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Fragment shell effect in low energy fission: Independent yields of technetium isotopes in the thermal-neutron-induced fission of ^{239}Pu

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The independent yields of ^{101}Tc, ^{103}Tc, ^{104}Tc, and ^{105}Tc have been determined in the thermal-neutron-induced fission of ^{239}Pu using fast radiochemical separation techniques followed by gamma-ray spectrometry. The most probable fragment mass A'_p and mass dispersion σ'_A are obtained from the measured independent yields as 108.35 ± 0.45 and 1.80 ± 0.25 mass units, respectively. A comparison of the elemental yield (Y_z) of technetium obtained from the present work and the literature for the thermal-neutron-induced fission of ^{235}U and ^{239}Pu and the spontaneous fission of ^{252}Cf shows that the Y_z of technetium in the case of fission of ^{240}Pu and ^{252}Cf is very high compared to that of ^{236}U. The enhancement of the yields in the case of ^{240}Pu and ^{252}Cf is attributed to the proximity of spherical-deformed neutron shells 82—88 in the complementary fragments antimony and cesium, respectively.

I. INTRODUCTION

The studies of nuclear charge distribution are ideally suited to examine the influence of nuclear structure such as nucleon pairing and spherical-deformed nuclear shells on the fission process. While the investigations of the effects of nucleon pairing[1-8] are quite extensive the influence of nuclear shells on charge distribution has so far been limited. In recent years a few studies have been reported in the literature[8-11] where the influence of the closed 50-proton shell has been discussed, but the role played by neutron shells is not considered in these studies. The influence of neutron shells was observed by Unik *et al.*[12] in a study of mass-yield systematics in the low energy fission of actinides. These authors observed that there is a strong preference for splits containing 52—58 protons for the heavy fragment. However, since proton numbers 52—58 do not correspond to any closed shell configuration it was argued that neutron numbers (80—88) associated with these proton configurations may have a strong influence on yield systematics. Wilkins *et al.*,[13] in their scission point model, which is based on the assumption of statistical equilibrium among the collective degrees of freedom at the scission point, have clearly shown that the stability brought by the spherical-deformed neutron shell is stronger than that associated with the 50-proton shell. Vine and Wahl,[10] using a fast radiochemical separation method, determined the fractional independent yields (FIY) of ^{104}Tc and ^{105}Tc isotopes in the thermal-neutron-induced fission of ^{235}U and ^{239}Pu, and attributed the enhanced yields of technetium isotopes in the case of ^{239}Pu to the proximity of the 50-proton shell configuration in the complementary fragment. However, it was not possible to arrive at the values of A'_p and σ'_A from only two FIY values. Fassbender *et al.*[9] measured the ratio of fractional independent to fractional cumulative yields (FCY) of ^{104}Tc to ^{108}Tc in the thermal-neutron-induced fission of ^{235}U and ^{239}Pu. Since these values are relative, and also since the FIY values of the earlier members of the isotopic chains were not determined, these data could not be used to obtain the values of A'_p and σ'_A. In order to evaluate the relative importance of neutron-proton shells it is essential to obtain the values of A'_p and the associated most probable values of neutron (N'_p) numbers. With this in view the fractional independent yields of ^{101}Tc, ^{103}Tc, ^{104}Tc, and ^{105}Tc in the thermal-neutron-induced fission of ^{235}U and ^{239}Pu were determined using fast radiochemical separation methods followed by gamma spectrometry. The fractional independent yields of technetium isotopes are less than 0.01 in the thermal-neutron-induced fission of ^{235}U and hence are not mentioned in the text. Only the FIY's of the Tc isotopes in the thermal-neutron-fission of ^{239}Pu are reported. The yields of ^{101}Tc and ^{103}Tc have been measured for the first time. This permitted the evaluation of A'_p and N'_p so that the effects of a spherical-deformed neutron shell could be examined in the present work.

II. EXPERIMENTAL

The targets containing $\sim 12\ \mu$g of plutonium (94.1% of ^{239}Pu) in solution form were sealed in polypropylene tubes and irradiated in a neutron flux of 1×10^{13} n/sec/cm^2 using the pneumatic carrier facility of the CIRUS reactor. The time of irradiation was varied from 10 to 30 sec depending on the half-lives of the fission products of interest and their precursors. After irradiation, the technetium was separated from the rest of the fission products as tetraphenyl arsonium pertechnate by exchange on a preformed precipitate of tetraphenyl arsonium chlorate.[14] The radioactivity of the separated samples was followed on a precalibrated 45-cm^3 intrinsic Ge detector coupled to a 4 K analyzer. The sample-detector distance was chosen in such a way as to keep the dead time losses at a minimum (10%). Technetium was separated at various cooling times ranging from 40 to 100 sec. The peak

TABLE I. Nuclear data used in the present work.

