Non-equilibrium processes in Heavy-ion induced Fission Reactions

R.K.Choudhury and R.G.Thomas
Nuclear Physics Division, Bhabha Atomic Research Centre, Trombay, Mumbai - 400085, India
E-mail: rkc@barc.gov.in

Abstract. Heavy ion induced reactions offer a wide range of possibilities in exploring the collective behavior of nuclei and nuclear processes. Recent years have seen a spurt in the experimental and theoretical studies to understand the mechanism of the fission process in heavy target projectile systems, arising from the interest to produce elements in the superheavy region. A number of new features are observed experimentally with respect to the kinetic energy, mass and angular distributions of fission fragments and their correlations, implying prevalence of non-equilibrium phenomena in the fission process in many target projectile systems. There are many theoretical attempts both in terms of static potential energy considerations and dynamics to understand the important degrees of freedom that govern the evolution of the nuclear system from initial interaction to the final fission stages. Depending on the degree and nature of equilibration, various processes such as fast-fission, quasi-fission and pre-equilibrium fission have been invoked that can compete with the fully equilibrated compound nuclear fission process. In what we call a C-T Fissility plot, we show the remarkable dependence of the onset of these non-equilibrium processes on the entrance channel parameters.

1. Introduction

Heavy ion induced reactions offer a wide range of possibilities in exploring the collective behavior of nuclei and nuclear processes [1]. The time evolution of the composite system formed after the initial interaction, and the parameters on which the dynamics depends are still not fully understood. A systematic study of these reactions can reveal information about the complex processes involved in the heavy ion collisions. Worldwide there are considerable efforts in the synthesis of heavy and superheavy elements using heavy ion collisions [2]. The success of synthesising heavier and heavier elements, therefore depends crucially on the accurate estimation of the non-equilibrium processes which take away the flux from the formation of superheavy evaporation residues.

A major challenge in superheavy element production is the presence of non-equilibrium processes such as quasifission [3, 4, 5, 6]. In terms of reaction time scales, quasifission lies between deep inelastic collisions (DIC) and Compound Nucleus (CN) formation. DIC reactions do not involve capture but represent the energy relaxation mode, exhibiting a wide spectrum of kinetic energy losses while mass distributions show little relaxation that peak around the projectile and target masses. On the other hand, CN formation and subsequent fission and/or ER formation is characterized by full equilibration in all degrees of freedom. Quasifission is a fission like process that proceeds without the formation of a compact mono-nuclear system and is characterized by total energy relaxation and mass division ranging from initial entrance
channel mass asymmetry and all the way to symmetry. This process by its very own nature is critically dependent on the entrance channel conditions. The probability of this process in turn determines the probability of formation of evaporation residues in heavy and superheavy systems.

2. Characteristics of equilibrium fission

Equilibrium fission is characterised by the formation of a compound nucleus (CN) which is equilibrated in all degrees of freedom. The mass, energy and angular distribution of fragments are expected to exhibit properties which are independent of the entrance channel [8].

2.1. Mass-TKE distributions

Figure 1. The Mass-TKE correlation plot for a system undergoing compound nucleus fission. The average kinetic energy shows a remarkable dependence on the charge and mass of the composite system and is independent of the entrance channel parameters. Figures taken from [16] and [15] respectively.

There have been extensive measurements on the mass and kinetic energy distribution of fission fragments in heavy ion induced reactions. Based on these systematics, the mass distributions from the fission of a compound nucleus formed at moderate excitation energies ($E^* = 30 - 60 MeV$) are well described by Gaussian peaking around symmetric mass division, and with the variance depending linearly on the temperature and the square of the angular momentum [9, 10, 11, 12, 13, 14].

\[
\sigma_A^2 = \mu T + \nu < \hat{l}^2 >
\]  

where $T$ is the transition state temperature and $l$ the angular momentum.

