

Electron-capture delayed fission properties of ^{244}Es

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Electron-capture delayed fission was observed in 37-s ^{244}Es produced via the $^{237}\text{Np}(^{12}\text{C},5n)^{244}\text{Es}$ reaction at 81 MeV (on target) with a production cross section of $0.31 \pm 0.12 \mu\text{b}$. The kinetic energies of coincident fission fragments were measured with our rotating wheel detection system and the average pre-neutron-emission total kinetic energy of the fragments was found to be $186 \pm 19 \text{ MeV}$. The mass-yield distribution of the fission fragments is predominantly asymmetric. Based on the ratio of the number of fission events to the measured number of α decays from the electron-capture daughter ^{244}Cf (100% α branch), the probability of delayed fission was determined to be $(1.2 \pm 0.4) \times 10^{-4}$. This value for the delayed fission probability fits the experimentally observed trend of increasing delayed fission probability with increasing Q value for electron capture.

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I. INTRODUCTION

Electron-capture delayed fission (ECDF) is a nuclear decay mode whereby a parent nucleus undergoes electron-capture (EC) decay, populating excited states in the daughter nucleus, which then fission. The ECDF decay mode is of special interest because it allows study of the fission properties of the daughter nucleus, which would normally have a ground state spontaneous fission (SF) branch too small for detailed investigation. Delayed fission (DF) is also thought to play an important role in determining the yields of heavy elements produced in multiple neutron capture processes such as the astrophysical r process and in nuclear weapons tests [1–5]. For a more complete description of the DF process including a theoretical derivation, see Refs. [6–9], and the references therein.

The probability of undergoing ECDF (P_{DF}) is defined as the ratio of the number of EC decays resulting in fission N_{ECDF} to the total number of EC events N_{EC} :

$$P_{\text{DF}} = \frac{N_{\text{ECDF}}}{N_{\text{EC}}}.$$

ECDF has been previously reported in neutron deficient neptunium [10,11], americium [7,8,12,13], berkelium [10,13,14], and einsteinium [10,13,15–17] isotopes. This decay mode is expected to have measurable branches in nuclides where the electron-capture Q value (Q_{EC}) is compa-

able to the height of the fission barrier in the daughter nucleus. Nuclides that meet this requirement are found in neutron-deficient actinides that have odd numbers of protons and neutrons. These odd-odd nuclei have enhanced Q_{EC} values associated with EC decay to their more stable even-even daughter nuclei. The Q_{EC} for ^{244}Es is calculated to be 4.36 MeV [18], which approaches the estimated fission barrier heights of 5–7 MeV for this region [19]. Previous experiments have shown that the P_{DF} increases with increasing Q_{EC} [9,11,14,16,17].

^{244}Es was first identified by Eskola [20] during an experiment in which ^{233}U was bombarded with ^{15}N projectiles. In this preliminary report, ^{244}Es was reported to decay with a 100% EC branch and a half-life of $40 \pm 5 \text{ s}$. Furthermore, no α particles from the decay of ^{244}Es were observed during an experiment in which ^{241}Am was bombarded with ^{12}C projectiles to look for isotopes of mendelevium and their einsteinium daughters [21]. A subsequent paper by Eskola *et al.* [22] reported α particles from the decay of ^{244}Es produced via the $^{233}\text{U}(^{15}\text{N},4n)^{244}\text{Es}$ reaction at projectile energies of 77–82 MeV. They assigned an α energy of $7.57 \pm 0.02 \text{ MeV}$, an α branch of $4^{+3}\%$, and a half-life of 37 $\pm 4 \text{ s}$ to ^{244}Es .

