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THE SEARCH FOR SPONTANEOUS FISSION FROM THE ^{250}No ISOMERIC STATE VIA PROMPT NEUTRON MULTIPLICITY DISTRIBUTION ANALYSIS

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Поиск спонтанного деления изомерного состояния ^{250}No с помощью анализа распределения множественности мгновенных нейтронов

Исследовались две активности ^{250}No, получаемого в реакции полного слияния ^{204}Pb(^{48}Ca, $2n$)^{250}No. Всего 1357 событий получено с использованием детектирующей установки для регистрации нейтронов SFiNx. Исследование было направлено на поиск различий в распределениях кратностей мгновенных нейтронов спонтанного деления, отнесенных к каждой наблюдаемой активности. Однако согласно статистическим тестам два распределения оказались статистически неразличимыми. Среднее число испущенных нейтронов на акт спонтанного деления ^{250}No получено равным $4{,}1 \pm 0{,}1$. С использованием метода статистической регуляризации Тихонова восстановлена и впервые получена форма распределения кратности мгновенных нейтронов спонтанного деления для ^{250}No. Несмотря на значительное количество экспериментов, список открытых вопросов касательно ^{250}No все еще обширен и требует продолжения исследований.

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The Search for Spontaneous Fission from the ^{250}No Isomeric State via Prompt Neutron Multiplicity Distribution Analysis

The complete fusion reaction ^{204}Pb(^{48}Ca, $2n$)^{250}No was used to study two activities of ^{250}No with distinct half-lives. A total of 1357 events were observed in the SFiNx neutron detection system. The study aimed to differentiate prompt neutron multiplicity distributions of ^{250}No for the ground and the K-isomeric states. However, the distributions for both activities were indistinguishable according to statistical tests. The average number of neutrons emitted per spontaneous fission of ^{250}No was determined to be 4.1 ± 0.1. Using the Tikhonov regularization method, a prompt multiplicity distribution was presented for the first time. Despite significant efforts, numerous questions about the ^{250}No isotope remain, emphasizing the need for continued research.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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INTRODUCTION

Experimental investigation of heavy and superheavy nuclei (so-called transfermium nuclei with $Z > 100$) is an advanced task due to the small production cross sections and short half-lives of these nuclei. The investigation of structural properties on the nanobarn and subnanobarn scale requires longer irradiation times followed by effective separation and detection methods. A systematic study of the decay properties of these exotic nuclei will bring us closer to the answer to the question concerning the limit of nuclear existence in terms of mass and charge.

Most of the known nuclei in the transfermium region decay via α decay or spontaneous fission (SF). SF is one of the most complex processes in nuclear physics, mainly due to the variety of possible output configurations. In the SF process, it is a frequent practice to divide the whole process into two parts. The first one consists of the mother nucleus overcoming multiple fission barriers by quantum tunneling, which determines the SF half-life of the nucleus. The second one is the evolution of a fission system on the potential energy surface from the penetration to the fission point. It defines the properties of fission fragments such as kinetic and excitation energies.

Our experimental investigation is to study the multiplicity distribution of prompt neutrons emitted by excited fission fragments in the SF process. The average number of neutrons per SF event helps to describe the static part of the process, i.e., the configuration of the statistical equilibrium. In addition, it was previously shown [1, 2] that an asymmetrical shape of the neutron multiplicity distribution could indicate the existence of additional SF modes.

The classical approach to describing the SF process considers fission from the ground state. At the same time, the existence of long-lived K-isomeric states is a common feature [3] and the question about the possibility of observing spontaneous fission directly from the high-K states arises. However, theoretical investigations predict high hindrances for such events [4, 5] mostly due to wider fission barriers for the isomeric state in comparison with the ground state.

