Regular Article – Experimental Physics

Mass distribution in the quasi-mono-energetic neutron-induced fission of $^{\rm 232}{\rm Th}$

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Received: 29 June 2014 / Revised: 10 August 2014

Published online: 19 September 2014 – © Società Italiana di Fisica / Springer-Verlag
 2014 Communicated by H. Miyatake

Abstract. The cumulative yields of various fission products in ²³²Th(n, f) with average neutron energies of 6.35, 8.53 and 10.09 MeV have been determined using an off-line γ -ray spectrometric technique. The neutron beam was produced from the ⁷Li(p, n) reaction. From the cumulative fission yields, the mass chain yields were obtained by using charge distribution correction of medium energy. The peak-to-valley (P/V)ratio, the average value of light mass $(\langle A_{\rm L} \rangle)$, heavy mass $(\langle A_{\rm H} \rangle)$ and the average number of neutrons $(\langle \nu \rangle)$ at the three different neutron energies of the present work and at other energies from the literature in the ²³²Th(n, f) reaction were obtained from the mass yield data. The present and the existing literature data in the ²³²Th(n, f) reaction at various excitation energies were compared with similar data in the ²³⁸U(n, f) reaction. The fine structure in the mass yield distribution was interpreted from the point of nuclear structure effect such as shell closure proximity and even-odd effect. The role of standard I and standard II asymmetric mode of fission was discussed. The different types of mass-yield distributions between ²³²Th(n, f) reactions were explained from different types of the potential energy between the two fissioning systems. The role of excitation energy was also investigated.

1 Introduction

Measurements of nuclear data such as neutron capture cross-sections, fission cross-sections and fission yields are required for many reactor calculations. Sufficient data on neutron capture cross-section and fission cross-section over a wide range of neutron energies from thermal to MeV range are available in the literature [1–3]. Similarly, the fission yields data in the neutron-induced fission of actinides, ranging from 227 Th to 257 Fm, are available from different compilations [4–8]. Among the nuclear data, the fission yields in the neutron-induced fission of the isotopes of actinides, ranging from Th to Cm are important for their applications in conventional light- and heavy-water reactors as well as fast reactors [9–13], advanced heavy-water reactors (AHWR) [14,15] and accelerator-driven subcritical systems (ADSs) [16–21].

Besides the above applications, basic studies in the fission yield distribution in the low-energy fission of actinides provide important information about the nuclear structure effect (e.g., shell closure proximity and even-odd effect) and descent dynamics from the point of saddle to scission [2,3]. It is a well-known fact that the mass yield distributions in the spontaneous and neutron-induced fission of heavy-Z actinides (e.q., Es to Lr) are symmetric in nature [2,3]. On the other hand, the mass yield distributions in the neutron-induced fission of medium-Z actinides (e.q., U to Cf) are asymmetric with double hump, whereas for light-Z actinides (e.g., Ac, Th, Pa) are asymmetric with triple hump [2,3]. Other than this, the fine structures due to the even-odd effect and shell closure proximity have been observed in the low-energy neutron-induced fission of light and medium even-Z actinides. This was also seen for the fission fragments yield data of neutron-deficient lighter even-Z actinides such as $^{220-229}\rm{Th}$ and $^{231-234}\rm{U}$ in the excitation energy range of the GDR region due to electromagnetic fission in inverse kinematics [22–24]. Similarly, the nuclear structure effect in the reactor [25– 28] and mono-energetic [29–82] neutron-induced fissions of 232 Th and 238 U can be seen. The fission yield data in the

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mono-energetic neutron-induced fission of ²³⁸U are available in the literature [54–82], within the neutron energy of 1.5 to 17.7 MeV. However, systematic fission yield data in the mono-energetic neutron-induced fission of 232 Th are available in the literature [29–52], only within the neutron energy of 2 to 8 MeV and at 14.8 MeV. Within the neutron energy of 8 to 14.8 MeV, sufficient fission yield data for 232 Th(n, f) are not available in the literature to examine the effect of nuclear structure. At higher neutron energies, the experimental results in the 33-60 MeV quasimono-energetic neutron-induced fission of ²³²Th and ²³⁸U are available from physical measurements [53], where the effect of the nuclear structure is not expected. In view of this, in the present work, the yields of various fission products in the 232 Th(n, f), with average neutron energies of 6.35, 8.53 and 10.09 MeV, have been determined using an off-line γ -ray spectrometric technique. From the yields of the fission products, their mass chain yields were obtained by using charge distribution correction [8,83]. The fission product yields data in the three neutron energies of the present work and at other neutron energies from the literature [29-52] in ²³²Th(n, f) are compared with similar data in 238 U(n, f) [52–82] to examine the role of the excitation energy in the nuclear structure effect. The role of the excitation energy in the peak-to-valley (P/V) ratio was examined. The variation of average light $(\langle A_{\rm L} \rangle)$ and heavy $(\langle A_{\rm H} \rangle)$ masses as well the average neutron number $\langle \langle v \rangle \rangle$ as a function of the excitation energy has been also discussed.

2 Experimental details

The quasi-mono-energetic neutron beam used in the present experiment was obtained from the ${}^{7}Li(p, n){}^{7}Be$ reaction by using the proton beam from the 14UD BARC-TIFR Pelletron facility at Mumbai, India [84,85]. A collimator of 6 mm diameter was used before the Li target to avoid the energy spread of the proton beam. The lithium foil used was made up of natural lithium with thickness of $3.7 \,\mathrm{mg/cm^2}$, sandwiched between two tantalum foils of different thickness. The front tantalum foil facing the proton beam is $3.9 \,\mathrm{mg/cm^2}$ thick, in which the degradation of proton energy is only 50-80 keV [86]. On the other hand, the back tantalum foil is 0.025-0.1 mm thick, which is sufficient to stop the proton beam. Behind the Ta-Li-Ta stack, the $^{232}\mathrm{Th}$ metal foil wrapped with a $0.025\,\mathrm{mm}$ thick super pure aluminum foil was used for irradiation. The aluminum wrapper was used as a catcher foil to stop fission products recoiling out from the 232 Th metal foil during the irradiation. The size of each 232 Th metal foil was about $1.0 \,\mathrm{cm}^2$ with thickness of about $323.7 - 325.2 \,\mathrm{mg/cm}^2$. The sample was mounted at zero degree angle in the forward direction with respect to the beam direction at a distance of 2.1 cm from the location of the Ta-Li-Ta stack. Different sets of Ta-Li-Ta stacks and Al-wrapped ²³²Th samples were made for different irradiations at various neutron energies. The sample was then irradiated by neutrons generated by impinging the proton beam on the lithium metal foil through the thin tantalum foil of the Ta-Li-Ta metal