ECDF in ^{244}Es was first reported in 1980 by Gangrskii *et al.* [13]. The nuclide was produced both via the $^{233}\text{U}(^{14}\text{N},5n)^{244}\text{Es}$ and $^{237}\text{Np}(^{12}\text{C},5n)^{244}\text{Es}$ reactions at projectile energies of 82–86 MeV. The production cross section was reported to be $1 \mu\text{b}$ but it was not specified with which reaction this cross section was associated. A P_{DF} of 10^{-4} was determined by comparing the number of fission events observed in a solid-state fission track detector to the number of α -decay events from the ^{244}Cf EC daughter. The total number of ^{244}Es EC events was determined from the number of daughter events by assuming a 100% EC branch in ^{244}Es . No errors were given for this reported P_{DF} value. Also, the fission properties of the ^{244}Cf daughter were not determined. Therefore, we decided to measure the ECDF of ^{244}Es in order to better evaluate its P_{DF} value and to determine the fission properties of its EC daughter.

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II. EXPERIMENTAL TECHNIQUES

A. Targets and irradiation

An aqueous solution containing 1.61 mg of ^{237}Np was sorbed onto a 7.5 mm by 27.5 mm anion exchange column (AG 1X-8 resin, 200–400 mesh) and rinsed with concentrated HCl to remove lead and other impurities. A small amount of ^{239}Pu that was present in the original solution was removed from the column by eluting with a 7:1 solution of concentrated HCl:HI. Any residual HI was removed by rinsing the column with concentrated HCl, and the ^{237}Np was eluted with 2 M HCl. The resulting solution, which contained 480 μg of ^{237}Np , was evaporated to dryness and dissolved in 1 mL of isopropyl alcohol (IPA) to yield a solution that was approximately 0.5 mg/mL in ^{237}Np . Successive target layers were produced by electroplating aliquots containing 25 μg of ^{237}Np from 1.25 mL of IPA in a 6-mm diameter circle (area of 0.28 cm^2) on a 0.5-mil (2.32 mg/cm^2) Be foil at 300 V (0.7 mA) for 30 min. The ^{237}Np was then converted to the oxide by baking each layer in a 450 $^\circ\text{C}$ oven for 30 min. The amount of ^{237}Np in the target was determined by counting the α emissions from the ^{237}Np in an α -spectrometer system utilizing a Si(Au) solid state surface barrier detector operated under vacuum with a detection efficiency of $34 \pm 5\%$. From the measured ^{237}Np α decay rate of $(2.2 \pm 0.3) \times 10^5$ disintegrations per minute the number of ^{237}Np atoms electroplated on the Be foil was calculated to be 3.5×10^{17} atoms using a ^{237}Np half-life of 2.14×10^6 y, resulting in 0.49 ± 0.02 mg/cm^2 of ^{237}Np in the target.

A 3.0- μA $^{12}\text{C}^{4+}$ beam (81 MeV in the lab system at the entrance to the target) was provided by the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory. The target chamber configuration has been described elsewhere [23]. During bombardment, reaction products were swept from the target chamber, attached to KCl aerosols from a He/KCl gas jet, and then transported via a 1.4-mm i.d. Teflon capillary to our rotating wheel detection system [24] for α and fission measurements.

B. Measurements of α and fission activity

Online measurements of α particles and fission fragments were made in our merry-go-around (MG) rotating wheel collection and detection system [24]. This rotating horizontal wheel, online continuous collection and detection system, has been previously described by Hoffman *et al.* [24]. The activity-laden KCl aerosols were deposited via the He/KCl gas-jet successively onto 80 thin polypropylene foils (40 ± 10 $\mu\text{g}/\text{cm}^2$) supported on 0.63-mm i.d. rings positioned around the periphery of a 51-cm diameter fiberglass wheel. There were 80 collection sites on each wheel, but only 79 were used during a given experiment. The transport efficiency of the gas-jet system was estimated to be $60 \pm 20\%$ based on previous experiments [25]. Six pairs of passivated ion implanted silicon (PIPS) detectors were situated directly above and below the wheel to measure the kinetic energy of α particles and coincident fission fragments. The horizontal wheel was rotated every 30 s so as to move the first foil from the collection site into position for counting between the first

detector pair while collection proceeded concurrently on a new foil. Each step of the wheel moved a new foil into the collection position and the collected samples were moved successively between the six pairs of detectors so that each collection foil was counted for a total time equivalent to 180 s. With this system, collection and counting are essentially continuous since the time required to move the wheel (~ 0.1 s) is much less than the stepping interval. The efficiency in any given detector was approximately $32 \pm 3\%$ for α particles and $64 \pm 6\%$ for fission fragments.