The existence of a long-lived K-isomeric state in combination with a large neutron deficit makes ^{250}No an interesting candidate for research. An SF activity of 36 μs was first reported for ^{250}No in [6]. Subsequently, two activities (5.6 and 54 μs) were observed in [7], but the longer-lived activity was wrongly attributed to ^{249}No. Later on, using the mass analyzer FMA [8], it was confirmed that both activities belonged to ^{250}No. An interesting result was then reported in [9], in which the average multiplicities of prompt neutron emission in the SF corresponding to each activity were measured for the first time. The difference between the average number of neutrons per SF for the short- and long-lived states was found to be quite large (4.38 ± 0.13 and 3.9 ± 0.2), but still statistically insignificant (the difference $\approx 2\sigma$). Nevertheless, this result strongly hinted at a possible SF directly from the isomeric state. In [10], the internal decay branch from the isomer was observed for the first time, allowing for a first estimate of the excitation energy of the high-K isomer and indicating that the longer-lived SF activity could be due to the delayed SF of the ground state. The dependence of the isomer population probability on the excitation energy was first shown in [11] and finally, experimental evidence for the existence of a second isomeric state in ^{250}No as well as arguments against SF of the lower-lying isomer were given in [12].

The main aim of the current work was to verify the experimental results of [9] using a more advanced experimental setup and larger sample size to clarify if there is evidence of K-isomer fission due to difference in prompt neutron multiplicity distributions.

1. EXPERIMENTAL DETAILS

The complete fusion reaction $^{204}\mathrm{Pb}(^{48}\mathrm{Ca}, 2n)^{250}\mathrm{No}$ was used to produce nuclei of interest. The beam of ^{48}Ca ions was delivered from the U-400 cyclotron with an energy of (226.5 ± 2.0) MeV at the FLNR of JINR. Isotopically enriched ^{204}PbS targets (450 μg/cm^2 thickness, 99.94% enrichment and with 1.5 μm thick Ti backing) were used. The estimated energy in the middle of the target was (215 ± 2) MeV. The desired evaporation residues (ER) were separated from other reaction products and the primary beam moved out of the target using the velocity filter SHELS (Separator for Heavy ELement Spectroscopy) [13].

The ERs pass the time-of-flight (TOF) system that is placed just after the separator. The TOF system consists of mylar foils covered in a 30–40 μg/cm^2 thick gold layer and micro-channel plates (MCP) [14]. An external magnetic field is used to guide the emitted electrons to the sensitive part of the MCPs. The TOF signals help to distinguish whether the signal in the main detection system has been caused by ER implantation or decay inside the detector.

The detection system SFiNx [15] was used in the experiment. It consists of a box-like assembly of Double-sided Silicon Strip Detectors (DSSD) surrounded by 116 neutron counters filled with ^{3}He at 7 atm pressure. The high granularity of the neutron detector allows it to detect multiple neutrons simultaneously. The average neutron lifetime in the setup is (18.4 ± 0.2) μs. The single neutron detection efficiency calibration was carried out using the source of ^{248}Cm. It was assumed that the energy spectrum of prompt SF neutrons of ^{250}No does not differ significantly from ^{248}Cm [16]. The rapid dependency between the single neutron detection efficiency and the geometry of the fission fragment emission was noted. In the case where only one fission fragment was detected in the DSSD box, the efficiency was reduced. It could be easily described by a correlation between neutron emission direction and the axis of SF in the laboratory frame. To achieve higher detection efficiency with smaller uncertainty, only SF events for which both fragments were detected in the DSSD box were taken into account during the calibration. The single neutron detection efficiency was $(56 \pm 1)\%$.

The DSSD detector which is placed at the focal plane of SHELS consists of 128×128 strips, and its area is 100×100 mm^2, while the eight side 16×16 DSSD detectors of 50×60 mm^2 are located around the focal plane DSSD in the backward direction. The DSSD energy calibration at α-particle energies was carried out using the ^{174}Yb(^{48}Ca$, xn)^{222-x}$Th reaction.

