Mass distribution in 36.2 MeV alpha induced fission of $^{232}\text{Th}$

D. Banerjee$^{1,a}$, T. N. Nag$^2$, R. Tripathi$^2$, S. K. Wasim Raja$^1$, S. Sodaye$^2$, P. K. Pujari$^2$, A. Chakrabarti$^3$, M. Bhattacharjee$^1$, L. K. Doddi$^3$, V. Naik$^3$

$^1$ Radiochemistry Division (BARC), Variable Energy Cyclotron Centre, 1/AF, Bidhan Nagar, Kolkata 700064, India
$^2$ Radiochemistry Division, Bhabha Atomic Research Centre, Mumbai 400085, India
$^3$ Radioactive Ion Beam Group, Variable Energy Cyclotron Centre and HBNI-Kolkata, 1/AF, Bidhan Nagar, Kolkata 700064, India

Abstract

Mass distribution of fission products has been determined in $^\alpha+^{232}\text{Th}$ reaction at $E_{\text{lab}} = 36.2$ MeV using $\alpha$ particles from the cyclotron at the Variable Energy Cyclotron Centre (VECC), Kolkata. Yields of 64 fission products having half-lives in the range of about ~1 min to several days have been measured using gamma ray spectrometry. The mass distribution shows a clear triple humped structure indicating the contribution from both asymmetric and symmetric modes of fission. The experimental mass distribution was well reproduced by the calculation from the GEF code, which takes into account the multi-chance fission.

1 Introduction

Historically, nuclear fission has been described using a macroscopic approach, where the potential energy of a liquid drop is traced as a function of deformation of the nucleus undergoing fission [1–3]. However, the observed asymmetric mass distribution in the fission of actinides, especially in thermal neutron induced fission of $^{235}\text{U}$ at low excitation energies, could only be explained by incorporating the shell effects in the macroscopic liquid drop approach [4]. The spherical shell corresponding to $N = 82$ and deformed shell corresponding to $N = 88$ appear to play a major role in the asymmetric split. The shell effect is expected to play a role only at low excitation energies and one would expect a gradual washing out of the shell effect with increasing excitation energy [5–7], resulting in a gradual shift from asymmetric to a completely symmetric split. The asymmetric peak to symmetric valley ratio, 600, for thermal neutron induced fission changes to 6 when neutron energy is increased to 14 MeV. More studies are required to understand the role of shell effects in governing the mass distribution with variation in excitation energy of the compound nucleus. The compound nucleus, $^{236}\text{U}$, can be produced in $n + ^{235}\text{U}$ as well as in $\alpha + ^{232}\text{Th}$ reaction. Due to the availability of $\alpha$-particles of different energies from different types of accelerators, a number of studies on fission product mass distribution at higher excitation energies of the $^{236}\text{U}$ compound nucleus have been carried out by different groups using the $\alpha + ^{232}\text{Th}$ reaction [8–12]. In two of the earlier studies involving $\alpha + ^{232}\text{Th}$ reaction, the mass distributions were found to have three peaks/humps [8,9]. Roginski et al. [8] observed a triple humped mass distribution, corresponding to asymmetric and symmetric splitting for incident $\alpha$-particle energies of 33 and 39 MeV, whereas for incident energy of 22.9 MeV, a double humped asymmetric mass distribution was observed. Chakrabarti et al. [9] observed a three peaked (or triple humped) structure in the mass distribution over a wide range of $\alpha$-particle energy from 28.5 to 71.4 MeV. However, there are studies that observed either double humped mass distribution or completely symmetric mass distribution. For example, Guin et al. [10] reported double humped asymmetric mass distribution for the same system for incident $\alpha$-particle energy of 28.5 MeV, while Chaudhuri et al. [11] reported the complete washing out of shell effects beyond 40 MeV leading to a symmetric mass distribution. Also, a recent study carried out for a number of actinide fissioning systems close to $^{236}\text{U}$ (e.g. $^{237–240}\text{U}$, $^{239–242}\text{Np}$, etc.) over an excitation energy range of 10–60 MeV, shows pronounced asymmetric component up to 60 MeV of excitation energy [12]. Three peaked mass distributions were, however, consistently observed in the fission of $^{228}\text{Ra}$ at low excitation energies [13], which signifies the co-existence of multi-mode fission of the same fissioning nucleus. Only rather recently, a suitable explanation for this observation has been put forward by Moller et al. by describing the shape evolution of

---

*e-mail: dbanerjee@vecc.gov.in* (corresponding author)
the fissioning nucleus on a five dimensional potential energy surface [14]. Apart from elongation, neck diameter and mass asymmetry, the authors considered two more shape parameters, viz., the deformations of the left and right nascent fragments. With this prescription they were able to explain the observed two-mode fission in 228Ra. But their calculations do not predict bi-modal fission for uranium nuclei at similar excitation energy above the fission barrier. The probability of multi-chance fission [15] increases with the excitation energy of the compound nucleus. Depending upon the initial excitation energy, the evaporation of few neutrons would bring down the excitation energy of the fissioning system to such a level that shell effect becomes operational, leading to an asymmetric mass distribution. Multi-chance fission can, thus, explain the survival of asymmetric mass distribution at ∼60–70 MeV, as observed in earlier experimental studies [9,12].

