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Seventy-five years of nuclear fission

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Abstract. Nuclear fission process is one of the most important discoveries of the twentieth century. In these 75 years since its discovery, the nuclear fission related research has not only provided new insights in the physics of large scale motion, deformation and subsequent division of a heavy nucleus, but has also opened several new frontiers of research in nuclear physics. This article is a narrative giving an overview of the landmarks of the progress in the field.

Keywords. Nuclear fission; nuclear shell effects; superheavy nuclei.

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There is no doubt that Einstein's theory of relativity and the development of quantum mechanics have been the most revolutionary advances of the 20th century in physics, as these theories have laid the foundation of our understanding of the physical phenomena occurring at all levels from the microscopic subatomic world to the macroscopic Universe. But it is the epoch-making discovery of nuclear fission in 1939 by Otto Hahn and Fritz Strassmann [1] which had a direct impact on human life and heralded the onset of a new era in the history of human civilization by changing the world forever. Soon after the discovery of neutron in 1932 by Chadwick, Fermi recognized that due to the absence of Coulomb barrier, neutron was the ideal projectile to induce successive transformations in all the nuclei up to uranium. Thus, Fermi and coworkers started experiments with the goal of extending the periodic table by bombarding thorium and uranium with neutrons anticipating that the neutron capture in the uranium nucleus followed by β decay will lead to the formation of an element with the next higher atomic number, thus producing a transuranic element. But the observed decay curves of the induced radioactivity did not conform with such a simple picture and the intense research activities carried out over the years in trying to resolve the transuranic puzzle ultimately led to the discovery of nuclear fission. It did not occur to anyone, that a heavy nucleus of uranium which is so strongly bound, can be split in two nuclei with the bombardment of a low-energy neutron. Yet, when such a conclusion was forced by the compelling experimental evidence of the radiochemical work, soon thereafter Meitner and Frisch worked out a physical solution to the transuranic puzzle by proposing that the particles in a heavy nucleus would move in a collective way with some resemblance to the motion of a charged liquid drop, which can divide itself into smaller drops if movement is violent enough. They also calculated the fragment kinetic energy release to be as large as about 200 MeV, which was soon confirmed by Frisch using the ionization chamber experiment. In their paper, Meitner and Frisch have described the process as 'nuclear fission' drawing analogy with the division of a biological cell [2]. Frisch told Niels Bohr about this new discovery, just when Bohr was leaving for USA on 7 January 1939. Subsequently on reaching Princeton, Bohr and Wheeler developed a basic theory of nuclear fission primarily based on the liquid-drop model (LDM) which continues to be the basic framework to study the fission process [3].

The discovery of nuclear fission can be considered amongst the most important discoveries of the 20th century for several reasons. It was not a confirmation of a theoretical prediction but an unanticipated experimental discovery, which brings out the importance of experimental science. The large-scale energy release in the fission process in accordance with the Einstein's celebrated mass–energy equivalence equation $E = mc^2$ and the fact that two to three neutrons are also emitted in fission making a chain reaction possible, led to a way to tap a large amount of nuclear energy that is locked inside the nuclei in the form of nuclear binding energy. The subsequent developments that led to the Manhattan project in the USA to build an unsurpassed nuclear military might are now part of history. Further, the harnessing of nuclear energy for generating electricity and many applications of radioisotopes and nuclear radiations started a new era of peaceful applications of nuclear energy for the benefit of mankind. The realization that the science and technology can play such a vital role in a nation's development subsequently motivated the governments to make substantial investments in science and technology. It can be said that it was the discovery of nuclear fission 75 years back which triggered the exponential growth of not only nuclear science, but also science and technology in general, that we have subsequently witnessed. Discovery of nuclear fission is a very good example of how curiosity-driven basic research can lead to a totally unpredictable discovery of immense benefit to the society.

