Eleven new heaviest isotopes of elements Z=105 to Z=117 identified among the products of ^{249}Bk+^{48}Ca reactions

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Eleven new heaviest isotopes of elements $Z = 105$ to $Z = 117$ identified among the products of ^{249}Bk+^{48}Ca reactions

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The heaviest isotopes of elements Z=117 to Z=105, ^{294}117, ^{293}117, ^{290}115, ^{289}115, ^{286}113, ^{285}113, ^{282}Rg, ^{281}Rg, ^{278}Mt, ^{274}Bh and ^{270}Db, were identified by means of the Dubna Gas-Filled Recoil Separator among the products of the ^{249}Bk+^{48}Ca reaction. The details of the observed six decay chains indicating the production and decay of isotopes ^{293}117 and ^{294}117 are presented and discussed. The decay energies and resulting half-lives of these new nuclei show a strong rise of stability with increasing neutron number validating the concept of the island of enhanced stability for superheavy nuclei [Yu. Ts. Oganessian *et al.*, Phys. Rev. Lett. **104**, 142502 (2010)].

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I. INTRODUCTION

The liquid-drop model, which treats the atomic nucleus as a drop of incompressible fluid of nucleons, was formulated over 70 years ago [1]. It was particularly successful in the reproduction of measured nuclear masses. However, the liquid drop model predicts that all nuclei having over 100 protons cannot form a bound system, and will disintegrate by fission. A proper description of the binding energy of the heaviest nuclei requires careful consideration of the quantum structure of the nucleus; applying the so-called "shell correction" [2] enhances the stability of the heaviest nuclei against the spontaneous fission (SF) process.

There is still an open question as to how many protons and neutrons can be held inside a nucleus. This depends on the properties of the single-particle states in the heaviest nuclei. Practically all advanced nuclear structure models predict the existence of an "Island of Stability" of nuclides around a new spherical doubly-magic nucleus (see, e.g., [3, 4] and references therein). However, while the neutron shell closure at N=184 is a common result of model calculations, there is no consensus about the identity of the next magic proton number among the different models. Progress in understanding the structure of the heaviest nuclei can be achieved through the studies of production and decay of superheavy elements (SHE) [5] as well as through γ-spectroscopy studies of $Z > 100$ nuclei [6, 7].

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The syntheses of the heaviest known and most neutron-rich superheavy elements have involved the fusion of doubly magic ^{48}Ca ions with radioactive actinide targets. These experiments were pioneered at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. The first superheavy elements that were synthesized in Dubna were the even-Z nuclei (with $Z = 114$ and 116) produced in fusion reactions of ^{244}Pu and ^{248}Cm targets with ^{48}Ca projectiles [8, 9]. In subsequent experiments of ^{48}Ca-induced reactions with the long-lived even-Z target nuclei ^{238}U, ^{242}Pu, ^{245}Cm and ^{249}Cf, other even-Z nuclides with $Z = 112$ - 118 have been synthesized [10–12]. Eight of the isotopes of elements 114 and 116 with $N = 172$ - 177 and the isotope ^{294}118 produced in these reactions decay primarily through α-emission ($T_\alpha \leq T_{SF}$). The decay chains of consecutive emissions are terminated by the spontaneous fission of descendant even-even or even-odd nuclei with $Z = 114$, 112 or 110 ($T_\alpha > T_{SF}$). The total decay times T are in the range of about 0.1 s to 1 min depending on the number of neutrons in the superheavy nucleus (see review [5] and references therein).

The probabilities of formation and the decay properties of all 18 new nuclides with $Z = 104$ - 118 produced in these reactions provide evidence for a considerable increase in nuclear stability with increasing neutron number in the nucleus. At the limits of Coulomb stability, the experimental data support the prediction that shell stabilization lowers the ground-state energy, creates a fission barrier, and thereby enables SHE to exist. Thus, one of the fundamental outcomes of the nuclear shell model, namely the existence of an "Island of Stability" in the domain of the superheavy elements has received strong experimental support. Parts of these data were

recently confirmed in independent experiments [13–19] for elements with $Z = 112$ and $Z = 114$.