In view of these new theoretical developments and experimental findings, the accurate determination of the mass distribution at comparatively higher excitation energies in the fission of actinides has gained renewed interest. Also, the mass distribution data at higher excitation energies is important for ADS development. Therefore, a fresh effort to accurately determine the mass distribution of actinide fissioning systems at different excitation energies is necessary. In view of the varying results on the nature of the mass distribution in α-induced fission of 232Th [8–12,16], it would be important to measure the mass distribution and compare it with the calculations based on the present theoretical understanding. The present study deals with the determination of the mass distribution in α-induced fission of 232Th for the incident α particle energy of 36.2 MeV, which corresponds to 31 MeV excitation energy of the compound nucleus 236U. The fission yields (FYs) of 64 fission products (FPs) were measured, which allowed a reliable determination of the mass distribution. The experimental mass distribution has been compared with the calculation of GEF code (version 2014/2.1) [17].

2 Experimental details

A self-supporting target of 232Th (thickness: 8mg/cm2) was irradiated with 40 MeV α beam from the K = 130 AVF Cyclotron at Variable Energy Cyclotron Centre (VECC), Kolkata, India. The projectiles entered into the cylindrical irradiation chamber (diameter: 65 mm, length along the beam direction: 35 mm) through a 25 µm thick Havar window and then passed through a 25 µm super pure aluminum foil placed upstream of the target before bombarding the target. The energy loss of the projectile in the Havar window, the aluminum foil, and the target was calculated using the software SRIM [18]. The energy of the α-beam was 36.2 MeV after traversing half the thickness of the 232Th target. The irradiations were performed for three different durations: 2 min, 10 min and 2 h which will be referred to as R-I, R-II and R-III respectively. All through the irradiation, the beam intensity was monitored at an interval of 10 s to determine the beam intensity during irradiation.

The reaction products recoiling out of the target were transported using He gas jet (1 atm. pressure) transport system at VECC [16]. The fission products were transported using a Tygon capillary tube with inner diameter of 1/16 inch and length of about 15 metre to a low background detection site and were implanted on a graphite catcher foil. The catcher foil was then counted with a pre-calibrated high purity germanium (HPGe) detector for a period of about 30 min for R-I, ∼2 h for R-II and for a few days for R-III. Different irradiation times helped in determining the yields of fission products with half-lives spanning over a very wide range. In order to account for any possible variations in the transport efficiency for different fission products, an irradiation for a duration of 4 h was carried out with a catcher foil placed immediately after the target in the same setup. The ratio of yields of fission products obtained from the direct measurement (without transport) to that obtained after the transport has been used to correct for the difference in transport of the fission products.

The data acquisition was performed by PC-based, software controlled PCI-bus multichannel analyzer FAST Com Tec MCA-3 [19]. The typical γ-ray spectra for 10 min and 2 h irradiations are shown in Figs. 1 and 2 respectively. For short irradiation, the spectrum has been obtained after a cooling time of 1 min and has an acquisition time of 120 s. The spectrum corresponding to the long irradiation was acquired for 3000 s after a cooling time of 5 h. The decay data of the FPs used in the present study was taken from the literature.
The yield of a mass chain $Y(A)$ is obtained from experimentally measured independent yield $IN(A, Z)$ or cumulative yield $CY(A, Z)$ of a fission product with mass number $A$ and atomic number $Z$, using the following equations:

\[
Y(A) = \frac{IN(A, Z)}{FIY(A, Z)} \tag{1}
\]

\[
Y(A) = \frac{CY(A, Z)}{FCY(A, Z)} \tag{2}
\]

where $FIY(A, Z)$ and $FCY(A, Z)$ are fractional independent and cumulative yields respectively and are calculated using the equations:

\[
FCY(A, Z) = \frac{1}{\sqrt{2\pi\sigma_Z^2}} \int_{-\infty}^{z+0.5} e^{-\frac{(z-z_p)^2}{2\sigma_Z^2}} \, dz \tag{3}
\]

\[
FIY(A, Z) = \frac{1}{\sqrt{2\pi\sigma_Z^2}} \int_{z-0.5}^{z+0.5} e^{-\frac{(z-z_p)^2}{2\sigma_Z^2}} \, dz. \tag{4}
\]