2. RESULTS

The search for SF events was conducted through the identification of time-correlated ER–SF chains. The energy of implanted recoils detected at the focal plane, coinciding with the TOF signal, is required to be in the range of 1–20 MeV. Subsequent to an ER implantation, fission events exhibiting energies exceeding 40 MeV were examined in the same detector pixel in the time window of 500 μs. The analysis yielded 1357 ER–SF correlations, which equates to an approximate total of $6 \cdot 10^{17}$ incident ^{48}Ca ions passed through the ^{204}Pb target.

The ER–SF time difference distribution histogram is shown in Fig. 1. The time-logarithmic scale was used to extract half-lives according to [17–19]. The dead time of the detection system was 4.5 μs (or ≈ 1.5 in log scale). The

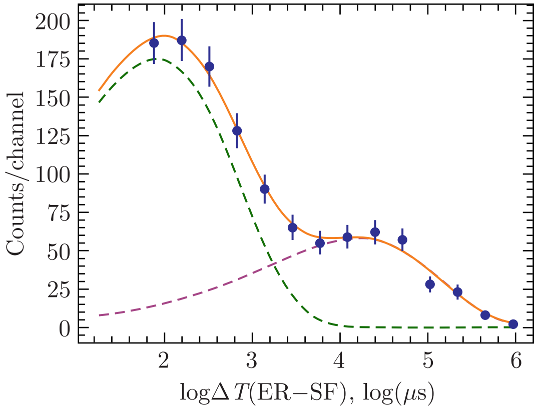

Fig. 1. ΔT(ER–SF) time distribution. The experimental data (blue circles) are fitted by a two-component exponential curve (orange solid line). The short-lived (green dashed line) and the long-lived (purple dashed line) components were extracted from the fitting

decay curve for a single activity f_s as a function of time t, decay constant λ, and the initial number of radioactive nuclei n is presented in Eq. (1), as well as the fitting function for double decay curve f_d as a function of two sets of parameters and the background constant C.

$$\begin{cases} \theta = \log(t), \\ f_s(\theta, n, \lambda) = \left|\dfrac{\Delta N}{\Delta\theta}\right| = n\,\exp(\theta + \log\lambda)\exp(-\exp(\theta + \log\lambda)), \\ f_d(\theta, n_1, \lambda_1, n_2, \lambda_2, C) = f_s(\theta, n_1, \lambda_1) + f_s(\theta, n_2, \lambda_2) + C. \end{cases} \tag{1}$$

The fitting quality has been verified using the chi-square test ($\chi^2 = = 4.7$ p-value $= 0.79$). The half-lives of the two activities extracted from the fitting are (4.7 ± 0.2) and (46 ± 4) μs. By extrapolating the fitting curve to the zero-time moment, it is possible to estimate the total number of events for each activity. For the short- and long-lived components, the expected numbers of nuclei are 1510 ± 50 and 500 ± 50, respectively. Using these numbers, the isomer population probability was estimated to be 0.25 ± 0.02. This value is in agreement (Fig. 2) with previously published results [8, 10–12] for the same reaction. The background constant was found to be 0.

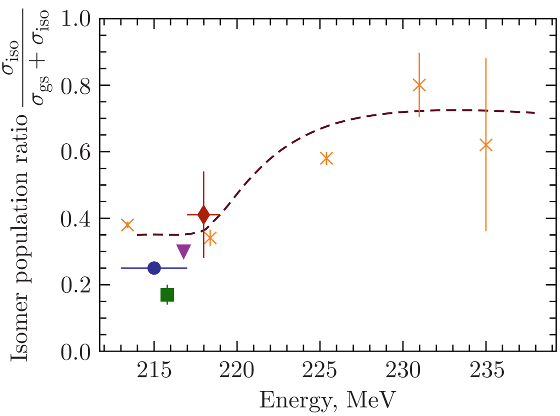

Fig. 2. The isomer population probability dependence on the energy of ^{48}Ca ions in the middle of the ^{204}Pb target. The circle represents the results of the current work. Other depicted experimental data include the triangle from Peterson et al. [8], the rhombus from Kallunkathariyil et al. [10], crosses from Tezekbayeva et al. [11], and the square from Khuyagbaatar et al. [12]. The dashed line corresponds to the theoretical predictions for isomer energy $E_{\rm iso} = 1.2$ MeV taken from [10]. See text for details

