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Overview of research by the fission group in Trombay

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Abstract. Nuclear fission studies in Trombay began nearly six decades ago, with the commissioning of the APSARA research reactor. Early experimental work was based on mass, kinetic energy distributions, neutron and X-ray emission in thermal neutron fission of ^{235}U, which were carried out with indigenously developed detectors and electronics instrumentation. With the commissioning of CIRUS reactor and the availability of higher neutron flux, advanced experiments were carried out on ternary fission, pre-scission neutron emission, fragment charge distributions, quarternary fission, etc. In the late eighties, heavy-ion beams from the pelletron-based medium energy heavy-ion accelerator were available, which provided a rich variety of possibilities in nuclear fission studies. Pioneering work on fragment angular distributions, fission time-scales, transfer-induced fission, γ-ray multiplicities and mass–energy correlations were carried out, providing important information on the dynamics of the fission process. More recently, work on fission fragment γ-ray spectroscopy has been initiated, to understand the nuclear structure aspects of the neutron-rich fission fragment nuclei. There have also been parallel efforts to carry out theoretical studies in the areas of shell effects, superheavy nuclei, fusion–fission dynamics, fragment angular distributions, etc. to complement the experimental studies. This paper will provide a glimpse of the work carried out by the fission group at Trombay in the above-mentioned topics.

Keywords. Neutron and heavy-ion-induced fission; experimental observables; superheavy nuclei.

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1. Introduction

Research on nuclear fission in Trombay began nearly 60 years ago as part of the nuclear energy programme in the country. There were parallel programmes in both the physics and chemistry aspects of actinide nuclei and the fission process. These groups were established by Drs Raja Ramanna and H D Sharma, respectively. Over the years, many members have contributed to these research programmes, and also the synergy in the two fields have progressed in a harmonious manner, intensifying the fission research in a significant manner. Presently, nuclear fission research in Trombay can claim to have international recognition with the pioneering work carried out in many frontier areas in this field. Early research up to the mid-eighties were based mainly on thermal neutron-induced fission of the fissile nuclei. The neutron beams were provided by the APSARA and CIRUS reactors. Some important studies were also carried out using fast neutrons produced by the Van-de-Graaff accelerators. During the last three decades, emphasis has been given to heavy-ion-induced fission of a large range of nuclei using the beams from the pelletron accelerators at Mumbai and Delhi. Nuclear fission process has been envisioned to proceed through various stages of overcoming a fission barrier. Many observed features have been explained by the macroscopic–microscopic description comprising the liquid-drop and shell models. The three stages in the fission process are: the pre-saddle, saddle-to-scission and post-scission phases [1]. There has been significant progress in understanding these various stages both theoretically and experimentally. However, the study of fission process has been predominantly through empirical and phenomenological methods. The experimental studies can be classified into the following observables, which provide information to a certain extent on the above three stages:

(1) Mass, kinetic energy of fission fragments
(2) Angular distribution
(3) Neutron, gamma and charged particle emission
(4) Angular momentum of fission fragments
(5) Spectroscopy, level scheme of fragments
(6) Mass, kinetic energy correlations
(7) Mass, angle correlations
(8) Fission cross-section and excitation function
(9) Ternary fission/light charged particle (LCP) accompanied fission

In this overview, it will not be possible to do enough justice to all the different types of contributions made by the various members of the group. However, a selective glimpse of the important results obtained over the years by the fission physics group of Trombay will be provided in the following sections.

2. Early research using neutron beams

Experimental work in the early 1950s and 1960s were carried out using the neutron beams from the APSARA reactor. The highlights of these studies are the information obtained on neutron and γ-ray emissions in the fission process. For the first time, it was shown from fragment-neutron angular correlation measurements that a small fraction ($\sim$10%) of neutrons is emitted in the pre-scission stage, corresponding to isotropic emission in the CM frame of the fissioning nucleus [2]. The experimental set-up used in the neutron measurement is shown in figure 1.

