PRAMANA
— journal of
physics

© Indian Academy of Sciences

Vol. 85, No. 2
August 2015
pp. 291–301

Fusion–fission dynamics studies using mass distribution as a probe

T K GHOSH[1,*], A CHAUDHURI[1], K BANERJEE[1], S BHATTACHARYA[1], C BHATTACHARYA[1], S KUNDU[1], G MUKHERJEE[1], R PANDEY[1], T K RANA[1], P ROY[1], T ROY[1], V SRIVASTAVA[1] and P BHATTACHARYA[2]
[1]Variable Energy Cyclotron Centre, 1/AF Bidhan Nagar, Kolkata 700 064, India
[2]Saha Institute of Nuclear Physics, 1/AF Bidhan Nagar, Kolkata 700 064, India
*Corresponding author. E-mail: tilak@vecc.gov.in

DOI: 10.1007/s12043-015-1043-3; ***e*Publication:** 2 August 2015

Abstract. Study of quasifission reaction mechanism and shell effects in compound nuclei has important implications on the synthesis of superheavy elements (SHE). Using the major accelerator facilities available in India, quasifission reaction mechanism and shell effects in compound nuclei were studied extensively. Fission fragment mass distribution was used as a probe. Two factors, viz., nuclear orientation and direction of mass flow of the initial dinuclear system after capture were seen to determine the extent of quasifission. From the measurement of fragment mass distribution in α-induced reaction on actinide targets, it was possible to constrain the excitation energy at which nuclear shell effect washed out.

Keywords. Fission dynamics; mass distribution; shell effects.

PACS No. 25.85

1. Introduction

One of the major areas of thrust in contemporary nuclear physics research is the synthesis and study of superheavy elements (SHE) [1]. From the liquid-drop model (LDM) of nucleus [2], it is known that elements beyond atomic number 104 cannot survive as the fission barrier vanishes. Shell effects in nuclei are responsible for the existence of SHE with $Z > 104$. Although LDM fission barrier vanishes for these SHEs, the addition of shell correction [3] with LDM energies alters the fission barrier appreciably so as to develop a large barrier that can increase the fission half-lives by several orders of magnitude. It is known that with the increase in excitation energy, nuclear shell effect decreases. One of the burning question is "what is the temperature for which nuclear shell effect washes out for actinide nuclei which are used as target nuclei for the production of SHE"?

While shell effects play crucial roles in the survival of heavy elements, the initial production of SHE requires fusion of a target and projectile nuclei. The target–projectile system must overcome the repulsive Coulomb potential and reach attractive nuclear potential trap. The dinucleus thus formed should equilibrate in all degrees of freedom (e.g., charge, energy, mass, shape, etc.) to form a compound nucleus (CN). The CN, produced in heavy-ion-induced reaction, is usually excited with a large angular momentum. This excited CN can either decay by light particle emission leaving behind an evaporation residue (ER) or undergo binary fission because of its high fissility.

However, in some of the cases, it is found that the initial target–projectile dinuclear system may not equilibrate in all degrees of freedom. Depending on the entrance-channel mass asymmetry, energy and angular momentum transfer, the dinuclear system separates out without equilibration in one or more degrees of freedom. These processes include quasifission [4], fast fission [5] and pre-equilibrium fission [6].

Quasifission which is a dynamical process, is a serious competitor to CN formation (and hence synthesis of SHE) in some target–projectile systems. It seems to have a very strong entrance-channel correlation. Thus, efforts are on to find a suitable target–projectile system which would give an optimum yield for the formation of SHE at reasonably low excitation energies. This is really important, because the projected cross-section of SHE is very small and any further loss of cross-section owing to entrance-channel effects is undesirable.

From the series of measurements, using the major accelerator facilities (Cyclotron at Kolkata, Pelletron at Delhi and Mumbai) in India, we report the factors that affect quasifission and fusion–fission dynamics. In particular, we study the role of the target and the projectile nuclei that influences the fusion of the two nuclei. From the measurement of fragment mass distribution, we could constrain the excitation energy at which nuclear shell effect washes out for an actinide nucleus.