The average kinetic energy of the fragments is found to depend on the charge and mass of the compound nucleus and independent of the entrance channel properties. The Viola systematics gives remarkably close estimate of the experimentally obtained $<TKE>$ over the entire range of fissility [15]. As a typical example, Figure 1 shows the mass-$TKE$ correlations the fission fragments in the $^{238}U + ^{16}O$ reaction [16] (left). The $<TKE>$ of a number of systems along
with the Viola systematics [15] is plotted in the right part of Figure 1. Figure 2 shows the variance of the mass distribution as a function of excitation energy for different systems, along with the fit to the above expression. The mass distribution and the mass-angle correlation for \(^{16}O + ^{186}Os\) are shown in Figure 2(right), which demonstrate the general features observed for the compound nuclear fission reactions.

**Figure 2.** The dependence of the variance of the mass distribution on the temperature of the fissioning system. From [9]. The mass angle distribution showing no dependence of angle of emission and mass split in the case of compound nucleus fission. Figure from [17]

### 2.2. Angular distributions

**Figure 3.** Fission fragment angular distributions well described by the statistical model [8] in case of compound nucleus fission. Figure taken from [23].
For compound nucleus fission, the transition state model (TSM) assumes that statistical equilibrium is achieved at the saddle point transition state and the K degree of freedom (projection of total angular momentum onto the symmetry axis) of the system is a good quantum number during the fission process. The angular distribution of fragments is described by the rotor wave function (|$D_{M,K}^J(\theta)$|) and the angular anisotropy reduces to

$$A \simeq 1 + \frac{\langle \hat{\rho}^2 \rangle}{4K_0^2}$$

where, $K_0^2$ is the variance of the Gaussian K distribution peaked at K=0. It can also be shown that $K_0^2$ is proportional to the effective moment of inertia at the saddle point and the temperature at the saddle point [8]. The model, often known as statistical saddle point model (SSPM) successfully explains the angular distribution of a large number of systems at moderate excitation energies [8].

3. Characteristics of non-equilibrium fission (fission-like processes)

3.1. Strong mass angle correlations

The dynamical models proposed in early eighties predicted the onset of quasifission process for heavier systems, when the product $Z_PZ_T > 1600$ (where $Z_P$ and $Z_T$ are the atomic charges of the projectile and target, respectively) [4, 5, 6]. This has been experimentally verified in the mid eighties by a number of experimental groups [16, 18, 19] where strong mass angle correlations were observed for systems satisfying the above mentioned criterion. However, there are very recent measurements where strong mass angle correlations are also observed for systems with $Z_PZ_T < 1600$.

Figure 4. Mass angle correlation observed in the $^{48}Ti + ^{154}Sm$ reaction. Figure taken from [17].

3.2. Broader mass distributions

It has been observed in a number of cases that even when there is no apparent mass angle correlation, the variance of the mass distributions for a near symmetric entrance channel reaction is higher than that for more asymmetric entrance channel for the same composite system at same excitation energy and angular momentum [17, 20, 21]. The apparent broadening of mass distributions may be caused by the presence of a small fraction of non-equilibrated mass asymmetry degree of freedom.
3.3. Highly anisotropic angular distributions

In the mid-eighties Back et al. [9, 25, 26, 27] have reported the observation of anomalously large anisotropies in systems where the charge product is much less than 1600. These results indicated the occurrence of fission-like events which are occurring in a time scale which is comparable to or less than that of the characteristic $K$ equilibration time $\tau_K$. This non-equilibrium feature is ascribed to the presence of quasifission. However, for highly fissile systems when the temperature becomes comparable to the temperature of the CN, a new process called pre-equilibrium fission (PEF) has been also invoked to describe the angular distribution results [28].

4. Macroscopic variables defining dynamics

Extensive experimental investigations carried out by Toke et al. [16] Bock et al. [18] and Shen et al. [19] using U and Pb projectiles on a number of targets ranging from O to Cr in the mid eighties had brought out the dependence for the onset of quasifission on various entrance channel parameters. For completeness, in the following we describe them briefly. According to the macroscopic model of Swaitecki and others, [4, 5, 6], the dynamic evolution of the di-nuclear system after contact is principally governed by the multi dimensional potential energy surface (PES) of the combined system described by the three variables viz, the radial separation, the neck radius and the mass asymmetry of the fragments. While the choice of the variables may vary with models [7], the dynamics is depicted by the three milestone configurations viz, the contact configuration, the conditional saddle point and the un-conditional saddle point. The location of these points on the PES, and the injection point onto this multi dimensional surface are the governing factors for the subsequent evolution of the composite system. From the experimental observations in the eighties, Blocki et al. have parameterized the onset of quasifission using a macroscopic variable called mean fissility given by 