The γ-ray measurements provided the evidence for large angular momentum generation in fission fragments during the fission process [3]. These results have important implications on the saddle-to-scission stages of the fissioning nucleus. Further detailed studies on the neutron emission in fission were carried out using the larger flux available from the CIRUS reactor. The workhorse of the various neutron-based experiments was the back-to-back gridded ionization chamber shown in figure 2. Using this set-up in

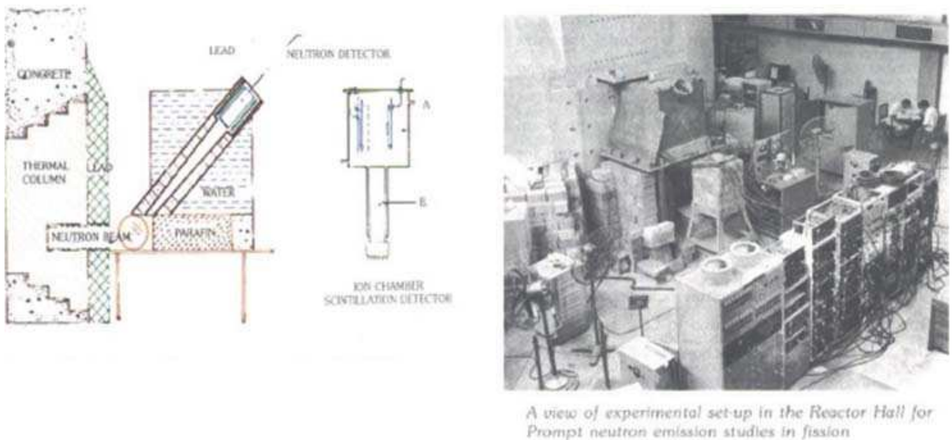

Figure 1. Experimental set-up for fission studies at APSARA reactor.

association with neutron and charged particle detectors, extensive studies were carried out on the mass–energy correlations in binary and ternary fission, and also in neutron emission in the fission process. The gridded ionization chamber provides the energy, mass, and angle information of the fission fragments [4–7]. Some of the important results of these investigations are:

(1) The fraction of pre-scission multiplicity (ν_{pre}) to total neutron multiplicity increases with increasing total fragment kinetic energy (E_{k}) for all mass splits.
(2) At the highest fragment kinetic energies, the neutron emission is predominantly from the pre-scission stage.
(3) In case of ternary fission, α-particles are the dominant third particle (LCP), followed by triton, proton, deuteron, helium-6, etc.
(4) Most of the LCPs are emitted perpendicular to the fragment emission; with a small fraction (2–5%) emitted along the fragment direction (polar emission).

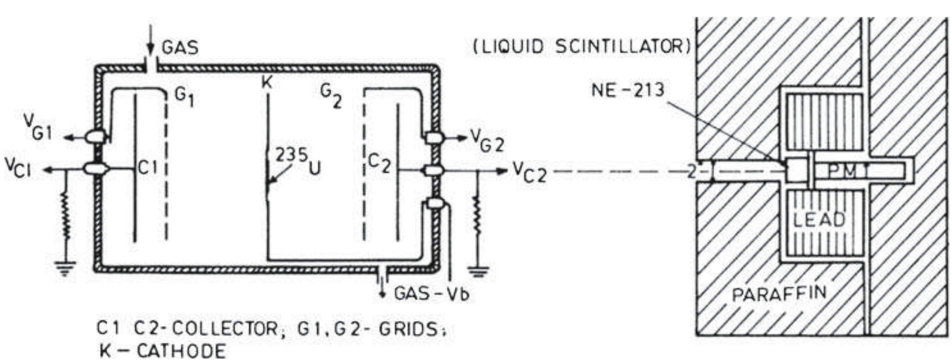

Figure 2. Schematic diagram of the experimental set-up showing the neutron detector shielding and back-to-back gridded ionization chamber.

(5) A rare mode of fission with two LCPs along with two fragments (quarternary fission) takes place in the thermal neutron fission of ^{235}U, with a very small probability ($\sim 10^{-5}$–10^{-6}) with respect to binary fission.

Recently, new investigations have been initiated in the thermal neutron fission for studying the γ-ray spectroscopy of the neutron-rich fragment nuclei, the results of which will be discussed later.

3. Heavy-ion-induced fission studies

With the setting up of the 14 MV pelletron tandem accelerator in the end of 1980s, heavy-ion beams in the low and medium mass regions are available for fission research. The energy range of the heavy ions cover from subbarrier to above barrier regions depending on the target–projectile combination. In this energy range, the dominant reaction mechanisms are a few-nucleon transfer and compound nucleus formation. Fission occurs when the excitation energy is close to and above the fission barrier. The description of nuclear fission based on the rotating LDM and shell effects can be validated by the experimental observations of the mass, energy correlation, angular distribution and fission probabilities. In this regard, experiments have been planned at the pelletron accelerator by the fission group in the last two or more decades on different target–projectile systems. In the following sections, a selective list of experiments and results obtained therein will be discussed.