2. Why is the production cross-section for superheavy elements small?

In a pioneering experiment of production of $^{293,294}117$, through the fusion of ^{48}Ca on ^{249}Bk required 70 days of beam time (intensity $\sim 50\ \mu$A) to produce one element [7]. The reported cross-section is less than a picobarn (pb). The main reason for the hindrance of fusion cross-section is attributed to quasifission. In a dinuclear model the evaporation residue cross-section may be given as

$$\sigma_{\text{ER}}(E_{\text{c.m.}}) = \sigma_{\text{capture}}(E_{\text{c.m.}}) \cdot P_{\text{CN}}(E_{\text{c.m.}}) \cdot P_{\text{survival}}(E_{\text{c.m.}}),$$

where σ_{capture} is the partial capture cross-section for the formation of dinucleus system in competition with other peripheral reactions like quasielastic processes. The partial capture cross-section for the formation of dinuclear system overcoming the Coulomb barrier with transition probability $\tau(E_{\text{c.m.}}, J)$ is given by

$$\begin{aligned} \sigma_{\text{capture}}(E_{\text{c.m.}}) &= \Sigma\sigma_{\text{capture}}(E_{\text{c.m.}}, J), \\ \sigma_{\text{capture}}(E_{\text{c.m.}}, J) &= \pi\lambda^2(2J+1)\tau(E_{\text{c.m.}}, J), \end{aligned} \qquad (1)$$

where P_{survival} is the survival probability of the ER that is determined by the competition between fission and neutron evaporation of the excited CN. P_{CN} is the probability of complete fusion after the capture stage in the dinuclear system. P_{CN} is heavily dependent on the extent of quasifission in the reaction. For the production of SHE, currently the challenge is to understand the factors that influence P_{CN}, or in other words the quasifission process.

3. Quasifission process

Fission fragment angular anisotropy measurements for heavy-ion-induced fission reactions showed considerable deviation from that predicted by the statistical saddle-point model (SSPM), particularly for systems which have high $Z_{\mathrm{T}}Z_{\mathrm{P}}$ (multiplication of target and projectile atomic numbers) values. These anomalously high angular anisotropies were attributed to the presence of non-compound nuclear processes, viz., fast fission, pre-equilibrium fission and quasifission.

Fast fission dominates at higher excitation energies where the angular momentum-dependent fission barrier vanishes [5]. At moderate excitation energies that are used for the synthesis of superheavy elements, the presence of fast fission can be ruled out.

The idea of pre-equilibrium fission was proposed by Ramamurthy and Kapoor [6]. They argued that a highly mass asymmetric system with mass asymmetry greater than a critical mass asymmetry (known as Businero–Gallone (BG) critical mass asymmetry (α_{BG})) would tend to amalgamate into a more asymmetric system, thus eventually forming a CN. On the other hand, for a symmetric target–projectile, the initial dinuclear system will undergo fission before the system is equilibrated in shape (or K, the projection of the total angular momentum on the symmetry axis of the fissioning nucleus). This results in smaller values in K_0^2 and thus larger angular anisotropies but does not affect the formation of the evaporation residue. Therefore, pre-equilibrium fission process does not hinder the production of SHE.

In case of quasifission, the composite system breaks before complete equilibration in mass degrees of freedom, hindering the production cross-section of the evaporation residue or heavy elements. It was found that almost all systems involving actinide targets show anomalous anisotropies irrespective of the entrance-channel mass asymmetry. This was attributed by Hinde *et al* [4] to orientation-dependent quasifission. It was argued that for a highly deformed actinide target nucleus quasifission is more probable for a projectile hitting the polar region of the target nuclei rather than the equatorial region. This is due to the latter configuration leading to a more compact dinuclear system and thus preferentially equilibrates in all degrees of freedom into a CN.

It is evident from the discussions that among the above-mentioned non-compound nuclear fission process (fast fission, pre-equilibrium fission and quasifission), it is the quasifission process that has direct impact on the production of superheavy elements. It is to be mentioned that, from the dynamical point of view, fusion–fission and quasifission are different.

For heavier target–projectile systems, merely overcoming the Coulomb barrier does not ensure the formation of a CN. Swiatecki [8], in his dynamic model, proposed an extra push energy for a target–projectile system to form a mononucleus and an extra extra push for this mononucleus to equilibrate in all degrees of freedom and form a CN. The requirement of this extra push to form CN is mainly seen for systems with $Z_{\mathrm{T}}Z_{\mathrm{P}} > 1600$. The non-availability of this extra energy (extra extra push) leads to quasifission.