The most probable $Z$ ($Z_p$) for a given mass chain $A$ and the width of the isobaric yield distribution ($\sigma_z$) are the two parameters which are required to carry out the correction for the charge distribution to obtain the mass yield $Y(A)$ from the experimentally measured yields of the respective fission products [25]. In this work, $\sigma_z = 0.7$ has been used for the charge distribution correction as was used by Umezawa et al. [26] for the fission of different actinide nuclei including $^{236}$U in the similar excitation energy range. The $Z_p$ value for a mass chain with mass number $A$ was calculated as $A/\left[(A_{CN} - \nu_T)/Z_{CN}\right]$, where $A_{CN}$ and $Z_{CN}$ are the mass number and proton number of the compound nucleus (CN) respectively and $\nu_T$ is the average number of neutrons emitted per fission calculated using the prescription of Kozulin et al. [27].

In the present experiment, determination of the yields of the fission product involves transport through He gas jet system which can affect the fission product mass distribution due to the possible variation of transport efficiency with fission product mass. Also, the large thickness of the target may result in a variation in the relative emission probability of the fission products from the target. The details of the correction for these effects are as follows. First the relative emission probabilities of the fission products with respect to the central mass (115) were determined. In order to determine the relative emission probabilities, the ranges of a few selected fission products, viz., $^{78}$Ga, $^{88}$Br, $^{102}$Nb, $^{115}$Pd, $^{128}$Sb, $^{142}$La and $^{152}$Pm (covering the mass range of fission products measured in the present study) were calculated using the code SRIM [18]. In order to calculate the ranges of the fission products, their kinetic energies were calculated using the prescription given in Ref. [28]. The target was divided into 100 layers and the range of a given fission product was used to estimate the cut-off angle for every layer beyond which the fission product wouldn’t be emitted from the target. These cut-off angles for different fission products were used to estimate their relative emission probabilities for each layer. These values were added up and normalised with respect to the emission probability for the symmetric mass 115 to obtain the correction factor for the variation in the emission probability as a function of fission product mass. The correction factor (with respect to the symmetric mass) changes from 1.08 for mass 78 to 0.83 for mass 152.

A correction for the variation in the transport efficiency of fission products as a function of their mass was also carried out. During the present experiment, an irradiation was carried out for a period of 4 h with the catcher foil placed immediately after the target for the direct measurement of fission products. The activity of the fission products in this foil was measured after a cooling time of $\sim 5$ h. Yields of only comparatively longer-lived fission products were obtained from this run. In order to investigate the variation of transport efficiency for different fission products, a ratio of directly measured yields to the corresponding yields obtained after He gas jet transport
### Table 1

Decay data [20] and yields of fission products identified in the present study for $\alpha + ^{232}\text{Th}$ reaction at $\alpha$ particle energy of 36.2 MeV (see text for determination of % yields)