4. Pre-scission and near scission emission of light charged particles in heavy-ion-induced fission reactions

Light charged particles (mostly α-particles) are emitted during the different stages of the fission process. The multiplicity of α-particles emitted in the fission process corresponds to pre-scission, neck emission and post-fission components. Experiments were conducted to determine these components by measuring the angular correlation of the α-particles and fission fragments in the ^{11}B + ^{232}Th reaction at $E_{\text{lab}} = 45$ MeV. The angular correlation data were fitted by the multisource emission to differentiate the various components. The results are: α_{pre} including neck emission = $(5.7 \pm 0.1) \times 10^{-3}$, $\alpha_{\text{post}} = (0.16\pm 0.02) \times 10^{-3}$. The fraction of neck emission component to the total pre-scission multiplicity is about 10$\pm$3%, which is about the same as observed from all the available literature data for a large variety of target–projectile systems [8]. This implies a universal behaviour of neck emission of the LCP corresponding to the statistical emission from the neck of the fissioning nucleus at scission point. The neck emission multiplicity in the heavy-ion fission (compound nucleus ^{243}Am) is, however found to be much lower than that observed in the systematics of the low energy and spontaneous fission data, as shown in figure 3. There is a clear deviation in the behaviour of neck emission of α-particles at high excitation energies as compared to low energy, providing clear evidence for a transition from superfluid to dissipative behaviour of the fission process as the excitation energy increases.

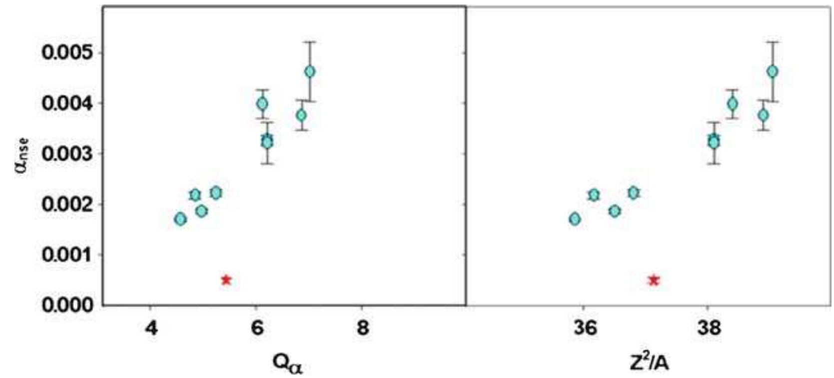

Figure 3. The comparison of the near-scission α emission data for $^{11}B + ^{232}Th$ at $E_{lab} = 45$ MeV ($\star$) with low-energy data from spontaneous and thermal neutron-induced fissions ($\circ$).

5. Emission of neutron as a tool for measuring fission delay

An experiment in $^{11}B + ^{237}Np$ and $^{12}C + ^{232}Th$ reactions (CN $= ^{244}Cm$) was conducted to examine the entrance channel dependence of fission time-scales in systems corresponding to opposite side of the Businaro–Gallone (BG) mass asymmetry. The pre-scission neutron multiplicities were determined from fragment-neutron angular correlations by multisource fit. From the present data along with the available literature data of many other systems, the different components of the time-scales corresponding to CN formation time, saddle–scission time and pre-saddle transient delay were determined [9].