4. Mass distribution as a probe to study quasifission

At least four experimental probes are used to explore the presence or absence of quasifission:

(1) Measurement of ER cross-section gives a clue about the presence/absence of quasifission in a fusion reaction. As quasifission hinders the ER cross-section, the measured ER cross-section will be less compared to the statistical model for reactions where quasifission is present. However, for fissile targets, where the ER cross-section is very small, measurements are very challenging.

(2) Fission fragment angular anisotropy, which is defined as the ratio of the fission yields at 0° (or 180°) to 90°, is another probe that is used to detect the presence or absence of quasifission. In quasifission reaction mass asymmetry degree of freedom may not be equilibrated. As per the statistical saddle-point model, angular anisotropy $A = 1 + \langle l^2 \rangle / 4K_0^2$, where $\langle l^2 \rangle$ is the second moment of the CN spin distribution. The variance of the K-distribution K_0^2 is given as $I_{\text{eff}} T / h^2$, where I_{eff} is the effective moment of inertia and T is the temperature of the nucleus at the saddle point. Mass asymmetry degree of freedom equilibrates more rapidly than shape or K-equilibration. Thus, the fact that K-equilibration may not occur in quasifission (implying smaller value of K_0^2) will result in a larger angular anisotropy compared to the statistical model calculation.

(3) Pre-scission neutron multiplicity is also considered as a useful probe for studying fission dynamics. As the time-scales of quasifission (saddle to scission time) and fusion–fission (pre-saddle time + saddle to scission time, typically 30×10^{-21}s) are different, the appearance of quasifission at near-barrier energies should also be reflected in pre-scission neutron multiplicity data.

(4) However, variance of mass distribution of fission fragments has been a quite successful probe for studying quasifission, particularly near the Coulomb barrier [9–11]. Statistical fusion–fission is assumed to be undergoing fission through a mass symmetric unconditional saddle. Thus, the mass distribution of fission fragments are typically peaked at symmetry and the variance of mass distribution is expected to behave as follows:

$$\sigma_m^2 = \left(\frac{\partial \sigma_{\text{m}}^2}{\partial T} \right)_{(L=0)} T + \left(\frac{\partial \sigma_{\text{m}}^2}{\partial L} \right)_{(T=0)} \langle L^2 \rangle,$$

where T is the temperature of the CN at the saddle point and $\langle L^2 \rangle$ is the mean square angular momentum. It is observed that sensitivity of variance to angular momentum is very weak. $(\partial \sigma_{\text{m}}^2 / \partial T)_{(L=0)}$ is related to the stiffness parameter k of the mass asymmetry degree of freedom.

Ideally, for pure fusion–fission reactions, the width of the mass distribution should be narrow and increase uniformly with temperature; an equivalent reaction containing an admixture of fusion–fission and quasifission should be wider. Therefore, if the proportion of the quasifission reaction increases with change in the excitation energy, there will be an anomalous increase in the width of mass distribution. Such an increase in the width of fragment mass distribution with a decrease in energy has been reported recently around the Coulomb barrier [10]. We have observed similar anomalous increases in the width of

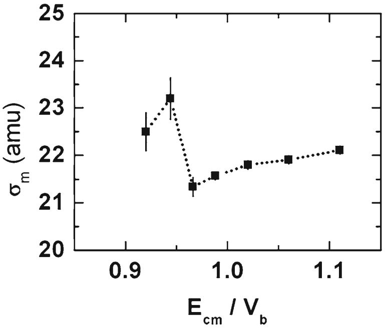

Figure 1. Variation of mass distribution (σ_m) as a function of E_{CM}/V_B for the ^{16}O $+^{238}U$ system. The dotted line is to guide the eye [14].

mass distribution with decrease in beam energy in fusion of systems with the deformed ^{232}Th target [12].