| S. no. | Nuclide | Half-life | $E_\gamma$ (keV) | % Yield | $FIY \ or \ FCY$ |
|--------|---------|-----------|-----------------|---------|-----------------|
| 1      | $^{84}\text{Se}$ | 3.26 min | 408.2           | 1.09±0.23 (C) | 0.772 |
| 2      | $^{89}\text{Rb}$ | 15.4 min | 1031.9          | 1.03±0.11(C) | 0.989 |
| 3      | $^{90}\text{mRb}$ | 4.3 min | 831.7           | 1.34±0.13 (C) | 0.958 |
| 4      | $^{90}\text{gRb}$ | 2.63 min | 831.7           | 3.95±0.62 (C) | 0.995 |
| 5      | $^{91}\text{Sr}$ | 9.63 h | 749.8           | 3.81±0.20 (C) | 0.980 |
| 6      | $^{92}\text{Sr}$ | 2.71 h | 1383.9          | 3.46±0.17 (C) | 0.933 |
| 7      | $^{93}\text{Sr}$ | 7.43 min | 590.2           | 3.79±0.46 (C) | 0.998 |
| 8      | $^{93}\text{Y}$ | 10.18 h | 266.9           | 3.79±0.39 (C) | 0.991 |
| 9      | $^{94}\text{Sr}$ | 1.255 min | 1427.7        | 2.59±0.23 (C) | 0.991 |
| 10     | $^{95}\text{Sr}$ | 9.63 h | 749.8           | 3.81±0.20 (C) | 0.980 |
| 11     | $^{95}\text{Y}$ | 10.3 min | 954             | 5.42±0.77 (C) | 0.966 |
| 12     | $^{95}\text{Zr}$ | 64.02 day | 756.7           | 4.03±0.54 (C) | 0.999 |
| 13     | $^{96}\text{Zr}$ | 16.744 h | 657.9           | 4.21±0.10 (C) | 0.985 |
| 14     | $^{99}\text{Mo}$ | 65.94 h | 140.5           | 4.21±0.10 (C) | 1.000 |
| 15     | $^{101}\text{Mo}$ | 14.61 min | 590.9           | 3.57±0.38 (C) | 0.997 |
| 16     | $^{103}\text{Ru}$ | 39.25 day | 497.1           | 3.14±0.12 (C) | 1.000 |
| 17     | $^{105}\text{Ru}$ | 4.44 h | 724.2           | 2.76±0.08 (C) | 1.000 |
| 18     | $^{105}\text{Rh}$ | 35.36 h | 318.9           | 2.93±0.10 (C) | 0.979 |
| 19     | $^{105}\text{Tc}$ | 7.6 min | 143.26          | 2.17±0.46 (C) | 0.929 |
| 20     | $^{106}\text{Tc}$ | 0.59 min | 270.1           | 1.55±0.46 (C) | 0.991 |
| 21     | $^{107}\text{Ru}$ | 3.75 min | 194.3           | 1.78±0.31 (C) | 0.991 |
| 22     | $^{107}\text{Rh}$ | 21.7 min | 302.8           | 1.84±0.15 (C) | 1.000 |
| 23     | $^{109}\text{Rh}$ | 1.35 min | 326.9           | 2.06±0.31 (C) | 0.966 |
| 24     | $^{111}\text{Ag}$ | 7.45 day | 342.1           | 2.32±0.26 (C) | 1.000 |
| 25     | $^{112}\text{Pd}$ | 21.05 h | 617.4           | 2.62±0.09 (C) | 0.993 |
| 26     | $^{113}\text{Ag}$ | 5.37 h | 298.6           | 2.42±0.11 (C) | 0.999 |
| 27     | $^{114}\text{Pd}$ | 2.42 min | 126.7           | 2.32±0.85 (C) | 0.910 |
| 28     | $^{115}\text{Ag}$ | 20 min | 229             | 2.23±0.23 (C) | 0.987 |
| 29     | $^{115}\text{Cd}$ | 2.23 day | 336.2           | 2.19±0.09 (C) | 1.000 |
| 30     | $^{117}\text{mCd}$ | 3.36 h | 552.9           | 1.10±0.05 (C) | 0.995 |
| 31     | $^{117}\text{Cd}$ | 2.49 h | 273.3           | 1.07±0.05 (C) | 0.925 |
| 32     | $^{119}\text{mCd}$ | 2.2 min | 1025            | 1.50±0.15 (C) | 0.992 |
| 33     | $^{123}\text{Sn}$ | 40.06 min | 160.32         | 2.28±0.26 (C) | 0.982 |
| 34     | $^{125}\text{Sn}$ | 9.52 min | 331.9           | 0.96±0.18 (C) | 0.836 |
| 35     | $^{126}\text{Sb}$ | 12.46 day | 666.3           | 0.56±0.04 (C) | 0.307 |
| 36     | $^{127}\text{Sn}$ | 4.13 min | 490.6           | 0.25±0.04 (C) | 0.444 |
| 37     | $^{127}\text{Sb}$ | 3.85 day | 685.7           | 2.49±0.09 (C) | 0.904 |
| 38     | $^{128}\text{Sn}$ | 59.07 min | 482.3           | 0.98±0.14 (C) | 0.242 |
| 39     | $^{128}\text{Sb}$ | 9.01 h | 754             | 1.53±0.09 (C) | 0.773 |
| 40     | $^{129}\text{mSb}$ | 17.7 min | 759.8           | 0.81±0.06 (I) | 0.575 |
| 41     | $^{129}\text{gSb}$ | 4.36 h | 914.96          | 1.79±0.16 (C) | 0.355 |
| 42     | $^{130}\text{mSb}$ | 6.3 min | 793.4           | 1.38±0.22 (C) | 0.355 |
| 43     | $^{130}\text{gSb}$ | 39.5 min | 793.4           | 0.74±0.02 (C) | 0.355 |
| 44     | $^{130}\text{mI}$ | 8.84 min | 536.1           | 1.33±0.38 (I) | 0.135 |
Table 1 continued