Nuclide	Half-life	Gamma ray (keV)	Abundance (%)
^{101}Tc	14.2 min	306.8	88
^{103}Tc	54.2 sec	346.4	16.2
^{104}Tc	18.4 min	358	89
^{105}Tc	7.7 min	159.3	7.0
^{105}Ru	4.44 h	724.2	46.7
^{99}Mo	66 h	739.4	12.1
^{99}Tc^{m} [a]	6.01 h	140.5	87.7
^{106}Ru [a]	371.6 d	511.9	86.4

[a] Used as tracer for chemical yield determination.

area of each technetium gamma ray was obtained by linear subtraction of Compton background. The chemical yields for separated samples were obtained using ^{99}Tc^{m} as a tracer. To obtain the FCY of ^{105}Tc, ruthenium was separated as ruthenium tetroxide from the rest of the fission products using the cold distillation procedure.[14]

The fission rate for each irradiation was obtained from the activity of the fission monitors ^{92}Sr, ^{97}Zr, and ^{139}Ba. Table I shows the half-lives, gamma-ray energies, and abundances[15] of the nuclides used in the present work. The activity of the nuclide of interest was obtained from the measured activity after making the corrections for dead time losses, chemical yield, and the total number of fissions for each cooling time.

III. CALCULATIONS

The isobaric decay chains involved in the present work are the following:

$$\underset{14.6\text{ min}}{^{101}\text{Mo}} \xrightarrow{\beta^-} \underset{14\text{ min}}{^{101}\text{Tc}} \xrightarrow{\beta^-} \underset{\text{stable}}{^{101}\text{Ru}} ,$$

$$\underset{62\text{ sec}}{^{103}\text{Mo}} \xrightarrow{\beta^-} \underset{54\text{ sec}}{^{103}\text{Tc}} \xrightarrow{\beta^-} \underset{39.35\text{ d}}{^{103}\text{Ru}} \xrightarrow{\beta^-} \underset{\text{stable}}{^{103}\text{Rh}} ,$$

$$\underset{1.1\text{ min}}{^{104}\text{Mo}} \xrightarrow{\beta^-} \underset{18\text{ min}}{^{104}\text{Tc}} \xrightarrow{\beta^-} \underset{\text{stable}}{^{104}\text{Ru}} ,$$

$$\underset{40\text{ sec}}{^{105}\text{Mo}} \xrightarrow{\beta^-} \underset{7.6\text{ min}}{^{105}\text{Tc}} \xrightarrow{\beta^-} \underset{4.4\text{ h}}{^{105}\text{Ru}} \xrightarrow{\beta^-} \underset{36.5\text{ d}}{^{105}\text{Rh}} \xrightarrow{\beta^-} \underset{\text{stable}}{^{105}\text{Pd}} .$$

Since the last members of the mass chains 101, 103, and 104 are either long lived or stable, their independent formations were assumed to be negligible and therefore the fractional independent yields (FIY) of ^{101}Tc, ^{103}Tc, and ^{104}Tc were obtained as (1-FCY), where FCY refers to the fractional cumulative yield of precursor molybdenum. In the case of ^{105}Tc, the FIY was obtained from the difference between the FCY's of ^{105}Tc and ^{105}Mo with the assumption that the chain terminates at ^{105}Rh. The activity of the daughter nuclide measured experimentally was corrected for dead time losses as follows:

$$A=\frac{A_0 \Delta T e^{\lambda_D T}\lambda_D}{\text{LT}(1-e^{-\lambda_D \Delta T})} , \tag{1}$$

where A_0 is the peak area and ΔT and LT are the clock and live time, respectively. The daughter's activity (A) at the time of separation (T_s) is related to the initial number of daughter atoms N_D (formed independently) and parent atoms N_p by the following relation:[14]

$$Y=EXN_p+EN_D , \tag{2}$$

where

$$Y=A/(1-e^{-\lambda_D t})e^{-\lambda_D T} , \tag{3}$$

$$X=1+\frac{\lambda_D[(e^{-\lambda_D t}-e^{-\lambda_p t})e^{-\lambda_D T}+(1-e^{-\lambda_p t})(e^{-\lambda_p T}-e^{-\lambda_D T})]}{(\lambda_D-\lambda_p)(1-e^{-\lambda_D t})e^{-\lambda_D T}} , \tag{4}$$

where E is the net counting efficiency of the gamma ray, λ_p and λ_D are the decay constants of the parent and the daughter, respectively, T is the cooling time, t is the time of irradiation, and A is the peak area corrected for the chemical yield and fission rate. From the value of activity (A) for different cooling times T, X and Y were obtained and fitted to a straight line using a least squares analysis. Figure 1 shows a typical plot of X and Y for ^{104}Tc for six different cooling times ranging from 40 to 100 sec. Similar plots were made for other technetium