The first question regarding neutron multiplicities is if there is a significant difference in neutron multiplicities for SF events associated with short- and long-lived activities. Figure 1 shows that the activities are strongly mixed. To solve this issue, the following likelihood functions attributed to the short-lived activity $P(A|\theta)$ and the long-lived activity $P(B|\theta)$ were used:

$$\begin{cases} P(A|\theta) = \dfrac{f_s(\theta, n_1, \lambda_1)}{f_s(\theta, n_1, \lambda_1) + f_s(\theta, n_2, \lambda_2)}, \\ P(B|\theta) = 1 - P(A|\theta). \end{cases} \tag{2}$$

The first way to build two prompt neutron multiplicity distributions corresponding to the two activities was to attribute every SF event to both activities with weights equal to the corresponding likelihoods (Eq. (2)). The resulting distribution is shown in Fig. 3, *a*. The following list of statistical tests has been conducted: t-test for the mean value comparison (p-value $= 0.20$)

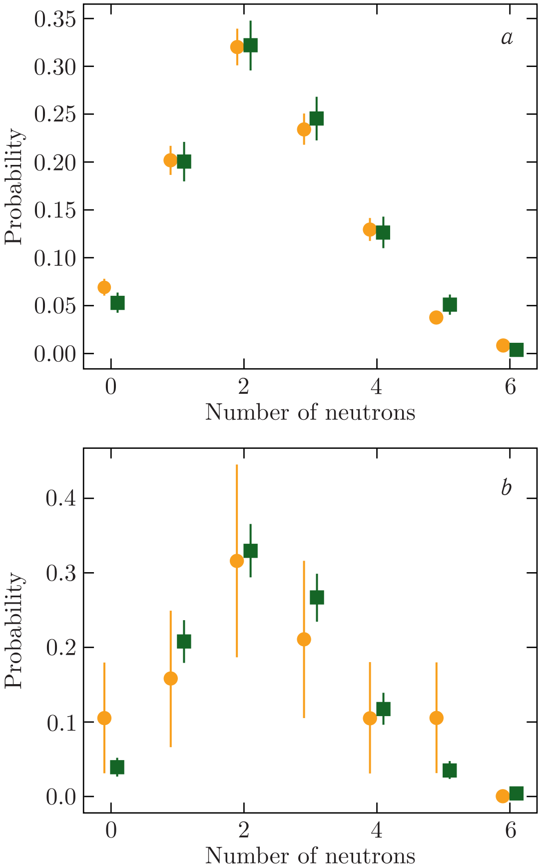

Fig. 3. Prompt neutron multiplicity distributions attributed to short-lived (orange circles) and long-lived (green squares) activities. *a*) All SF events have been used with weight coefficients. *b*) Only SF events that could be surely attributed to the activity have been used. See text for details. Markers have been shifted along the horizontal axis for better visual comparison

and the Kolmogorov–Smirnov (p-value = 0.57) test for the comparison of the shapes of distributions. According to both tests, no evidence was found to suggest dissimilarities between the distributions of Fig. 3, *a*.

An alternative approach involves considering only those SF events that can be confidently attributed to the specific activity, i.e., with likelihood value (Eq. (2)) not less than 95%. While this method simplifies the process, it also reduces the sample size and increases uncertainties. The resultant distributions are displayed in Fig. 3, *b*. The same series of statistical tests were conducted, including the t-test (p-value = 0.91) and the Kolmogorov–Smirnov test (p-value = 0.99). Once again, no evidence was found to indicate a significant difference between the two distributions. As there is no significant difference between activities concerning prompt neutron multiplicity distribution, all SF events will be considered in combination in the following discussion.