In the reactions of actinide nuclei with ^{48}Ca the decay properties of the neighboring odd-Z nuclei have also been investigated [20–22]. For the neighboring odd-Z elements, especially in the odd-odd isotopes, the probability of α-decay with respect to spontaneous fission should increase due to the strong hindrance of SF caused by unpaired nucleons. For instance, the isotopes of elements 113 and 115 synthesized in the ^{237}Np, ^{243}Am + ^{48}Ca reactions [20–22], have longer consecutive α-decay chains that were terminated by the SF of neutron-rich isotopes with $Z = 105$.

The decay patterns of odd-Z nuclei with larger neutron excess are of particular interest for nuclear theory. According to theoretical models, the increased stability of superheavy nuclei is determined by spherical proton and neutron shells located at $Z = 114$ (or 120, or 126) and at $N = 184$. In the course of a series of α-decays, the effect of these shells should gradually become weaker for descendant isotopes with smaller proton and neutron numbers. However, the nuclei at the end of the decay chains are located near the deformed shells at $Z = 108$ and $N = 162$, and their stability against spontaneous fission should again increase. Indeed, the decays of ^{282}113, ^{287}115 and ^{288}115 after four or five α-decays, end in spontaneous fission of the isotopes $^{266-268}$Db ($N = 161$ - 163) with long half-lives of 0.3 h, 1.2 h and 29 h, respectively [20–22] or end with the β^+/EC decay followed by the SF of presumably short-lived even-Z Rf isotopes. In these chains all the nuclei with odd atomic numbers $Z = 107$ - 115 located in the transition area between the shells at $N = 184$ and $N = 162$ mainly undergo α-decay ($T_\alpha < T_{SF}$).

A further increase in the neutron excess of the parent nucleus with odd Z should result in a considerable increase in the stability of superheavy nuclei with $Z \geq 111$ when approaching the shell at $N = 184$. On the other hand, in the neutron-rich nuclei of lighter elements (with $Z = 105$ - 109), the extra neutron excess would result in a decrease in their spontaneous-fission half-lives due to their increased distance from the deformed shell at $N = 162$. In this situation the decay chain could be terminated at an earlier stage and spontaneous fission would dominate for nuclei characterized by a minimum shell effect. In a new domain of superheavy nuclei whose stability is determined entirely by nuclear shell effects, such a picture would give an indication as to the limits of the influence of closed nuclear shells and the areas where they overlap.

Unfortunately, in the absence of the appropriate target material, more neutron-rich nuclei with $Z \leq 115$ can be obtained only as products of the α-decay of heavier nuclei, such as the decay daughters of element 117 isotopes that can be produced as evaporation residues in the ^{249}Bk + ^{48}Ca reaction. However, synthesizing a new superheavy element in this reaction is complicated by certain technical challenges. First, it requires the production of a considerable amount of special target material, the rare isotope ^{249}Bk ($T_{1/2}$=320 d), in a high flux reactor. In addition, one would expect a low cross section for the production of nuclei with $Z = 117$. This requires a highly sensitive experiment. The maximum cross section of the ^{249}Bk(^{48}Ca, xn)$^{297-x}$117 reaction is expected to be at the level of 1.0-1.5 pb for x=3, 4. This estimate is based on experimentally measured cross sections for the formation of the superheavy nuclides in reactions with ^{48}Ca, in particular, for the neighboring nuclei with $Z = 116$ [10, 11] and $Z = 118$ [12]. These extrapolated values agree well with calculated cross sections [23–25].

