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Charge distribution in the thermal neutron fission of ^{245}Cm: Fractional cumulative yields of ^{135}I and ^{140}Ba

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Fractional cumulative yields for ^{135}I and ^{140}Ba have been determined in the thermal neutron induced fission of ^{245}Cm following the respective daughter product activities. The fractional cumulative yield values obtained are 0.951 ± 0.014 and 0.969 ± 0.006 for ^{135}I and ^{140}Ba, respectively. The fractional cumulative yield value for ^{135}I has been compared with the one reported earlier and an attempt has been made to correlate the value to the neutron to proton ratio of the fissioning nucleus. The data have been analyzed to examine the effect of shells on the width of the charge distribution. This indicates a narrower width of charge distribution for mass chain 135 and a broader width for mass chain 140 compared to normal width of the distribution.

[NUCLEAR REACTIONS, FISSION Thermal neutron fission of ^{245}Cm, measured FCY, data analyzed in terms of neutron to proton ratio, odd-even effect and shell effect.]

INTRODUCTION

Charge distribution studies in the low energy fission of actinide elements have indicated the effect of the 82 neutron shell on the width parameter (σ) of the distribution. Recent studies by Tokay *et al.*[1] have revealed the isotopic mass dispersion discontinuities in various fission processes which have been attributed to the neutron shell effect. In ^{235}U(n_{th},f), several authors[2-6] have pointed out that the width of the distribution in the mass region 132–136 is smaller than the normally observed σ value (0.56 ± 0.06) with minimum around mass 136. In ^{252}Cf(sf), a low σ was indicated for the 135 mass chain, as reported earlier from this laboratory.[7] It was therefore considered interesting to study the influence of shells in the mass region 130–140 in different fissioning systems of comparable excitation energies.

In ^{245}Cm(n_{th},f) the fractional independent yield (FIY) of ^{135}Xe has been determined by Harbour and Troutner[8] using direct gamma counting. However, the width of the charge distribution for the mass chain 135 in ^{245}Cm(n_{th},f), as one would expect from the reported[8] ^{135}I fractional cumulative yield (FCY), is higher in contrast to the low value observed for the same mass chain in other fissioning systems. Further, an analysis of the ^{135}I FCY values in terms of neutron to proton ratio in different fissioning systems indicates that the FCY value for ^{135}I as reported in Ref. 8 is low. Therefore, a redetermination of the FCY of ^{135}I in ^{245}Cm(n_{th},f) was considered worthwhile. In addition, the FCY value for ^{140}Ba has also been determined for the first time in ^{245}Cm(n_{th},f).

EXPERIMENTAL

Curium-244 obtained from the Oak Ridge National Laboratory was analyzed mass spectrometrically in this laboratory and the isotopic composition (atom %) was found to be as follows: ^{244}Cm—93.86%, ^{245}Cm—2.01%, ^{246}Cm—4.13%. As the fission cross section of ^{245}Cm is about 1660 times that of ^{244}Cm, more than 98% of the fissions were due to ^{245}Cm. Electrodeposited target of ^{244}Cm(~ 80 μg) was prepared from isopropyl alcohol medium on gold plated aluminum foil.[9] The target was covered with a 0.0025 cm thick superpure aluminum foil and was irradiated in the swimming pool type reactor "Apsara" in a flux of $\sim 1.2 \times 10^{12}$ n/sec cm^2. The length of the irradiation was varied from 1 to 6 h depending on the nuclide of interest. After each irradiation, the catcher foil was mounted on a 0.06 cm thick aluminium plate and was counted in a fixed geometry on a 60 cc Ge(Li) detector coupled to a 4096 channel analyzer. The resolution of the detector was 1.5 keV at 122 keV and 3.9 keV at 1332 keV. The direct gamma counting of the 249.6 keV and 1596 keV gamma rays of ^{135}Xe^{g} and ^{140}La, respectively, was carried out for optimum time at suitable intervals. To avoid pile up the counting rate was maintained low enough to keep the dead time below 5%. The linear compton subtraction was done visually. In order to confirm the methodology used in the determination of ^{140}Ba FCY in the present work, the same was also redetermined in ^{233}U(n_{th},f). The obtained ^{140}Ba FCY value in ^{233}U(n_{th},f) was in agreement with the earlier reported value.[10]

CALCULATIONS

The isobaric mass chains 135 and 140 are as follows:

15.5% $^{135}Xe^m$(15.3 min)

$^{135}Te \xrightarrow{\beta^-} {}^{135}I \xrightarrow{\beta^-} {}^{135}Xe^g \xrightarrow{\beta^-} {}^{135}Cs$

18 s 6.59 h 84.5% 9.17 h 2×10^6 y

$^{140}Cs \xrightarrow{\beta^-} {}^{140}Ba \xrightarrow{\beta^-} {}^{140}La \xrightarrow{\beta^-} {}^{140}Ce$

64 s 12.8 h 40.2 h stable.