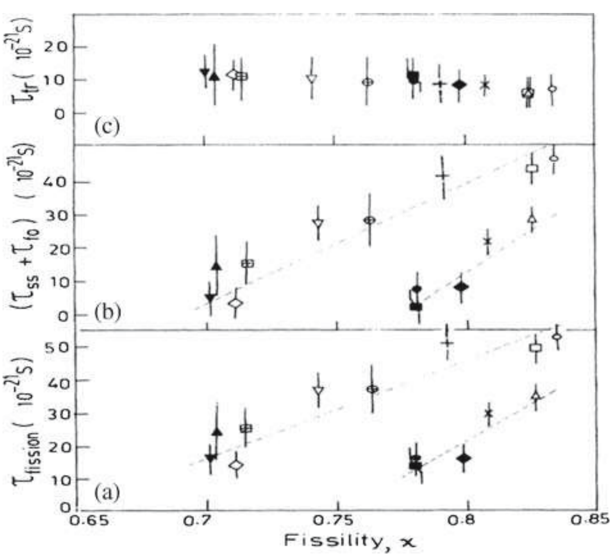

Figure 4. (**a**) Total dynamical fusion–fission delay deduced for $B_f/T = 1.0$, against the fissility parameter x. (**b**) Sum of the saddle-to-scission delay τ_{ss} and formation delay τ_{fo} as a function of x. (**c**) Transient delay τ_{tr} at $B_f/T = 1.0$ vs. fissility parameter x.

This experiment showed, for the first time, a clear transition of the CN nuclear formation dynamics as the projectile–target mass asymmetry is varied across the BG asymmetry (see figure 4).

6. Fission fragment mass and kinetic energy correlations

One of the areas where extensive work has been carried out is the measurement of fragment mass energy correlations to study the variance of mass distribution as a function of various quantities such as the compound nucleus fissility, temperature, angular momentum, etc. These results can be found in [10–12]. In one of the studies, where the mass variance is plotted as a function of temperature of the CN at the scission point for all systems measured in our experiments, along with the data reported in [13], it is found that the mass variance shows a universal behaviour for all fissilities with temperature at scission point, implying that the mass split and kinetic energy are decided at a later stage in the fission process, close to the scission point.

7. Angular distribution of fission fragments at near and sub-barrier energies

The statistical saddle point model based on the rotating LDM and shell model provides a good description of the angular distribution of fission fragments in heavy-ion-induced fission reactions. According to this model, the fragment angular distribution is a sensitive tool to investigate the saddle point shape, temperature and mass and K-equilibration time-scales. Experimental work on the measurement of the fragment angular distributions began with the establishment of the pelletron accelerator resulting in extensive studies of different fissioning systems. During the last 25 years of research in this area, many new findings have been reported. Some of the highlights of these studies are:

(1) Entrance channel effects across the BG mass asymmetry on the angular anisotropy of fission fragments at the above-barrier energies [14].
(2) Observation of peak-like structure in the angular anisotropy at sub-barrier energies for the actinide target nuclei [15].
(3) Fragment mass–angle correlations as indicators of the presence of non-equilibrium fission or quasifission [16].
(4) Effect of ground-state spin of the target and projectile nuclei on the fragment angular anisotropy at sub-barrier energies [17].

Presently, the study of quasifission and compound nuclear fission in very heavy mass regions is an important area of research signifying the production of the doubly closed shell stable super-heavy nuclei by heavy-ion-induced reactions.

8. Surrogate reaction method: For measuring neutron-induced fission cross-section of unstable nuclei

Nuclear fission research has direct relevance to the nuclear energy programme in the country. During the fuel burn-up inside the reactor, a large number of unstable actinide nuclei

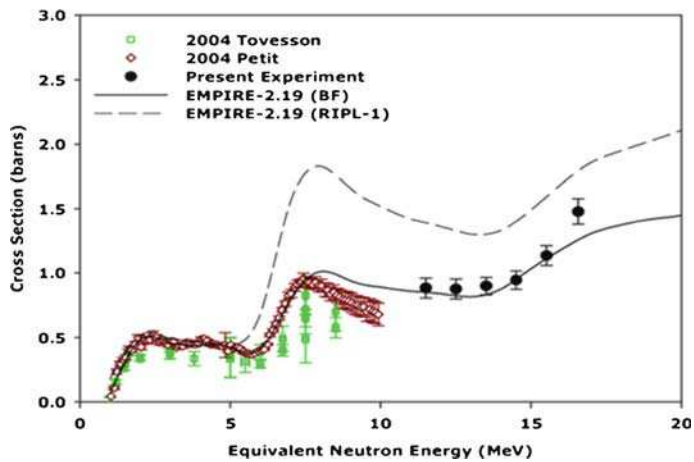

Figure 5. ^{233}Pa(n, f) excitation function. ^{233}Pa(E_{ex}, n, f)=^{233}Pa$((233/234)E_n +$ S_n(^{234}Pa)$, n, f)$.