Similarly we can estimate the decay properties of the unknown nuclei from the measured decays of nearby nuclides. From our estimates and the calculations by A. Sobiczewski [26], the nuclei ^{293}117 and ^{294}117, in the absence of competition with spontaneous fission, will undergo seven sequential decays leading to the very neutron-rich isotopes ^{265,266}Lr with half-lives T_α, and $T_{\beta^+/\mathrm{EC}}$ that presumably would exceed ten days. The point where these α decay chains will be terminated by SF depends on increasing fissionability (weakening of the effect of nuclear shells) as the products of the sequential α-decays move away from the closed neutron shells at $N = 162$ and $N = 184$. In addition, bearing in mind the ratio of the decay probabilities (α/SF) in the neighboring even-Z nuclei with various neutron numbers, one would expect that the daughters and grand-daughters of the decay of $Z = 117$ nuclei will most likely undergo α-decay. Their half-lives are expected to be in the range of tenths of a second ($Z = 115$) to seconds ($Z = 113$).

These expected longer half-lives can open up unique opportunities for studying the chemical properties of the heaviest elements, in particular for determining their chemical behavior relative to that of their lighter homologues - Bi and Tl. Investigations of the differences in chemical behavior caused by "relativistic effects" in the electronic structure of superheavy atoms, that have been observed in the Cn/Hg pair [13, 15] as well as the 114/Pb pair [17], could be extended to elements with $Z = 113$ and possibly $Z = 115$.

The heaviest nuclei are synthesized in small quantities and selected from many events recorded during long experiments lasting often over one hundred days. Therefore, the evidence for new decay chains has to be carefully verified against potential random sequences. Here, in more detail, we present the synthesis of the isotopes ^{293,294}117 in ^{249}Bk+^{48}Ca reactions as well as the extended discussion of the measured yields and decay properties that were presented concisely in an earlier publication [27]. As predicted, the eleven new more neutron-rich isotopes produced in these reactions and their decay products are found to have longer half-lives providing further support for the increased stability of isotopes as they approach $N = 184$.

II. EXPERIMENT

The ^{249}Bk was produced at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL) by intense neutron irradiation of seven targets (see Fig. 1) containing ∼50 grams of actinides, predominately Cm (42 grams), Am (5 grams), and Pu (3 grams). Target fabrication and assembly was accomplished remotely in the hot cells at the Radiochemical Engineering Development Center (REDC) at ORNL. Each target incorporated a maximum thirty-five actinide pellets of nominal 0.25-inch diameter and 0.571-inch length, with each pellet consisting of a tubular aluminum pellet liner and cermet core blend of CmO_2 microspheres and aluminum powder compacted to ∼80% theoretical density. Actinide pellets and aluminum spacer components were assembled into a finned aluminum tube with coolant shroud for insertion into the hexagonal target array in the HFIR flux trap. A diagram of an assembled target is shown in Fig. 1. In the course of the 18-months campaign, which ended in December 2008, the neutron fluence was about $4.5{\cdot}10^{22}$ cm^{-2}.

