The study of the properties of spontaneously fissioning transfermium nuclei synthesized in the complete fusion reactions with heavy ions

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Abstract. This report describes a series of experiments aimed on investigating characteristics of spontaneous fission of nuclei in the exotic region of short-lived neutron-deficient isotopes with \(Z \geq 100\). Unique data on the decay properties of neutron-deficient isotopes \(^{244,246}\)Fm, \(^{250}\)No, \(^{252,254}\)No, \(^{256}\)Rf produced in the complete fusion reactions with accelerated heavy ions \(^{40}\)Ar + \(^{206,208}\)Pb \(\rightarrow\) \(^{246,248}\)Fm\(^*\), \(^{48}\)Ca + \(^{204,206}\)Pb \(\rightarrow\) \(^{252,254}\)No\(^*\), \(^{50}\)Ti + \(^{206,208}\)Pb \(\rightarrow\) \(^{256,258}\)Rf\(^*\) were obtained in experiments at the VASSILISSA (SHELS) separator combined with a neutron detector.

1. Introduction

Fission of heavy nuclei – both induced and spontaneous – is one of the most important fields of nuclear physics. The interest in fission has been driven by the fact that this process is associated with the radical rearrangement of nuclear structure and the movement of a large amount of nuclear matter. According to modern concepts, it is spontaneous fission (SF) that defines the boundaries of Mendeleev’s Periodic Table for heavy nuclei.

Partial half-lives have now been measured experimentally for most of the known SF nuclei with \(Z \geq 100\) [1]. The total kinetic energy (TKE) of fragments has been measured fairly accurately, and the mass and energy distributions of fragments have been built for several isotopes [1,2,3]. However, measurements of such an important characteristic as the average number of prompt SF neutrons have been made only for long-lived isotopes in the interval from uranium to californium [4,5], for a number of neutron-rich isotopes of fermium [5,6], and for \(^{252}\)No [7,8], \(^{259}\)Md and \(^{260}\)Md [9].

As a rule, SF nuclei, the properties of which have been described in a number of works, have half-lives long enough to investigate them using the so-called "off-line" techniques. Complications emerging while studying the properties of spontaneous fission of heavy nuclei can be explained by the difficulty
in obtaining sufficient quantities of isotopes to study, as well as by the impossibility to use "off-line" methods for measuring the characteristics of spontaneous fission of short-lived nuclei.

2. Experimental setup
A significant improvement in the efficiency of heavy nuclei studies has been made using detector systems allowing one to obtain inclusive data on the characteristics of decays of the studied nuclides. Even greater progress in the investigation of nuclei with small production cross-section has been made by means of combined detector systems located in the focal plane of separators suppressing background products by several orders of magnitude.

![Figure 1. Schematic view of the focal-plane detector surrounded by the neutron counters and moderator.](image)

Such an approach to experiment performance has been implemented at the electrostatic recoil separator VASSILISSA [10] (after deep modernization – SHELS [11, 12]) used to study production cross-sections and radioactive decay properties of the nuclei - evaporation residues (ER), observed in complete fusion reactions with heavy ions accelerated in the U400 cyclotron of FLNR, JINR. The combined detector system, placed at the focal plane of separator, consists of 54 $^3$He-filled neutron counters mounted in a plastic moderator around the assembly of multi-strip silicon detectors, providing detection of SF fragments and alpha particles in the geometry close to $4\pi$ (see figure 1) [13]. Neutron detectors with $^3$He-filled counters are typically used for experimental studies of prompt spontaneous-fission neutrons because of their constant high efficiency in a broad range of neutron energy (in thick detectors). The main advantages of the $^3$He-based neutron detector system are a practically zero energy threshold, the absence of cross-talk and a low sensitivity to gamma rays. They have stable parameters during long measurements with low intrinsic background. The geometry of the detectors can be easily chosen for various experimental demands. A $^{248}$Cm neutron source was used to measure the efficiency of the single neutron registration at the beginning and at the end of the experiment. Depending on the configuration of the counters and moderator, the efficiency in the experiments varied within 36-45%.

The focal-plane detector assembly was housed in a cylindrical vacuum chamber of 120mm in diameter. Neutron counters were placed around this chamber in three concentric rings (see figure 1). When the heavy ($A > 40$) ion beam intensity was about 0.5 pμA on the target, the counting rate of background neutrons in the detector room was equal to 100 Hz. The location of the detectors in the separator focal plane allows studying the properties of radioactive decay of short-lived isotopes ($T_{1/2} > 2\ \mu$s, which is determined by the ER time of flight through the separator). The focal-plane semiconducting Si detector system allows measuring the energy of both fission fragments from the spontaneous fission of the implanted ER. The detector array consists of five identical 16-strip silicon wafers. The active area of a single silicon strip detector is 60×60 mm. As for the stop detector, each of its strips is position sensitive in the vertical direction with a position resolution of 0.5mm (FWHM) for sequential $\alpha$-$\alpha$ decays, 0.8mm for ER-$\alpha$, and 1.0mm for ER-SF events. The average energy resolution is about 25 keV for the standard $\alpha$-source. Four wafers are mounted in the backward hemisphere facing
the stop detector. They measure escaping alphas or fission fragments, and the total geometrical efficiency is 85% of 4π [13].