are produced. This is more so, when one deals with fast reactors and ADSS systems. Also, during the Indian nuclear energy programme based on Th–U fuel cycle, many short-lived protactinium nuclei such as ^{233}Pa ($T_{1/2} = 27$ d), ^{234}Pa ($T_{1/2} = 1.2$ min) are produced, for which the neutron-induced fission cross-sections cannot be measured directly. Similar situation arises in U–Pu fuel cycle, where many neptunium isotopes have very short half-lives, e.g., ^{240}Np ($T_{1/2} = 7.22$ min). In order to experimentally estimate the fission cross-sections of such short-lived actinides, a new programme was started based on the hybrid surrogate reaction method. In this method, loosely bound heavy ions (^{6}Li, ^{7}Li) are bombarded on ^{232}Th, ^{238}U target nuclei at suitable energies to measure the projectile break-up-fusion–fission events in the complementary break-up channels. From the ratio of the fission cross-sections in the two channels, one can estimate the fission cross-sections at equivalent neutron energies in the MeV range. For example, the published results on ^{233}Pa(n, f) excitation function are shown in figure 5 [18]. Measurements for other nuclei have also been carried out [19] and efforts are being made to measure more nuclei.

9. Fission fragment spectroscopy using reactor neutrons

Fission fragment nuclei are on the neutron-rich side of the β stability line, and are expected to exhibit varieties of structural properties in different mass regions. Currently, spectroscopic study of neutron-rich nuclei is of great interest, as well as quite challenging. There are very few experimental facilities in the world to study the spectroscopy of fission fragment nuclei produced in reactors. With the availability of high efficiency, high resolution γ-ray clover detectors, this field has opened up new possibilities for experimental investigations. The fission group at Trombay has recently started a spectroscopy programme using thermal neutron beams from CIRUS reactor to produce fission and capture reactions on different nuclei. Of particular interest is the thermal neutron fission of ^{235}U,

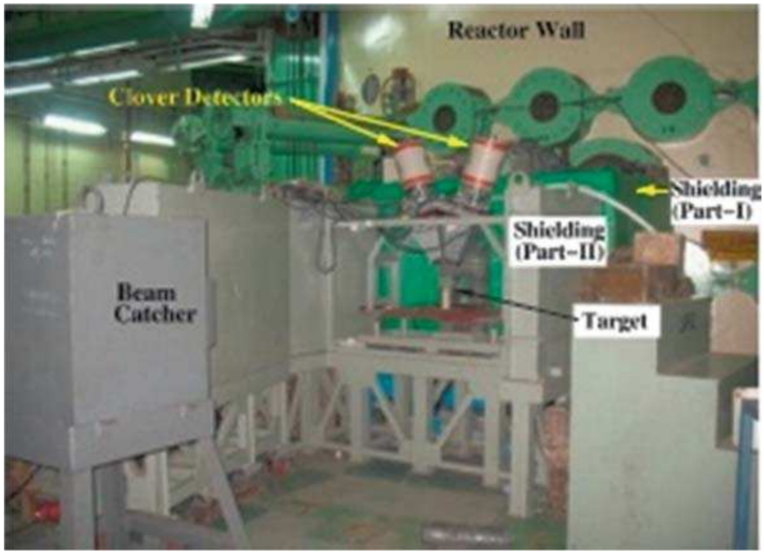

Figure 6. Experimental set-up at the CIRUS reactor facility for studing prompt γ–γ coincidence spectroscopy.

where a large number of neutron-rich fragment nuclei can be investigated. The experimental set-up is shown in figure 6 [20]. Four HPGe clover detectors are placed around a fission chamber, containing the ^{235}U target. The neutron flux at the target is about 10^7, which produces sufficient number of fragments for carrying out the γ–γ coincident analysis to study the γ-ray transitions in the fragment nuclei.

A detailed paper has been published recently discussing quite new and interesting results of fragment nuclei in different mass regions [21]. After the CIRUS reactor was shut down in 2011, the spectroscopy set-up was moved to DHRUVA reactor with further augmentation of the number of clover detectors.