$$\chi_m = \frac{1}{3}\chi_{eff} + \frac{2}{3}\chi_{CN},$$

which incorporates the target-projectile and compound nucleus fissilities:

$$\chi_{eff} = (Z^2/A)_{eff}/(Z^2/A)_{crit}$$

with

$$(Z^2/A)_{eff} = 4Z_1Z_2/(A_1^{4/3}A_2^{1/3}(A_1^{1/3} + A_2^{1/3}))$$

while the compound nucleus fissility is

$$\chi_{CN} = (Z^2/A)/(Z^2/A)_{crit}$$

where

$$(Z^2/A)_{crit} = 50.883(1 - 1.7826I^2), I = (A - 2Z)/A$$

From the analysis of Blocki et al., it was found that $\chi_m = 0.72$ marks the onset of the need for extra push energy (excess energy over the Coulomb barrier) to achieve fusion. It was also noticed that the manifestation of the non-equilibrium nature of the re-separation process on the mass and spatial distributions of the fragments depends critically on the sticking time $\tau_s$ of the di-nuclear system compared to the characteristic equilibration time of various degrees of freedom. As an example, Figure 5 shows the evolution of the mass asymmetry degree with time for various systems. The characteristic equilibration time for mass asymmetry is seen to be $\tau_M = (5.3 \pm 1)10^{-21}$ s.

5. Non-equilibrium fission at different fissility regions

In the following we try to classify the observation of non-equilibrium fission based on two quantities, one describing the entrance channel ($\chi_{eff}$) and the other corresponding to the compound system ($\chi_{CN}$).
Figure 5. Characteristic equilibration time ($\tau_M$) of mass equilibration obtained by Shen et al. [19]. Figure taken from [19].

5.1. The C-T Fissility Plot

Figure 6 shows the plot of entrance channel fissility ($\chi_{\text{eff}}$) vs the compound nucleus fissility ($\chi_{\text{CN}}$) (referred to as C-T fissility plot). Different targets are characterized by arcs whereas projectiles are described by lines cutting the arcs for various target projectile combinations. $Z_1Z_2 \geq 1600$ (thick orange line) and $\chi_m = 0.72$ (solid blue line) are also shown in the plot. The onset of non equilibrium fission can be very well classified into different regions in this plot. The locus of $\alpha = \alpha_{\text{BG}}$ for different composite systems are shown by the thick black curve. $\alpha_{\text{BG}}$ is calculated by the following equation [24],

$$\alpha_{\text{BG}} = p \sqrt{x - x_{\text{BG}}} / (x - x_{\text{BG}}) + q$$ \hspace{1cm} (7)

where $x_{\text{BG}} = 0.396, p = 1.12$ and $q = 0.24$.

For this calculation, following Blocki et al. [6], we have taken the charge to mass ratio of target and projectile to lie along the valley of beta stability as approximated by Green’s formula,

$$Z/A = \frac{1}{2}[1 - 0.4A/(200 + A)]$$ \hspace{1cm} (8)

For a given pair of nuclei with mass numbers $A_1$ and $A_2$, we first calculate the corresponding atomic numbers according to the above equation and the sum of these numbers is the total atomic number $Z$, which is then divided between the two fragments in the ratio of their masses, viz. $Z_i = A_i(Z/A)$.