The half-lives of the isotopes have been taken from Ref 11. The branching ratio of ^{135}I to $^{135}Xe^m$ is 15.5%.[12] Since the half-lives of the precursors of ^{135}I and ^{140}Ba are short compared to the irradiation times as well as the intervals between the end of irradiation and the time of the first counting, the isobaric chains could be assumed to consist of ^{135}I, $^{135}Xe^m$ and $^{135}Xe^g$ for mass chain 135 and ^{140}Ba and ^{140}La for mass chain 140. In the case of ^{135}I, the decay to $^{135}Xe^g$ is partly through $^{135}Xe^m$, hence the growth and decay of $^{135}Xe^m$ and $^{135}Xe^g$ during the irradiation and in the time interval up to the mean counting time depend on their branching ratios, relative independent yields, and their half-lives. The ratio of the independent yields of metastable and ground states for ^{135}Xe has been assumed as 1.0 in the absence of any data. Since 84.5% of ^{135}I decays to $^{135}Xe^g$ and the half-life of $^{135}Xe^m$ is short compared to the time of irradiation and the mean counting time, the error in FCY of ^{135}I due to the above assumption is negligible. The growth of the activity of the daughter product $^{135}Xe^g$ (249.6 keV) was used to determine the ^{135}I FCY value taking into account the contribution of $^{135}Xe^m$ to $^{135}Xe^g$. Detailed calculations are given elsewhere.[7] For the mass chain 140, consisting of only ^{140}Ba and ^{140}La, the peak area A of 1596 keV gamma ray of ^{140}La was related to the initial number of atoms of ^{140}Ba (N^0_{Ba}) and that of $^{140}La(N^0_{La})$ independently formed at the end of the irradiation by Eq. (1):

$$Y = K\,X\,N^0_{Ba} + K\,N^0_{La}\,, \tag{1}$$

where

$$Y = \frac{A\,t'}{t''}\,\frac{\lambda_2 e^{\lambda_2 T}}{(1-e^{-\lambda_2 t})(1-e^{-\lambda_2 t''})} \tag{2}$$

and

$$X = 1 - \frac{\lambda_2}{\lambda_2-\lambda_1}\left[1 - \frac{\lambda_2}{\lambda_1}\left(\frac{1-e^{-\lambda_1 t}}{1-e^{-\lambda_2 t}}\right) \times\left(\frac{1-e^{-\lambda_1 t''}}{1-e^{-\lambda_2 t''}}\right)e(\lambda_2-\lambda_1)T\right], \tag{3}$$

where K is the net counting efficiency for 1596 keV gamma ray, λ_1 and λ_2 are the decay constants of ^{140}Ba and ^{140}La, respectively, t is the time of irradiation, and T is the interval between the end of the irradiation and the starting time of counting. t' is the clock time while t'' is the live time of counting. Measurements with varied t were carried out and the calculated values of X and Y were fitted into a straight line using least-squares analysis (as shown for ^{135}I in Fig. 1).

The FCY of ^{140}Ba is given by Eq. (4):

$$\mathrm{FCY}\ (^{140}\mathrm{Ba}) = \frac{N^0_{Ba}}{N^0_{Ba}+N^0_{La}}. \tag{4}$$

According to Eq. (1), it is given by

$$\mathrm{FCY\ of\ }^{140}\mathrm{Ba} = \frac{\mathrm{Slope}(N^0_{Ba})}{\mathrm{Slope}(N^0_{Ba}) + \mathrm{Intercept}(N^0_{La})}. \tag{5}$$

RESULTS

Table I shows the data on the measurements of the FCY of ^{135}I and ^{140}Ba. According to Wahl *et al.*,[2] the charge dispersion in an isobaric mass chain follows the Gaussian distribution as given by

$$Y_Z^{\mathrm{cum}} = (2\pi)^{-1/2}\sigma^{-1}\int_{-\infty}^{Z+0.5} \exp[-(Z-Z_p)^2/2\sigma^2]\,dZ \tag{6}$$

where Y_Z^{cum} is the cumulative yield up to $Z+0.5$, Z_p is the most probable charge for the mass chain, and σ is the width parameter of the distribution. The normal value of σ was observed to be 0.56 $\pm$0.06 by the same author.[2] The probability plot of FCY vs $(Z-Z_p)$ for different mass chains for a given fissioning nucleus or, for a given mass chain in different fissioning nuclei should be a straight line for a defined σ value. Figure 2 shows such a plot consisting of two straight lines drawn for $\sigma = 0.50$ and 0.62 as limits of the normal value of $\sigma = 0.56 \pm 0.06$. The FCY values for ^{135}I in different fissioning nuclei and in $^{245}Cm(n_{th},f)$ as ob-

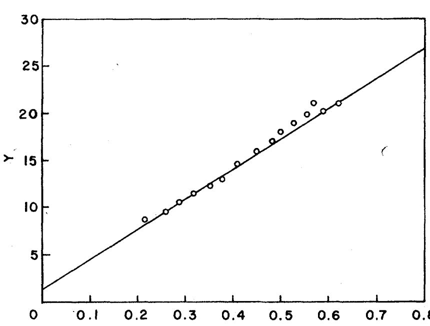

FIG. 1. Plot of Y vs X for ^{135}I–$^{135}Xe^g$ system.

