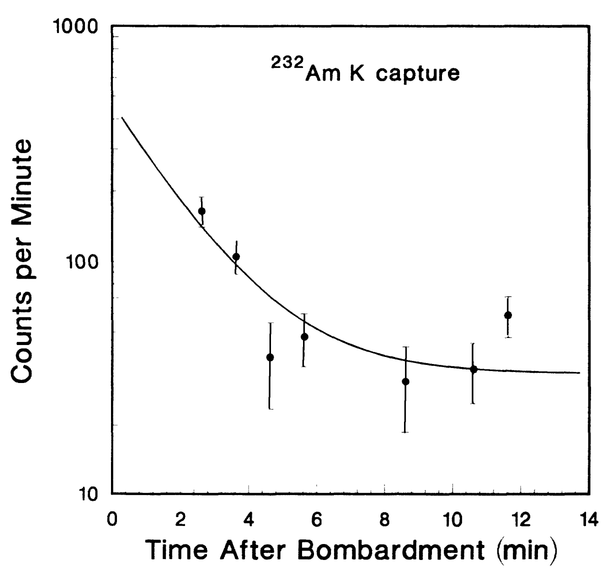

FIG. 6. Representative half-life fit for the plutonium $K_{\alpha1}$ x ray observed in the chemically purified ^{232}Am sample.

TABLE II. Individual measurements of P_{DF} for ^{232}Am.

$D_{0,\epsilon}$	I_f[a]	P_{DF} (10^{-4}) (%)
2894±35	1.0±0.7	2.71±78
8480±16	1.5±0.9	1.39±62
2777±42	0.5±0.5	1.41±108
4201±26	4.5±1.5	8.39±42
64±1700	1.0±0.7	1.22±1701
3480±31	1.5±0.9	3.38±68
3734±33	3.0±1.2	6.29±52
1150±144	1.5±0.9	10.2±156
226±53	3.5±1.3	12.1±65
2340±153	2.5±1.1	8.37±159
53±3000	3.5±1.3	518±3000
3382±33	4.5±1.5	10.4±47
3383±25	5.0±1.6	11.6±41
2066±81	3.5±1.2	13.3±89
7021±24	3.0±1.2	3.35±47
5614±52	2.5±1.1	3.49±68
745±153	2.5±1.1	26.0±159
2086±46	1.0±0.7	3.75±84
9500±19	1.0±0.7	0.82±73
6287±27	1.0±0.7	1.25±75
3234±53	2.5±1.1	6.05±69
4854±28	1.5±0.9	2.42±66
1355±57	2.5±1.1	14.5±72
1521±114	1.0±0.7	5.15±134

[a]I_f for the P_{DF} measurement of ^{232}Am is the average of the preceding and succeeding fission measurements.

of σ_ϵ through the plutonium K x rays. Of course, this method of measuring P_{DF} is sensitive to K conversion of γ rays, but it would require a cascade of 20 γ rays that are 100% K converted *per electron capture* to account for the discrepancy with Habs. It seems much more likely that the evaporation codes and decay systematics become unreliable this far from stability. It should be noted that one cannot take our lower limits on the fission and EC cross sections and directly calculate P_{DF}. If one were to do so, one would obtain a value of 2×10^{-3}, which is closer to the value reported by Habs.[16] However, this number would need to be scaled by the different gas-jet efficiencies since these were separate measurements with separate capillaries and collection equipment. We have no way of quantifying the overall gas-jet yield directly in these irradiations, which is why we used the alternating-measurements technique. ^{232}Am was formed by the ^{237}Np(α,9n) reaction in the study by Habs *et al.*,[16] and the data used by Kuznetsov[44] involved the ^{230}Th(^{10}B,8n)^{232}Am reaction. Our limits and values are summarized in Table III.

TABLE III. Summary of the cross-section lower limits and decay branches for ^{232}Am.

