Delayed fission of atomic nuclei

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Abstract. The history of the discovery of delayed nuclear fission is presented, and the retrospective of investigations into this phenomenon that were performed at various research centers worldwide is outlined. The results obtained by measuring basic delayed-fission features, including the fission probability, the total kinetic energy of fission fragments, and their mass distributions, are analyzed. Recommendations concerning further studies in various regions of nuclear map with the aim of search for atomic nuclei undergoing delayed fission are given. Lines of further research into features of delayed fission with the aim of solving current problems of fission physics are discussed.

1. Introduction
At present, we know five types of atomic nuclei fission: induced by neutron and charged particles, spontaneous fission, spontaneously fissionable shape isomers, delayed fission, and quasi-fission. The fission of uranium nuclei was discovered in 1938 by O. Hahn and F. Strassman. They were able to establish that during the bombardment of uranium by neutrons, elements of the middle part of the periodic system are formed — barium, krypton, etc. [1]. L. Meitner and O. Frisch correctly interpreted this fact. They explained the appearance of these elements by the decay of the uranium nuclei after the neutron capture into two approximately equal parts. This phenomenon is called fission of nuclei, and the nuclei formed are fragments of fission. Two years after the discovery of the fission, in 1940, G. N. Flerov and K. A. Petrizhak observed that uranium nuclei can spontaneously undergo fission [2]. The half-life for spontaneous fission of $^{238}$U was about $8 \times 10^{15}$ years.

The activities of the Flerov Laboratory of Nuclear Reaction (FLNR), JINR have been focused on synthesizing new elements and isotopes and on studying their properties. In order to synthesize new elements, a few the setups were originally made based on detecting nuclides produced in fusion reactions upon their alpha decay and spontaneous fission. Techniques for detecting decays of nuclei that may undergo spontaneous fission, the respective lifetime ranging between several milliseconds to several minutes or more, were developed. The first experiments for search and synthesis of transuranium elements possibly undergoing decay through the spontaneous fission channel were performed via detecting fission fragments by means of proportional chambers [3]. Some discoveries concerning new properties of nuclei synthesized in heavy ion reactions were made when solving this problem. The phenomenon of spontaneous fission of nuclei in isomeric states was discovered in Dubna in 1961 [3]. The americium isomers with mass numbers 240, 242, and 244 have half-lives of 0.6, 14, and 0.9 ms, respectively, and they are formed in complex transfer reactions upon irradiation of curium, plutonium, and uranium by heavy ions [4]. After the discovery of spontaneously fissioning isomers whose lifetimes...
2. Discovery of delayed fission and its study

Nuclei whose production cross sections were so small could be observed by irradiation of the target in the internal heavy-ion beam of the U-300 cyclotron. The intensity of heavy ions was high in that case (up to 100 eμA for B and Ne). Moreover, the energy of the accelerated beam incident to the target could readily be varied by moving the target along the radius of the cyclotron magnetic pole [7, 8]. This made it possible to obtain rather high currents and to detect fission fragments with the aid of movable low-background solid-state trace detectors of efficiency \(0.90 \pm 0.05\). The angle of inclination of the target with respect to the beam axis was \(12^\circ\) (Fig. 1).

![Figure 1](image)

**Figure 1.** Layout of the test setup involving a sloped target: (1) sloped target, (2) screen electrically connected to the target, (3) ion beam incident on the target, (4) assembly of solid-state detectors (4.1-4.4 are the successive positions of each detector above the target), and (5) motion of the detectors toward the irradiated target and away from it. The target was manufactured from aluminum (the cooling system is not shown).