However, the identification of quasifission reaction mechanism is not always unambiguous using these experimental probes. In fact, measurement of ER cross-section and fragment angular anisotropy provided contradictory conclusions regarding the presence/absence of QF in the $^{16}O + ^{238}U$ reaction. The system is highly fissile and target is deformed and therefore is a probable candidate for quasifission at near-barrier energies. Anomalous increase of fission fragment angular anisotropy compared to statistical model has been observed for this system at near-barrier energies, which indicates a significant contribution from quasifission reaction. By assuming that the effect of quasifission is predominant in the sub-barrier region, where the orientation of the deformed target–projectile system is crucial to determine the fusion trajectory, Hinde *et al* [4] explained the anomalous energy dependence of fragment anisotropy for the $^{16}O + ^{238}U$ system and concluded that there is a quasifission reaction at sub-barrier energies. On the contrary, the cross-sections of the evaporation residues measured for the same system at the near and sub-barrier energies were reported [13] to be consistent with the statistical theory, indicating that the contribution from quasifission is not significant.

We measured the mass distributions of fission fragments and pre-scission neutron multiplicity for the $^{16}O +^{238}U$ system [14]. While the pre-scission neutron multiplicity was found to be consistent with the statistical model prediction, we found anomalous increase in the width of mass distribution near the Coulomb barrier energies (figure 1), signifying the presence of quasifission reaction. It is evident from the extensive study of the $^{16}O + ^{238}U$ system that mass and angular distributions of fission fragments are more sensitive probes than the to ER cross-section or neutron multiplicity measurement in concluding the presence/absence of quasifission in heavy-ion-induced fission reaction with highly fissile targets.

5. Measurement procedure

We have extensively investigated the fusion–fission and quasifission dynamics for a number of target–projectile systems. The experiments mainly involved measurements of mass distribution of fission fragments (FFMD) at near-Coulomb barrier energies. Masses were determined from the time-of-flight measurement of the fission fragments.

It is evident that accurate measurement of FFMD through time-of-flight or velocity distributions requires detectors which provide good timing and position information. Indigenously developed large area position-sensitive multiwire proportional counters (MWPC) [15] were used in the experiments. The detectors provide information on timing, position (X-Y information through delay line) and energy loss. The flight times for each event were measured through the fast anode pulse with respect to the pulsed beam. Two MWPCs were kept at folding angles. The target detector distance was decided by keeping in mind the objectives of the experiments. Even though keeping the distance close to the target increases the solid angle coverage and thus the count rate, it will deteriorate the time-of-flight (mass) resolution. The masses of the fission fragments were determined event-by-event by precise measurement of the time-of-flight difference of the complementary fission fragments. The typical mass resolution achieved was ∼3–6 amu units.

6. Role of target deformation on quasifission processes

The first set of our experiments involved a spherical ^{209}Bi target with ^{16}O beams [17]. In the second set of experiments we bombarded the same beams on a deformed ^{232}Th target [16]. The measurements led us to understand the role of target deformation on fusion–fission and quasifission processes.

When ^{16}O projectile was bombarded on spherical ^{209}Bi nuclei, it was seen that the variance of mass distributions varies smoothly against the excitation energy (figure 2). This is in agreement with the statistical model calculations. The fragment angular anisotropy is also consistent with SSPM [18,19]. Therefore, it was concluded that the spherical nuclei equilibrate in all degrees freedom to attain unconditional equilibrium forming a CN.

When a deformed target nucleus ^{232}Th was bombarded with ^{16}O at different excitation energies, significant deviations were seen near and below the Coulomb barrier energies. It was observed that for both the systems σ_{m}^2 decreases smoothly with decrease in energy at the above-barrier energies (figure 3). Near the barrier, however the variance of mass distribution increases. This anomalous increase has been attributed to the orientation-dependent quasifission that dominates at sub-barrier energies.

7. Role of entrance-channel on quasifission

To explore the role of entrance channel mass asymmetry on fusion–fission reaction cross-section, ^{246}Bk was populated using two pathways, viz., ^{11}B on ^{235}U and ^{14}N on ^{232}Th at the same excitation energies [22]. While the target deformations were similar for these systems (β_2 for $^{235}\mathrm{U} = 0.215$ and for $^{232}\mathrm{Th} = 0.207$), the entrance channel-mass asymmetry parameters ($\alpha = 0.911$ and 0.886, respectively) were on either side of the Businero–Gallone mass parameter ($\alpha_{\mathrm{BG}} = 0.893$).