stack. The irradiations time were for 5–15 h depending upon the proton energy faced by the front tantalum foil. The proton energies used in the present work were 10, 14 and 18 MeV, respectively. The proton current during the irradiations varied within 200 to 400 nA. The corresponding maximum energies of the neutron beam impinging on the 232 Th samples were 8.12, 12.12 and 16.12 MeV, respectively. After each irradiation, the samples were cooled for 2–3 h.

The irradiated ²³²Th targets along with aluminum catcher were mounted on different Perspex plates. The γ rays counting of the fission products from the irradiated samples were done in a energy- and efficiency-calibrated $80 \,\mathrm{cm}^3$ HPGe detector coupled to a PC-based 4K channel analyzer. The dead time of counting was always less than 5% by placing the irradiated sample at a suitable distance from the end cap of the detector to avoid pileup effects. The γ -ray counting of the sample was done in live-time mode and was followed as a function of time. The resolution of the detector system during counting was 1.8 keV FWHM at 1332.5 keV of ⁶⁰Co. The energy and efficiency calibration of the detector system was performed by using the 121.8 to 1408.01 keV γ -rays from standard ¹⁵²Eu source at same geometry, where the summation error was negligible. This was checked by comparing the efficiency obtained from γ -ray counting of standards such as 241 Am (59.5 keV), 133 Ba (80.9, 276.4, 302.9, 356.0, and 383.8 keV), 137 Cs (661.7 keV), 54 Mn (834.6 keV), 60 Co (1173 and 1332 keV). The detector efficiency was 20% at 1332.5 keV, relative to 3" diameter \times 3" length NaI(Tl) detector. The uncertainty in the efficiency was 2–3%. For each irradiated samples, several sets of measurements were done with increasing counting time to cover the different fission products, from the half-life of 50 minutes to 33 days. The $\gamma\text{-ray}$ counting of the irradiated ^{232}Th samples was done up to few months to check the half-life of the fission products of interest.

3 Calculation and results

3.1 Calculation of the neutron energy

The neutron flux in the present experiment were produced from the ⁷Li(p, n) reaction [87-89] of the natural occurring ⁷Li with isotopic abundance of 92.41%. The Q-value for the ${}^{7}\text{Li}(p, n){}^{7}\text{Be}$ reaction to the ground state is $-1.644 \,\mathrm{MeV}$, whereas the first excited state is $0.431 \,\mathrm{MeV}$ above ground state leading to an average Qvalue of -1.868 MeV. However, the threshold value to populate the ground state of ⁷Be is 1.881 MeV. Thus, for the proton energies of 10, 14 and 18 MeV, the resulting peak energies of first group of neutrons (n_0) will be 8.12, 12.12 and 16.12 MeV, respectively. The corresponding neutron energies of second group of neutrons (n_1) , for the first excited state of 7 Be will be 7.63, 11.63 and 15.63 MeV, respectively. This is because, above 2.37 MeV, the n_1 group of neutron is also produced. Above the proton energy of 4.5 MeV, the fragmentation of 8Be to $^4He + ^3He + n$ $(Q = -3.23 \,\mathrm{MeV})$ takes place, which causes continuous

neutron energy distribution besides n_0 and n_1 groups of neutrons. Meadows and Smith [88] have given experimental neutron distributions from the break-up channels and also parameterized these distributions. We have generated the neutron spectrum using the neutron energy distribution given by C.H. Poppe *et al.* [89] and shown in our earlier work [84,85]. From those neutron spectrums [84, 85], the flux-weighted average neutron energies were calculated as 6.35, 8.53 and 10.09 MeV, respectively. The energy spread for the above average neutron energies range from around 0.3 to 0.7 MeV [84,85].

3.2 Calculation of fission product yields

The numbers of detected $(N_{\rm obs}) \gamma$ -rays for the fission products of interest were obtained by subtracting the linear Compton background from the total peak areas. From $N_{\rm obs}$ of an individual fission product, the cumulative yields $(Y_{\rm R})$ relative to ⁹²Sr were calculated by using the usual decay equation [27,28],

$$Y_{\rm R} = \frac{N_{\rm obs}({\rm CL/LT})\lambda}{n\sigma_f \varphi I_{\gamma} \varepsilon (1 - e^{-\lambda t}) e^{-\lambda T} (1 - e^{-\lambda {\rm CL}})}, \qquad (1)$$

where *n* is the number of target atoms, φ is the neutron flux and σ_f is the neutron fluxed average fission crosssection of ²³²Th at 6.35, 8.53 and 10.09 MeV. I_{γ} is the γ -ray intensity or γ -ray emission probability, ε is the detection efficiency of the γ -rays in the detector system and λ is the decay constant of the fission-product of interest $(\lambda = \ln 2/T_{1/2})$. *t* and *T* are the irradiation and cooling times, whereas, CL and LT are the real time and the live time of counting, respectively.