Before starting the chemical processing, the irradiated targets were stored for three to four months to allow ^{131}I to decay to an insignificant level. The first chemical processing step was the dissolution of the aluminum matrix of the targets using a heated solution of NaOH and $NaNO_3$. The insoluble actinides (and most fission products) were removed by filtration and then dissolved in strong HNO_3. After the dissolution, the actinides were separated from most metallic impurities (e.g., Al, Cs, Sr, Zr, Ru, Mo) using a batch solvent extraction step [28]. In this process the starting HNO_3 aqueous solution was adjusted to low acidity (∼0.02 NH^+), the actinides were extracted into an organic solvent containing di-2-ethylhexylphosphoric acid (HDEHP), the solvent was washed with dilute HCl, and the purified actinides were then back-extracted with 6 *M* HCl. For the next step the actinide solution was adjusted to 12 *M* LiCl-0.1 *M* H^+ and pumped through a column of anion exchange resin [29]. The lanthanide and actinide elements were both loaded on the column and were then chromatographically separated using different eluant solutions. The transcurium elements were then precipitated using LiOH, filtered, washed with demineralized water, and re-dissolved in HNO_3. The transcurium elements were then pumped through a column of very small diameter cation resin using a high-pressure pump [30]. The transcurium elements were chromatographically separated by pumping solutions with alpha-hydroxyisobutyric (AHIB) acid of different concentrations and pH through the column. The ^{249}Bk fractions from this run contained ≤1% of the ^{252}Cf but this was still orders of magnitude higher than what can be safely taken from the hot cells. The final separation step [31] involved adjusting the feed solution to 8 *M* HNO_3, oxidizing the berkelium to the Bk^{+4} valence with $NaBrO_3$, extracting the Bk^{+4} into a solvent containing HDEHP, washing with 8 *M* HNO3, and finally reducing the berkelium to Bk^{+3} valence and back-extracting with a solution of 8 *M* HNO_3 - 1 *M* H_2O_2. Overall the hot cell processing effort took two months of round the clock effort. The final solution that was taken to the glove box facilities for final processing was ∼500 mL in volume and contained $\sim 5{\cdot}10^{-7}$ g of ^{252}Cf, $<1.9{\cdot}10^7$ Bq of ^{106}Ru, $<7.4{\cdot}10^7$ Bq of ^{144}Ce which represent overall decontamination factors for the hot cell processing of $\sim 3{\cdot}10^5$, $>6{\cdot}10^4$, and $>2{\cdot}10^5$, respectively.

After initial operations to remove organics remaining in the solution from the solvent extraction process and to remove the excess acid, the material was loaded on a small cation resin column to wash metallic and chemical impurities from the material. The material was then pressured through a column of very small diameter cation resin using inert gas. The berkelium and californium elements were chromatographically separated by pressuring AHIB solutions of different concentrations and pH through the column. The ^{249}Bk material was then loaded on a small cation resin column to wash chemical impurities from the AHIB separation away from material and to concentrate the material in a very small volume of ultra-pure HCl. The final solution was 5.1 mL in volume and contained $2.22{\cdot}10^{-2}$ g of ^{249}Bk and $7.45{\cdot}10^{-10}$ g of ^{252}Cf, $<4.1{\cdot}10^4$ Bq of ^{141}Ce, $<1.7{\cdot}10^5$ Bq of ^{144}Ce and $<8.1{\cdot}10^4$ Bq of ^{106}Ru. After the berkelium chloride solution was dried, the solids were dissolved using concentrated ultra-pure HNO_3 and the resulting solution was dried. The solids were dissolved a second time using concentrated ultra-pure HNO_3 and dried to ensure complete conversion of the material to the nitrate form. After the second conversion sequence, the material was dissolved in 5 mL of ultra-pure HNO_3, and the solution was equally divided into five quartz shipping cones that were taken to dryness before shipment packaging. The glove box processing and preparations for shipment lasted 7 days.

At the end of June 2009 the material was delivered to Research Institute of Atomic Reactors at Dimitrovgrad for making the target. Six targets were manufactured from this material, by depositing BkO_2 oxide onto 0.74 mg/cm^2 Ti foils to a thickness of 0.31 mg/cm^2 of ^{249}Bk. Each target had an area of 6.0 cm^2 in the shape of an arc segment with an angular extension of 60 degree and an average radius of 60 mm. The segments were mounted on a disk that was rotated at 1700 rpm such that the target was perpendicular to the direction of the incoming beam direction while the beam was moved up and down vertically over the target actinide deposit in order to dissipate heat over the whole target. In the course of the bombardment with an intense beam of ^{48}Ca, the target layers of BkO_2 were systematically monitored by counting particles from the decay of the ^{249}Cf - the β-decay daughter of ^{249}Bk. At the start of the experiment 37 μg/cm^2 of the ^{249}Cf daughter was present in the target.