The background influence was measured in two different ways. The first one is the average load of the detector during the experiment. The value of ≈ 100 neutron/s (or 0.0128 neutron/window) for the whole neutron detection system was found. The second way is to search for background neutrons in coincidence with observed SF events but far enough in time so as not to include neutrons emitted in the fission process. To do so, the search for background neutrons was performed by analysis of 20 time windows of length 128 μs for each event of SF in the range of 2440–5000 μs. The average lifetime of the neutron in the setup is (18.4 ± 0.2) μs [15], so the overlapping with neutrons emitted in SF is negligible. The total number of observed time windows was 27 140 and the average background neutron multiplicity was 0.015 neutron/window. The results are shown in the table and are in good agreement with the average load of the detection system. The background neutron multiplicity distribution given in the table was taken to perform the background correction.

The observed number of events in the experiments N, the probability of detecting background neutrons of the given multiplicity f_b, and the restored emission probability ν for the prompt neutron multiplicity k for ^{250}No. See text for details

k	N	f_b	ν
0	32	0.9852	0.009 ± 0.007
1	100	0.0145	0.004 ± 0.006
2	169	$2.7 \cdot 10^{-4}$	0.102 ± 0.015
3	125	0	0.226 ± 0.018
4	63	0	0.281 ± 0.019
5	18	0	0.232 ± 0.020
6	3	0	0.120 ± 0.017
7	0	0	0.024 ± 0.013
8	0	0	0.002 ± 0.003
9	0	0	0.001 ± 0.003

The prompt neutron multiplicity distribution obtained in the experiment is highly distorted due to the detection efficiency (which is far from 100%) and due to the background influence. To achieve accurate results, only SF events that were observed in coincidence with the signal in side detectors ($\approx 40\%$ of a total amount of events) were taken into account while restoring the true distribution. The average number of emitted prompt neutrons per spontaneous fission of ^{250}No after background and efficiency correction was 4.1 ± 0.1.

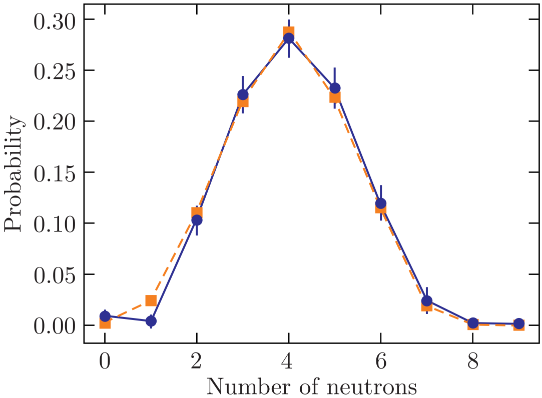

Fig. 4. Restored prompt neutron multiplicity distribution of ^{250}No obtained in the experiment (circles with uncertainty bars) and theoretical predictions (squares)

The distortion of the prompt neutron multiplicity distribution shape caused by the single neutron detecting efficiency has been corrected with the statistical Tikhonov regularization technique. The method adds extra information to the system like the smoothness or non-negativity of the resulting distribution. The description of the technique and the usage examples can be found in [20–22]. The restored prompt neutron multiplicity distribution is presented in the table and in Fig. 4.

3. DISCUSSION

^{250}No is the most neutron-deficient nobelium isotope for which the prompt neutron multiplicity distribution is analyzed. The shape of the distribution is symmetrical, no additional fission mode has been observed (Fig. 4). In the row of neighbor isotopes ^{252,254}No [15, 23], the SF properties do not look exotic and follow the systematic of an average number of neutrons per SF event: $\overline{\nu}_{250} = 4.1 \pm 0.1$, $\overline{\nu}_{252} = 4.25 \pm 0.09$ [15], $\overline{\nu}_{254} = 4.9 \pm 0.5$ [23].