10. Fission fragment spectroscopy using heavy ions

The fission group has participated in the Indian national gamma array (INGA) programme in which an array of HPGe clover detectors is employed to carry out the spectroscopic study of nuclei by γ-ray transitions. In one of the studies, γ-ray spectroscopy information was used to determine the isotopic mass distributions in the fission of ^{18}O+^{238}U system at 100 MeV bombarding energy. The mass of even–even fission fragments were identified by the $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$ transitions. It was shown for the first time that the isotopic mass yield in the fission of ^{256}Fm exhibits fine-structure dips at specific masses, corresponding to shell closures of the fragment nuclei, as shown in figure 7. The dips in the present system also coincided with the similar dips observed earlier for the ^{18}O+^{208}Pb system, implying a universal effect in the mass splitting mechanism during the fission process. This result was explained [22] intuitively as a hindrance of certain mass splits at scission, corresponding to the spherical shape of one of the fragments due to shell closure.

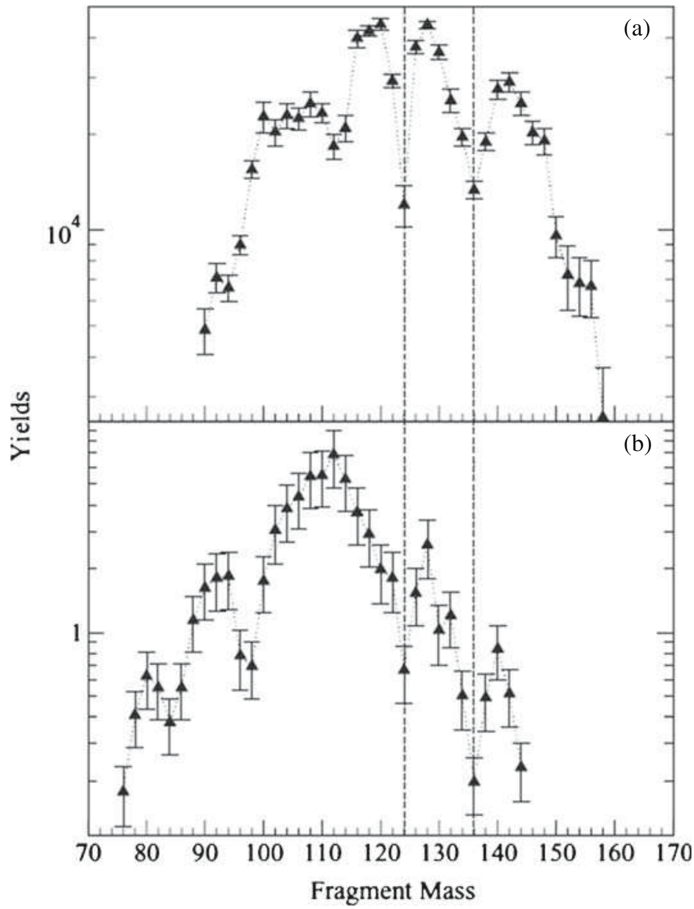

Figure 7. Fission fragment mass distribution obtained in (**a**) $^{18}O + ^{238}U$ at 100 MeV and (**b**) $^{18}O + ^{208}Pb$ at 85 MeV [22].

Further experiments have also been carried out using the INGA to study the γ-ray spectroscopy of neutron-rich fragment nuclei using heavy-ion reactions. The level schemes of the fragment nuclei in different mass regions are determined to understand the evolution of their structures as *Z* and *N* of the nuclei are varied.

11. Theoretical studies

The fission group at Trombay has published many papers on theoretical studies of different aspects of fission and related topics. It is impossible to describe all these in this overview. Some of the important areas of study are:

(1) Fragment mass division as nucleon exchange by Markov process [23].
(2) Washing out of shell effects in nuclei at high temperature [24].

(3) Nuclear level density as a function of excitation energy [25].
(4) Dynamical trajectory model description of heavy-ion collisions [26].
(5) Fragment angular distribution analysis and pre-equilibrium fission [27].
(6) Entrance channel-dependent *K*-state model for fragment angular distribution at sub-barrier energies [28].
(7) Production cross-section of superheavy nuclei by rare-earth nuclear collisions [29].

12. Summary and conclusions

In this review, an attempt has been made to provide a glimpse of the variety of research problems taken up by the fission group in Trombay. Nuclear fission has still remained an empirical science, and many new features are being unfolded by the experiments. Future research will focus on the synthesis of superheavy nuclei through different routes, including the RIBs. Understanding the fusion–fission dynamics in heavy-ion collisions in the multidimensional domain of CN fissility, target–projectile mass asymmetry, excitation energy and angular momentum are quite challenging.

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