In these past 75 years of nuclear fission research, it is well realized that the nuclear fission process which involves massive motion and division of a heavy nucleus into two fragments with a broad mass distribution is a unique nuclear phenomenon involving large-scale nuclear dynamics. Thus, from the point of view of basic nuclear physics research, this process provides a way to study large-scale nuclear dynamics and nuclear structure of highly deformed nuclear configurations. Several aspects of the fission process were brought out in the articles published in the special volume of *Pramana – J. Phys.* on the occasion of 50 years of the discovery of fission [4]. In the past, several theoretical models have been put forward to explain the asymmetric mass and charge distributions in the low energy and spontaneous fission of actinide nuclei and how these distributions become symmetric for nuclei lighter than thorium and at high excitation energies. While these models differ in detail, in all the models the observed asymmetry arises basically due to nuclear shell structure. Recently, it is observed that the fission of even a lighter nucleus namely ^{180}Hg, results in an asymmetric mass distribution [5].

It is well established that the potential energy landscape around the fission transition state (saddle point) involving a fission barrier height controls the fission probability and the fission fragment angular distributions with respect to the direction of the energetic projectile-inducing fission. Further, the dynamics from the transition state to the scission point also appears to be involved in governing the fragment mass, charge and kinetic energy distributions. The time of passage from the saddle point to the scission point is expected to be of the order of 10^{-21}–10^{-20} s depending on the magnitude of the nuclear viscosity, which controls this time. The Coulomb energy of the two fragments at the scission point subsequently gets converted into the fragment kinetic energy. But, depending upon the magnitude of nuclear viscosity, the fragments may already acquire some kinetic energy up to the scission point, which will also get added to the Coulomb energy. While almost all the prompt neutrons are emitted from the fragments after they have acquired their full velocities on Coulomb repulsion, a small fraction of about 10% appears to be emitted around the scission point. Some fragments after β decay have an excitation energy greater than the neutron binding energy, and emit what is called the delayed neutrons in the characteristic times of β decay which range from a fraction of a second to several tens of seconds for different fragment groups. These delayed neutrons in fission are the nature's boon as controlled release of nuclear fission energy in the nuclear reactors would not have been possible without the presence of these delayed neutrons. Another characteristic of fission is the occasional emission of a light charged particle (mostly an α-particle, say about 1 in 300 fissions) and these charged particles have served as probes to investigate the dynamics of the fission process.

An important landmark in fission was the realization by Bohr that the passage of a fissioning nucleus over the saddle point is slow enough for it to exhibit quasistationary quantum states similar to the collective states of any deformed nucleus [6]. Consequently, the angular distribution of the fragments with the direction of the incident projectile causing fission will depend on the quantum states through which the nucleus passes at the saddle point. This model successfully tested for fission induced by γ-rays and other low-energy projectiles was subsequently extended for medium and high-energy projectiles by Halpern and Strutinsky to develop a very successful statistical theory of the fragment angular distributions [7]. In the framework of this theory, it became possible to deduce the effective moment of inertia of the fissioning nucleus at the saddle point by analysing the fragment angular distributions. It turned out that these deduced effective moments of inertia, and hence nuclear shapes of the fissioning nuclei at the saddle point agree well with the saddle point shapes predicted by the advanced LDM calculations. Thus starting from around 1960s, the studies on the fragment angular distributions with a variety of projectile beams including heavy-ions became a very active and fertile area of fission research.

Following the discovery of fission, the unfinished task of extending the periodic table of elements beyond uranium was pursued with renewed vigour. In the period up to 1960s, many transuranic elements extending to atomic number $Z \sim 104$, were discovered and synthesized in nuclear reactions with accelerated charged particle beams. As spontaneous fission results from the tunnelling of the fission barrier, the observed general behaviour of exponential decrease of the spontaneous fission half-lives with increasing Z^2/A of these transuranic nuclei reflected the general trend as expected from LDM. The half-lives of these very heavy nuclei are found to be in the range of fractions of a second and further extrapolation on the LDM basis would suggest that spontaneous fission would terminate the periodic table around $Z \sim 104$ as superheavy nuclei with higher Z may not exist even momentarily.