TABLE I. Fractional cumulative yields of ^{135}I and ^{140}Ba in the thermal neutron fission of ^{245}Cm.

	^{135}I			^{140}Ba		
Ser. no.	Slope	Intercept	FCY	Slope	Intercept	FCY
1	31.115	1.722	0.947	1 536 580	38 888	0.975
2	6.825	0.310	0.956	2 184 400	82 979	0.963
3	31.757	1.250	0.962	261 663	10 025	0.963
4	35.410	2.695	0.930	318 367	7833	0.975
5	59.356	2.232	0.963	436 373	13 190	0.970
Average FCY value		0.951 ± 0.014			0.969 ± 0.006	

tained in the present work and the one reported by Harbour and Troutner[8] are plotted in the same figure where Z_p values for the 135 mass chain in different fissioning systems are obtained using the empirical relation as given by Wahl *et al.*,[2]

$$Z_p - Z_{UCD} = \mp(0.45 \pm 0.10) \qquad (7)$$

for heavy or light fragments, as the case may be.

Z_{UCD} is the most probable charge for the fission product mass chain calculated using the unchanged charge density (UCD) hypothesis. In $^{245}Cm(n_{th}, f)$ system the Z_p value for the mass chain 135 ($A' = 136.4$), as obtained using Eq. (7), is 52.78. Using this Z_p value in the plot of Fig. 2, the observed FCY (0.951) indicates $\sigma \sim 0.45$. In a similar fashion a Z_p value (54.88) for the mass chain 140 was obtained using $A' = 141.80$ as calculated based on Ref. 8. The Z_p for the 140 mass chain was also calculated following the prescription of Nethaway[13] (54.84) and Coryell (54.51).[14] The ^{140}Ba FCY value (0.969) for the average Z_p value (54.75) shows a σ value ~0.85.

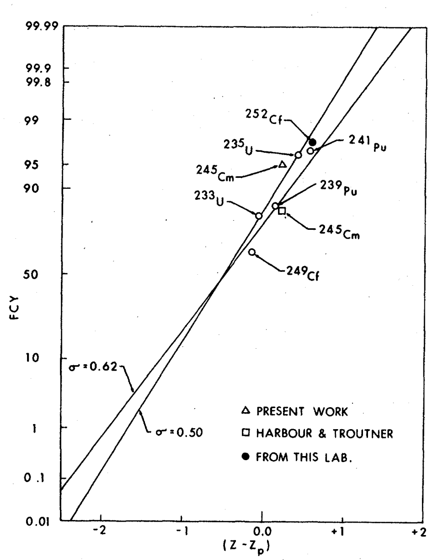

FIG. 2. Plot of ^{135}I FCY vs $(Z - Z_p)$ in different fissioning nuclei.

DISCUSSION

The value of FCY for ^{135}I obtained in this work is 0.951 ± 0.014 which is different from the value (0.83 ± 0.02) reported by Harbour and Troutner[8] for $^{245}Cm(n_{th}, f)$. The value of Z_p obtained using Eq. (7) is 52.78. In an attempt to examine the reason for the above difference, a comparison of the reported FCY values of ^{135}I in different fissioning nuclei of comparable excitation energies is made as shown in Table II. The values are plotted in Fig. 3. It is seen that the FCY of ^{135}I increases with the increase of neutron to proton ratio (N/P) of the fissioning nucleus. This trend is understandable since with increasing N/P ratio of the fissioning nucleus, the Z_{UCD} value and hence the corresponding Z_p value decreases for a given mass chain. Thus, for ^{135}I, the FCY increases with decreasing Z_p value [or with increasing $(Z - Z_p)$ value] for a given σ value as apparent from Eq. (6). However, the calculated Z_p values are dependent on the adequacy of Eq. (7) for different fissioning nuclei as well as on the knowledge of the prompt neutron number for calculation of the A' values for ^{135}I. Eventually, the close correspondence of the calculated Z_p values using Coryell's[14] method with those obtained using Eq. (7) (as shown in Table II) gives credence to the use of the latter. Hence for

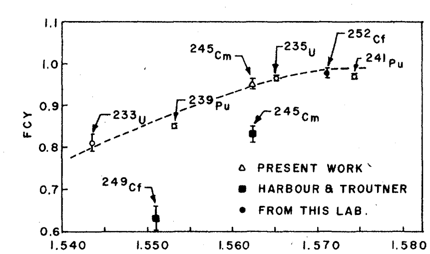

FIG. 3. Plot of ^{135}I FCY vs N/P ratio of different fissioning nuclei.