After 80 min of continuous measurement (two complete revolutions of the wheel), the last six collections were stopped under the detector pairs and counted while the wheel was stationary for an additional 40 min. During this time interval, the longer-lived daughter activity was measured after the shorter-lived interfering activities had decayed away. After that time, the wheel was replaced with a clean one to prevent the buildup of KCl on the foils to avoid degradation of the α resolution during the experiment, and to prevent the buildup of any longer-lived fission activities. This entire process was continually repeated over the course of 36 h of beam time.

Data were collected using the GOOSY data acquisition system [26]. Calibrations were performed before the experiment using a ^{212}Pb source, which provided 6.062-MeV and 8.784-MeV α particles. A ^{252}Cf source was used for calibration of fission fragment energies. Fission fragment energy calibrations were based on the SF of ^{252}Cf using the method of Schmitt, Kiker, and Williams [27] and the constants of Weissenberger *et al.* [28]. 78.4 and 102.6 MeV were used for the most probable post-neutron low and high fragment kinetic energies for ^{252}Cf . The ^{252}Cf calibration source was measured on the same kind of polypropylene collection foils used on the MG wheels during the experiment, so no correction was made for energy degradation of fission fragments as they traveled through the foils to the bottom detectors. No correction was made for the approximately 10–15 $\mu\text{g}/\text{cm}^2$ of KCl aerosol [29] deposited on each foil by the gas-jet transport system because typical fission fragments only lose 0.2–0.4 MeV of energy [30] as they travel through this amount of KCl. The energy resolution [full width at half maximum (FWHM)] of the detectors positioned above the wheel was approximately 0.04 MeV and the detectors below the wheel had a resolution of approximately 0.1 MeV due to energy degradation of the α particles as they traveled through the polypropylene foil. The fission background was measured prior to the start and at the end of the experiment and was less than one fission event per single detector per day.

III. RESULTS AND DISCUSSION

A. Half-life and fission properties

A total of 13 pairs of coincident fission fragments was detected over the course of the entire experiment. Subsequent analysis of the data showed that at some point during the experiment the first detector pair had stopped working. Only two coincident fission events were detected in the first pair instead of the approximately ten we would expect based on the subsequent decay curve of observed fission events.

The two events from detector pair one were removed from the half-life analysis but were included in determining fission properties. The half-life of the coincident fission events was determined from a one-component fit using a maximum likelihood decay by the simplex method, the MLDS computer code [31]. This resulted in a half-life of 38 ± 11 s for the fission events. The population of states in the fissioning daughter nucleus occurs with the half-life of the EC parent, and the subsequent fission is instantaneous compared to this half-life [32]. Therefore, the fission events decay with the characteristic half-life of the parent nucleus. Even though it is based on only a few coincident fission events, our half-life of 38 ± 11 s is consistent with the best-reported value of 37 ± 4 s for ^{244}Es by Eskola *et al.* [22]. We attribute the observed fissions to ECDF in ^{244}Es based on this half-life and the observed α decay of ^{244}Cf (its EC daughter) which would be seen in the α spectrum if ^{244}Es were present (see Sec. III B). In addition, there are no other known spontaneous fission or ECDF activities that would have been produced in the reaction of ^{12}C with ^{237}Np . The only other nuclide that could have been produced with a half-life close to that of ^{244}Es is ^{243}Es ($t_{1/2} = 21 \pm 2$ s [30]). It is unlikely that the fission events could have come from ^{243}Es due to its lower Q_{EC} of 4.0 MeV [18]. From previous data [7,8,11,16,17,19], the P_{DF} of ^{243}Es (based on a Q_{EC} of 4.0 MeV) is estimated to be $< 5 \times 10^{-5}$, too low to account for the number of fissions

