

Electron-capture delayed fission properties of ^{242}Es

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Electron-capture delayed fission of ^{242}Es produced via the $^{233}\text{U}(^{14}\text{N},5n)^{242}\text{Es}$ reaction at 87 MeV (on target) was observed to decay with a half-life of 11 ± 3 s, consistent with the reported α -decay half-life of ^{242}Es of 16_{-4}^{+6} s. The mass-yield distribution of the fission fragments is highly asymmetric. The average pre-neutron emission total kinetic energy of the fragments was measured to be 183 ± 18 MeV. Based on the ratio of the measured number of fission events to the measured number of α decays from the electron-capture daughter ^{242}Cf (100% α branch), the probability of delayed fission was determined to be 0.006 ± 0.002 . This value for the delayed fission probability fits the experimental 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. This 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 study. Delayed fission (DF) is also thought to affect the production yields of heavy elements produced via multiple neutron captures followed by successive beta decays in the stellar r process and in nuclear weapons tests [1–5]. For a more complete description of the DF process, see Refs. [6–9], and references therein.

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

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

ECDF has been previously reported in the neutron deficient neptunium [10,11], americium [7,8,12,13], berkelium [10,13,14], and einsteinium [10,13,15] regions. This decay mode is expected to have measurable branches in nuclides that have electron-capture Q values (Q_{EC}) approaching the height of the fission barrier of the daughter nucleus. Q values meeting this criterion are found in neutron deficient actinides, which have odd-proton, odd-neutron nuclei. These nuclides show enhanced Q_{EC} values associated with EC de-

cah to their more stable even-even daughters. The Q_{EC} for ^{242}Es is 5.35 MeV [16], which approaches the estimated fission barrier heights of 5–7 MeV for this region [17]. ^{242}Es was chosen for this study of fission properties because its relatively large Q value is greater than in any other system where ECDF has been reported, and should have a relatively large P_{DF} . Previous experiments have shown that the P_{DF} increases with increasing Q_{EC} [9,11,14], and according to systematics, ^{242}Es could have a P_{DF} as large as 1%.

^{242}Es was first identified in 1984 by Hingmann *et al.* [15]. Five α particles, as well as three delayed fission events, were attributed to this unknown isotope produced via the $^{205}\text{Tl}(^{40}\text{Ar},3n)$ reaction. From these fission events, a P_{DF} of $(1.4 \pm 0.8) \times 10^{-2}$ was estimated for ^{242}Es . In 1994, Hofmann *et al.* [18] observed an unreported number of α particles from the decay of ^{242}Es as a decay product of ^{246}Md produced via the $^{209}\text{Bi}(^{40}\text{Ar},3n)$ reaction. They measured a half-life of 40_{-20}^{+40} s and an α decay energy of 7.910 MeV. In 1996 Ninov *et al.* [19] published different values for the half-life and α -decay energy. They reported a half-life of 16_{-4}^{+6} s and an α -decay energy of 7.920 ± 0.020 MeV. Neither the fission properties of ^{242}Cf , the EC daughter of ^{242}Es , nor the α to EC branching ratio of ^{242}Es were measured in these experiments.

II. EXPERIMENTAL TECHNIQUES

A. Targets and irradiation

A solution of ^{233}U was purified to remove lead contamination, and was then dissolved in 0.5 mL of isopropyl alcohol (IPA) to yield a solution that was approximately 0.5 mg/mL in ^{233}U and contained 4.2 ppm ^{232}U [11]. Successive target layers were electroplated on 0.5-mil (2.32 mg/cm²) Be from aliquots containing 25–50 μg of ^{233}U . The ^{233}U was converted to the oxide by baking each layer in a 500 °C oven for 20 min. The target area was 0.28 cm². The thickness of the ^{233}U target used during irradiations was 0.502 mg/cm². The target configuration has been described previously [20].

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A $3 \mu\text{A}^{14}\text{N}^{4+}$ beam (97 MeV) was provided by the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory. After passing through a 1.8 mg/cm^2 HAVAR vacuum window, N_2 cooling gas, and the Be target backing, the beam energy entering the ^{233}U target was 87 MeV (lab system). The reaction products were swept from the target chamber with a He/KCl aerosol gas jet, which transported the activities via a 1.4-mm i.d. capillary tube to our rotating wheel detection system [21] for α and fission measurements.