Therefore, the beam intensity and irradiated matter were distributed over an area 4.81 times as large as that in the case of the orthogonal orientation of the target. Another advantage of the slope target was that recoil nuclei produced in nuclear reactions were decelerated in the surface layer of the target material. The detectors viewed a thin target-material layer, which led to a low energy loss for fission fragments of reaction products and did not prevent their escape from the target. After the irradiation of the target with an ionic flow for a fixed time interval, the cyclotron beam was interrupted, whereupon four solid-state detectors were placed by an automated system above the target for a time interval close to the lifetime of the isotope being studied. Before the beginning of the next target exposure run, the detector assembly was positioned, in just the same way as earlier was done in the case of the semiconductor detectors, within the chamber at a distance from the target as long as to ensure the protection of the detectors from scattered particles and background neutrons. As soon as the detectors were arranged in the shielded chamber, the beam was again switched on in order to irradiate the target. This procedure of target irradiation and exposure of the detectors was repeated periodically. So the bombardment was done on a sample with a slope target allowing high currents to be obtained, and the fission fragments were recorded by means of movable, low background, solid-state track detectors. Delayed fission was discovered in the FLNR JINR (Dubna) in 1966. Fission products with half-lives of the order of minutes were observed. In bombarding \(^{209}\text{Bi}\) by an intense beam of \(^{22}\text{Ne}\) ions, a new “spontaneous” fission activity with \(T_{1/2} \approx 60 \pm 5\) s was observed (Fig. 2), which was attributed to \(^{228}\text{Np}\) [7].
Shortly afterwards, two additional “emitters” — fission fragments with half-lives $2.6 \pm 0.2$ and $1.4 \pm 0.3$ min — were detected and identified as $^{234}$Am and $^{232}$Am in reaction channels $^{230}$Th($^{10}$B, $8n$)$^{232}$Am and $^{236}$Th($^{10}$B, $6n$)$^{234}$Am, respectively. The analysis of the available experimental evidence has shown that the observed fission fragment with half-lives of several minutes are due to fission from excited nuclear states of daughter nuclei populated via electron capture, rather than by nuclear shape isomerism. In work [9], it was shown that the above experiments with heavy-ion beams make it possible to synthesize for the first time $^{228}$Np, $^{232}$Am, and $^{234}$Am parent nuclei whose daughter products formed after electron capture, $^{238}$U, $^{232}$Pu, and $^{234}$Pu, undergo fission from excited states based on the comparison of the K-capture energy in a predecessor nucleus and the fission-barrier height in the respective daughter nucleus.

It would be natural to expect delayed fission in two nuclear-map regions — the region of neutron-deficient nuclei and the region of neutron-rich nuclei. However, this requires fulfillment of two conditions: first, the existence of a nonzero branch of the K-capture or β-decay and, second, the energy value $Q_{EC(\beta)}$ in a parent nucleus should be comparable with or greater than the fission barrier height value $B_f$ in the respective daughter nucleus. In moving away from the β-stability line (as neutron deficit or excess grows), the β-decay energy $Q_{\beta}$ of heavy nuclei becomes comparable to the fission barrier height of the daughter nuclei and can even exceed it. At high values of the total energy $Q_{EC(\beta)}$, a wide spectrum of excited states in the daughter nucleus is populated, and the levels lying near the fission barrier top obviously possess large fission widths. Hence, the half-life measured directly for the fission fragments branch is governed by the slow β-decay process, while the fission itself occurs on a time scale of the order of $10^{-14}$–$10^{-16}$ s.

The work on search for new emitters and investigation into their delayed fission was continued by two groups in FLNR JINR. The studies of Gangrsky et al. revealed the delayed fission of $^{244}$Es, $^{246}$Es, $^{246}$Bk, and $^{250}$Md [10, 11]. The region of nuclei with $N < 126$ and $Z < 82$ deserves particular attention. For $N < 126$ nuclei, the alpha-decay energy $Q_\alpha$ decreases sharply, whereupon it grows slowly as the neutron deficit increases. However, the role of alpha decay becomes less pronounced owing to the enhanced competition of the electron-capture-induced decay. The energy $Q_{EC}$ begins to exceed the barrier height $B_f$ in the daughter nuclei — first of all, in even–even nuclei. This opens the possibility of observing delayed fission in this neutron-deficient region. The first experiments in this region of nuclei were performed by the group headed by Oganessian [12]. Three nuclides that underwent delayed fission, $^{186}$Tl, $^{188}$Bi, and $^{196}$At, were synthesized in studying heavy-ion reactions on various targets [11, 12].