As both the targets being actinides and are similarly deformed, it can be said that the probability of the initial dinucleus forming an elongated shape and undergoing

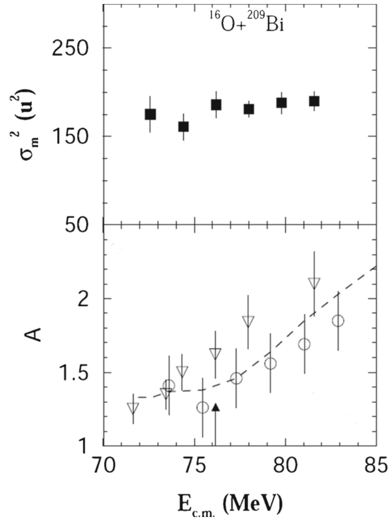

Figure 2. Variance of mass distributions (σ_{m}^2) as a function of beam energy for spherical ^{209}Bi target [17]. The arrow represents the Coulomb barrier. The solid lines show smooth variation of σ_{m}^2 with excitation energies. Fragment anisotropy A (○▽) and SSPM calculations (- - - - -) are also shown.

orientation-dependent quasifission are expected to be similar. Thus, any deviations of fission fragment mass distribution variances for different energies will be, to a large extent, due to the direction of mass flow in the initial dinucleus system. A system with mass

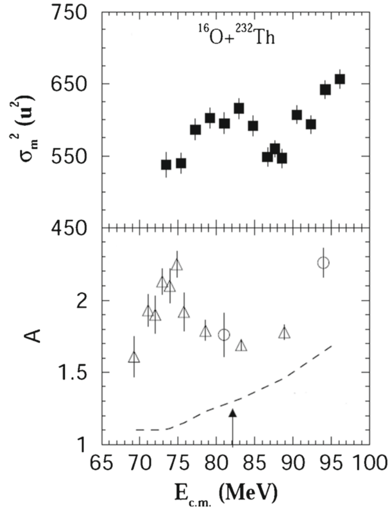

Figure 3. Variance (σ_{m}^2) as a function of beam energy for the deformed ^{232}Th target [12]. The arrow represents the Coulomb barrier. Fragment anisotropy A and SSPM predictions (- - - - -) are also shown [20,21].

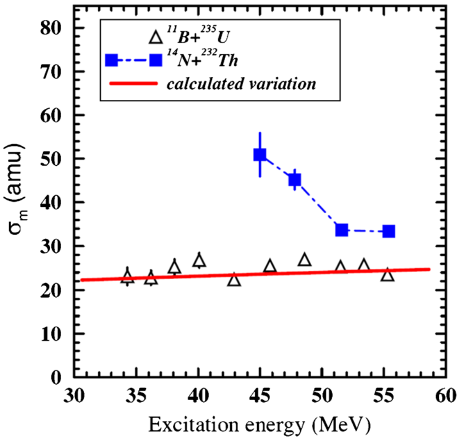

Figure 4. Variation in standard deviation ($\sigma_{\rm m}$) of mass distributions as a function of excitation energies [22]. The solid curve shows the expected $\sigma_{\rm m}$ from the theoretical calculations.

asymmetry α larger than the critical mass asymmetry $\alpha_{\rm BG}$ will experience mass flow from the projectile to the target in the initial dinuclei, thus establishing a mononuclear compact shape which facilitates equilibrium in all degrees of freedom. Thus, quasifission is not expected for such systems. On the other hand, for a more symmetric mass pair ($\alpha < \alpha_{\rm BG}$) the tendency of mass flow is towards the projectile creating a symmetric dinucleus before evolving into a mononucleus and fissioning through an asymmetric conditional saddle leading to quasifission.

The variation of standard deviation of mass distribution with excitation energies (figure 4) shows that for the more asymmetric system ^{11}B +^{235}U ($\alpha > \alpha_{\rm BG}$), the widths of the mass distribution increase linearly with increase in excitation energy, a trend indicative of fusion–fission reactions from an equilibrated compound nucleus. However, for the ^{14}N + ^{232}Th system, there is a sudden increase in the width of mass distribution near the Coulomb barrier energies. As the ground-state deformation of the two targets are similar, it can be concluded that the anomalous increase of the widths of mass distribution in the ^{14}N +^{232}Th reaction ($\alpha < \alpha_{\rm BG}$) near the Coulomb barrier is due to entrance-channel effects. In particular, the entrance-channel mass asymmetry was found to affect the fusion process sharply.