From eq. (1), the $n\sigma_f \varphi$ term was first obtained from the photo-peak activity of 1383.9 keV γ -line and by assuming the cumulative yield $(Y_{\rm R})$ of ⁹²Sr as 1.0. Thus the individual terms, such as n, σ_f and φ , are not necessary to be used separately. The nuclear spectroscopic data, such as the γ -ray energies, the half-lives $(T_{1/2})$, and the γ -ray intensity (I_{γ}) of the fission products were taken from the literature [90,91]. Then, by using the $n\sigma_f \varphi$ term in eq. (1), the relative cumulative yields $(Y_{\rm R})$ of other fission products were obtained from the photo-peak activities of the γ -lines of corresponding fission products. From the $Y_{\rm R}$ values of the fission products, their relative mass-chain yields $(Y_{\rm RA})$ were calculated after correcting for charge distribution [8,83]. According to Wahl's prescription [8], the fractional cumulative yield $(Y_{\rm FCY})$ of a fission product in an isobaric mass chain is given as

$$Y_{\rm FCY} = \frac{\rm EOF^{a(Z)}}{\sqrt{2\pi\sigma^2}} \int_{-\infty}^{Z+0.5} \exp[-(Z-Z_{\rm P})^2/2\sigma^2] dZ, \quad (2)$$

$$Y_{\rm RA} = Y_{\rm R} / Y_{\rm FCY},\tag{3}$$

where $Z_{\rm P}$ is the most probable charge and σ_z is the width parameter of an isobaric-yield distribution. EOF^{*a*(*Z*)} is the even-odd effect with a(Z) = +1 for even-*Z* nuclides and -1 for odd-*Z* nuclides. From the above equations it can be seen that in an isobaric mass chain, it is necessary to have knowledge of $Z_{\rm P}$, σ_z and EOF^{a(Z)} to calculate the $Y_{\rm FCY}$ value of a fission product and thus the mass-chain yield $(Y_{\rm RA})$. The EOF^{a(Z)} values in the medium-energy fission is negligible. It was shown by Umezawa *et al.* [83] that the σ_z value in medium-energy proton- and alpha-induced fission of ²³²Th and ²³⁸U is 0.70 ± 0.06 . The $Z_{\rm P}$ values of the individual mass chain (A) in the neutron-induced fission of ²³²Th at the average energy of 6.35, 8.53 and 10.09 MeV were calculated based on the prescription of Umezawa *et al.* [83] as

$$Z_{\rm P} = \eta Z_{\rm F} \pm \Delta Z_{\rm P}, \quad \eta Z_{\rm F} = Z_{\rm UCD} = (Z_{\rm F}/A_{\rm F})(A + v_{\rm post}),$$

$$\eta = (A + v_{\rm post})/(A_{\rm C} - v_{\rm pre}), \quad A_{\rm F} = A_{\rm C} - v_{\rm pre}, \quad (4b)$$

where $Z_{\rm C}$ and $A_{\rm C}$ are the charge and mass of the compound nucleus. $Z_{\rm F}$ and $A_{\rm F}$ are the charge and mass of the fissioning system (²³³Th^{*}). $Z_{\rm UCD}$ is the most probable charge based on the unchanged charge-density distribution as suggested by Sugarman and Turkevich [92]. A is the mass of the fission product and $\Delta Z_{\rm P}$ (= $Z_{\rm P} - Z_{\rm UCD}$) is the charge-polarization parameter. The + and - signs for the $\Delta Z_{\rm P}$ value are applicable to light and heavy fragments, respectively. $\nu_{\rm pre}$ and $\nu_{\rm post}$ are pre- and post-fission neutrons at scission, which were calculated from the excitation energy (E^*) of the compound nucleus using the following relations [83]:

$$\nu_{\rm pre} = \frac{E^*}{7.5 \pm 0.5} + \frac{Z_{\rm C}}{2A_{\rm C}} - (19.0 \pm 0.5), \ 1.0 \ \text{for} \ A > 88,$$
(5a)

$$\nu_{\text{post}} = 1.0 \text{ for } A > 88, 1.0 + 0.1(A - 88) \text{ for } 78 < A < 88, 0 \text{ for } A < 78.$$
(5b)

For the average neutron energies of 6.35, 8.53 and 10.09 MeV, the excitation energy (E^*) of the compound nucleus is 10.96, 13.14 and 14.7 MeV, respectively. The E^* values were used in eq. (5b) to calculate the $\nu_{\rm pre}$ values at three different neutron energies. The $\nu_{\rm pre}$ and $\nu_{\rm post}$ values obtained from eqs. (5b) and (5b) were used in eqs. (4a) and (4b) to calculate the value of $Z_{\rm UCD}$ as a function of mass number for the different fission products. The $\Delta Z_{\rm P}$ value was then calculated from the following relation [83]:

$$\Delta Z_{\rm P} = 0, \text{ for } I\eta - 0.5I < 0.04,$$
 (6a)

$$\Delta Z_{\rm P} = (20/3)(I\eta - 0.5I - 0.04),$$

for 0.04 < I\eta - 0.5I < 0.085. (6b)

The $Z_{\rm P}$ value as a function of mass number and the average width parameter (σ_z) of 0.7 were used in eq. (2) to obtain the $Y_{\rm FCY}$ values for individual fission products. The relative mass-chain yield ($Y_{\rm RA}$) of the fission products from their relative cumulative yield ($Y_{\rm R}$) were obtained in eq. (3) by using the $Y_{\rm FCY}$ values of different fission products. The relative mass-chain yields ($Y_{\rm RA}$) of the fission products obtained were then normalized to a total yield of 200% to calculate the absolute mass-chain yields ($Y_{\rm A}$). Page 4 of 11