Later Hall et al. [11], Andreev et al. [13] observed delayed fission of nuclei in neutron-deficient and neutron-rich regions of the nuclide map. Hall et al. [14] studied the delayed fission $^{256m}$Es isomer synthesized in the nuclear reaction $^{254}$Es($t$, $p$)$^{256m}$Es. The delayed fission activity of $^{256m}$Es after...
population of the level 1425 keV of the $^{256}$Fm nucleus with spin and parity $7^-$ was apparently the first case, when the delayed fission was registered as fission from the isomeric state of the first well of the potential-energy surface. The delayed fission activity of $^{188m_{1,2}}$Bi was also studied at the ISOLDA facility (CERN) [15].

There are direct experiments on checking the delayed fission hypothesis, in particular, coincidences of $K$ X-rays and fission fragments from the resulting daughter nuclei have been observed. After electron capture of $^{232}$Am and $^{234}$Am, spectra of X-rays in coincidence with fission fragments from the daughter nuclei $^{232}$Pu, $^{234}$Pu, and energy distributions of the fission fragments have been measured. The isotopes $^{232}$Am and $^{234}$Am were produced in the reactions $^{237}$Np($\alpha$, $9n$)$^{232}$Am and $^{237}$Np($\alpha$, $7n$)$^{234}$Am, respectively. Hall et al. [14] confirmed the half-lives observed by us [7, 8] for the $^{232}$Am and $^{234}$Am delayed-fission emitters and branching ratios for fission. A radiochemical separation into fractions revealed that fission is observed in the americium fraction and that measured X-ray radiation accompanying $K$-capture is associated with the fission of the product daughter plutonium nucleus. Data on the energy distributions of fragments originating from the delayed fission of $^{232}$Am and $^{234}$Am were also obtained. These experiments revealed that fission fragments of $^{232}$Pu and $^{234}$Pu daughter nuclei with a low excitation energy (not more than the $K$-capture energy in the parent nucleus) have an asymmetric mass distribution and that the total kinetic energy (TKE) had nearly the same values as in the case of spontaneous fission.

These and subsequent experiments devoted to synthesizing delayed-fission emitters employed radiochemical separation methods; later on, methods of laser-induced ionization and electromagnetic separation come into use [13, 14]. Energy and mass distributions of fragments originating from delayed fission were also measured in addition to the charge identification of parent nuclei and measurements of delayed-fission probabilities. At the present time, quite comprehensive data from measurements of fission-fragment energies and masses are known for 12 predecessors in the neutron-deficient region of nuclei from $^{178}$Tl to $^{244}$Es.

More than 27 isotopes undergoing delayed fission are known at the present time in four regions of nuclides: neutron-deficient isotopes in the region around lead, neutron-rich isotopes in the region of actinium and protactinium, neutron-deficient isotopes of transuranium elements, and neutron-rich isotopes of transuranium elements [13]. However, information about some nuclei undergoing delayed fission is scanty in almost all of the regions. It is clear that delayed-fission probabilities ($P_{DF}$) will essentially depend on both the total $K$- or $\beta$-decay energy $Q_{EC(\beta)}$ and the height and the shape of the fission barrier ($B_f$).

3. Qualitative theory of delayed fission

The delayed-fission probability is equal to the ratio $P_{DF} = \frac{N_{DF}}{N_i}$, where $N_i$ is the total number of EC($\beta$)-decays of the parent nucleus and $N_{DF}$ is the number of decays accompanied by delayed fission. One of the approximations frequently used in theoretical calculations [8, 11] is given by

$$P_{DF} = \frac{\int_0^{Q_i} W_i(Q_i - E) \frac{\Gamma_f(E)}{\Gamma_{tot}(E)} dE}{\int_0^{Q_i} W_i(Q_i - E) dE}.$$ 

Here $W_i(Q_i - E)$ is the transition probability function, $\Gamma_f(E)$ is the ratio of the fission width of excited levels of the daughter nucleus to the total decay width of these states, $E$ is the excitation energy of the daughter nucleus, and $Q_i$ is either $Q_{EC}$ or $Q_{\beta}$.  