The experiments on the synthesis of element 117 were performed employing the Dubna Gas-Filled Recoil Separator (DGFRS) [32, 33] and the heavy-ion cyclotron U-400, FLNR (JINR). The experimental setup is shown in

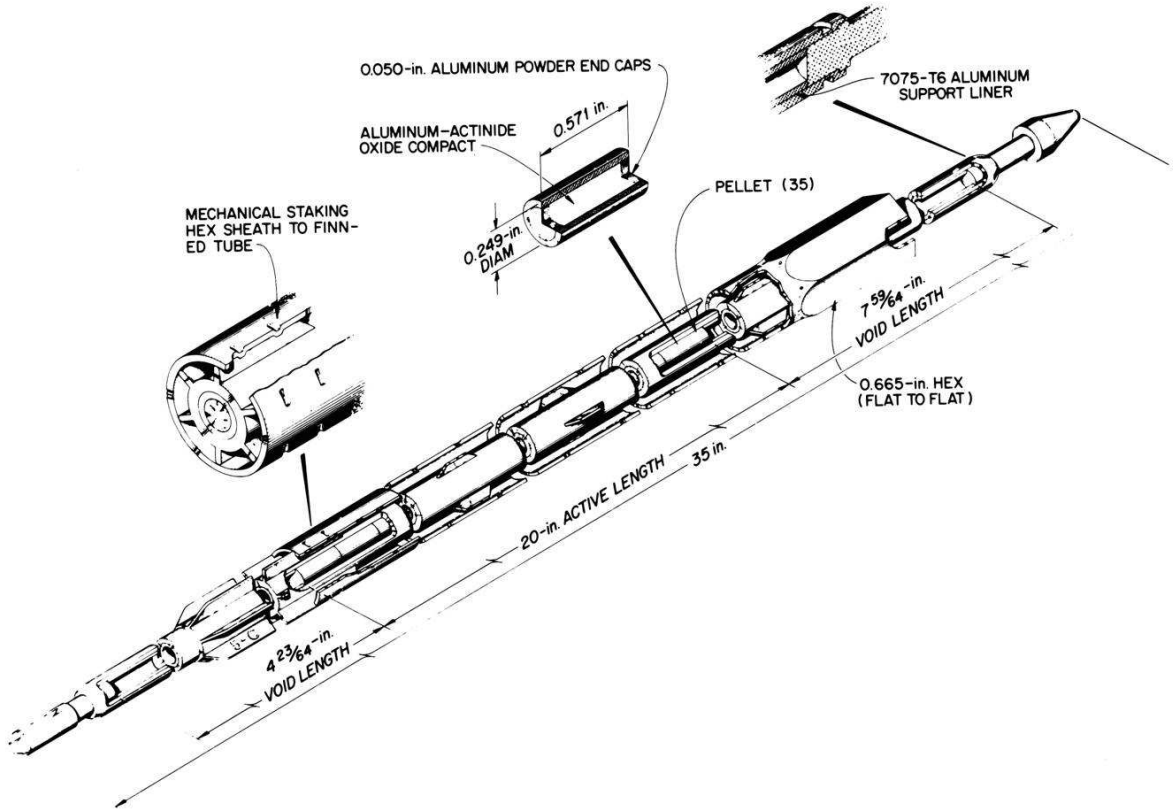

FIG. 1: Diagram of a recycle Cm target for HFIR irradiation.

Fig. 2. The products of the complete fusion reactions recoiled from the target and were separated in flight from beam particles and other reaction products before being implanted in detectors mounted at the focal plane of the separator at a distance of about 4 m from the target. The flight time trough the separator was ~ 1 μs. The transmission efficiency of the separator for Z=117 nuclei is about 35% [33] whereas full-energy ^{48}Ca projectiles, projectile-like ions, and target-like nuclei were suppressed by factors of $3{\cdot}10^{15}$, $3{\cdot}10^{13}$, and 10^4-10^6, respectively.