Nuclide	Half-life	γ -ray energy	γ -ray intensity	$6.35\mathrm{MeV}$	$6.35\mathrm{MeV}$	$8.53\mathrm{MeV}$	$8.53\mathrm{MeV}$	$10.09\mathrm{MeV}$	$10.09{ m MeV}$
		(keV)	(%)	$Y_{\rm C}~(\%)$	$Y_{\rm A}~(\%)$	$Y_{\rm C}~(\%)$	$Y_{\rm A}~(\%)$	$Y_{\rm C}~(\%)$	$Y_{\rm A}~(\%)$
⁸⁵ Kr ^m	4.48 h	151.2	75.0 ± 0.4	3.870 ± 0.304	3.870 ± 0.304	2.956 ± 0.297	2.956 ± 0.297	3.225 ± 0.082	3.225 ± 0.082
		304.9	14.0 ± 0.5	3.846 ± 0.375	3.846 ± 0.375	2.919 ± 0.343	2.919 ± 0.343	3.571 ± 0.177	3.571 ± 0.177
87 Kr	76.3 min	402.6	49.6 ± 2.0	5.970 ± 0.710	5.988 ± 0.712	5.970 ± 0.710	5.988 ± 0.712	5.864 ± 0.268	5.882 ± 0.273
⁸⁸ Kr	2.84 h	196.3	25.98 ± 1.7	5.336 ± 0.126	5.423 ± 0.128	5.336 ± 0.126	5.423 ± 0.128	5.268 ± 0.268	5.354 ± 0.273
$^{91}\mathrm{Sr}$	$9.63\mathrm{h}$	749.8	23.61 ± 1.7	6.268 ± 0.556	6.268 ± 0.556	5.621 ± 0.449	5.621 ± 0.449	5.577 ± 0.610	5.577 ± 0.610
		1024.3	33.0 ± 0.7	6.202 ± 0.617	6.202 ± 0.617	6.478 ± 0.723	6.478 ± 0.723	5.677 ± 0.359	5.677 ± 0.359
92 Sr	$2.71\mathrm{h}$	1384.9	90.0 ± 0.3	6.005 ± 0.479	6.017 ± 0.480	5.258 ± 0.199	5.259 ± 0.199	5.248 ± 0.569	5.259 ± 0.570
⁹³ Y	10.18 h	266.9	7.3 ± 0.4	5.233 ± 0.261	5.233 ± 0.261	4.954 ± 0.167	4.954 ± 0.167	4.640 ± 0.505	4.640 ± 0.505
95 Zr	$64.02\mathrm{d}$	724.2	44.17 ± 1.3	7.171 ± 0.584	7.171 ± 0.584	6.256 ± 0.097	6.256 ± 0.097	6.077 ± 0.428	6.077 ± 0.428
97 Zr	16.91 h	743.4	93.0 ± 0.9	5.138 ± 0.664	5.143 ± 0.665	4.634 ± 0.421	4.639 ± 0.422	4.318 ± 0.177	4.322 ± 0.177
^{99}Mo	$65.94\mathrm{h}$	140.5	89.43 ± 2.3	3.286 ± 0.252	3.286 ± 0.252	3.179 ± 0.255	3.179 ± 0.255	2.939 ± 0.301	2.939 ± 0.301
		739.5	12.13 ± 1.2	3.305 ± 0.299	3.305 ± 0.299	3.244 ± 0.292	3.244 ± 0.292	3.303 ± 0.296	3.303 ± 0.296
103 Ru	$39.26\mathrm{d}$	497.1	90.9 ± 1.0	0.620 ± 0.062	0.622 ± 0.062	0.656 ± 0.083	0.622 ± 0.083	0.969 ± 0.164	0.969 ± 0.164
105 Rh	$35.36\mathrm{h}$	319.1	$19.0 \pm 1.0^{(a)}$	0.218 ± 0.019	0.218 ± 0.019	0.366 ± 0.051	0.366 ± 0.051	0.446 ± 0.041	0.466 ± 0.041
^{112}Ag	$3.13\mathrm{h}$	617.5	$43.0 \pm 2.2^{(a)}$	0.266 ± 0.033	0.266 ± 0.033	0.477 ± 0.037	0.477 ± 0.037	0.623 ± 0.150	0.624 ± 0.150
$^{115}\mathrm{Cd^g}$	$53.46\mathrm{h}$	527.9	27.45 ± 1.8	0.237 ± 0.024		0.417 ± 0.083		0.562 ± 0.102	
115 Cd ^{total}				$0.277 \pm 0.028^{(b)}$	$0.277 \pm 0.028^{(b)}$	$0.487 \pm 0.097^{(b)}$	$0.487 \pm 0.097^{(b)}$	$0.655 \pm 0.123^{(b)}$	$0.655 \pm 0.123^{(b)}$
$^{127}\mathrm{Sb}$	$3.85\mathrm{d}$	685.7	$37.0 \pm 1.9^{(a)}$	0.532 ± 0.043	0.532 ± 0.043	0.639 ± 0.056	0.639 ± 0.056	0.964 ± 0.187	0.964 ± 0.187
128 Sn	59.07 min	482.3	59.0 ± 0.3	1.145 ± 0.103	1.218 ± 0.109	1.163 ± 0.174	1.237 ± 0.185	1.578 ± 0.214	1.679 ± 0.217
^{129}Sb	$4.32\mathrm{h}$	812.8	$43.0 \pm 2.2^{(a)}$	1.699 ± 0.080	1.710 ± 0.081	1.603 ± 0.189	1.613 ± 0.190	2.035 ± 0.163	2.047 ± 0.164
^{131}I	$8.02\mathrm{d}$	364.5	81.7 ± 0.6	3.182 ± 0.256	3.182 ± 0.256	2.901 ± 0.236	2.901 ± 0.236	2.980 ± 0.237	2.980 ± 0.237
$^{132}\mathrm{Te}$	$3.2\mathrm{d}$	228.2	88.0 ± 1.8	4.136 ± 0.304	4.136 ± 0.304	3.815 ± 0.345	3.846 ± 0.348	3.416 ± 0.232	3.440 ± 0.234
^{133}I	$20.8\mathrm{h}$	529.9	87.0 ± 1.7	5.598 ± 0.394	5.603 ± 0.394	5.166 ± 0.412	5.172 ± 0.412	5.013 ± 0.301	5.018 ± 0.302
^{134}I	52.5 min	847.3	95.4 ± 0.3	6.569 ± 0.588	6.582 ± 0.589	6.692 ± 0.412	6.705 ± 0.412	6.615 ± 0.817	6.628 ± 0.819
		884.1	64.9 ± 1.9	7.123 ± 0.640	7.137 ± 0.641	7.123 ± 0.640	7.137 ± 0.641	6.674 ± 0.790	6.687 ± 0.792
^{135}I	$6.57\mathrm{h}$	1131.5	22.7 ± 1.4	4.927 ± 0.301	4.977 ± 0.304	5.822 ± 0.528	5.881 ± 0.533	5.147 ± 0.621	5.199 ± 0.628
		1260.4	28.9 ± 1.7	5.157 ± 0.155	5.209 ± 0.157	5.244 ± 0.523	5.297 ± 0.528	5.174 ± 0.554	5.227 ± 0.560
139 Ba	83.06 min	165.9	23.7 ± 2.4	7.465 ± 0.556	7.465 ± 0.556	7.197 ± 0.343	7.197 ± 0.343	6.892 ± 0.732	6.892 ± 0.732
140 Ba	$12.75\mathrm{d}$	537.3	24.4 ± 0.7	8.358 ± 0.551	8.358 ± 0.551	8.299 ± 0.579	8.299 ± 0.579	7.579 ± 0.491	7.579 ± 0.491
141 Ce	$32.5\mathrm{d}$	145.4	48.2 ± 0.3	7.304 ± 0.423	7.304 ± 0.423	6.983 ± 0.598	6.893 ± 0.598	6.811 ± 0.828	6.811 ± 0.828
142 La	91.1 min	641.3	47.0 ± 2.0	6.383 ± 0.551	6.383 ± 0.551	6.265 ± 0.403	6.265 ± 0.403	6.251 ± 0.851	6.251 ± 0.851
¹⁴³ Ce	33.04 h	293.3	42.8 ± 1.3	6.777 ± 0.399	6.777 ± 0.399	6.780 ± 0.459	6.780 ± 0.459	6.669 ± 0.741	6.669 ± 0.741
¹⁴⁷ Nd	$10.98\mathrm{d}$	531.0	13.1 ± 0.7	2.197 ± 0.019	2.203 ± 0.019	2.412 ± 0.134	2.419 ± 0.134	2.118 ± 0.313	2.124 ± 0.314
150 Pm	2.68 h	333.97	$68.0 \pm 3.4^{(a)}$	0.502 ± 0.104	0.503 ± 0.104	0.472 ± 0.055	0.473 ± 0.056	0.528 ± 0.051	0.529 ± 0.051