$\sigma_\epsilon > 1.3$ μb
$\sigma_{\epsilon f} > 2.5$ nb
$P_{DF}=(6.9\pm1.0)\times10^{-4}$
$t_{1/2}=1.31\pm0.04$ min

D. X-ray-fission results

Samples were collected from the gas-jet system at 2-min intervals, and these samples were placed in the counting chamber for the correlation studies. Figure 7(a) shows the x-ray and γ spectrum of those events in prompt coincidence with the fission signal. The data in Fig. 7(c) are the logarithm of a maximum-likelihood function[10] L for finding an idealized x-ray spectrum [shown in Fig. 7(b)] in the observed data as a function of the $K_{\alpha 1}$ position.

From the likelihood functions, the most probable $K_{\alpha 1}$ energy was found to be 103.8±0.3 keV for ^{232}Am ϵDF, in excellent agreement with the plutonium $K_{\alpha 1}$ energy[43] of 103.76 keV. The total number of K x rays was found to be 42±8 by allowing the intensity of the ideal spectrum to vary within the maximum-likelihood analysis. Observed and expected x-ray intensities are given in Table IV.

The number of x-ray-fission coincidences relative to the total number of fissions was consistent with the detector geometries, indicating that all of the observed fissions were due to ϵDF. The number of fissions in coincidence with prompt γ rays from the deexcitation of fission fragments relative to the total number of fissions observed indicate that the γ multiplicity of ^{232}Am ϵDF is similar to that of spontaneous fission in ^{252}Cf. No evidence was observed for fission-delay times longer than the best timing resolution of this experiment, about 8 nsec. The fact that plutonium x rays can be seen requires that the lifetime of the fissioning state be longer than the time it takes the orbital electrons to cascade down and fill a K vacancy. The time required for this is on the order of 10^{-17} sec.[45] We can therefore set boundaries on the excited-state half-lives of $10^{-8} < t_{1/2} < 8$ nsec for ^{232}Pu. If the nucleus is truly heavily damped in the second well for low energies (as is commonly assumed,[9,10,16] and supported by experimental measurements[46]), then these limits are also limits on the lifetimes of the shape isomer ^{232f}Pu. These limits are consistent with the half-life systematics of plutonium-shape isomers (see Fig. 3 of Ref. 47), from which one would expect the half-life of ^{232f}Pu to be in the range of 1–10 psec. The observation of x-ray fission correlations in this experiment unequivocally proves that the decay is indeed EC-delayed fission. The coincidence γ data in Fig. 7 also show what appear to be photopeaks at about 77, 87, 111, 240, 247, and 283 keV. These peaks are very weak, but prompt γ rays from fission fragments do not display such structure. It is possible that these γ rays are a result of the level structure of ^{232}Pu in the second well. If this is the case, the correlation of these γ

TABLE IV. Observed and expected x-ray intensities from the correlated x-ray fission data for ^{232}Am. Expected x-ray intensities are taken from the Table of Isotopes (Ref. 43).

X ray	E (keV)	I_{theo}	No. observed[a]	I_{obs}
Pu $K_{\alpha 2}$	99.55	0.299	19	0.33±0.09
Pu $K_{\alpha 1}$	103.76	0.479	23	0.40±0.10
Pu $K_{\beta 1'}$	116.9	0.162	11	0.19±0.06
Pu $K_{\beta 2'}$	120.6	0.060	4	0.07±0.04

[a]Approximately 15±4 of the observed events are attributable to the prompt γ-ray continuum for the ^{232}Am study.