B. Measurements of fission and α activity

Fission fragments and α particles were measured in our merry-go-around (MG) rotating wheel system [21]. The activity-laden KCl aerosols were deposited via the He gas-jet transport system on 80 thin polypropylene foils ($40 \pm 10 \mu\text{g/cm}^2$ thick) positioned around the periphery of a 51-cm diameter rotating fiberglass wheel. There were 80 collection sites on each wheel, but only 79 were used at any given time. The transport efficiency of the gas jet was estimated to be $60 \pm 20\%$ based on previous experiments [22]. Three separate experiments were performed to study ECDF in ^{242}Es . During the first two experiments the MG wheel was stepped at 10-s intervals to position the foils between six pairs of passivated ion-implanted silicon (PIPS) detectors that were positioned directly above and below the wheel. These detectors measure the kinetic energy of α particles and fission fragments. The detection efficiency in any given detector was 32% for α particles and approximately 64% for fission fragments. After one hour of continuous collection and counting (four and a half wheel revolutions), the wheel was removed and replaced with a new, clean wheel and the process was repeated. This prevented the buildup of KCl on the foils, which would worsen the α resolution during the experiment, and also prevented the buildup of any longer-lived fission activities. During the third experiment the wheel was stepped at 20-s intervals for one hour (two and a quarter wheel revolutions), and then the wheel was stopped so that the last six collections were positioned between the detectors. These six collections were counted for an additional 40 min without moving the wheel. After that time, the wheel was replaced with a clean one and the process was repeated. Data were collected using the CHAOS data acquisition system [23]. Calibrations were performed before the experiment using a ^{212}Pb source, which provided 6.062-MeV and 8.784-MeV α particles, and a ^{252}Cf source was used for fission fragment energies. The energy resolution (FWHM) of the detectors above the wheel was approximately 0.040 MeV, while the detectors below the wheel had a resolution of approximately 0.1 MeV due to energy degradation of the α particles in the polypropylene collection foil. The fission background was less than one fission per detector per day based on background measurements taken prior to the experiment.

III. RESULTS AND DISCUSSION

A. Fission properties and half-life

A total of 48 pairs of coincident fission fragments were detected over the course of the three experiments. Figure 1

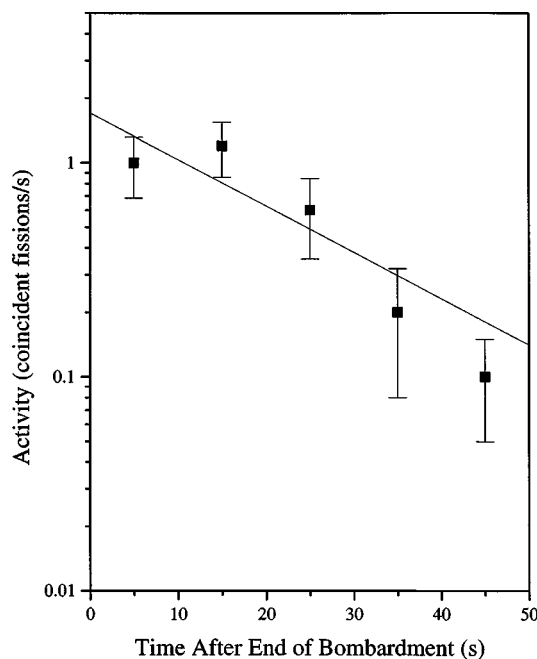


FIG. 1. Nonlinear least-squares fit to the fission activity vs time after bombardment for the decay of ^{242}Es as measured with the MG. These data were from the third ^{242}Es experiment, which used a 20-s stepping interval in the MG.