We also show on the right hand side of Figure 6, the Coulomb barrier obtained using the parametrization following Swaitecki [29],

$$V_0 = 0.85247z + 0.001361z^2 - 0.00000223z^3 \text{MeV}.$$ \hspace{1cm} (9)

where the Coulomb parameter, $z$ is given by $z = Z_1Z_2/(A_1^{1/3} + A_2^{1/3})$. On the top along the x axis, the liquid drop fission barrier is also shown for the corresponding fissility [30].
5.2. Less fissile systems ($\chi \sim 0.7 - 0.8$): Evidence of fission before mass and K equilibration for $l > l_{B_f=0}$

It was predicted in the early eighties that when the angular momentum $l$ of the system exceeds the angular momentum at which fission barrier vanishes ($l_{B_f=0}$), the di-nuclear system re-separates prematurely giving rise to fission like fragments [31]. This non-equilibrium process known as fast fission (fission occurring at a much shorter time scale) is characterized by a sudden broadening of mass widths (Figure 7) and highly anisotropic angular distributions [9]. This phenomenon, occurring due to the vanishing fission barrier is different from quasifission process, predicted by Swaitecki, where the dynamical trajectories end up outside the unconditional saddle point even for zero angular momentum.

While fast fission is predominant at very high energies, quasifission is predicted to occur when the mean fissility, $\chi_m$, exceeds 0.72. As can be seen in the C-T plot, in this fissility range, the systems above the $\alpha = \alpha_{BC}$ line are expected to evolve to a di-nuclear complex and a re-separation before reaching the compound nucleus may be probable. However, it must be noted that the fission barrier is considerably larger than the temperature ($T \sim 1$MeV) in this region, thereby reducing the probability of PEF considerably. Nevertheless, evidence of non-equilibration is reported in a number of systems, e.g. Mg, Si, S + W [20, 21, 22] in this region though they lie way below the $\chi_m = 0.72$ line (blue solid line). The two factors that

![C-T Fissility Plot](image_url)
Figure 7. Sudden rise in the widths of the mass distributions when $l$ exceeds $l_{B_f=0}$ clearly indicating the presence of fast fission. Figure taken from [9].

Figure 8. Strong mass angle correlations observed in less fissile systems above barrier (a) and below barrier (b) for the $^{48}$Ti+$^{170}$Er reaction.

are facilitating early re-separation may be the elongated contact configuration [17, 21] and the lowering of fission barrier at higher angular momentum. The mass distribution is symmetric for both CN and quasifission reactions.

5.3. Fissile systems (trans-actinide region): Fission before $K$ equilibration

As we move to the fissile systems where the fissility $\chi \sim 0.8-0.9$, anomalously large anisotropies were reported in several cases were actinide targets were involved [32, 33, 34, 35, 36] even for systems that lie way below the red and blue lines (right-bottom region of the C-T plot). However, no mass angle correlation or anomalously large variances were observed in these systems [16]. This points to the fact that the sticking time of the di-nuclear complex in this region is considerably larger that the characteristic rotation time and mass equilibration time, $\tau_M$. Anomalously large anisotropies observed in very asymmetric systems like $B, C + Th, B, C + Th$ [36], can only be understood if we assume a fraction of events re-separating before the characteristic $K$ equilibration time $\tau_K \sim 10^{-20}$s, a non-equilibrium process known as pre-
Figure 9. Anomalously large anisotropies irrespective of the entrance channel mass asymmetry in the trans-actinide region showing evidence of fission before K equilibration. Figure taken from [36].

equilibrium fission (PEF). This also implies the evolution towards a di-nuclear system, due to an apparent shift of the Businaro-Gallone point towards larger mass asymmetries due to the elongated touching configuration (large static deformation of actinide targets in this region indicated by the green thick dashed line in Figure 6.). It should also be noted that in this region the temperature (∼ 1 MeV) becomes comparable to the fission barrier and a substantial portion of the flux can diffuse over the barrier before reaching the compound nucleus phase. This is schematically illustrated in Figure 10 [37].

Figure 10. Figure illustrating the shift of the Businaro-Gallone mass barrier towards higher mass asymmetries for elongated contact configurations and the consequent occurrence of PEF even in very asymmetric systems at energies close to the Coulomb barrier.