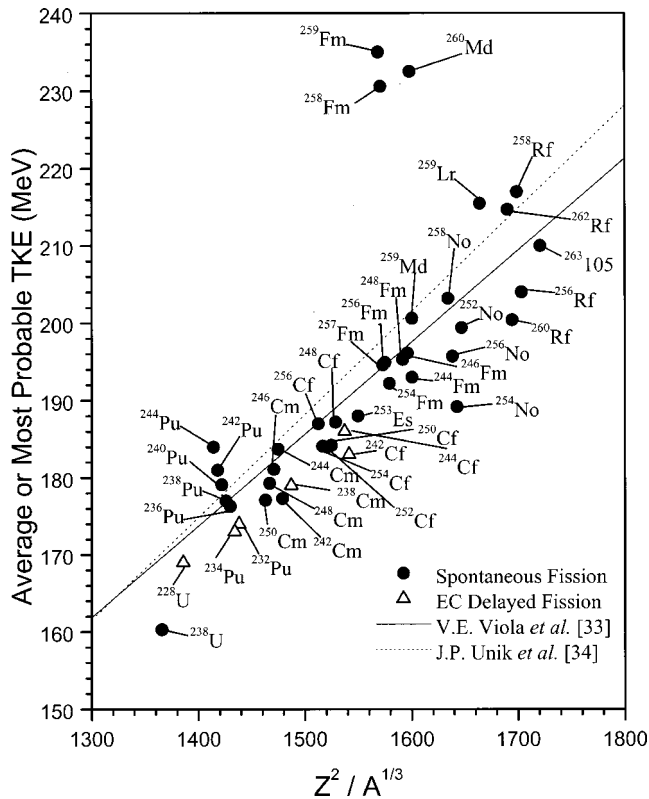


FIG. 1. The average or most probable TKE vs $Z^2/A^{1/3}$ for known cases of spontaneous or delayed fission is shown. The solid line is the linear fit of Viola *et al.* [33] and the dashed line is from Unik *et al.* [34]. All of the TKE values have been corrected to be consistent with the calibration parameters of Weissenberger *et al.* [28].

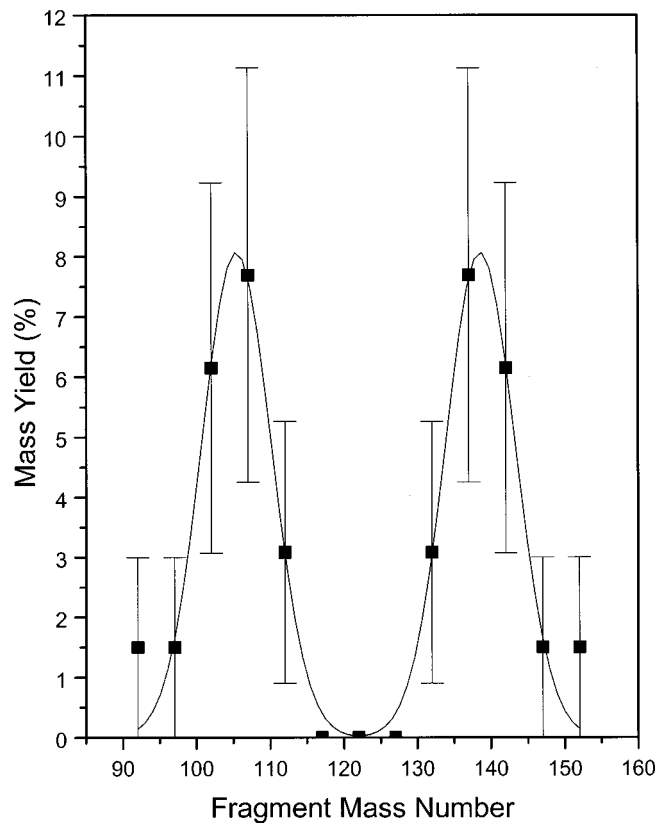


FIG. 2. Pre-neutron-emission mass-yield distribution for the ECDF of ^{244}Es . The fissioning species is ^{244}Cf . The data were averaged over five mass units. Mass yield (%) is expressed as yield per fragment mass number normalized to 200% total fragment yield.

detected during the experiment. Also, the cross section of the $^{237}\text{Np}(^{12}\text{C},6n)$ reaction would be expected to be much lower than the corresponding $5n$ reaction, resulting in an even lower number of fissions. Since no other nuclide produced in this reaction has a fission branch large enough to account for 13 fissions and their half-life of 38 ± 11 s is consistent with that previously reported for ^{244}Es , we have assigned these events to ECDF of ^{244}Es .