shows the decay curve for fission events from the third experiment, which used a 20-s stepping interval in the MG. This decay curve represents a majority of the fission events (approximately half) and therefore has better counting statistics than the decay curves from the other two experiments. Two components were evident in the decay curves from all three experiments, the shorter ^{242}Es component, and a long-lived constant activity (0.1 counts/s). A nonlinear least-squares two component fit to the decay curves of coincident fission events measured in each of the three experiments resulted in a half-life of 11 ± 3 s for ^{242}Es . Our value is lower than that reported by Ninov *et al.* [19] (16_{-4}^{+6} s), but within error the two are consistent. This half-life implies that the fission events we observed came from the ECDF of ^{242}Es . The fission process is very fast compared to the initial EC decay, which means that the fission events decay with the half-life of the EC parent. The only other nuclide in this region with a similar half-life is ^{243}Es , but based on the relationship between Q_{EC} and P_{DF} from Refs. [9,11,14], it should have a delayed fission probability of only about 10^{-5} , resulting in a fission rate much lower than we observed during these experiments.

Based on the fission fragment energy calibration from the spontaneous fission of ^{252}Cf , the mass-yield and kinetic energy distributions were determined for all of the coincident fission pairs. The fission-fragment energy calibrations were obtained using the method of Schmitt, Kiker, and Williams [24] using the constants of Weissenberger *et al.* [25]. The average neutron emission function, $\bar{\nu}(A)$, 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. [26]). Since fission events in the ECDF process are preceded by EC

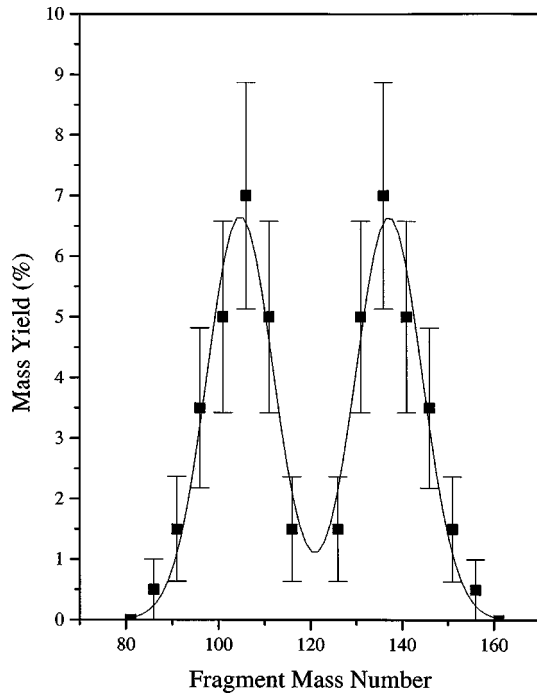


FIG. 2. Pre-neutron-emission mass-yield distribution for the ECDF of ^{242}Es . The fissioning species is ^{242}Cf . The data were averaged over 5 mass units.

decays, the fission properties measured during the experiment are for ^{242}Cf , the EC daughter. Figure 2 shows the mass-yield distribution for ^{242}Cf .

The total kinetic energy (TKE) distribution for coincident fission fragments from ^{242}Cf is shown in Fig. 3. The average

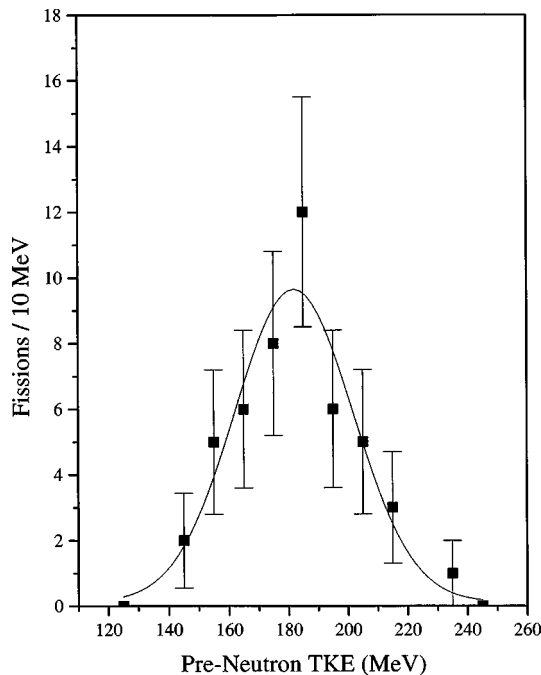


FIG. 3. Pre-neutron-emission total kinetic energy (TKE) distribution for the ECDF of ^{242}Es . The fissioning species is ^{242}Cf . The data are in groupings of 10 MeV.