Table 1. Nuclear spectroscopic data and yields of fission products in the 6.35, 8.53 and 10.09 MeV neutron-induced fission of 232 Th. $Y_{\rm R}$ – Cumulative yields; $Y_{\rm A}$ – Mass yields; 92 Sr – Fission rate monitor.

 $^{(a)}$ Assumed value of 5%.

^(b) The yields of ¹¹⁵Cd^{total} is based on the ratio of ¹¹⁵Cd^g/¹¹⁵Cd^m = 6 from ref. [71].

The absolute cumulative yields $(Y_{\rm C})$ of the fission products in the 6.35, 8.53 and 10.09 MeV neutron-induced fission of ²³²Th were then obtained from the mass-chain yield data and $Y_{\rm FCY}$ values by using eq. (7), which is the modified form of eq. (3),

$$Y_{\rm C} = Y_{\rm A} \times Y_{\rm FCY}.$$
 (7)

The absolute cumulative yield $(Y_{\rm C})$ and mass-chain yield $(Y_{\rm A})$ of the fission products in the 6.35, 8.53 and 10.09 MeV neutron-induced fission of ²³²Th along with the nuclear spectroscopic data from refs. [90,91] are given in table 1. The uncertainty shown in the measured cumulative yield of the individual fission products shown in table 1 is the statistical fluctuation of the mean value from two determinations. The overall uncertainty is the contributions from both random and systematic errors. The random error in the observed activity is due to counting statistics and is estimated to be 5–10%, which can be determined by accumulating the data for the optimum period of time, depending on the half-life of the nuclide of interest. The overall systematic errors is about 3.8– 10.5%, which comes from the uncertainties in irradiation time (0.2%), detector efficiency calibration (~ 3%), halflife of the fission products (~ 1%), and γ -ray intensity (2–10%) [90,91].Thus for the yields of fission-products an upper limit of error of 6.3–34.7% was obtained based on 5–10% random error and a 3.8–10.5% systematic error.

4 Discussion

The fission products yields for 232 Th(n, f) at average neutron energy of 8.53 and 10.09 MeV shown in table 1 are determined for the first time. At 6.35 MeV, the yields for 232 Th(n, f) are the redetermined values and are in agreement with the literature data at 6.4 MeV [46]. The literature data [29–51] are based on mono-energetic neu-



Fig. 1. Plot of the mass yields distribution in log scale from the quasi-mono-energetic neutron-induced fission of 232 Th at 6.35, 8.53 and 10.09 MeV and of 238 U at 6.0, 8.1 and 10.09 MeV.

trons, whereas the present data are for flux-weighted average neutron energies based on quasi-mono-energetic neutrons. The mass-chain yield data in 232 Th(n, f) from the present work at average neutron energies of 6.35, 8.53 and 10.09 MeV are plotted as a function of their mass number in log and in linear scale in fig. 1 and fig. 2, respectively. In the same figures, the fission products yields in $^{\bar{2}38}$ U(n, f) at neutron energies of 6.0, 8.1 and 10.09 MeV from the literature [70,82] are also plotted for comparison. It can be seen, from fig. 1, that within the neutron energy of 6.4–10 MeV of present work, the mass-yield distribution in ²³²Th(n, f) is triple-humped, unlike in 238 U(n, f), where it is doublehumped. The fission fragments yield distribution in the excitation energy range of the GDR region due to electromagnetic fission in inverse kinematics [22-24] for the neutron-deficient $^{220-229}$ Th is also triple-humped and, for $^{231-234}$ U, it is double-humped. This is due to the different type of potential energy surface in $^{220-233}$ Th* compared to $^{231-239}$ U* [93].