Since fission events in ECDF are preceded by EC decays, the fission properties measured during our experiment are for the EC daughter ^{244}Cf . The average neutron emission function for ^{244}Cf , $\bar{\nu}(A)$, was assumed to be similar to that of ^{252}Cf , normalized to an average neutron emission, $\bar{\nu}_i = 2.6$, estimated from systematics in Ref. [33], and was used to calculate pre-neutron-emission total kinetic energy (TKE) values from the measured post-neutron-emission TKE values. The average pre-neutron-emission TKE for coincident fission fragments from ^{244}Cf was determined to be 186 ± 19 MeV. The most probable light fragment pre-neutron-emission energy was determined to be 79 ± 10 MeV and the most probable heavy fragment energy was 107 ± 10 MeV. Figure 1 shows the average or most probable TKE versus $Z^2/A^{1/3}$ for all known spontaneous fission and delayed fission isotopes, as well as the empirical fits of Viola *et al.* [34] and Unik *et al.* [35]. The average TKE of 186 ± 19 MeV agrees, within the statistical error, with these empirical predictions

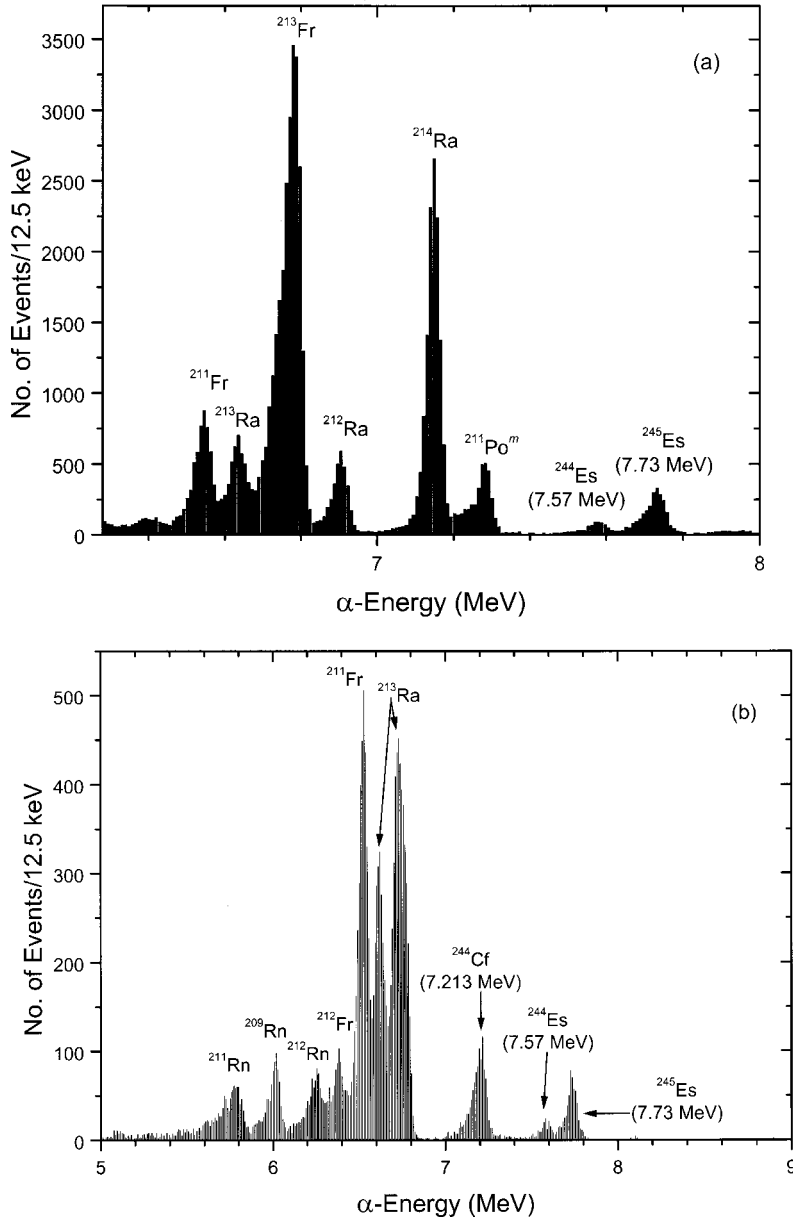


FIG. 3. Summed α spectra for the $^{237}\text{Np} + ^{12}\text{C}$ reaction at a beam energy of 81 MeV. (a) Spectrum from the first top detector recorded while the wheel was stepping for a total of 36 h. (b) Spectrum from the sixth top detector recorded while the wheel was stationary representing approximately 13 h.

and appears to follow the trend of TKE values measured in other ECDF systems.