However, it was also well established in the 1960s that the atomic nucleus has a shell structure similar to that of the atom and consequently the nuclei show significant increase in stability at the shell closures corresponding to the so-called magic numbers at Z or $N = 2, 8, 20, 28, 50, 82$ and also $N = 126$. It was therefore realized that the LDM cannot be quite valid for accurately determining the nuclear stability particularly in the vicinity of the magic numbers. During the late 1960s, it became possible to determine the nuclear binding energies as a function of proton number Z, neutron number N and nuclear deformation to within an accuracy of about 1–2 MeV using a new theoretical approach proposed by Myers and Swaitecki [8] and generalized by Strutinsky [9]. This approach synthesized the nuclear macroscopic behaviour as given by the LDM with the nuclear microscopic behaviour as predicted by the nuclear shell model.

One of the predictions of these theoretical studies was that the fission barriers of the actinide nuclei are not single-humped as given by the LDM but are double-humped in nature with a secondary minimum due to the nuclear shell effect. The early 1960s also saw the discovery of the spontaneously fissioning isomers in the actinide nuclei, which could be later explained on the basis of this double-humped nature of the fission barriers. In fact, the measurements of these fission isomers provided a way to quantitatively determine the parameters of the double-humped barrier and also carry out spectroscopy of the highly deformed nuclear configurations corresponding to the second well [10]. These spontaneously fissioning isomers are now also known as shape isomers. The double-humped nature of the fission barrier also led to the significant revision in the evaluated nuclear data which consequently had an impact on the design of fast reactors.

The other prediction of the above theoretical studies is that the fission barrier height of a doubly closed shell nucleus, such as ^{208}Pb, becomes significantly increased with respect to the prediction of the LDM due to the shell correction energy of its ground-state spherical configuration. This, then also implies that as a result of incorporating shell effects, even superheavy nuclei can have sizeable barrier heights to permit significant stability against spontaneous fission, if such nuclei have closed proton or/and neutron shells (magic numbers). The shell effects will also increase their α decay half-lives. The question then arises as to where are the closed shells in the superheavy region? Most macro–microscopic calculations which have been performed over the years correspond to the theoretically predicted closed shells at $Z = 114$, $N = 184$ [11]. Relativistic mean-field calculations suggest magic numbers of $Z = 120/126$ and $N = 184$. Certain Hartree–Fock calculations with Skyrme interactions predict $Z = 126$, $N = 184$. Also, additional regions of deformed shell-stabilized nuclei in the superheavy region are expected. Of course, the theoretical calculations can only be taken as a guide in the search for the region of enhanced stability in the superheavy region as it is difficult to make accurate theoretical prediction on the precise location of the closed shells in this region.

For over 30 years or so, scientists have sought to synthesize superheavy nuclei at or near the above regions of closed proton and neutron shells. In the recent years, active research has been in progress globally to synthesize the closed shell superheavy nuclei, through fusion of two nuclei by the suitably chosen heavy-ion reactions. In order to choose suitable reactions for optimum cross-section of the production of a superheavy nucleus, one needs to acquire a good understanding of the role of: (i) nuclear dynamics, (ii) entrance-channel parameters (mass asymmetry, neutron richness of projectile–target combination, bombarding energy) and (iii) washing out of the stabilizing effects of the closed nuclear shells with nuclear excitation energy on the mechanism of fusion, compound nucleus formation, fission and de-excitation processes.