| S. no. | Nuclide | Half-life | $E_\gamma$ (keV) | % Yield | $\text{FIY}$ or $\text{FCY}$ |
|--------|---------|-----------|------------------|---------|-------------------|
| 45     | $^{131}\text{I}$ | 8.02 day  | 364.5            | 3.68±0.11 (C) | 0.975             |
| 46     | $^{133g}\text{I}$ | 20.8 h   | 529.9            | 3.12±0.10 (C) | 0.802             |
| 47     | $^{133m}\text{Te}$ | 55.4 min | 334.3            | 1.47±0.23 (C) | 0.254             |
| 48     | $^{134g}\text{I}$ | 52.5 min | 847              | 2.21±0.11 (C) | 0.613             |
| 49     | $^{134m}\text{I}$ | 3.52 min | 272.1            | 1.07±0.14 (I)  |                   |
| 50     | $^{138m}\text{Cs}$ | 2.91 min | 463              | 3.17±0.77 (I)  | 0.828             |
| 51     | $^{139}\text{Ba}$ | 83.06 min| 165.8            | 3.06±0.19 (C) | 0.967             |
| 52     | $^{140}\text{Ba}$ | 12.75 day | 537.3         | 4.07±0.28 (C) | 0.899             |
| 53     | $^{141}\text{Ce}$ | 32.5 day | 145.4            | 4.41±0.23 (C) | 1.000             |
| 54     | $^{141}\text{Ba}$ | 18.27 min| 190.3            | 2.89±0.21 (C) | 0.763             |
| 55     | $^{142}\text{Ba}$ | 10.6 min | 255.3            | 3.41±0.25 (C) | 0.562             |
| 56     | $^{142}\text{La}$ | 91.1 min | 641.2            | 3.56±0.26 (C) | 0.946             |
| 57     | $^{145}\text{Ce}$ | 3.01 min | 724.33           | 2.18±0.41 (C) | 0.915             |
| 58     | $^{146}\text{Ce}$ | 13.49 min| 316.7            | 2.18±0.27 (C) | 0.792             |
| 59     | $^{147}\text{Pr}$ | 13.4 min | 314.7            | 4.41±0.85 (C) | 0.956             |
| 60     | $^{148}\text{Pr}$ | 2.01 min | 697.5            | 3.72±0.85 (I)  | 0.493             |
| 61     | $^{149}\text{Pr}$ | 2.26 min | 138.5            | 1.69±0.34 (C) | 0.721             |
| 62     | $^{150}\text{Pm}$ | 2.68 h   | 333.9            | 0.18±0.03 (I)  | 0.068             |
| 63     | $^{151}\text{Nd}$ | 12.44 min| 116.8            | 0.46±0.09 (C) | 0.520             |
| 64     | $^{152}\text{Nd}$ | 11.4 min | 250.2            | 0.53±0.09 (C) | 0.639             |

The symbols ‘C’ and ‘I’ represent cumulative and independent yields respectively. Values in the last column are fractional independent yield ($\text{FIY}$) or fractional cumulative yield ($\text{FCY}$) calculated using the charge distribution parameters which were used to convert fission product yields into respective mass yields from run R-III was calculated. The ratio was again normalized with respect to the value expected for symmetric mass ($A = 115$) to obtain relative transport efficiency or correction factors. The relative transport efficiency showed a systematic increase with fission product mass and became nearly constant beyond $\sim 105$. Thus, due to the opposite dependence of the emission probability of the fission products from the target and their transport in the He gas jet on their mass, the overall transport is the highest in the symmetric region and decreases on either side of the symmetry as seen from the plot of the overall correction factor in Fig. 3. This variation was approximated as a Gaussian and was fitted with the centroid fixed at fission product mass of 115. The determination of absolute yields (cumulative/independent yields) requires the absolute transport efficiency of the He gas jet system. However, in order to avoid the uncertainties regarding the constancy of He-jet transport efficiency in between the different runs, fission product yields from different runs were normalized using the areas under the respective mass distribution curves to obtain relative yields. These scaled yields were further normalized to make the total fission yield (obtained as the area of the fitted curve to the final mass distribution, which will be discussed later) to be 200%. The relative yields in %, obtained after correcting for the variation in the transport, has been given in Table 1. The uncertainties quoted on the yields of the fission products include the propagated uncertainty on peak areas obtained from the multiple rounds of counting or their standard deviation, whichever was higher. For the fission products, whose yields were taken from two different runs, their difference is quoted as the uncertainty if it