Figure 2 shows the predominantly asymmetric pre-neutron-emission mass-yield distribution of fission fragments from ^{244}Cf . The mass-yield data were averaged over five mass numbers, but are expressed as yield (%) per mass number with the fragment yield normalized to 200%, and are derived based on conservation of momentum considerations from the ratio of the kinetic energies of both fragments for each coincident fission fragment pair. From the most probable fragment energies given above, it was determined that the mass (A) of the light fragment was 103 while the heavy fragment had a mass of $A = 141$.

According to the static fission model of Wilkins *et al.* [36], actinides with neutron number greater than 140 should have asymmetric mass splits until the heavy Fm region is reached. The heavy fragment in the split should remain nearly constant around either the $N = 82$ (spherical) or N

≈ 88 (deformed) neutron shell. If the heavy fragment is located near the spherical neutron shell, then the complementary fragment is forced to be highly deformed. In order to maintain the N/Z ratio of the fissioning nucleus, the heavy fragment ($A = 141$) in the fission of ^{244}Cf would be nearly spherical with $N = 82$, $Z = 59$, and $\beta = 0.2$ where β is the nuclear deformation parameter from Ref. [36]. Its complement would therefore be highly deformed with $N = 64$, $Z = 39$, and $\beta \approx 0.9$ [36]. A symmetric split would result in two fragments with $Z = 49$ and $N = 73$. The presence of the $Z = 50$ spherical proton shell might suggest a symmetric component in the fission of ^{244}Cf , but there are no corresponding neutron shells around $N = 73$, which means both fragments would have deformations greater than $\beta = 0.25$. This in turn removes the protons from the spherical shell, causing the fragments to become more deformed. A symmetric split would then consist of two deformed fragments, resulting in a lower TKE than in the case of one nearly spherical fragment