$W_i(Q_i - E)$ for the transition to the level with $E$ can be represented as the product of the Fermi function $f(Q_i - E, Z)$ and the beta-decay strength function $S_{\beta}(E)$; that is, $W_i(Q_i - E) = f(Q_i - E, Z) \cdot S_{\beta}(E)$.

In this expression, the Fermi function $f(Q_i - E, Z)$ determines the kinematics of $K$-capture ($\beta$-decay) and may be represented as $f = (Q_{EC} - E)^2$ in the case of $K$-capture and $(Q_{\beta} - E)^5$ in the case of $\beta$-decay. The strength function $S_{\beta}(E)$ is the excitation energy distribution of the squares of the beta-decay type matrix elements for Fermi and Gamow–Teller transitions.

The dependence of $P_{DF}$ on the energy of levels and on the fission-barrier structure is primarily due to the fission width which appears in the expression $\frac{\Gamma_f}{\Gamma_f + \Gamma_{\gamma}}$ and depends on the excitation energy of the daughter nucleus. In the electron-capture case, an analysis of the expressions for $\Gamma_f$ and $\Gamma_{\gamma}$ makes it possible to obtain a simple equation for the probability $P_{EC}$ of delayed fission after electron capture

$$P_{EC} = N_{EC}(A) \int_{C} \left[ (Q_{EC} - E)^2 \right] \frac{\Gamma_f}{\Gamma_f + \Gamma_{\gamma}} dE,$$

where $N_{EC}(A)$ is a normalizing function equal to $3(Q_{EC} - 26A^2)^{-3}$.

Delayed fission was observed for odd-odd neutron-deficient nuclei. A large value of $Q_{EC}$ compared to the respective values for neighboring nuclei because of the odd-even effect leads to the comparable values of $Q_{EC}$ and $B_{f_{even}}$ even at a relatively small distance from the beta-stability line.

A significant value of the neutron binding energy $B_n$ in the even-even daughter nucleus gives sufficient grounds to disregard the competition of the delayed and neutron channels. The density of low-lying levels of even-even daughter nuclei in quite low. This circumstance and a weaker energy dependence of the Fermi function in the $K$-capture case favors the population of high-lying levels. As a result, the fissility of the even-even daughter nucleus is higher than that of odd daughter nuclei.

4. Prospects for studies of delayed fission

Substantial advances can be expected in all four regions of the nuclides undergoing delayed fission upon developing and employing new setups for producing isotopes and for studying their decay properties. Efforts should primarily be focused on precise measurements of the half-lives of isotopes undergoing delayed fission, as well as on measuring $P_{DF}$, energy and mass distributions of fission fragments. For fissile isotopes in the lead region, it is necessary to continue direct measurements of $A$ and $Z$ distributions of fission fragments with the aim of reliably establishing their mass distributions and clearly separating the symmetric and asymmetric regions — that is, finding a transition from asymmetric fission to symmetric fission in the mass region $A = 180–204$.

The possibility for observing shape isomers may arise for transuranium nuclei having a high $Q_{EC}$ and a two-humped fission barrier (Fig. 3). Upon electron capture, levels both in the first and in the second potential well can then be populated in the daughter nucleus. Observing gamma transitions accompanying the population of levels in the second well, one can calculate the nuclear shape deformation upon fission. Moreover, the observation of daughter-nucleus fission, along with gamma radiation accompanying it, may provide information about the shape of the fission barrier and about the probability for tunneling through the barrier at various excitation energies of this daughter nucleus.
Figure 3. Scheme of the population of levels of the daughter nucleus. The horizontal arrows indicate (1) delayed and (2) spontaneous fission of nuclei. The deformation of the daughter nucleus is schematically illustrated below (3).