TABLE II. Fractional independent yield of Tc isotopes in the thermal-neutron-induced fission of ^{239}Pu.

Mass	$Y_{\text{FI}}(Z)$	Y_A	$Y_I(A,Z)$	Ref.
101	0.0023±0.0015	5.95±0.08	0.014±0.009	This work
102			0.050±0.025	Interpolated
103	0.0346±0.015	6.95±0.14	0.241±0.097	This work
104	0.090 ±0.014	5.96±0.12	0.536±0.083	This work
	0.077 ±0.012			10
105	0.195 ±0.045	5.57±0.11	1.086±0.251	This work
	0.264 ±0.021			10

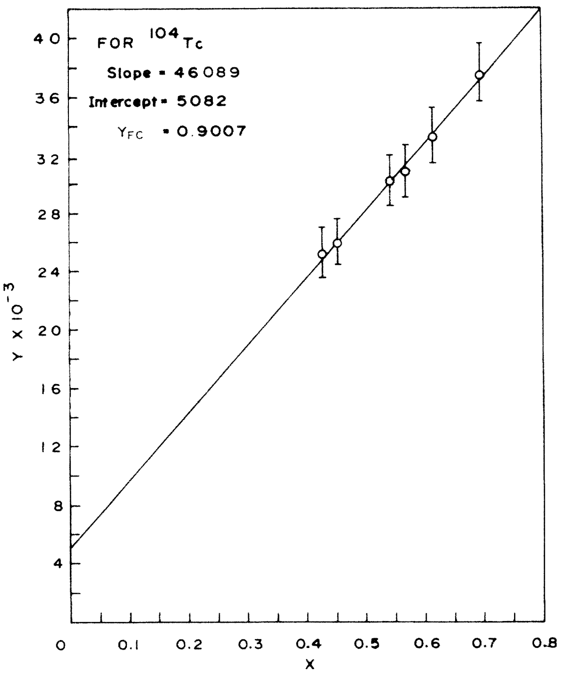

FIG. 1. Plot of X-Y for ^{104}Tc in the thermal-neutron-induced fission of ^{239}Pu.

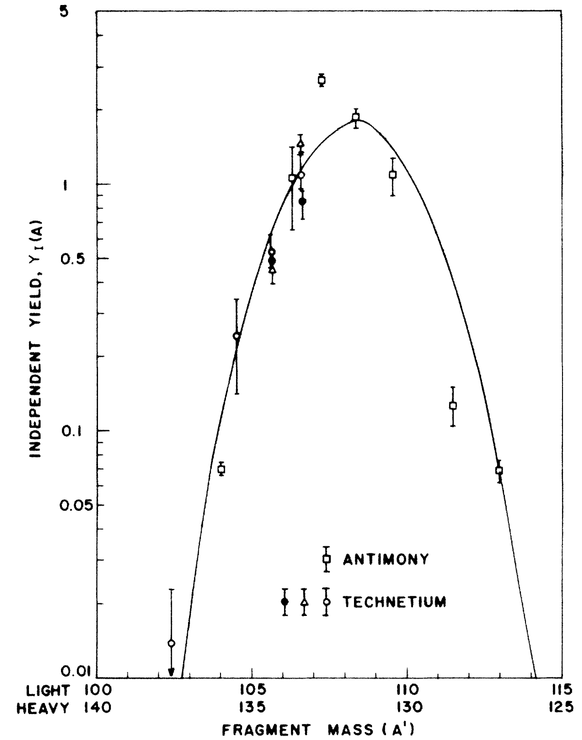

FIG. 2. Independent yield $Y_I(A)$ of Tc and Sb isotopes in the thermal-neutron-induced fission of ^{239}Pu.

isotopes. From the value of slope and intercept of the X-Y plot, the fractional cumulative yield was obtained as

$$Y_{\mathrm{FC}}(\text{parent}) = \frac{\text{slope}(N_p)}{\text{slope}(N_p) + \text{intercept}(N_D)} \ . \tag{5}$$

The above procedure has been adopted for the calculation of FCY's for ^{101}Mo, ^{103}Mo, ^{104}Mo, and ^{105}Tc.