Experimental efforts to synthesize superheavy nuclei have been vigorously pursued by collaborating scientific teams utilizing the heavy-ion beams at GSI, Darmstadt, and JINR, Dubna [12–15]. In the GSI facility, synthesis of superheavy nuclei up to $Z = 112$ has been observed through the bombardment of heavy-ions of ^{54}Cr, ^{58}Fe, ^{64}Ni, ^{62}Ni and ^{70}Zn on ^{208}Pb and ^{209}Bi target nuclei. In such reactions with smaller mass asymmetry in the entrance channel and with the heavy-ion energies close to the fusion barrier, the cross-section for the formation of a compound nucleus is rather small. But this is off-set by the advantage that a compound nucleus formed in the above type of fusion reactions has a relatively low excitation energy (the so-called cold-fusion reactions). This decreases the chance of its decay by fission in the de-excitation process and correspondingly increases the probability of formation of an evaporation residue of a superheavy nucleus. In JINR, Dubna facility synthesis of superheavy nuclei with Z up to 118 has been reported by bombarding neutron-rich ^{48}Ca projectiles on various transuranic nuclei. This approach involving larger mass asymmetry in the entrance channel leads to higher compound nucleus excitation energies (the so-called hot fusion reactions), but has the advantage of larger fusion cross-sections at the fusion barrier energies as compared to the equivalent cold fusion reaction approach. As was expected on the basis of cross-sections, in the above experiments, only a few events of superheavy nuclei production have been detected in several days of bombardment. The half-lives of the superheavy nuclei observed so far are still quite small, being in the range of msec to a fraction of a second. This is expected as the superheavy nuclei produced so far are neutron-deficient with respect to the predicted closed neutron shell in this region. One way to also reach the closed neutron shell region is to achieve heavy-ion fusion with more neutron-rich projectile and target. One can expect that with the availability of intense neutron-rich rare ion beams (RIB) in future, it may become possible to achieve such doubly-closed shell configurations of much longer half-lives in the superheavy region.

Recent years have seen dramatic progresses in accelerator technology for the development of high-energy and high-current accelerators needed for basic research in high-energy physics and for developing RIB accelerator facilities. The development of such high-energy (about 1 GeV proton) and high-current (10 mA) accelerators is also needed to build accelerator-driven subcritical reactor systems (ADS) for the incineration of minor actinides of spent fuel and thorium utilization [16]. The already established reactor technology based on nuclear fission will remain the only way to harness nuclear energy for a long time to come. But in harnessing nuclear energy through the critical fission reactors, disposal of nuclear waste is an important issue of concern which needs to be addressed satisfactorily. Recent years have seen considerable research and developmental activities to tackle this problem by finding a suitable way to incinerate the long-lived transuranic and fission product nuclei of the spent fuel waste. For this reason there is much interest in the ADS. The possibility to operate a reactor core generating hundreds of megawatts of fission-based thermal power at a value of k less than 1 is a great advantage as it opens up possibilities of having reactor cores of any composition of fissile fuels, even consisting of mainly plutonium and/or minor actinides which are not permitted in the critical reactors due to their small delayed neutron fractions. In India, exploitation of nuclear power is considerably more dependent on an efficient utilization of the available nuclear fuel resources. Our country has abundant reserves of thorium which makes ADS very attractive because one can also exploit its potential to design hybrid reactor systems that can produce nuclear power by using thorium as the main fuel [17,18].