3. Experimental results and discussion

At the experimental campaigns of 2008 – 2010 years using VASSILISSA separator and 2014-2017 years using SHELS separator this methodology was realized for collecting the decay properties of short-lived neutron-deficient SF-isotopes with Z ≥ 100.

In the year 2008, the properties of SF neutron-deficient short-lived isotope $^{246}$Fm produced in the complete fusion reaction $^{40}$Ar + $^{208}$Pb = 2n + $^{246}$Fm were investigated. More than 100 events identified as spontaneous fission of $^{246}$Fm isotope were obtained. For the first time in the experiment, the mean number of neutrons per SF was measured $\bar{\nu} = 3.6 \pm 0.5$ and neutron multiplicity distribution was restored [14].

In 2010, two experiments were carried out [15]. These experiments measured TKE of fission fragments and neutron yields of $^{252}$No and $^{244}$Fm SF isotopes. In this experiment, nuclei of $^{252}$No were synthesized in the complete fusion reaction $^{48}$Ca + $^{206}$Pb = 2n + $^{252}$No with cross-section about 200 nb at the maximum of the excitation function. As a result, within one day of exposure, about 2000 events of $^{252}$No spontaneous fission were detected, which allowed making high-precision measurements of neutron yields and testing the operability of the entire detector system. The sufficiently high rate of $^{252}$No spontaneous fission events was used to calibrate the “fission section” of the focal Si detector array. The post-neutron TKE value of 198.7±1.2MeV is taken from ref. [16]. For the study of the decay properties of $^{244}$Fm isotope the complete fusion reaction $^{40}$Ar + $^{206}$Pb = 2n + $^{244}$Fm was used. The production cross-section of the reaction was about 3 nb, more than 200 events associated with the spontaneous fission of $^{244}$Fm were detected. It was the first time when SF neutron yields were measured for this neutron-deficient isotope. The average number of neutrons per spontaneous fission of $^{244}$Fm was $\bar{\nu} = 3.3 \pm 0.3$. Furthermore, for the $^{244}$Fm nucleus it became possible to estimate the fragment TKE calculated by summing the signals in the focal and lateral semiconducting detectors. Taking into account the calibration data obtained from $^{252}$No TKE, this value was approximately 198 ± 15 MeV. It was in good agreement with the earlier findings [17]. In addition, the half-life of $^{244}$Fm nucleus was measured. The value of 3.47 ± 0.26 ms consistent with the value obtained in [1].

The experience accumulated over the years of operating the VASSILISSA separator has allowed summing up the requirements to build a new separator, make ion-optical calculations and design the necessary equipment. In the year 2013, the new kinematic separator named «SHELS» (Separator for Heavy ELement Spectroscopy) underwent all tests and was prepared for the physics experiments. For the registration of heavy ER in the focal plane of the separator, a new improved system with a 48x48 double-sided silicon stripped detector (DSSSD) assembly, 60x60mm in size, and surrounded by backward position sensitive detectors was developed [18].

In 2014 the isotope $^{256}$Rf had been investigated after production in the reaction $^{50}$Ti + $^{208}$Pb = 2n + $^{256}$Rf. In total, there were more than 1500 events of spontaneous fission of this isotope detected [19]. It was the first time when the average number of prompt neutrons per spontaneous fission ($\bar{\nu} = 4.47 \pm 0.09$) for $^{256}$Rf isotope was determined. The measured half-life was 5.75 ± 0.17 ms, which was consistent with the previously published results [1]. In the data processing not a single α-decay related to the $^{256}$Rf decay was found. Therefore, the probability of spontaneous fission for this isotope is close to 100%, which is also consistent with the existing data [1].

Next experiment was aimed at investigating the properties of $^{250}$No nucleus synthesized in the complete fusion reaction $^{48}$Ca + $^{208}$Pb → $^{250}$No + 2n. Previously, the decay properties of this isotope had been studied in two experiments using the same reaction of complete fusion [20, 21]. Both experiments featured two spontaneously fissioning activities with half-lives different by one order of magnitude. In [20], the longer-lived activity was referred to the fission of $^{249}$No isotope, in [21] - to the fission of $^{250}$No from the isomeric state. Our experiment, as well as the previously published studies, registered two short-lived activities ($t_{1/2} = 5.1 \pm 0.3$ μs and $t_{1/2} = 36 \pm 3$ μs) that can be related to the spontaneous fission of neutron-deficient nobelium isotopes. Having compared the values of the ER yields with the estimated
excitation function, we came to the same conclusion as the authors of [21] – the registered activities can be referred to the spontaneous fission of $^{250}$No nucleus from the ground ($t_{1/2} = 4.86 \pm 0.25$ μs) and isomeric ($t_{1/2} = 60 \pm 4$ μs) states [22].