![Fig. 3](image-url)
was higher than the uncertainty obtained from peak areas. In addition, the uncertainty due to the transport correction factor has also been included in the final uncertainty values given in Table 1. The fission product yields were corrected for the charge distribution to obtain the mass yields using Eqs. (1)–(4). The $Z_p$ values, calculated using the unchanged charge distribution (UCD) hypothesis, were corrected for charge polarization which results from the tendency of attaining minimum potential energy by the fragments during the fission process [29]. The $Z_p$ values obtained using UCD hypothesis were increased by an amount $\Delta Z_p = (A_f/2 - A)/x$, where $x$ was a variable parameter which was fixed by minimizing the chi square of the fit. The $\Delta Z_p$ values for the lowest and the highest masses, whose yields have been measured in the present studies, were +0.31 and −0.37 respectively. The last column in Table 1 gives the fractional independent yield (FIY) or fractional cumulative yield (FCY) which were calculated using the charge distribution correction parameters. Experimentally measured fission product yields were divided by their respective FIY or FCY values to obtain the corresponding mass chain yields. As seen from the table, these correction factors are close to unity for most of the fission products and, therefore, the overall nature of the mass distribution will not be affected within the uncertainty of the parameters used for the charge distribution correction. Mass yields obtained after the charge distribution correction are shown in Fig. 4a. As seen from the figure, the mass distribution is triple humped with a pronounced asymmetric component. In order to reduce the scattering in the data points, pronounced in some of the mass regions, and also to better constrain the fit to the experimental mass distribution, the following procedure was adapted. The complementary fragment mass to have the same yield was determined to be $(A_{CN} - \nu_T - A_f)$, where $A_{CN}$ is the compound nucleus mass, $\nu_T$ is the number of neutrons evaporated in the fission process, which was calculated using the prescription of Kozuline et al. [27] and $A_f$ is mass of the fission product whose yield has been measured. The complementary masses were rounded off to the nearest integer. For several masses, multiple yields were available which were obtained to attain the final mass yield value. The mass distribution, thus obtained including the complimentary mass yields, is shown in Fig. 4b. It can be seen from this figure that the inclusion of complementary yields significantly improves the quality of the mass distribution. The final mass distribution was fitted to a sum of three Gaussian functions, two for asymmetric fission and one for symmetric fission. The centroid of the symmetric fission was obtained as 115.0 ± 0.6, which is close to the value of 115.1, calculated using the $\nu_T$ value of 5.8, as obtained using the prescription of Kozuline et al. [27]. The asymmetric peaks are centered at 93.0 ± 0.7 and 136.9 ± 0.7 respectively. The present study clearly shows that the mass distribution has a three-peaked structure due to the contribution from both asymmetric and symmetric fission modes.

A comparison of the present mass distribution with those obtained in earlier studies on $\alpha + ^{232}$Th reaction [8] at close by excitation energies is shown in Fig. 5. Excitation energies of the compound nucleus $^{236}$U are also given in the figure. Based on PACE2 [30] calculations, the average angular momentum was about 10 $\hbar$ for all the fissioning systems and was not strongly dependent on neutron evaporation. The areas of the mass distributions taken from the literature were normalized with respect to the area of the mass distribution obtained in present study for comparison. It can be seen from the figure that the overall nature of the mass distribution obtained from the present study is similar to that obtained in Ref. [8] showing the dominant contribution from the asymmetric fission. A triple humped mass distribution was also reported at an excitation energy of 34.5 MeV in Ref. [9], though the number of data points was much less compared to that of the present study. Recently, Hirose et al. [12] measured mass distributions for a large number of actinide nuclei with wide range of excitation energies populated through transfer reactions.
Fig. 5 Comparison of mass distribution from the present work with those obtained in Ref. [8] for the $\alpha + ^{232}$Th reactions at close by excitation energies. The data from literature has been normalized with respect to the area of the mass distribution obtained in the present study for comparison.

In this study, the asymmetric peaks observed in the fission of $^{237}$U (very close to the present fissioning system) persist even up to the excitation energy of 50 MeV. Thus, the result of present study is in qualitative agreement with most of the earlier studies in literature.