Evaporation residues passing through the separator were registered by a time-of-flight (TOF) system with a detection efficiency of 99.9% and were implanted in a 4 cm by 12 cm semiconductor detector array with 12 vertical position-sensitive strips. The detection efficiency of these focal-plane detectors for α particles with $E_\alpha \approx$ 10 MeV emitted from the decays of the implanted nuclei is about 52% of 4π. To detect escaping α's, the strip detector was surrounded with eight 4 cm by 4 cm side detectors without position sensitivity, forming a box open to the front (beam) side (see Fig. 2). In this geometry, the position-averaged detection efficiency for full-energy particles from the decays of implanted nuclei increases to 87% of 4π.

The detection system was calibrated by registering the recoil nuclei and decays (α or SF) of known isotopes of No and Th and their descendants produced in the reactions ^{206}Pb(^{48}Ca,2n) and ^{nat}Yb(^{48}Ca,3-5n), respectively. The full-width-at-half-maximum (FWHM) energy resolution for α-particles implanted in the focal-plane detector was 60-140 keV, depending on the strip and the position within the strip. Alpha particles that escaped the focal-plane detector at different angles and were subsequently registered in a side detector had an energy resolution of 160-230 keV for the summed signals (side detector plus residual focal-plane detector). If the energy deposited by an α-particle as it recoiled out of the focal-plane detector was lower than the detection threshold of 0.9-1.1 MeV (such that its position was also lost) and it was detected only by a side detector, its total energy was estimated as the sum of the energy measured by the side detector and half of the threshold energy ($\approx$ 0.5 MeV) with an uncertainty in determining the total energy increased to $\approx$0.4 MeV (68% confidence limit *c.l.*). The assignment of these α-particles to the observed decay chains was made using the calculated probability of random correlations based on the decay rate in the side detectors associated with the actual experimental conditions. The FWHM position resolutions from the signals of correlated decays of nuclei implanted in the detectors were 1.1-1.3 mm for ER-α signals and 0.4-0.8 mm for ER-SF signals. If an α-particle was detected by both the focal-plane ($E_{\alpha 1}$) and

a side detector ($E_{\alpha 2}$) (i.e., E_α=$E_{\alpha 1}$+$E_{\alpha 2}$), the position resolution was dependent on the amplitude of ($E_{\alpha 1}$) (see, e.g., Fig. 5 in [34]) but was generally inferior to that obtained for the full-energy signal.

Fission fragments F1 and F2 from the decay of ^{252}No implants produced in the ^{206}Pb+^{48}Ca reaction were used for the fission total kinetic energy (TKE) calibration. The measured fragment energies (E_{tot}=E_{F1}+E_{F2}) were not corrected for the pulse height defect of the detectors, the energy loss in the detector's entrance windows or dead layers, or the energy loss in the pentane gas filling the detection system (at a pressure of about 1.7 Torr) of the escaping fragment (E_{F2}). The average sum energy loss of fission fragments from the SF decay of ^{252}No [35] was measured to be about 23 MeV. Therefore the TKE of nuclei with Z>102 was determined to be the sum E_{tot}+23 MeV. The systematic uncertainty in estimating the TKE value through this method is about 5 MeV when both fission fragments are detected.

From model calculations [26] and the available experimental data for neighboring odd-Z and even-Z nuclei, one can estimate the expected α-particle energies for the isotopes of element 117 and their descendant nuclei that could be produced in the ^{249}Bk+^{48}Ca reaction. This allowed us to employ a special low-background detection scheme for the nuclei to be investigated [36]. In the bombardment of ^{249}Bk, the beam was switched off after a recoil signal was detected with parameters of implantation energy expected for Z=117 evaporation residues, followed by an α-like signal with an energy of 10.7≤E_α ≤ 11.4 MeV, in the same strip, within a 2.2-mm-wide position window (2 FWHM). The triggering ER-α time interval was set at 132 ms for E_α=10.7-10.8 MeV, linearly dependent on energy for E_α=10.8-11.3 MeV and was 18 ms for E_α =11.3-11.4 MeV. The beam-off interval was initially set at 3 min. In this time interval, if an α-particle with E_α =9.0-10.6 MeV was registered in any position of the same strip, the beam-off interval was automatically extended to 15 min. During this 15-min period, if other α-particles with energies expected for heavy nuclei were observed, we could prolong the beam-off pause further. This experimental running condition was a compromise between the inevitable loss of beam dose (5% of the beam time was lost) and the necessity of a beam pause for the registration of long-duration sequential decays of the daughter nuclides with $Z \leq 115$ under very low background conditions.