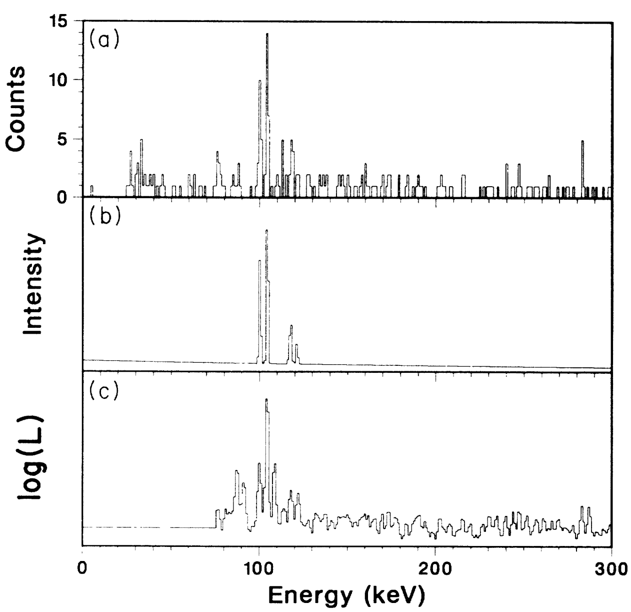

FIG. 7. X-ray-fission correlation results for ^{232}Am. (a) X rays and γ rays in coincidence with delayed fission from ^{232}Am. (b) An idealized plutonium K x-ray spectrum, based on the measured detector resolution and prompt γ-ray continuum. (c) The likelihood function for the position of the ideal spectrum (b) in the data (a), as a function of the $K_{\alpha 1}$ position.

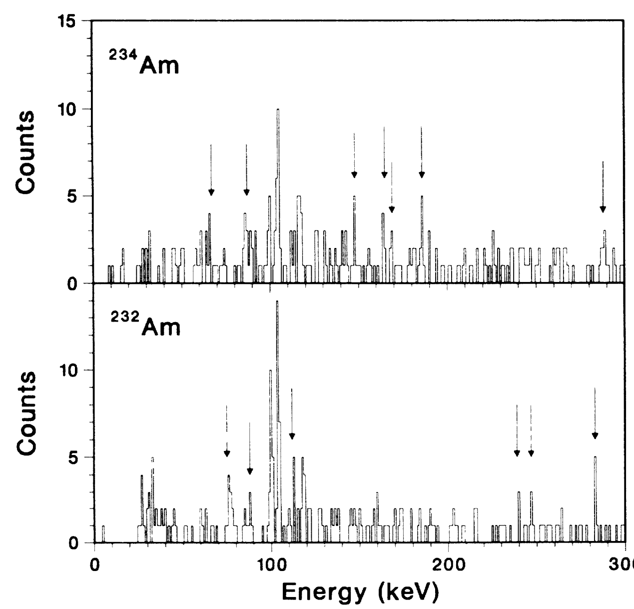

FIG. 8. The spectra of x and γ rays in coincidence with fissions from ϵDF in ^{232}Am and ^{234}Am. Anomalous background peaks which could arise from γ transitions in the second well are marked with arrows.

rays supports the hypothesis that the second well is strongly damped. Unfortunately, the very low statistics and lack of γ-γ-fission-time correlation data precludes constructing a level scheme for ^{232f}Pu. Similar anomalous γ lines[10,30] also appear in the x-ray-fission data for ^{234}Am, as shown in Fig. 8. More investigation is required to understand these phenomena.

IV. CONCLUSIONS

^{232}Am was produced using multiple ^{237}Np targets irradiated with α particles in the energy range 98–94 MeV at the Lawrence Berkeley Laboratory 88-inch cyclotron. The half-life was determined to be 1.31 ± 0.04 min using the MG-RAGS rotating-wheel system. The fission properties of the ^{232}Am ϵDF mode were measured. It was found to have highly asymmetric mass division. The total-kinetic-energy distribution displayed only one component, and had an average of TKE of 174 ± 5 MeV. The mass-yield distribution showed no evidence of the radium anomaly observed in lighter-Z nuclei with the same neutron number.

Radiochemical measurements of the electron-capture branch of ^{232}Am yielded a cross section of 1.3 ± 0.2 μb for the ^{237}Np(α,9n) reaction. The delayed-fission probability was measured experimentally and found to be $(6.9\pm1.0)\times10^{-4}$.

Finally, the observation of coincidences between the plutonium K x rays and fission provided direct proof that the fissions observed in this experiment are the result of americium K capture followed promptly (≤ 8 nsec) by fission of the daughter plutonium nucleus. These data show that ϵDF is a true decay mode of ^{232}Am, and offers the intriguing prospect of observing γ transitions in the decay of nuclear-shape isomers. The unequivocal evidence for ϵDF in ^{232}Am given here and in the earlier[10] work on ^{234}Am shows that delayed fission is general mode of decay for heavy nuclei with sufficiently large Q_ϵ values. This supports the assignment of other observed fission activities in neutron-deficient regions (see Ref. 10 for a tabulation of these activities) to delayed fission.

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