8. Manifestation of shell effects on fragment mass distribution

Fission fragment mass distribution for actinide nuclei at low excitation energies has been found to be predominantly asymmetric, with one peak at $A \sim 132$–140. Liquid-drop model (LDM) predicted the symmetric mass distribution of fission fragments but could not explain the asymmetric mass distribution for spontaneous (or low-energy) fission of actinide nuclei. The asymmetric shape of mass distribution has been attributed to the preference of the fragment nuclei mass towards doubly magic spherical ^{132}Sn nuclei. Shell effects in deformed nuclei show preference for a mean mass of about 140. Thus, the symmetric mass distribution, as predicted by the LDM, is suppressed due to shell effects in the fissioning heavy nuclei.

As a result of incorporating Strutinsky's shell correction energies to nuclear potential for heavy nuclei, the potential shows double-humped character as a function of deformation. Potential energy surface calculation shows [23] that the saddle point corresponding to the second barrier has an asymmetric shape for heavy nuclei. Thus, fission fragment mass distribution should be asymmetric if the fragments pass over the shell-corrected fission barrier.

It has been generally accepted that nuclear shell effects wash out at higher temperature. Ramamurthy *et al* [24] have shown that shell effects on nuclear level density parameter disappear at excitation energies close to 40 MeV for actinide nuclei. As the asymmetry in fission fragment mass distribution is due to shell effects, a change-over of the mass distribution from asymmetric to symmetric shape with increasing excitation energies would directly signify washing out of shell effects.

We measured the fission fragment mass distribution in α-induced reaction on a ^{232}Th target at the K-130 cyclotron facilities at Variable Energy Cyclotron Centre, Kolkata. The measurements were carried out at a wide excitation energy range with a close energy interval.

The experimental results indicate that the shape of the mass distributions changes gradually from symmetric to asymmetric as the excitation energy is lowered. FFMDs at four representative energies are shown in figure 5. At the lower excitation energy of 21 MeV (figure 5a), the mass distributions could be fitted by three Gaussians, one with peak position at the symmetric mass ($A \sim 118$) and the other two at around $A \sim 132$ and 100. In this fitting procedure, the widths of the distributions were varied but the intensities of the two asymmetric distributions (peaking at $A \sim 100$ and 132) were constrained to be equal to obtain a best fit (lowest χ^2) to the experimental mass distribution. The asymmetric and symmetric components are represented by dot–dashed and dashed line, respectively. The solid line is the overall fitting of the measured mass distribution. It was found that the

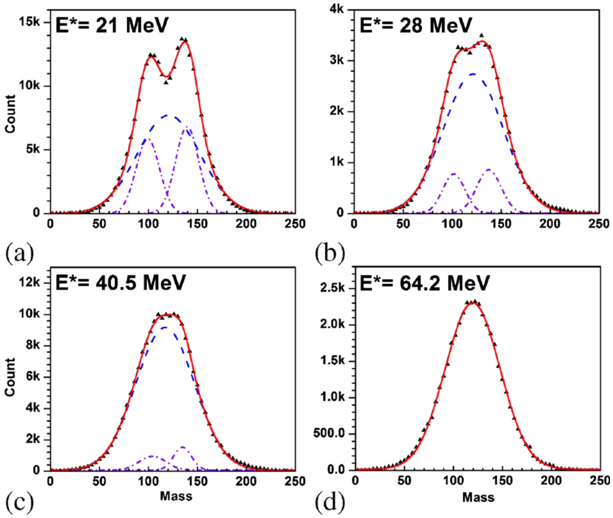

Figure 5. Mass distribution in fission of ^{236}U at different excitation energies.

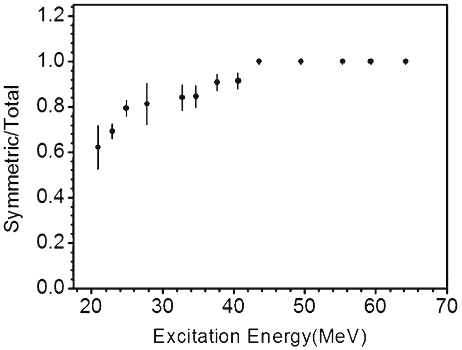

Figure 6. The variation of the ratio (relative unit) of the symmetric fission yield to the total fission yield at different excitation energies.

mass distributions with $E^* > 40$ MeV (figure 5d) are best fitted with a single Gaussian having peak position around the symmetric mass division.