The impact of the discovery of nuclear fission heralded the arrival of a new scientific era in the 1940s. This nuclear era saw a keen competition amongst the major nations of the world to see that they are not left behind in the pursuit of nuclear physics and related nuclear science and technology. In those early days, India was fortunate to have the scientific leadership of the great scientist and visionary Homi J Bhabha, who having established himself as a theoretical physicist of great international distinction, chose to subsequently devote his time to the task of developing a viable nuclear programme in India. Under his leadership, India's first reactor APSARA became operational as early as 1956 and India became the first country in Asia to have indigenous capability in designing and building a nuclear reactor. In India, nuclear fission research started quite early in the mid-1950s under the inspiring leadership of Raja Ramanna and I was fortunate to join the fission group soon thereafter. In the early 1960s, Raja Ramanna and his team carried out landmark experimental studies on the emission of prompt neutrons and γ-rays in the thermal neutron-induced fission of ^{235}U using neutron beams from the then commissioned APSARA reactor [19,20]. In these studies, from a detailed analysis of prompt neutron spectra measured at different angles with respect to the fragment direction using a novel experimental technique based on a gridded ion chamber, emission spectra and angular distributions of the neutrons from selected fragments were deduced. Among other results, a detailed analysis of the data also provided experimental evidence that a small fraction of the prompt neutrons (now known as pre-scission neutrons) are not emitted from the moving fragments. From the measured γ-ray anisotropy with respect to the fragment direction, it was inferred that a significant angular momentum in each pair of fragment nuclei is produced in the nuclear dynamics from saddle to scission. These experimental studies provided information on the saddle-to-scission time which, in the later years has been related to nuclear viscosity in the large-scale motion of nuclear matter on its way to scission.

A few years later, a 5.5 MV Van de Graaff accelerator was commissioned at Trombay, which enabled the Trombay fission group to carry out experimental studies of correlation between fragment anisotropy and mass asymmetry in fast neutron-induced fission. Ramanna was not the one to follow a beaten track and in the early 1960s came out with a model proposing nucleon exchange between the two nascent fragments to explain asymmetric mass distributions [21]. A decade later, in the heavy-ion deep inelastic collisions, nucleon exchange between two nuclei in proximity became a common topic of research. With the discovery of double-humped fission barrier for actinide nuclei, one would have expected that the effective saddle point moment of inertia deduced from the fragment angular distributions should have corresponded with the second barrier shape. But the studies of energetic α-induced fission showed that it follows liquid-drop barrier shapes. This anomaly was resolved by showing that the shell effects melt away with increasing excitation energy [22]. When the Pelletron accelerator at Mumbai came into operation in the late 1980s, we could experimentally demonstrate [23] the existence of pre-equilbrium fission in heavy-ion-induced fusion–fission reactions as conjectured earlier in our heavy-ion-induced fragment angular distribution theory [24]. Another significant contribution of Trombay group was the analysis of the fragment-neutron angular correlations using pre-scission neutron clock to deduce time-scales involved in the different stages of the fusion–fission process, namely the stages of compound nucleus formation, compound nucleus to saddle point and saddle to scission time [25]. In recent years, fission fragment spectroscopy has also been carried out at Trombay by measuring prompt γ-rays using high-resolution clover germanium detectors to study the properties of neutron-rich fragment nuclei [26]. The above-mentioned contributions are only a few examples of the fission-related work at Trombay. There are many other important contributions, some of which resulted from the radiochemical studies at Trombay, and several others from the studies carried out at other research centres in India.

Thus, in BARC, Trombay and also at various other research centres in India, research programmes in nuclear fission process including heavy-ion-induced fission reactions have continued to be actively pursued in the last five decades or so theoretically and experimentally using physical as well as radiochemical techniques. These include studies of the binary and ternary fission accompanied by light charged particle in low-energy fission, charged particle including heavy-ion-induced fission and also spontaneous fission. From the studies of the fragment mass, charge, kinetic energy, excitation energy angular distributions and also mutual correlations among various observables, a great deal of information has been obtained about the fission process in the research work carried out in India.

I may conclude by saying that even after 75 years, nuclear fission research continues to be a contemporary area of research globally. In fact with the availability of rare ion beams (RIB) of high intensity, much research is expected to be focussed in the coming years on the studies of fission of exotic nuclei far away from the line of β stability in the nuclear chart. It speaks for the richness and complexity of the fission process that even after 75 years we do not have a rigorous theory to predict the fragment mass and charge distribution of the fragments for a given mass, charge and excitation energy of the fissioning nucleus. This continues to be a big challenge and open problem for nuclear theorists. Nuclear fission process continues to be a rich and rewarding area of nuclear physics research.

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