From fig. 2, it can be seen that both in 232 Th(n, f) and 238 U(n, f), the yields of fission products for A = 133-134, 138–140, 143–144 and their complementary products are higher than the other fission products. The oscillation of fission yields in the interval of five mass units around mass region of 133–144 is due to the even-odd effect [94]. The higher yields of fission products for A = 134-134 and 143-144 can also be explained from the point of view of the standard I and standard II asymmetric fission modes as mentioned by Brossa et al. [95], which arise due to shell effects [96]. Based on standard I asymmetry, the fissioning system is characterized by spherical heavy fragment with mass numbers 133-134 due to the spherical 82n shell and a deformed complementary light-mass fragment. Based on standard II asymmetry, the fissioning system is characterized by a deformed heavy-mass fragment near the mass

numbers of 143–144 due to a deformed 86-88n shell and slightly deformed light-mass fragment. Thus, the higher yields of fission products for A = 133-134 and 143-144 are due to the presence of spherical 82n and deformed 86-88n shells, respectively.

Further, it can be seen from fig. 2 that the yields of fission products for A = 133-134 and 143-144 are lower than for A = 138-140 in 232 Th(n, f). On the other hand, in 238 U(n, f), the yields of fission products for A = 133-134are higher than for A = 138-140 and 143-144. In order to examine this aspects, the yields of fission products for A = 133, 139 and 143 from the present work (table 1) and literature data in the 232 Th(n, f) [29–52] and $^{238}U(n, f)$ [54–82] and are plotted in fig. 3 as a function of excitation energy. From fig. 3, it can be seen that at all excitation energy, the yields of fission products for A = 134 and their complementary products are comparable in 232 Th(n, f) and 238 U(n, f). On the other hand, the yields of fission products are higher marginally for A = 139 and significantly for A = 143 in ²³²Th(n, f) than in 238 U(n, f). This difference is based on the presence or absence of shells combination in the complementary pairs as explained in earlier work [82]. The variation of yields data for A = 133-134, 139-140 and 143-144 in ²³²Th(n, f) and 238 U(n, f) causes variation of the average heavy mass $(\langle A_{\rm H} \rangle)$. In order to examine this, the average heavy mass $(\langle A_{\rm H} \rangle)$ and light mass $(\langle A_{\rm L} \rangle)$ in the ²³²Th(n, f) from the present work at average neutron energies of 6.35, 8.53 and 10.09 MeV as well as at other lower-neutron energies [29– 52] were calculated from the mass-chain yields (Y_A) of the fission products within the mass ranges of 80–105 and 125–150, and by using the following relation:

$$\langle A_{\rm L} \rangle = \sum (Y_{\rm A} A_{\rm L}) / \sum Y_{\rm A}, \quad \langle A_{\rm H} \rangle = \sum (Y_{\rm A} A_{\rm H}) / \sum Y_{\rm A}.$$
(8)



Fig. 2. Plot of the mass yields distribution in linear scale from the quasi-mono-energetic neutron-induced fission of 232 Th at 6.35, 8.53 and 10.09 MeV and of 238 U at 6.0, 8.1 and 10.09 MeV.

The $\langle A_{\rm L} \rangle$ and $\langle A_{\rm H} \rangle$ values obtained from the above relation in the ²³²Th(n, f) along with their corresponding average excitation energy ($\langle E^* \rangle$) are given in table 2. The $\langle A_{\rm L} \rangle$ and $\langle A_{\rm H} \rangle$ values in the ²³²Th(n, f) from table 2 and literature data [54–82] in ²³⁸U(n, f) are plotted in fig. 4 as a function of excitation energy. It can be seen from fig. 4 that at all excitation energy, the $\langle A_{\rm L} \rangle$ values are significantly lower in ²³²Th(n, f) than in ²³⁸U(n, f), whereas the $\langle A_{\rm H} \rangle$ values are higher in ²³²Th(n, f) than in ²³⁸U(n, f). This is due to the favorable standard II asymmetric mode compared to standard I asymmetric mode of fission in ²³²Th(n, f) than in ²³⁸U(n, f). However, significantly lower $\langle A_{\rm L} \rangle$ values in ²³²Th(n, f) than in ²³⁸U(n, f) are due to the mass conservation based on the standard I and II asymmetric mode of fission. It can be also seen from fig. 4 that the $\langle A_{\rm H} \rangle$ values for both ²³²Th(n, f) and ²³⁸U(n, f) reactions decrease with the excitation energy, whereas the $\langle A_{\rm L} \rangle$ values almost remain constant or slightly increase with the excitation energy. This is most probably due to the probability of more neutron emission from the heavy fragments [46].

Table 2. Average light mass $(\langle A_{\rm L} \rangle)$, heavy mass $(\langle A_{\rm H} \rangle)$ and average neutron numbers $(\langle v \rangle_{\rm expt})$ in the neutron-induced fission of ²³²Th.

$E_{\rm n}~({\rm MeV})$	E^* (MeV)	$\langle A_{\rm L} \rangle$	$\langle A_{\rm H} \rangle$	$\langle \nu \rangle_{\rm expt}$	Ref.
2.0	6.61	90.9	139.8	2.3	[46]
3.0	7.61	91.2	139.4	2.4	[46]
4.0	8.61	91.3	139.1	2.6	[46]
5.9	10.51	91.9	138.4	2.7	[46]
6.35	10.96	91.64	138.44	2.93	Present work
6.4	11.01	91.1	138.9	3.0	[46]
6.9	11.51	90.5	139.1	3.4	[46]
7.6	12.21	90.8	138.9	3.3	[46]
8.0	12.61	90.9	138.8	3.3	[46]
8.53	13.14	91.31	138.39	3.4	Present work
10.09	14.36	91.44	138.08	3.48	Present work
11.0	15.61	_	—	—	[30]
14.7	19.31	_	_	_	[50]
14.8	19.41	93.3	135.8	3.9	[31]



Fig. 3. Plot of yields of fission products (%) as a function of excitation energy for A = 143, 139 and 134 in the ²³²Th(n, f) and ²³⁸U(n, f) reactions. The data of present work in ²³²Th(n, f) reaction are in green with the same symbol as the literature data.