TABLE I. Properties of the measured post-neutron-emission (post- n) and calculated initial pre-neutron-emission (pre- n) fragment kinetic energy and mass distributions for ^{242}Es ECDF and the ^{252}Cf standard measured in the same system. The fission properties are for ^{242}Cf , the EC daughter. Energies are given in MeV.

	^{242}Cf ECDF		^{252}Cf standard	
	pre- n	post- n	pre- n	post- n
Average TKE	183	182	180	177
σ	18	20	15	12
Most probable TKE	182	181	181	178
σ	20	18	14	13
FWHM ^a	47.0	42.3	32.9	30.6
Light fragment energy ^b	78.3	76.8	77.2	76.3
σ	9.6	10.3	9.7	9.8
Heavy fragment energy ^b	105	102	103	101
σ	12	12	8	8
Light fragment mass ^c	104		108	
Heavy fragment mass ^c	138		144	

^aFull width at half maximum, calculated from 2.35σ for Gaussian fit to the top half of the peak.

^bThese represent most probable values.

^cMasses calculated from most probable pre-neutron energies.

preneutron TKE is 183 ± 18 MeV. Table I gives a summary of the kinetic energy and mass properties for the ECDF of ^{242}Es , as well as for the ^{252}Cf standard. Since the ^{252}Cf calibration source was on the same kind of polypropylene foil that was used during the experiments, no correction is applied for energy degradation of fission fragments through the foil en route to the bottom detectors. There was also no correction needed to account for the $10 \mu\text{g}/\text{cm}^2$ [11] of KCl deposited on each foil because the amount of energy lost by fission fragments as they traveled through the KCl was only 0.2–0.4 MeV [27]. Figure 4 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.* [28] and Unik *et al.* [29], respectively. The value for ^{242}Cf follows the trend of TKE values seen in other ECDF systems.

According to the static fission model of Wilkins *et al.* [30] actinides with neutron number greater than 140 should have asymmetric mass splits until the Fm region is reached. The heavy fragment in the split should remain nearly constant around either the spherical neutron shell at $N=82$ or the deformed neutron shell at $N \approx 88$. If the heavy fragment is located at the spherical neutron shell, then the complement is forced to be highly deformed. In order to maintain the N/Z ratio of the fissioning nucleus, the heavy fragment in the ^{242}Cf system ($A=138$) would be nearly spherical with $N=82$ ($Z=56$, $\beta=0.2$), and its complement would therefore be highly deformed with $N=62$ ($Z=42$, $\beta \approx 0.85$ [30]). At first glance, it seems that the fission of ^{242}Cf might have a symmetric component, resulting in two fragments with $N=72$ and $Z=49$ because of the proximity to the $Z=50$ spherical proton shell. However, the deformation diagram in Ref. [30] shows that a neutron number of 72 is not close to

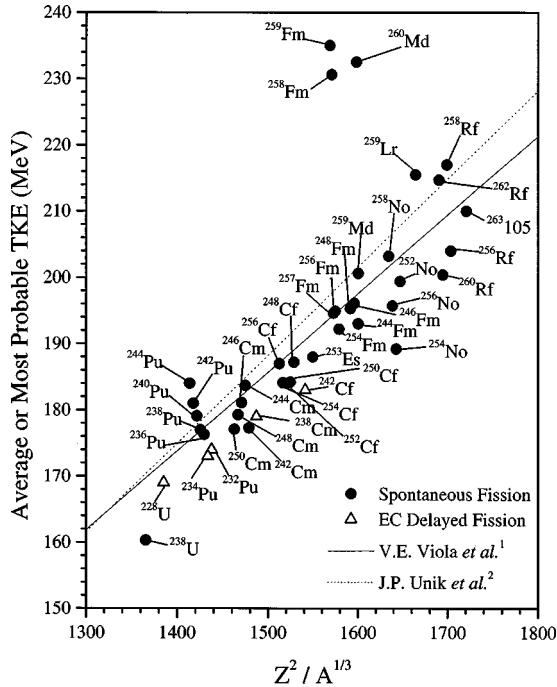


FIG. 4. The average or most probable TKE vs $Z^2/A^{1/3}$ for known cases of spontaneous or delayed fission. The solid line is the linear fit of Viola *et al.* [28] and the dashed line is from Unik *et al.* [29]. All of the TKE values have been corrected to be consistent with the calibration parameters of Weissenberger *et al.* [25].