In order to investigate further, the fission product mass yield distribution was calculated using GEF code [17]. In the GEF calculations, mass number, atomic number, excitation energy and root mean square angular momentum ($l_{\text{rms}}$) of the compound nucleus were supplied as inputs. The $l_{\text{rms}}$ value was calculated as $1\overline{1}$ $h$ from the spin distribution of the compound nucleus obtained from PACE2 [30] calculation. The post neutron emission mass distribution calculated using the GEF code is shown in Fig. 6 which also shows the individual contributions from the symmetric fission arising from super long mode and asymmetric fission components arising from standard I (corresponding to $N=82$) and II (corresponding to $N=88$) modes. It can be seen from the figure that the asymmetric fission is predicted as the dominant contribution in the GEF calculation too. Apparently, it is the contribution from multi-chance fission that results in a pronounced shell effect. The GEF code estimates the contribution from the first, second, third and fourth chance fission to be 30%, 34%, 31% and 5% respectively. The corresponding excitation energies of the fissioning nuclei were 31, 23, 16 and 8.5 MeV respectively. The different chance fission contributions were also estimated using the code PACE2 which predicted the contribution from first, second, third and fourth chance fission as 25%, 41%, 25% and 9% respectively with mean excitation energies of the fissioning systems as 31, 24, 17 and 9.6 MeV respectively. As predicted by the code PACE2, there was no significant change in the average angular momentum of the fissioning nucleus in different chance fission. Predictions by the two codes are in reasonable agreement, which suggests use of appropriate input parameters in the calculations. These calculations show that, about $\sim 78\%$ of fission occurs with the excitation energy of about $\sim 23$ MeV or less which can result in significant shell effects in the fission process. Thus, it is evident from both the experimental data and the theoretical calculations, that the shell effects are strongly pronounced in the fission of $^{236}$U, populated with initial excitation energy of 31 MeV, which result in a dominant contribution from the asymmetric fission.

A comparison of the experimental mass distribution (open triangles) with the calculated fission product yield distribution (open circles) is shown in Fig. 7. It can be seen from the figure that the experimental mass distribution is in reasonable agreement with the prediction of the GEF code. It can be seen from this figure that, in the fission product mass distribution calculated using the GEF code (denoted by open triangles), the shallow troughs around masses $\sim 108$ and $\sim 121$ have unequal depths. In order to investigate whether such a behavior results from the neutron evaporation from the primary fragments, the primary fragment yield distribution as calculated from the GEF code is also shown in Fig. 7. At this point, it is worth mentioning that the heavy and light mass wings in primary fragment mass distribution calculated using the GEF code (denoted by open triangles), the shallow troughs around masses $\sim 108$ and $\sim 121$ have unequal depths. In order to investigate whether such a behavior results from the neutron evaporation from the primary fragments, the primary fragment yield distribution as calculated from the GEF code is also shown in Fig. 7. At this point, it is worth mentioning that the heavy and light mass wings in primary fragment mass distribution calculated using the GEF code (denoted by open triangles), the shallow troughs around masses $\sim 98$ and $\sim 137$. This absence of mirror-symmetry can be attributed to the superposition of the multiple mass distributions arising from different fissioning.
nuclei due to the multi-chance fission as primary mass distributions arising from different chance fission were exactly mirror symmetric. However, the troughs on the either side of the central mass in the primary fragment mass distribution have equal depth. This observation suggests that the main reason for the unequal depth of the troughs observed in the calculated post neutron emission yield distribution is related to the varying neutron evaporation from primary fragments followed by superposition of symmetric and asymmetric fission components. In order to investigate this further, a plot of average neutron multiplicity as a function of primary fragment mass is shown in Fig. 8 which nicely shows a dip in the neutron emission curve around mass $\sim 132$ due to the shell effect. As seen from the figure, the maximum neutron evaporation from primary fragments around mass $\sim 120$ along with the rapid decrease in the neutron emission from heavier fragments would result in a larger depletion in the yields around the mass region 120 in the post neutron emission mass distribution arising from superposition of symmetric and asymmetric fission components. Neutron evaporation from primary fragments also appears to be responsible for the more pronounced peak like structure in the central mass region. Due to this, the fitting of the post neutron emission mass distribution, which is a convolution of symmetric and asymmetric fission components, yielded a significantly narrower symmetric component and underestimated the relative contribution from the symmetric fission. As GEF calculations separately give mass distributions for different fission modes as shown in Fig. 6, the contributions from symmetric and asymmetric fission were obtained by integrating these different mass distribution components. Based on the GEF calculations, the overall mass distribution contains $\sim 45\%$ contribution from symmetric fission and $\sim 55\%$ from asymmetric fission. The agreement between the calculated and the experimental post neutron emission mass distributions can be considered as a validation of the values obtained from GEF calculations. Thus, the contribution from the symmetric fission, as estimated from the present study, is much lower compared to the value of $\sim 80\%$ as observed in Ref. [11] at excitation energy of 32.7 MeV.