IV. RESULTS AND DISCUSSION

The fractional independent yields Y_{FI} of ^{101}Tc, ^{103}Tc, ^{104}Tc, and ^{105}Tc obtained in this work along with the literature values are given in Table II. The uncertainty quoted on the FIY's is the standard deviation of the average value based on the three independent measurements. The overall error on the measured values was estimated to be around 12%. In estimating these errors the possible uncertainty in the determination of the number of fissions, chemical yield, separation time, and counting statistics were taken into account. It is observed from Table II that the FIY values of ^{101}Tc and ^{103}Tc obtained in the present work are determined for the first time. Further the value of ^{104}Tc is in close agreement with the experimental value of the earlier workers[10] though there is a discrepancy between the FIY's of ^{105}Tc obtained in the present work and the literature value.[10] The FIY's obtained in the present work were used to arrive at the isotopic yield distribution parameters. Wahl[16] has shown that the isotopic yield distribution can be described by a Gaussian function, and the independent yield $Y_I(A,Z)$ is given by

$$Y_I(A,Z) = \frac{Y_Z}{\sqrt{2\pi\sigma_A'^2}} \int_{A'-0.5}^{A'+0.5} e^{-(A'-Ap')^2/2\sigma_A'^2} dA \ , \tag{6}$$

where A_p' is the most probable fragment mass, σ_A' is the width of the distribution, and Y_Z is the elemental yield. From the measured fractional independent yields the isotopic yield distribution parameters A_p' and σ_A' were

TABLE III. Fragment mass (A'), $Y_{\mathrm{FI}}(A)$, and $Y_{\mathrm{FC}}(A)$ of Tc isotopes in the thermal-neutron-induced fission of ^{239}Pu.

Nuclide	Fragment mass (A')	Independent yield (A,Z)	$Y_{\mathrm{FI}}(A)$	$Y_{\mathrm{FC}}(A)$
^{101}Tc	102.40	0.014±0.009	0.0018±0.0012	0.0018±0.0012
^{102}Tc[a]	103.44	0.050±0.025	0.0063±0.0033	0.0081±0.0035
^{103}Tc	104.48	0.241±0.097	0.0301±0.0131	0.0382±0.0136
^{104}Tc	105.56	0.536±0.083	0.0670±0.0152	0.1052±0.0204
^{105}Tc	106.62	1.086±0.251	0.1358±0.0389	0.241 ±0.0439

[a] From interpolation of $Y_I(A,Z)$ vs A' as shown in Fig. 2.

evaluated using the following relationships:

$$Y_I(A,Z)=Y_{FI}(Z)Y_A\ , \tag{7}$$

$$Y_{FI}(A)=Y_I(A,Z)/Y_Z\ , \tag{8}$$

$$Y_{FC}(A)=\sum_{i=1}^{n} Y_{FI}(A)\ , \tag{9}$$

where $Y_I(A,Z)$ is the independent yield, Y_A is the chain yield, Y_Z is the elemental yield, $Y_{FI}(A)$ and $Y_{FC}(A)$ are isotopic fractional independent and fractional cumulative yields, respectively. The Y_Z value was evaluated by summing the $Y_I(A,Z)$ values obtained from the smooth Gaussian plot of $Y_I(A,Z)$ as a function of fragment mass as shown in Fig. 2. The $Y_I(A,Z)$ values were obtained from the present work and the literature.[17] The fragment mass was obtained by correcting for prompt neutron emission using the experimental data of Apalin *et al.*[18] The Y_Z value was obtained as 8.01 ± 1.32 for Tc in the thermal neutron fission of ^{239}Pu. Table III gives the value of $Y_{FI}(A)$ and $Y_{FC}(A)$ obtained using Eqs. (8) and (9). A plot of $Y_{FC}(A)$ as a function of fragment mass is shown in Fig. 3. The value for ^{102}Tc was obtained from the interpolation of $Y_I(A,Z)$ as a function of fragment mass. From Fig. 3, A_p' and σ_A' values were obtained as 108.35 ± 0.45 and 1.80 ± 0.25, respectively. Table IV gives the Y_Z and A_p' values calculated for technetium and its complementary fragment in ^{235}U(n_{th},*f*) and ^{252}Cf(SF) from the literature data[11,14] as well as data obtained in the present work. It is clear from the Y_Z values given in Table IV that the formation probability of the isotopes of technetium and their complementary fragments is much higher in the case of ^{239}Pu(n_{th},*f*) and ^{252}Cf(SF) in comparison to the thermal-neutron-induced fission of ^{235}U. The values of the most probable neutron numbers (N_p') associated with these charge splits are also shown in Table IV. It is seen that N_p' for technetium in all three fissioning nuclei is 66, an expected deformed neutron shell, while N_p' for the complementary fragment in ^{236}U, being 78 is neither a spherical nor a deformed shell. In ^{240}Pu, N_p' for the complementary fragment is 81, closer to the 82 spherical neutron shell configuration while in ^{252}Cf, N_p' of the complementary fragment is 88 which is a major deformed neutron shell.[13] Hence from the point of view of neutron shell stabilization it is expected that the complementary charge split 43 and 55 (^{252}Cf) is most favored followed by 43 and 51 (^{239}Pu), while 43 and 49 (^{235}U) is not favored. As pointed out by Wilkins *et al.*,[13] the shell correction is more important when it reinforces the minimum in the liquid drop potential, which occurs at N/Z of the fragment close to that of N/Z of the fissioning nucleus, and the N/Z appropriate to actinide nuclei neutron shells (82,88) is more important than the 50-proton shell. Their calculations[13] show that the fragments having a spherical 82 N do not have the 50 protons due to the fact that the formation of such a "doubly magic" shell is hindered by strong preference in the liquid drop terms. To maintain the N/Z ratio of fragments close to that of the parent nucleus, $N=82$ would require 52 protons. A similar consideration for 88 N shell requires 55 and 56 protons. This gives a logical explanation why the charge yield peaking[19] occurs around 52—56 for heavy fragments, rather than at 50 for all the actinide nuclei, and the corresponding highest mass yields occurring around the 134—140 mass region. In the spontaneous fission of ^{252}Cf the presence of the 88-N deformed shell, together with the 66-N deformed shell in the complementary fragment, enables the minimization of the mutual Coulombic energy term in the total potential energy, and this could possibly be the

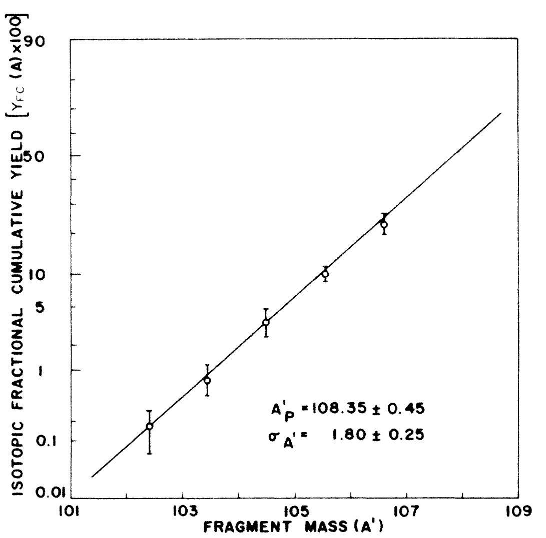

FIG. 3. Isotopic fractional cumulative yield $Y_{FC}(A)$ of Tc isotopes in the thermal-neutron-induced fission of ^{239}Pu.

TABLE IV. Most probable mass number A_p' and associated neutron number N_p' and total elemental yield Y_Z for charge split involving Tc and its complementary fragment charge in ^{236}U, ^{240}Pu, and ^{252}Cf.

Fissioning system	Element (Z)	A_p'	N_p'	Y_Z (%)	Ref.
^{236}U	Tc(43)	109.14±0.14	66.14±0.14	0.092±0.024	11
	In(49)	126.86±0.14	77.86±0.14		
^{240}Pu	Tc(43)	108.35±0.45	65.35±0.45	8.01±1.32	This work
	Sb(51)	131.65±0.45	80.65±0.45		
^{252}Cf	Tc(43)	109.10±0.50	66.10±0.50	16.90±2.09	This work
	Cs(55)	142.90±0.50	87.90±0.50		

reason for further enhancement of Tc yields in the spontaneous fission of ^{252}Cf. Very recently, while this paper was under preparation, further support to the important role played by the 88 deformed neutron shell was reported by Djbera *et al.*[20] in the charge distribution studies of ^{229}Th and ^{232}U.

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