Once the entrance channel is more symmetric beyond the red line, evidences are reported for strong mass angle correlation as well as the emergence of an asymmetric component particularly at sub-barrier energies [16, 38, 39] indicating a much shorter sticking time for complexes in this region. Figure 11 shows the mass angle correlation for $^{32}S + ^{232}Th$ system where the asymmetric component is clearly visible at sub-barrier energies [38].
The top-right hand side region of the C-T Fissility plot indicates the region of high fissility (also the superheavy region). It is quite evident that the region is heavily dominated by quasifission (above the thick red line). Several experimental studies have revealed the strong mass angle correlations and also the asymmetric nature of mass distribution in this region [38, 39, 40]. It is also interesting to note that the average kinetic energy of fission-like fragments produced in the $^{32}S+^{232}Th$ reaction shows consistently less value compared to the Viola systematics value indicating a much larger elongation at re-separation, than what is expected from the standard scission configuration for this fissility.

5.4. Highly fissile systems (superheavy region)

The top-right hand side region of the C-T Fissility plot indicates the region of high fissility (also the superheavy region). It is quite evident that the region is heavily dominated by quasifission (above the thick red line). Several experimental studies have revealed the strong mass angle correlations and also the asymmetric nature of mass distribution in this region [38, 39, 40]. It is also interesting to note that the average kinetic energy of fission-like fragments produced in the $^{32}S+^{232}Th$ reaction shows consistently less value compared to the Viola systematics value indicating a much larger elongation at re-separation, than what is expected from the standard scission configuration for this fissility.

**Figure 11.** Strong mass angle correlation observed in case of highly fissile systems ($^{32}S+^{232}Th$) [38]. Notice the appearance of the asymmetric component at sub-barrier energies. Figure taken from [38].

**Figure 12.** Highly asymmetric mass distributions observed in the superheavy region. Figure taken from [40].
6. Summary

It is quite apparent that the experimental observables in quasission reactions are found to crucially depend on the sticking time of the dinuclear complex with respect to the characteristic relaxation time of various degrees of freedom, before it decays into binary fission-like fragments. This interaction time in turn depends on the entrance channel parameters of the colliding nuclei. Thus a system that remains together for a time comparable or shorter than $\tau_M$ exhibits strong mass-angle correlations, large mass widths, and large anisotropies (the top-right region in C-T plot). The central region in the plot is dominated by systems whose sticking time is between $\tau_M$ and $\tau_K$ that may not show any significant correlation of mass with angle but can show broadened mass distributions and large angular anisotropies. The bottom-right of the plot indicates systems that stick together for time scales comparable to $\tau_K$ but re-separate before reaching a compound nucleus. These systems may not exhibit any signs of incomplete relaxation of mass but can still exhibit angular anisotropies larger than predicted by the TSM for compound nucleus fission.

It is evident that non-equilibrium fission is present in many more systems than that predicted by earlier dynamical models. True compound nucleus fission may be confined only to a small region bounded by the thick black line ($\alpha = \alpha_{BG}$) and the thick dashed green line (speculative) as shown in Figure 6.

Acknowledgments

The authors acknowledge Mr. Chandrabhan Yadav for his contribution in preparing the C-T Fissility plot.