To describe the obtained SF properties, the list of calculations has been performed. To calculate the neutron multiplicity, we assume that promptly after crossing a fission barrier, the fissile nucleus (Z, A) can be described as a superposition of binary systems specified by the mass, charge, and deformation of constituent fragments $n_i = (A_{1i}, Z_{1i}, \beta_{1i}, A_{2i}, Z_{2i}, \beta_{2i})$ with

$Z_{1i} + Z_{2i} = Z$ and $A_{1i} + A_{2i} = A$. Initially, the formed binary system evolves in deformation and mass(charge)-asymmetry degrees of freedom until it eventually decays in the relative distance. The potential energy of the binary system $U(n_i)$ as a function of masses, charges, and deformations of its fragments is calculated within the improved scission point (IMP) model [24]. The decay in relative distance is determined by the competition between repulsive Coulomb and attractive nuclear parts of the interaction potential between the fragments [25]. The evolution of the population of the state n with time is described by the master equation

$$\frac{dP(n)}{dt} = \sum_{n,n'} \left(\Lambda_{n,n'} P(n') - \left[\Lambda_{n',n} + \Lambda_d(n)\right] P(n)\right). \qquad (3)$$

The macroscopic transition probabilities are taken proportional to the level densities of the final states [26] $\Lambda_{n,n'}/\Lambda_{n',n} = \rho_{n'}(U_{CN} - U(n'))/\rho_n(U_{CN} - U(n))$, where U_{CN} is the energy of the fissile nucleus. The level densities are taken as for the Fermi gas with the level density parameter $a = A/12$ [2]. For the decay probability $\Lambda_d(n)$, the level density is taken at the top of the barrier in the interaction potential.

Solving Eq. (3), we obtain the distribution of the binary systems at the moment of decay. The fission observable is then determined by the properties of these binary systems [24].

The choice of the initial distribution of the binary systems is performed as explained in [2], by taking all the systems whose quadrupole moment lies in the 10% range around the fixed value Q_2. The Q_2 is fixed to give the best description of the average number of evaporated neutrons in spontaneous fission of nuclei with $Z \sim 100$ [27].

The model prediction for the neutron multiplicity distribution is in good agreement with experimental results (Fig. 4). This gives us confidence in the calculation results which we can use to discuss other SF properties that are impossible to measure in the present experimental setup: total kinetic energy (TKE) and fragment mass distributions which are shown in Fig. 5.

The next property observed in the current work is the K-isomer population probability (Fig. 2). As a result, it could be said that the population probability rapidly depends on the beam energy in the middle of the target in the range of 215–220 MeV. This range roughly corresponds to the 5–10 MeV excitation energy range of ^{250}No. Assuming the isomer to be K-isomeric 6^+ state [10] with an energy $E_{\rm iso} = 1.2$ MeV, we can estimate the isomer population probability as the ratio $\omega = W_{\rm sur}(E^*_{CN} - E_{\rm iso})/W_{\rm sur}(E^*_{CN})$ of the survival probabilities of the isomeric and ground states [28]. The results of the calculations are presented in Fig. 2 as a dashed line and do not contradict experimentally the observed values.