From the $\langle A_{\rm L} \rangle$, $\langle A_{\rm H} \rangle$ and compound nucleus mass $(A_{\rm C} = 233)$, the experimental average numbers of neutrons $(\langle v \rangle_{\rm expt})$ were calculated from the following relation:

$$\langle v \rangle_{\text{expt}} = A_{\text{C}} - (\langle A_{\text{L}} \rangle + \langle A_{\text{H}} \rangle).$$
 (9)

The $\langle v \rangle_{\text{expt}}$ values obtained from the above relation from the present work and literature data in 232 Th(n, f) [29–52] at different excitation energies are also listed in table 2. The $\langle v \rangle_{\text{expt}}$ values for 232 Th(n, f) from table 2 and liter-ature data [54–82] for 238 U(n, f) are plotted in fig. 5 as a function of excitation energy. It can be seen from fig. 5 that in the neutron-induced fission of 232 Th and 238 U, the value of $\langle v \rangle_{\rm expt}$ increases with excitation energy. From fig. 5, it can also be seen that, at the same excitation energy, the $\langle v \rangle_{\text{expt}}$ value in ²³²Th(n, f) is lower than in 238 U(n, f), which is due to the effect of fissility parameter besides excitation energy. However, at excitation energy around 11–13 MeV, the value of $\langle v \rangle_{\text{expt}}$ in ²³²Th(n, f) is comparable or higher than in ²³⁸U(n, f), which is due to GDR effect. Further, it can be seen from figs. 4 and 5 that the increasing trend of $\langle v \rangle_{\text{expt}}$ and $\langle A_{\text{L}} \rangle$ as well as decreasing trend of $\langle A_{\rm H} \rangle$ with excitation energy is not smooth in 232 Th(n, f) as in the case of 238 U(n, f). Within excitation energy of 11–13 MeV, the increasing trend of $\langle v \rangle_{\text{expt}}$ and $\langle A_{\rm L} \rangle$ as well as decreasing trend of $\langle A_{\rm H} \rangle$ with excitation energy is very sharp. This is due to major GDR effect in 232 Th(n, f) compared to 238 U(n, f) besides the effect of excitation energy.

The role of excitation energy on mass yield distribution at higher neutron energy fission of 232 Th and 238 U was clearly shown by I.V. Ryzhov *et al.* [53] from their data



Fig. 4. Plot of average values of heavy mass $(\langle A_{\rm H} \rangle)$ and average values of light mass $(\langle A_{\rm L} \rangle)$ as a function of excitation energy in the 232 Th(n, f) and 238 U(n, f) reactions. The data of the present work in 232 Th(n, f) reaction are in green with the same symbol as the literature data.



Fig. 5. Plot of yields of symmetric and asymmetric fission products (%) as a function of the excitation energy in the 232 Th(n, f) and 238 U(n, f) reactions. The data of the present work in the 232 Th(n, f) reaction are in green with the same symbol as the literature data.

based on physical measurements. As mentioned by them at the same neutron energy, the symmetric fission is more enhanced in the case of 232 Th(n, f) than in 238 U(n, f). In order to examine this aspect, the yields of symmetric products, high yield asymmetric products and the peakto-valley (P/V) ratios from the present work and literature data [29–52] in 232 Th(n, f) are shown in table 3.

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$E_{\rm n}~({\rm MeV})$	E^* (MeV)	$Y_{\mathrm{a}}(\%)$	$Y_{ m s}(\%)$	P/V ratio	Ref.
1.60 ± 0.02	6.21	_	_	218.9 ± 47.7	[45]
1.68 ± 0.02	6.29	-	_	205.1 ± 42.1	[45]
1.72 ± 0.02	6.33	_	_	292.7 ± 73.2	[45]
1.77 ± 0.02	6.38	_	—	241.5 ± 58.8	[45]
1.88 ± 0.02	6.49	_	_	238.2 ± 36.5	[45]
2.00 ± 0.02	6.61	—	—	283.5 ± 64.9	[45]
2.00	6.61	8.950 ± 0.250	0.005 ± 0.001	_	[46]
2.20 ± 0.02	6.81	—	_	212.3 ± 53.9	[45]
2.43 ± 0.02	7.04	—	—	214.5 ± 35.6	[45]
2.96 ± 0.41	7.57	-	_	118.5 ± 17.5	[45]
2.97	7.58	_	_	122.0	[41]
3.00	7.61	8.600 ± 0.230	0.023 ± 0.004	_	[46]
3.00	7.61	7.890 ± 0.094	0.045 ± 0.009	_	[31]
3.10 ± 0.15	7.71	_	_	63.0 ± 11.0	[47]
4.00	8.61	8.010 ± 0.200	0.099 ± 0.015	80.9 ± 12.3	[46]
4.03 ± 0.02	8.64	_	_	71.0	[41]
4.20 ± 0.11	8.81	_	_	27.2 ± 3.1	[45]
4.81 ± 0.02	9.52	_	_	51.0	[41]
5.20 ± 0.25	9.87	_	_	29.0 ± 3.0	[47]
5.30 ± 0.11	9.91	_	_	26.4 ± 2.1	[45]
5.90	10.51	8.080 ± 0.530	0.270 ± 0.040	29.9 ± 4.8	[46]
6.35	10.96	8.358 ± 0.551	0.277 ± 0.028	30.3 ± 4.4	Present work
6.40	11.01	8.410 ± 0.630	0.230 ± 0.040	36.6 ± 6.9	[46]
6.90	11.51	8.700 ± 0.340	0.200 ± 0.030	43.5 ± 6.7	[46]
7.60	12.21	8.380 ± 0.230	0.200 ± 0.030	41.9 ± 6.4	[46]
8.00	12.61	7.870 ± 0.350	0.290 ± 0.030	27.1 ± 3.9	[46]
8.53	13.14	8.299 ± 0.579	0.487 ± 0.097	17.1 ± 3.6	Present work
9.10 ± 0.30	13.71	(8.000 ± 0.500)	0.436 ± 0.014	18.3 ± 1.3	[32]
10.09	14.70	7.579 ± 0.491	0.655 ± 0.123	11.6 ± 2.6	Present work
11.00	15.61	8.100 ± 0.900	0.760 ± 0.015	10.7 ± 1.3	[30]
13.40 ± 0.17	18.01	(8.000 ± 0.500)	1.440 ± 0.020	5.60 ± 0.36	[32]
14.10 ± 0.16	18.71	(7.500 ± 0.500)	1.340 ± 0.020	5.60 ± 0.38	[32]
14.70 ± 0.30	19.31	(7.500 ± 0.500)	1.580 ± 0.050	4.75 ± 0.51	[50]
14.70 ± 0.30	19.31	_	1.400 ± 0.050	5.36 ± 0.41	[50]
14.70 ± 0.30	19.31	_	1.310 ± 0.140	5.73 ± 0.72	[38]
14.70 ± 0.30	19.31	_	1.380 ± 0.120	5.43 ± 0.59	[48]
14.80 ± 0.80	19.41	6.690 ± 0.325	1.720 ± 0.500	3.89 ± 1.15	[31]
14.80 ± 0.80	19.41	_	1.500 ± 0.200	4.46 ± 0.65	[34]
14.80 ± 0.80	19.41	_	1.240 ± 0.200	5.40 ± 0.92	[33]
14.90 ± 0.25	19.51	(6.500 ± 0.500)	1.280 ± 0.040	5.10 ± 0.42	[32]
18.10 ± 0.25	22.71	(6.500 ± 0.500)	1.920 ± 0.100	3.40 ± 0.31	[32]