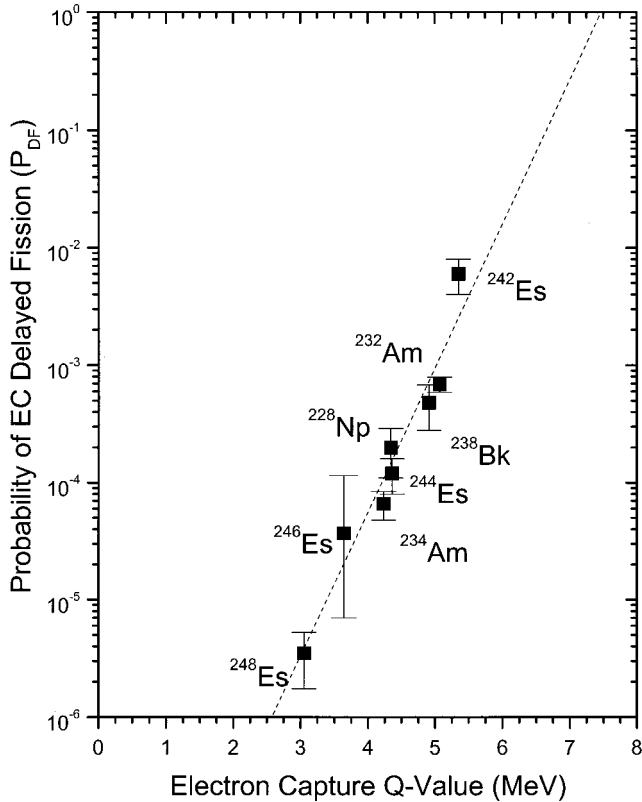


FIG. 4. Plot of the ECDF probability vs. electron-capture Q value for nuclides studied by our research group. The values for ^{232}Am and ^{234}Am are from Refs. [7,8], ^{228}Np is from Ref. [11], ^{238}Bk is from Ref. [19], ^{242}Es is from Ref. [16], and ^{246}Es and ^{248}Es are from Ref. [17].

and one highly deformed fragment. The mass-yield distribution in Fig. 2 shows no evidence of a symmetric component, indicating that the fission of ^{244}Cf prefers an asymmetric fragment configuration consisting of one nearly spherical fragment and one highly deformed fragment rather than two deformed fragments. However, more data are required to determine a limit on the amount of symmetric fission that occurs.

B. P_{DF}

Figure 3(a) shows the summed α spectrum from the top detector of the first detector pair taken from all of the MG wheels measured during a 36-h experiment. The interfering activities in the spectrum arise from the interaction of the ^{12}C beam with lead impurities in the ^{237}Np target. The peak observed at 7.580 MeV has been attributed to 37-s ^{244}Es based on the α energy of 7.57 MeV reported by Eskola *et al.* [22]. However, a 37-s component could not be identified in a MLDS analysis of the peak area over time. Instead, the half-life was 70 ± 1 s, about a factor of 2 larger than the reported 37-s half-life [22]. This α peak was also observed in a study of the α decay properties of light einsteinium isotopes by Hatsukawa *et al.* [37], but neither the half-life nor the identity of the α decay was given. This α decay cannot be due to the lowest energy α group of ^{245}Es (7.654 MeV, $I_\alpha = 3\%$ [30]) because the integrated number of counts in the 7.58-

MeV peak in Fig. 3(a) would have to be approximately 87 to contain 3% of the total number of ^{245}Es α decays while the peak at 7.58 MeV actually has 858 counts. Therefore, the most probable origin of the 7.58-MeV α peak is ^{244}Es based on the reported α -decay energy of 7.57 MeV [22]. The discrepancy in the two half-lives between the fission (or EC) decay and the α decay cannot be explained on the basis of the present data. Further research is required in order to determine the origin of this discrepancy and whether the identity of this α decay peak is actually ^{244}Es .

Due to this uncertainty in identifying the ^{244}Es α peak, we determined the P_{DF} of ^{244}Es by looking at the spectra recorded when the wheel was stationary to identify ^{244}Cf , the EC daughter of ^{244}Es , from which the amount of ^{244}Es present could be calculated. Figure 3(b) represents the summed spectrum of all measurements made in the top detector of the sixth detector pair while the wheel was stationary (approximately 13 h of counting.) The sample in detector pair six had the longest delay between collection and the start of counting (150 s), which allowed most of the shorter-lived interfering activities to decay before counting began. ^{244}Cf has a half-life of 19.4 min and α energies of 7.213 (75%) and 7.176 MeV (25%) with a 100% α decay branch [30]. By incorporating both α -particle energies in our analysis of the ^{244}Cf peak, the number of ^{244}Cf α particles detected is equal to the number of ^{244}Es EC decays (after applying a small correction for the 4% α branch in ^{244}Es [22]) in those collections counted while the wheel was stationary. We neglected the direct production of ^{244}Cf via the $^{237}\text{Np}(^{12}\text{C}, p4n)^{244}\text{Cf}$ reaction because of its low cross section. Based on information in Refs. [13] and [38] we concluded that the production of ^{244}Cf via the $p4n$ exit channel was less than 10% of the $5n$ exit channel, which is well within the standard deviation of our subsequent P_{DF} measurement.