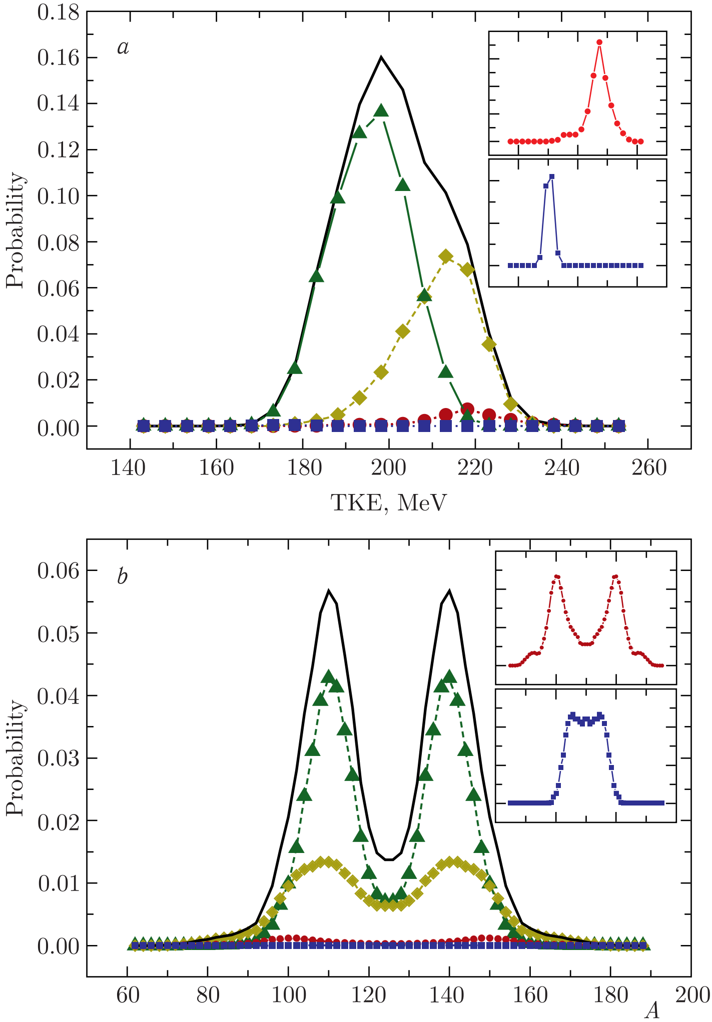

Fig. 5. Calculated total kinetic energy (*a*) and fragment mass (*b*) distributions. Solid line corresponds to the total distribution, while dashed lines correspond to decomposition per group of neutrons: circles — 0–1 neutrons; triangles — 2–3 neutrons; rhombuses — 4–7 neutrons; squares — 8–9 neutrons. Due to the low probability to emit 0–1 and 8–9 neutrons, the corresponding curves are shown in isolation in the same x-axis scale

CONCLUSIONS

The experimental study of spontaneous fission properties of ^{250}No nuclei produced in the hot-fusion reaction ^{204}Pb(^{48}Ca, $2n$)^{250}No has been carried out.

Two activities with half-lives of (4.7 ± 0.2) and (46 ± 4) μs were observed. The main aim of the work was to determine whether the analysis of the prompt neutron multiplicity distributions could provide strong evidence of the prevalence of spontaneous fission of the K-isomeric state of ^{250}No compared to the delayed fission of the ground state. To do this, 1357 SF events were analyzed. Two methods were used to attribute SF events to their rightful activity. According to statistical tests, the average values of the extracted distributions as well as the shapes of the distributions do not differ significantly. The achieved results can not give a clear answer as to whether the long-lived activity of ^{250}No is caused by the direct spontaneous fission from the K-isomeric state or by delayed fission of the ground state after electromagnetic transitions. It can be concluded that the analysis of the prompt neutron multiplicity distribution does not allow us to confidently answer the question as expected from the results of [9].

The average number of neutrons per SF of ^{250}No has been determined to be 4.1 ± 0.1. The prompt neutron multiplicity distribution emitted in SF of ^{250}No has been restored using the Tikhonov regularization method and published for the first time. In addition, the probability of populating the isomeric state has been estimated and compared with the previously published values.

Despite a large number of conducted experiments to study ^{250}No, the list of open questions is still comprehensive. For example, the energy of the K isomer as well as its configuration and the total number of isomeric states are still unclear. The ^{250}No isotope definitely creates challenges for experimental and theoretical groups and is worth further investigation.

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