References

[1] Yuri. Ts. Oganessian and Yuri. A. Lazarev, Heavy-ions and Nuclear Fission, Treatise in heavy-ion science, edited by D. A. Bromly, Vol. 4.
[2] Peter Armbuster, C. R. Physique 4 (2003) 571594.
[3] Peter Moller and Arnold J. Sierk, Nature 422, 485 (2003).
[4] W. J. Swiatecki, Phys. Scr. 24, 113 (1981).
[5] S. Bjornholm and W. J. Swiatecki, Nucl. Phys. A 391, 471 (1982).
[6] J. P. Blocki et al., Nucl. Phys. A 459, 145 (1986).
[7] P. Moller et al., Nature 409, 785 (2001).
[8] R. Vandenbosc and J. R. Huizenga, Nuclear Fission (Academic, New York, 1973).
[9] B. B. Back et al., Phys. Rev. C 53, 1734 (1996).
[10] R. K. Choudhury et al., Phys. Rev. C60, 054609 (1999).
[11] M. G. Itkis and A. Ya. Rusanov, Phys. Part. Nuclei 29, 160 (1998).
[12] G. D. Adeev, I. I. Gontchar, V. V. Pashkevich, N. I. Pischasov, O. I. Serdyuk, Phys. Part. Nuclei 19, 1229 (1988).
[13] E. G. Ryabov, A. V. Karpov and G. D. Adeev, Nucl. Phys. A765, 39 (2006).
[14] G. N. Knyazheva, E. M. Kozulin, R. N. Sagaidak, A. Ya. Chizhov, M. G. Itkis, N. A. Kondratiev, V. M. Voskresensky, A. M. Stefanini, B. R. Behera, L. Corradi, E. Fieretto, A. Gadea, A. Latina, S. Szilner, M. Trotta, S. Bhogini, G. Montagnoli, F. Scarlassara, F. Hass, N. Rowley, P. R. S. Gomes, and A. Szanto de Toledo Phys. Rev. C75, 064602 (2007).
[15] V. E. Viola, K. Kwiatkowski, and M. Walker, Phys Rev. C 31, 1550 (1985).
[16] J. Toke et al., Nucl. Phys. A440, 327 (1985).
[17] R. Rafiei, R. G. Thomas, D. J. Hinde, M. Dasgupta, C. R. Morton, L. R. Gasques, M. L. Brown, and M. D. Rodriguez, Phys. Rev C 77, 024606 (2008).
[18] R. Bock et al., Nucl. Phys. A 388, 334 (1982).
[19] W. Q. Shen et al., Phys. Rev. C 36, 115 (1987).
[20] A. C. Berriman, D. J. Hinde, M. Dasgupta, C. R. Morton, R. D. Butt, and J. O. Newton, Nature (London) 413, 144 (2001).
[21] R. G. Thomas, D. J. Hinde, D. Duniec, F. Zenke, M. Dasgupta, M. L. Brown, M. Evers, L. R. Gasques, M. D. Rodriguez, and A. Diaz-Torres, Phys. Rev C 77, 034610 (2008).
[22] E. Prasad et al., Phys. Rev. C 81, 054608 (2010).
[23] C. R. Morton et al., Phys. Rev. C 52, 243 (1995).
[24] M. Abe, KEK Report No. 86-26, KEK TH-28 (1986).
[25] B. B. Back, R. R. Betts, K. Cassidy, B. G. Glagola, J. E. Gindler, L. E. Glendenin, and B. D. Wilkins, Phys. Rev. Lett 50, 818 (1983).
[26] M. B. Tsang et al., Phys. Lett. B 129, 18 (1983).
[27] B. B. Back, R. R. Betts, J. E. Gindler, B. D. Wilkins, S. Saini, M. B. Tsang, C. K. Gelbke, W. G. Lynch, M. A. McMahan, and P. A. Baisden, Phys. Rev. C 32, 195 (1985).
[28] V. S. Ramamurthy and S. S. Kapoor, Phys. Rev. Lett. 54, 178 (1985).
[29] W. J. Swaitecki et al. Phys. Rev. C 71, 014602 (2005)
[30] A. J. Sierk, Phys. Rev. C33, 014602 (2005).
[31] C. Ngo et al. Nucl. Phys. A 400, 259c (1983).
[32] D. J. Hinde et al., Phys. Rev. Lett 74, 1295 (1995).
[33] D. J. Hinde, M. Dasgupta, J. R. Leigh, J. C. Mein, C. R. Morton, J. O. Newton, and H. Timmers, Phys. Rev. C 53, 1290 (1996).
[34] Z. Liu et al., Phys. Lett B 353, 173 (1995).
[35] J. C. Mein, D. J. Hinde, M. Dasgupta, J. R. Leigh, J. O. Newton, and H. Timmers, Phys. Rev. C 55, 1295 (1997).
[36] J. P. Lestone et al., J. Phys. G. 23, 1349 (1997)
[37] R. G. Thomas, R. K. Choudhury, A. K. Mohanty, A. Saxena, and S. S. Kapoor, Phys. Rev. C 67, 041601(R) (2003).
[38] D. J. Hinde et al., Phys. Rev. Lett 101, 092701 (2008).
[39] K. Nisho et al., Phys. Rev. C 82, 44604 (2010).
[40] R. G. Thomas et al., Phys. Rev. C 75, 24604 (2007).