Table 3. Yields of asymmetric (Ya) and symmetric (Ys) products and the P/V ratio in the neutron-induced fission of ²³²Th.



Fig. 6. Plot of the peak-to-valley (P/V) ratio as a function of the excitation energy in the 232 Th(n, f) and 238 U(n, f) reactions. The data of present work in 232 Th(n, f) are in green with the same symbol as the literature data.

The yields of high-yield fission products and symmetric products from table 3 in 232 Th(n, f) and similar data from the literature [54–82] in 238 U(n, f) are plotted in fig. 6 as a function of excitation energy. It can be seen from fig. 6 that in both 232 Th(n, f) and 238 U(n, f), the yields of asymmetric products decrease slightly, whereas the yields of symmetric products increase significantly with excitation energy. It can be also seen from fig. 6 that the yields of symmetric products in ²³²Th(n, f) increase sharply up to excitation energy of 10 MeV and then slightly decrease up to 13 MeV and thereafter again increase. On the other hand, in 238 U(n, f), the increase of yields of symmetric products is nearly smooth with excitation energy. The different behavior of 232 Th(n, f) compared to 238 U(n, f) within excitation energy of $10-12.5 \,\mathrm{MeV}$ is due to the greater effect of giant dipole resonance (GDR) in the former than in the later. Similar effect within excitation energy of 10-13 MeV was also seen in the proton-induced fission of 232 Th [97], which supports the present observation. Otherwise, above excitation energy of 7.5 MeV, the yields of symmetric products are higher in 232 Th(n, f) than in 238 U(n, f) and increase with excitation energy. The role of excitation energy can be also from the peakto-valley (P/V) ratio. Thus from table 3, the P/V ratios in 232 Th(n, f) and similar data from the literature [54–82] in ${}^{238}U(n, f)$ are plotted in fig. 7 as a function of excitation energy. It can be seen from fig. 7 that in both 232 Th(n, f) and 238 U(n, f), the P/V decreases with excitation energy. However, the P/V ratio in ²³²Th(n, f) decreases up to 10 MeV and then slightly increases up to 13 MeV and thereafter again decrease, which is due to the GDR effect. It can also be seen from fig. 7 that at all excitation energy, the P/V ratio in ²³²Th(n, f) is lower than in ²³⁸U(n, f). This is due to the different type of potential energy surface in 233 Th^{*} compared to 239 U^{*} [93] as mentioned before besides the role of excitation energy.



Fig. 7. Plot of the average neutron number $(\langle v \rangle)$ as a function of the excitation energy in the ²³²Th(n, f) and ²³⁸U(n, f) reactions. The data of the present work in ²³²Th(n, f) are in green with the same symbol as the literature data.

5 Conclusions

- i) The yields of fission products in the 6.35, 8.53 and 10.09 MeV quasi-neutron-induced fission of 232 Th were determined by using an off-line γ -ray spectrometric technique. From the yields of various products mass-chain yields were obtained by using charge distribution corrections.
- ii) In the average neutron energy of 6.35, 8.53 and 10.09 MeV, the mass-yield distributions in the 232 Th(n, f) is triple-humped unlike in 238 U(n, f), where it is double-humped. This is due to the different type of potential energy surface in 233 Th^{*} than in 239 U^{*}.
- iii) The yields of fission products for A = 133-134, A = 138-140, and A = 143-144 and their complementary products are higher than those of other fission products for 232 Th(n, f) and 238 U(n, f) reactions. This is due to shell closure proximity based on standard I and II asymmetric mode of fission besides the role of even-odd effect.
- iv) In ²³²Th(n, f) and ²³⁸U(n, f), the $\langle v \rangle$ and $\langle A_L \rangle$ values increase and $\langle A_H \rangle$ values decrease with excitation energies. However, within the excitation energy of 10–13 MeV, the increasing trends of $\langle v \rangle$ and $\langle A_L \rangle$ and the decreasing trend of $\langle A_H \rangle$ with excitation energy is sharp in ²³²Th(n, f) reaction. This is due to the major GDR effect in the ²³²Th(n, f) reaction than in the ²³⁸U(n, f) reaction.
- v) In ²³²Th(n, f) and ²³⁸U(n, f), the yields of high-yield asymmetric products decrease marginally, whereas for symmetric products increase sharply with excitation energies. In ²³⁸U(n, f) reaction, the yields of symmetric fission products increase smoothly with excitation energy. However, in ²³²Th(n, f), the yields of symmetric

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products increase sharply within excitation energy of 10 MeV and then decreases up to 12.5 MeV and thereafter again increase with excitation energy.

vi) The P/V ratios in ²³²Th(n, f) and ²³⁸U(n, f) decrease with excitation energy. This shows the role of excitation energy. However, at all excitation energies, the P/V ratio in the ²³²Th(n, f) are lower than in the ²³⁸U(n, f), which is due to the different type of potential surface between two fissioning systems.

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