any of the calculated neutron shells and prefers a deformation greater than 0.25. This in turn removes the protons from the spherical shell, causing the fragment to become more deformed. A symmetric split would therefore consist of two deformed fragments, resulting in an overall lower TKE than in the case of one nearly spherical fragment and one highly deformed fragment. This is consistent with the highly asymmetric mass-yield distribution seen in Fig. 2.

B. P_{DF}

In the α spectra recorded while the wheel was stepping, it was seen that ^{243}Es at 7.899 MeV (21 s) [27] interfered with the ^{242}Es peak, making it difficult to measure its α decay. We instead looked for ^{242}Cf , produced in the EC decay of ^{242}Es , during measurements when the wheel was not stepping. In these spectra, we measured ^{242}Cf α decay in the top detectors after most of the shorter-lived interfering activities had decayed. These interfering activities were produced by the interaction of the ^{14}N beam with lead impurities in the target, which formed decay chains of actinium, francium, radon, radium, astatine, and polonium isotopes. ^{242}Cf has a 3.5-min half-life and α energies of 7.385 and 7.351 MeV [27]. We neglected the direct production of ^{242}Cf via the $^{233}\text{U}(^{14}\text{N}, p4n)^{242}\text{Cf}$ reaction because of its low cross section. The SPIT code [32] predicts a production cross section for ^{242}Es of 32 nb using the $^{233}\text{U}(^{14}\text{N}, 5n)$ reaction at 87 MeV. Based on information in Refs. [13] and [33], we assumed that the cross section for production of ^{242}Cf via the $p4n$ exit channel was less than 10% of the $5n$ exit channel, which is well within the standard deviation of our P_{DF} measurement.

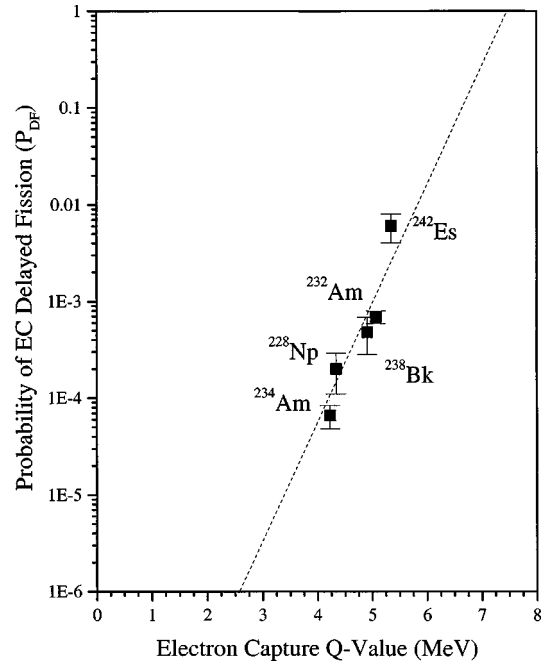


FIG. 5. Plot of the ECDF probability vs electron-capture Q value for nuclides studied by our research group. The values for ^{232}Am and ^{234}Am are from Refs. [7,8], ^{228}Np is from Ref. [11], and ^{238}Bk is from Ref. [19].