TABLE II. Z_p values for the 135 mass chain in various fissioning systems.

Fissioning system	A'	N/P	Z_p Coryell	Z_p	Z_p $\sigma = 0.56 \pm 0.06$	FCY
^{233}U	136.1	1.5430	52.99	53.06	53.00 ± 0.04	0.81 [c]
^{235}U	136.0	1.5652	52.53	52.57	52.48 ± 0.01	0.965 [c]
^{239}Pu	136.1	1.5532	52.79	52.86	52.93 ± 0.05	0.849 [c]
^{241}Pu	136.1	1.5745	52.37	52.42	52.47 ± 0.02	0.968 [c]
^{245}Cm	136.4	1.5625	52.70	52.78	52.57 ± 0.08	0.951 [a]
					52.97 ± 0.02	0.83 [c]
^{249}Cf	136.7	1.5510	53.01	53.13	52.25 ± 0.02	0.63 [c]
^{252}Cf	136.2	1.5714	52.21	52.40	52.40 ± 0.12	0.975 [b]

[a] From the present work.
[b] From Ref. 7.
[c] From Ref. 8.

^{135}I, FCY increases with the increase in N/P ratio of the fissioning nucleus due to decrease in the Z_p value. Figure 3 shows that the value obtained in the present work fits well with the expected trend (as shown by the dotted guideline in Fig. 3). The reported value of Harbour and Troutner[8] is appreciably lower than what one would expect from the trend. The value of FCY for ^{135}I obtained in this work gives a value of ~0.45 for σ which is lower than Wahl's normal value of $\sigma = 0.56 \pm 0.06$. It has been pointed out by several authors that the deviation in the σ value from the normally expected value (0.56 ± 0.06) could be due to a shell effect[15,16] and/or an odd-even effect.[17–21] In the ^{235}U(n_{th}, f) system, a minimum in the σ value exists around the mass region 132–136,[2–6] which was attributed to the 82-neutron shell effect by Wunderlich.[5] The observed low σ value in the mass chain 135 in ^{245}Cm(n_{th}, f) could therefore be attributed to the shell effect analogous to that in ^{235}U(n_{th}, f). However, the probable effect of nucleon pairing (odd-even effect) in modulating the σ value could not be ruled out as shown by several authors.[17–21] In fact, it was indicated by Clerc *et al.*[20] that the width parameter σ is an oscillating function of the fission product mass in the lighter mass region in the ^{235}U(n_{th}, f) system. The oscillatory trend of σ was attributed to the odd-even effect in general and also to the magic shell effect for nuclides at the proximity of magic shells.

The possible influence of nucleon pairing on the σ value of the fission product mass chain 135 was evaluated following the prescription of Amiel *et al.*[17,18] pertaining to a heavy mass chain as required in the present work. The normalized fractional independent yields of the isobaric nuclides of the fragment chain 136 (which leads to product the mass chain 135 following neutron evaporation) were calculated for various σ values with $Z_p = 52.78$. These yield values were corrected for the odd-even effect according to Eq. (8):

$$Y(A, Z) = Y^{(A,Z)}_{\text{normal}}(1 + 0.22\delta_P + 0.08\delta_N)\,, \qquad (8)$$

where $Y^{(A,Z)}_{\text{normal}}$ is the initial normalized fractional independent yield for fragment (A, Z), δ_P or δ_N is +1 or −1 for even or odd number of protons or neutrons as the case may be. The sum of the corrected fractional independent yields was normalized to unity. It was observed that for $\sigma \sim 0.50$, the odd-even effect corrected normalized FCY value for ^{135}I compared well with the determined FCY value. The low value of σ (~0.45) for the 135 mass chain could therefore be attributed to the odd-even effect as well as the shell effect.

In the 140 mass chain, the determined FCY value for ^{140}Ba (0.969 ± 0.006) indicates a high σ value ~0.85 for the calculated average Z_p value 54.75. Large σ values for the 140 mass chain were also observed in ^{233,235}U and ^{249}Cf.[10,22] The odd-even effect in the fragment mass chain 142 ($A' = 141.8$ for the 140 mass chain) was evaluated in a similar fashion as in the case of ^{135}I for various σ values with Z_p value 54.75. It was seen that the corrected normalized FCY of ^{140}Ba for $\sigma \sim 0.75$ agreed well with the determined FCY value. Therefore the high σ value for the 140 mass chain could be due to the odd-even effect and absence of the shell effect.

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