The ratio of the area of the symmetric to the total area of the fitted distributions is plotted as a function of excitation energy in figure 6. It can be seen that the probability of mass symmetric fission enhances with excitation energies. The asymmetry in mass distribution arising at lower excitation energies is due to shell effects and the gradual suppression of this mass asymmetric mode with increasing excitation energy is a direct evidence of washing out of shell effects. The change-over occurs at ∼40 MeV excitation energy.

9. Summary

Fission fragment mass distributions have been extensively studied using a series of experiments to understand the effect of entrance-channel dynamics on the formation of compound nucleus and constrain the excitation energy at which the nuclear shell effects wash out. It was found that fission fragment mass distribution is a sensitive probe to study quasifission. The role of target deformation and entrance-channel mass asymmetry on the quasifission reaction mechanism was explored. It is observed that, in addition to the effect of deformation, the entrance-channel mass asymmetry was found to play a crucial role in the reaction mechanism, particularly in energies close to the Coulomb barrier. The measurements indicate that for actinide nuclei, nuclear shell effects wash out at ∼40 MeV excitation energy.

Acknowledgements

The authors are thankful to collaborators J K Meena, A Saha, J K Sahoo, R Mondal Saha, J Sadhukhan, Santanu Pal from VECC, Kolkata; S Chattopadhayay, N Majumdar, T Sinha and S Pal from Saha Institute of Nuclear Physics, Kolkata; D C Biswas, A Saxena, L M Pant and P K Sahu from Bhabha Atomic Research Centre, Mumbai; K S Golda, R P Singh, P Sugathan and S K Datta from Inter University Accelerator Centre, New Delhi. Thanks are also due the operators in IUAC-Delhi, BARC-TIFR-Mumbai Pelletrons and VECC-Kolkata Cyclotrons.

References

[1] S Hofmann and G Münzenberg, *Rev. Mod. Phys.* **72**, 733 (2000)
[2] N Bohr and J A Wheeler, *Phys. Rev.* **56**, 426 (1939)
[3] V M Strutinsky, *Nucl. Phys. A* **95**, 420 (1967)
[4] D J Hinde *et al*, *Phys. Rev. Lett.* **74**, 1295 (1995)
[5] B B Back *et al*, *Phys. Rev. Lett.* **46**, 1068 (1981)
[6] V S Ramamurthy and S S Kapoor, *Phys. Rev. Lett.* **54**, 178 (1985)
[7] Yu Ts Oganessian *et al*, *Phys. Rev. Lett.* **104**, 142502 (2010)
[8] W J Swiatecki, *Phys. Scr.* **24**, 113 (1981)
[9] M G Itkis and A Y Rusanov, *Phys. Part. Nucl.* **29**, 160 (1998)
[10] R Rafiei *et al*, *Phys. Rev. C* **77**, 024606 (2008)
[11] R G Thomas *et al*, *Phys. Rev. C* **77**, 034610 (2008)
[12] T K Ghosh *et al*, *Phys. Lett. B* **627**, 26 (2005)
[13] K Nishio *et al*, *Phys. Rev. Lett.* **93**, 162701 (2004)
[14] K Banerjee *et al*, *Phys. Rev. C* **83**, 024605 (2011)
[15] T K Ghosh *et al*, *Nucl. Instrum. Phys. Res. A* **540**, 285 (2005)
[16] T K Ghosh *et al*, *Phys. Rev. C* **69**, 031603 (2004)
[17] T K Ghosh *et al*, *Phys. Rev. C* **70**, 011604 (2004)
[18] E Vulgaris *et al*, *Phys. Rev. C* **33**, 2017 (1986)
[19] A M Samant *et al*, *Eur. Phys. J. A* **7**, 59 (2000)
[20] H Zhang *et al*, *Phys. Rev. C* **49**, 926 (1994)
[21] N Majumdar *et al*, *Phys. Rev. C* **51**, 3109 (1995)
[22] T K Ghosh *et al*, *Phys. Rev. C* **79**, 054607 (2009)
[23] P Moller *et al*, *Nature* **409**, 785 (2001)
[24] V S Ramamurthy, S S Kapoor and S K Kataria, *Phys. Rev. Lett.* **25**, 386 (1970)