4 Conclusions

Measurement of the mass distribution has been carried out in $\alpha + ^{232}$Th reaction at $E_{\text{lab}} = 36.2$ MeV. The use of He gas jet transport and different irradiation times allowed the determination of yields of a large number (64) of fission products having half-lives spanning over a wide range from $\sim 1$ min to several days. This has allowed a comprehensive determination of the mass distribution. The mass distribution has been observed to be clearly triple humped with dominant contribution from asymmetric fission. This observation is consistent with some but not all of the earlier measurements carried out on the same system by various experimental groups. The experimental mass distribution has been observed to be in qualitative agreement with GEF calculations. The calculated mass distribution obtained from GEF code showed troughs with unequal depth on either side of the central mass, which could be correlated to the neutron evaporation from the primary fragments.

Acknowledgements The authors would like to thank Shri P. S. Chakraborty and operations team of K130 Cyclotron at VECC for their contribution in beam-delivery for the experiments.
Data Availability Statement  This manuscript has no associated data or the data will not be deposited. [Authors’ comment: All the relevant data including the representative gamma-ray spectra supporting the findings of the present study are given in the paper. Additional data such as gamma-ray spectra or other details can be provided by the corresponding author on request.]

References

1. L. Meitner, O.R. Frisch, Nature 143, 239 (1939)
2. N. Bohr, J.A. Wheeler, Phys. Rev. 56, 426 (1939)
3. S. Frankel, N. Metropolis, Phys. Rev. 72, 914 (1947)
4. V.M. Strutinsky, Nucl. Phys. A 95, 420 (1967)
5. A.S. Newton, Phys. Rev. 75, 209 (1949)
6. P.R. O’Connor, G.J. Seaborg, Phys. Rev. 74, 1189 (1948)
7. R.H. Goeckermann, I. Perlman, Phys. Rev. 76, 628 (1949)
8. T.C. Roginski, M.E. Davis, J.W. Cobble, Phys. Rev. C 4, 1361 (1971)
9. A. Chakrabarti, S.K. Saha, T. Mukhopadhyay, A. Bandyopadhyay, A. Roy, C. Bhattacharya, S.K. Basu, Z. Bikash Sinha, Phys. A 345, 401 (1993)
10. R. Guin, S.M. Sahakundu, S.B. Manohar, Satya Prakash, M.V. Ramaniah, Radiochimica Acta 48, 7 (1989)
11. A. Chaudhuri, T.K. Ghosh, K. Banerjee, S. Bhattacharya, C. Jhilam Sadhukhan, Bhattacharya S. Kundu, J.K. Meena, G. Mukherjee, R. Pandey, T.K. Rana, P. Roy, T. Roy, V. Srivastava, Phys. Rev. C 91, 044620 (2015)
12. K. Hirose et al., Phys. Rev. Lett. 119, 222501 (2017)
13. Y. Nagame et al., Radiochimica Acta 78, 3 (1997)
14. P. Möller et al., Nature 409, 785 (2001)
15. A. Turkevich, J.B. Niday, Phys. Rev. 84, 52 (1951)
16. A. Chakrabarti, D.P. Chowdhury, S. Gangadharan, J. Arunachalam, R.M. Iyer, Nucl. Inst. Methods A 263, 421 (1988)
17. K.H. Schmidt, B. Jurado, C. Amouroux, C. Schmitt, Nuclear Data Sheets 131, 107 (2016)
18. J.F. Ziegler, J.P. Biersack, TRIM code, SRIM-2013
19. https://www.fastcomtec.com
20. https://www.nndc.bnl.gov/. Accessed Aug 2016
21. P.K. Mukhopadhyay, in Proceedings of Symposium on Intelligent Nuclear Instrumentation, Mumbai, vol. 33 (2001), p. 307
22. P.K. Mukhopadhyay, INIS Repos. 33, 33001318 (2001)
23. R. Tripathi, S. Sodaye, K. Sudarshan, R. Guin, Phys. Rev. C 88, 024603 (2013)
24. R. Tripathi, S. Sodaye, K. Ramachandran, S.K. Sharma, P.K. Pujari, Int. J. Mod. Phys. E 27, 1850010 (2018)
25. T.N. Nag, R. Tripathi, S. Sodaye, K. Sudarshan, K. Ramachandran, B.K. Nayak, P.K. Pujari, Phys. Rev. C 96, 044608 (2017)
26. H. Umezawa, S. Baba, H. Baba, Nucl. Phys. A 160, 65 (1971)
27. E.M. Kozulin, A.Ya. Rusanov, G.N. Smirenkin, Phys. At. Nucl. 56, 166 (1993)
28. H.H. Rossner, J.R. Huizenga, W.U. Schröder, Phys. Rev. Lett. 53, 38 (1984)
29. W.J. Swiatecki, J. Phys. (Paris) 33, C5–45 (1972)
30. A. Gavron, Phys. Rev. C 21, 230 (1980)