The number of ^{242}Cf α particles detected during the experiment was equivalent to the total number of ^{242}Es EC decays (after making a correction for the number of EC events that resulted in fission) since ^{242}Cf undergoes α decay 100% of the time [27]. The P_{DF} was determined from the third experiment because the collections were counted long enough to detect ^{242}Cf . A total of 36 fission events and 70 ^{242}Cf α particles were detected from all of the MG wheels during the third experiment (these values were later normalized to the number of samples collected). The P_{DF} was determined using the equation given in Sec. I where N_{ECDF} was equal to the number of fission events and N_{EC} was equivalent to the number of ^{242}Cf α particles plus the number of fission events, which gives the total number of ^{242}Es EC events. From these values, a P_{DF} of 0.006 ± 0.002 was obtained. Because the α particles and fission fragments were measured from the same samples, experimental uncertainties in N_{ECDF} and N_{EC} were equal, and canceled each other 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 ultimately less than the standard deviation of our measurement. Therefore, only statistical uncertainties in the numbers of α particles and fission fragments need to be considered in the determination of the P_{DF} . Based on the relationship between P_{DF} and Q_{EC} shown in Fig. 5, our value for the P_{DF} of ^{242}Es seems reasonable. ^{242}Es has the largest P_{DF} of any system where ECDF has been studied, which is expected based on its large Q_{EC} of 5.35 MeV.

The EC to α -branching ratio of ^{242}Es could not be determined from the experiments because of the interfering ^{243}Es peaks. Using ^{242}Cf α particles as an indication of the number

of ^{242}Es EC decays and assuming a 100% EC branch in ^{242}Es , the lower limit of the production cross section for the $^{233}\text{U}(^{14}\text{N},5n)^{242}\text{Es}$ reaction was measured to be 22 nb at a beam energy of 87 MeV. Various experimental uncertainties were taken into account when determining this limit, including the yield of the gas-jet system, fluctuations in beam intensity, nonuniformity of target thickness, and detection efficiency. In a future experiment we plan to experimentally measure the EC branch of ^{242}Es by comparing the number of ^{242}Es α decays to the number of EC daughters produced during the experiment. A different production reaction, which may have a lower production rate but produces fewer interfering activities, would have to be used for this experiment. Knowing the EC branch will make it possible to determine the actual production cross section for the reaction used in these experiments.

IV. CONCLUSIONS

ECDF was studied in ^{242}Es produced via the $^{233}\text{U}(^{14}\text{N},5n)^{242}\text{Es}$ reaction using 87 MeV (on target) ^{14}N . The fission properties and half-life were measured using our rotating wheel detection system. The half-life was measured to be 11 ± 3 s and was based on more events than previously reported values. The mass-yield distribution for ECDF of ^{242}Es was predominantly asymmetric as expected for low-energy fission in this region. Based on the deformation diagrams of Wilkins *et al.* [30], the heavy fragment in the fission of ^{242}Cf is most likely nearly spherical, forcing the complementary light fragment to be highly deformed.

The average pre neutron emission TKE of the fission fragments was 183 ± 18 MeV. As shown in Fig. 4, the TKE values measured for ECDF systems are all lower than those reported for spontaneous fission isotopes. This may be due to the delayed fission process, which can impart excitation energy to the fissioning species up to the entire Q value [31]. Excitation energy tends to wash out shell effects in the fragments [34], which are normally very strong in the case of spontaneous fission. Therefore, the influence of the shell ef-

fects is weaker, which in the case of ^{242}Cf causes the heavy fragment to be more deformed than it normally would be in spontaneous fission. Increased deformation of the heavy fragment would result in an overall lower TKE than if the fragments were more spherical. The TKE values of Viola [28] and Unik [29] in Fig. 4 are greater than the experimentally determined ECDF values. The fact that the fits suggest larger TKE values than those measured in ECDF systems further implies that the lower ECDF values are due to weaker shell effects in the fission fragments.

A P_{DF} of 0.006 ± 0.002 was calculated from the delayed fission events and the α decay of ^{242}Cf . The line in Fig. 5 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} . As the Q value increases, the daughter nucleus is left in an excited state that is closer to the height of the fission barrier. Fission barrier heights in this region do not vary greatly with neutron number [17]. Therefore, the P_{DF} must have a strong dependence on the Q_{EC} since the fission barrier heights are not varying greatly. 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 one. Future experiments will try to determine the shape of the P_{DF} function in Fig. 5 at higher Q values. By looking at systems with even larger Q values, we will be able to determine whether this function keeps increasing toward a value of one, or whether it levels off to some maximum P_{DF} value.

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