Now, it is an open question, whether superheavy transifornium elements undergo delayed fission. Based on the foregoing, it can be concluded that the delayed fission process is usually observed for odd-odd nuclei if the K-capture energy $Q_{EC}$ is comparable with the height of the fission-barrier $B_f$ in the formed daughter nucleus.

There is an overview of theoretical and experimental studies devoted to the synthesis and “stability” of superheavy nuclei [16]; the dominant decay modes for $Z \sim 107, N \sim 162$ odd and odd-odd deformed nuclei and $Z \sim 110, N \sim 182$ spherical nuclei are K-capture in the first and beta decay in the second potential well.

Model calculations of $P_{DF}$ were performed in [17] for neutron-rich nuclei of transifornium elements (in particular, dubnium) in the vicinity of the $N = 184$ shell after their beta decay and of its effect on nucleosynthesis in the $r$-process. The authors of these studies state that, in this region of nuclei, delayed fission should have a substantial effect on nucleosynthesis.

The qualitative analysis discussed below and aimed at estimating $P_{DF}$ for neutron-deficient isotopes of superheavy transifornium nuclei relies on the assumption that, after electron capture in neutron-deficient odd-odd nuclei, for which the energy $Q_{EC}$ is high, delayed fission of even-even daughter nuclei is more probable. The line in Fig. 4 shows the logarithm of $P_{DF}$ as a linear function of the difference $(Q_{EC} - B_f)$ for the known of delayed fission nuclides (see Fig. 2 from [21]).

This dependence can be used to estimate $P_{DF}$ for isotopes of transifornium elements. The values of the K-capture energy are taken from [18], the fission-barrier heights are calculated based on various models for daughter nuclei and taken from [19, 20]. The differences $(Q_{EC} - B_f)$ are given for odd-odd transuranium nuclei and nuclei of superheavy elements in the vicinity of $N = 162$. Fig. 4 shows $P_{DF}$ values estimated for odd-odd isotopes of $Z = 103–107$ elements vs. the difference $(Q_{EC} - B_f)$. They show that for the new superheavy nuclei, $P_{DF}$ may reach values between $10^{-5}$ and $10^{-4}$. 


Figure 4. Delayed-fission probability $P_{\text{DF}}$ as a function of the difference of energy $Q_{\text{EC}}$ released upon $K$-capture in the parent nucleus and the height of the fission barrier $B_f$ in the daughter nucleus. $P_{\text{DF}}$ values with horizontal error bars (slightly shifted from the straight line for the picture to be clearer) indicate the expected $P_{\text{DF}}$ for nuclei of superheavy transfermium elements: (filled square) $^{252}\text{Lr}$, (filled pentagon) $^{254}\text{Lr}$, (filled triangle) $^{256}\text{Lr}$, (inverted filled triangle) $^{258}\text{Lr}$, (empty diamond) $^{256}\text{Db}$, (left-oriented filled triangle) $^{258}\text{Db}$, (right-oriented filled triangle) $^{260}\text{Db}$, (filled hexagon) $^{262}\text{Db}$, (star) $^{260}\text{Bh}$, (filled circle) $^{262}\text{Bh}$, and (empty square) $^{264}\text{Bh}$, and (empty circle) $^{266}\text{Bh}$.

For transuranium nuclei having a high $K$-capture energy and featuring a two-humped fission barrier, the possibility of observing shape isomerism in delayed fission may arise. Upon electron capture, levels can then be populated both in the first and the second potential well of the daughter nucleus (see Fig. 3). Observing gamma transitions upon the population of levels in the second well, one can calculate the deformation of the nuclear shape in the fission process.