From 20 single (noncoincident) fission events detected in 8607 total measurements and 380 ^{244}Cf α particles detected in 19 collections over the course of the experiment, a P_{DF} of $(1.2 \pm 0.4) \times 10^{-4}$ was determined using the equation given in Sec. I, where $N_{ECDF} = 2.32 \times 10^{-3}$ and $N_{EC} = 19.8$. Because the α particles and fission fragments were measured for the same samples, experimental uncertainties in N_{ECDF} and N_{EC} cancelled out in the calculation of the P_{DF} . Variations in beam intensity, target thickness, detection efficiency, and yield of the He gas-jet transport system were small from one collection to another and were much less than the standard deviation of our measurement. Therefore, only statistical uncertainties in the numbers of α particles and fission events were considered in the P_{DF} . Our value for the P_{DF} of ^{244}Es of $(1.2 \pm 0.4) \times 10^{-4}$ with a Q_{EC} of 4.36 MeV [18] for ^{244}Es fits the empirical relationship between P_{DF} and Q_{EC} shown in Fig. 4. Based on the number of ^{244}Es EC decays, a production cross section of $0.31 \pm 0.12 \mu\text{b}$ was calculated for the $^{237}\text{Np}(^{12}\text{C}, 5n)^{244}\text{Es}$ reaction at a beam energy of 81 MeV in the lab system (on target.) Experimental uncertainties, including the yield of the He gas-jet transport system ($60 \pm 20\%$), fluctuations in beam intensity (5%), nonuniformity of target thickness (7%), and detection efficiency (32

$\pm 3\%$), were all taken into account in the determination of this cross section and its standard deviation.

IV. CONCLUSIONS

ECDF was observed in ^{244}Es produced via the $^{237}\text{Np}(^{12}\text{C},5n)^{244}\text{Es}$ reaction using an 81-MeV ^{12}C beam (on target.) The fission properties were measured using our rotating wheel collection and detection system. The mass-yield distribution of fragments from the fission of ^{244}Cf was predominantly asymmetric as expected for low-energy fission in this region. Based on the deformation diagrams of Wilkins *et al.* [36], the heavy fragment in the fission of ^{244}Cf is probably nearly spherical, forcing the complementary fragment to be highly deformed.

The average pre-neutron-emission TKE of the fission fragments is 186 ± 19 MeV. As seen in Fig. 1, the TKE values measured for ECDF systems all appear to be lower than those reported for spontaneous fission isotopes. However, more precise measurements are needed to determine whether this is an actual phenomenon related to the delayed fission process.

A P_{DF} of $(1.2 \pm 0.4) \times 10^{-4}$ was calculated for ^{244}Es from the observed delayed fission events and the total number of ^{244}Es EC decays calculated from the α decay of its EC daughter ^{244}Cf . The line in Fig. 4 represents a nonlinear least-squares fit to the P_{DF} values that have been previously determined by our research group. It appears that the P_{DF} is directly dependent on the Q_{EC} . Based on theoretical considerations given in Refs. [6–9], the P_{DF} is dependent on both

the fission barrier height and the Q_{EC} . As the Q value increases or the fission barrier height decreases, the daughter nucleus is left in an excited state that is closer to the height of the fission barrier, resulting in a larger probability for undergoing ECDF. Fission barriers in this region do not vary greatly with neutron number [19]; therefore, the Q_{EC} values must have a stronger influence on the P_{DF} values in this region than the fission barriers since fission barrier heights are not changing enough to account for such a broad range of P_{DF} values. A larger Q_{EC} means that the daughter nucleus has a better chance to overcome its fission barrier, thereby increasing the probability that it will undergo fission. Since the P_{DF} is a measure of probability, it can never be greater than 1. Future experiments should be made to try to determine the shape of the P_{DF} function in Fig. 4 at higher Q values. By examining systems with larger Q values, it can be determined whether this function keeps increasing toward a value of one, or whether it levels off at some other maximum P_{DF} value.

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