Table 1. The results of calculating and measuring $\bar{\nu}$ and mean total kinetic energies.

| Nucleus | Estimated$^a$ $\bar{\nu}$ | Experimental $\bar{\nu}$ | Estimated$^a$ TKE (MeV) | Experimental pre-neutron TKE (MeV) |
|---------|-----------------|------------------|------------------|-------------------------------|
| $^{252}$Cf | 3.6             | 3.77±0.01$^c$    | 186              | 184.1±1.3$^c$                |
| $^{244}$Fm | 3.5             | 3.3±0.3$^b$      | 196              | 195±14$^b$                   |
| $^{246}$Fm | 3.6             | 3.6±0.5$^b$      | 196              | 199±4$^c$                    |
| $^{250}$No | 4.3             | 4.38±0.22$^b$    | 202              | 192±14$^b$                   |
| $^{252}$No | 4.1             | 4.06±0.12$^b$    | 201              | 198.7±1.2$^c$                |
| $^{254}$Rf | 4.7             | 3.87±0.34$^b$    | -                | -                            |
| $^{256}$Rf | 4.6             | 4.47±0.09$^b$    | 208              | 220±15$^b$                   |

$^a$ Using the model from [24].

$^b$ Our experimental results.

$^c$ From literature.

In a recent experiment performed at the SHELS separator, observed in an irradiation of $^{206}$Pb with $^{50}$Ti about 150 fission events identified as a spontaneous fission of $^{254}$Rf. For the first time the mean number of neutrons per SF was measured $\bar{\nu} = 3.87 \pm 0.34$ [23].

In the present work we used an improved scission-point model [24] which is powerful in describing various experimental data on binary and ternary fission of heavy nuclei: mass and charge distributions of fission fragments, their kinetic energies, prompt fission neutron multiplicities, etc. The fissioning nucleus at the scission point is considered as a dinuclear system consisting of two touching spheroidal fragments. The excitation energy of the nuclear system at scission is calculated as a difference between the potential energy of the fissioning nucleus in the ground state and the potential energy of the dinuclear system at the scission point. The potential energy of the system is a sum of the liquid-drop energies of each fragment, the deformation-dependent shell correction terms, and the energy of the Coulomb and nuclear interaction of the fragments. The model allows us to calculate the potential energy of the dinuclear system as a function of its parameters: mass and charge asymmetries and deformations of the fragments. The average values of these parameters are found using the statistical approach.

The deformations of the fragments play an important role in the model. Since the fragments are significantly deformed at the scission point, the deformation energy released after their separation increases the excitation energy of the fragments. The excitation energy is exhausted by the neutron evaporation and subsequent gamma emission. The number of evaporated neutrons is calculated on the assumption that each emitted neutron reduces the excitation energy by its separation energy and has on average a kinetic energy twice the temperature of the fragment. Averaging over the distributions of the parameters of the scission configurations, we obtained the average number of the emitted neutrons. As an improvement to [24], we took into account that some part of the excitation energy remained after the emission of neutrons is exhausted by gamma emission. We estimated this part of the excitation energy to be equal on average to the energy of emission of a half of a neutron that reduces the average neutron multiplicities by 0.5. The results of calculating and measuring $\bar{\nu}$ and mean total kinetic energies are summarized in table 1. The experimental data on the average number of neutrons per fission $\bar{\nu}$ (obtained for SF of Pu-to-Dp isotopes) make up the systematic of $\bar{\nu}$ depending on the atomic mass (see figure 2). Here, one can observe a considerable growth of $\bar{\nu}$ with an increase in the fissioning nucleus mass at a fixed Z. It starts at A > 242 and is most obvious for Cm and Cf isotopes, for which the increase is almost linear, and it continues up to A=254.

4. Conclusion
The methods implemented at the kinematic separator of FLNR, JINR, allow synthesizing large quantities of nuclei of exotic transuranic elements. After the launch of the Superheavy Element Factory of FLNR, where the intensity of the accelerated heavy-ion beams will be increased by several times,
these methods will significantly supplement the current data on the SF properties of superheavy isotopes with $Z > 100$ produced in complete fusion reactions, decaying mainly in spontaneous fission and having very short lifetimes.

The schedule of experiments using the SHELS separator and the detector system with a neutron detector includes continuation of detailed studies of the spontaneous fission of even-even isotopes of 104, 106 and 108 elements, for which it is possible to collect a reasonable amount of statistical data within one month. It is also planned to study the spontaneous fission of more neutron-rich isotopes of Fm, No, Rf and Sg using $^{238}$U, $^{242}$Pu, $^{248}$Cm targets and high-intensity beams of $^{22}$Ne accelerated ions. A one-month experimental period will be enough to collect a sufficient amount of statistical data to determine the decay properties with acceptable accuracy.

Acknowledgments
This work was supported by the Russian Foundation for Basic Research, contracts No 17-02-00867 and No 18-52-15004.

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