5. Conclusions

In addition to traditional regions of study of delayed fission of nuclei and features of this fission channel in traditional regions of nuclides (neutron-deficient nuclei in the lead region, neutron-rich nuclei in the region of actinium and protactinium, and neutron-deficient and neutron-rich nuclei of transuranium elements), the probability for delayed fission of atomic nuclei have been estimated for the neutron-deficient region of transfermium elements. Model calculations and rough estimates show that the probability of delayed fission in the considered regions of atomic nuclei ($Z \approx 103–107$) is available for experimental measurements at the new Super Heavy Elements (SHE) Factory at JINR. For the synthesis of nuclei in the neutron-deficient and neutron-rich regions, it is necessary to use both “cold” and “hot” methods for SHE synthesis. Investigation of properties of delayed fission, including measurements of mass and energy distributions of fission fragments and detection of gamma rays in the $\text{EC/\beta}$ decay of odd-odd nuclei, can be combined with the analysis of decay schemes for these superheavy nuclei.
Measurement of delayed-fission probabilities will make it possible to determine fission barriers in even-even daughter nuclei. This in turn will enable one to test model concepts underlying the calculation of fission barriers and the choice of beta-decay strength functions.

References
[1] Hahn O and Strassmann F 1939 Naturwissensch. 27 11.
[2] Petrzak K A and Flerov G N 1940 J. Phys. USSR 3 275; Flerov G N and Petrzak K A 1940 Phys. Rev. 58 89.
[3] Polikanov S M, Druin V A, Karnaukhov V A, MikheeV V L, Pleve A A, Skobelev N K, Subbotin V G, Ter-Akopyan G M, and Fomichev V A 1962 Sov. Phys. JETF 15 1016.
[4] Polikanov S M 1968 Sov. Phys. Usp. 11 22.
[5] Bjernholm S and Lynn J E 1980 Rev. Mod. Phys. 52 725.
[6] Druin V A, Skobelev N K, Fefilov B V, and Flerov G N 1964 Preprint P-1580 JINR Dubna.
[7] Kuznetsov V I, Skobelev N K, and Flerov G N 1966 Yad. Fiz. 4 279; 1967 Sov. J. Nucl. Phys. 4 202.
[8] Kuznetsov V I and Skobelev N K 1999 Phys. Part. Nucl. 30 666.
[9] Skobelev N K 1972 Yad. Fiz. 15 444; Sov. J. Nucl. Phys. 15 249.
[10] Gangrsky Yu P, Miller M B, Mikhailov L V, Kharisov I F 1980 Yad. Fiz. 31 306; 1980 Sov. J. Nucl. Phys. 31 162.
[11] Hall H L and Hoffman D C 1992 Annu. Rev. Nucl. Part. Sci. 42 147.
[12] Lazarev Yu A, Oganessian Yu Ts, Shirokovsky I V, Tretyakova S P, Utyonkov V K, and Buklanov G V 1987 Europhys. Lett. 48 93.
[13] Andreyev A N, Huyse M, and van Duppen P 2013 Rev. Mod. Phys. 85 1541.
Hall H L, Gregorich K E, Henderson R A, Gannett C M, Chadwick R B, Leyba J D, Czerwinski K R, Kadkhodayan B, Kreek S A, Hannink N J et al. 1990 Phys. Rev. C 42 1480; Hall H L, Gregorich K E, Henderson R A, Gannett C M, Chadwick R B, Leyba J D, Czerwinski K R, Kadkhodayan B, Kreek S A, Lee D M et al. 1990 Phys. Rev. C 41 618.
[14] Andel B 2018 Zakopane Conference on Nuclear Physics (August 26 – September 2, 2018, Zakopane, Poland) Book of Abstracts 101.
[15] Hofmann S 2015 J. Phys. G 42 114001.
[16] Panov I, Lutostansky Yu, and Thielemann F-K 2016 J. Phys.: Conf. Ser. 665 012060.
[17] Audi G, Bersillon O, Blachot J, and Wapstra A H 2003 Nucl. Phys. A 729 3.
[18] Möller P, Sierk A J, Ichikawa T, Iwamoto A, Bengtsson R, Uhrenholt H, and Eberg S 2009 Phys. Rev. C 79 064304.
[19] Jachimowicz P, Kowal M, and Skalski J 2017 Phys. Rev. C 95 014303.
[20] Skobelev N K 2018 Phys. At. Nucl. 8 500.