III. EXPERIMENTAL RESULTS

The Coulomb barrier for the ^{249}Bk+^{48}Ca reaction is $E_{C,lab}$=239 MeV [37]. With $E_{p,lab}$=252 MeV ^{48}Ca projectile energy in the middle of the target layer, the excitation energy of the compound nucleus ^{297}117 is E^*=39 MeV, corresponding to the expected maximum for the total ER cross section (sum of 3n- and 4n-evaporation channels [5, 8–12]. These channels result in the production of ^{294}117 and ^{293}117 with neutron numbers N = 177 and N = 176. The intensity of the ^{48}Ca-ion beam accelerated by the U400 cyclotron was 1.1-particle μA at the target. Irradiation of the ^{249}Bk target by 252-MeV ^{48}Ca projectiles was performed between July 27 and October 23, 2009. During the 70 days run, a beam dose of $2.4{\cdot}10^{19}$ projectiles was delivered to the target. The systematic uncertainty in the beam energy was 1 MeV. With the energy spread of the incident cyclotron beam, the small variation of the beam energy during irradiation, and the energy losses in the target (3.0 MeV), we expected the resulting ^{297}117 compound nuclei to have excitation energies between 37.2 and 41.4 MeV. Excitation energies of the compound nuclei are calculated using the masses found in [38, 39]. The beam energy losses in the separator's entrance window (0.74 mg/cm^2 Ti foil), target backing, and target layer were calculated using the Nuclear Data Tables [40, 41].

In this first ^{249}Bk+^{48}Ca experiment, we had 1512 beam interrupts, with a total beam-off time interval of 4748 min. The energy spectrum of the α-like signals registered by the front detector during 1680 hours of irradiating the ^{249}Bk target with 252 MeV ^{48}Ca, as well as the spectrum of α-particles registered during beam-off intervals is presented in Fig. 3a. As seen in Fig. 3, switching off the beam resulted in a considerable reduction of the counting rate in the detector. The background was mainly due to the decay of the known isotopes $^{211-214}$Po that are the daughters of heavier nuclei created from incomplete fusion reactions. In the energy range 8.8 MeV ≤ E_α ≤ 11.3 MeV where we expect the α-particles of the first five transitions in the chain 117→115→113→111→109→107 the counting rate was 0.17/s (with beam on) and 10^{-3}/s (beam switched off). Similar spectra of the fission-fragment-like signals with $E_F \geq$ 35 MeV measured under the same conditions are shown in Fig. 3b. In the energy range $E_F \geq$ 135 MeV the counting rates from the front detector were $1.2{\cdot}10^{-4}$/s (beam on) and $7{\cdot}10^{-5}$/s (beam off).

In four instances switching off the beam resulted in the observation of the decay chains presented in Table I. In the first decay chain, implantation of a 11.9 MeV recoil in the center of strip 10 (15.2 mm from the top of the strip) of the focal-plane detector was followed 17 ms later by an α-particle with $E_{\alpha 1}$=10.99 MeV and a position of 15.0 mm. Detection of this sequence caused the beam to switch off, and one more α decay and SF were detected over a total time interval of 56.4 s in the absence of a beam-associated background. The second particle was detected by the focal-plane detector with an energy E_α=9.72 MeV and a position of 15.4 mm, 16.2 s after emission of the first particle. The SF decay of the final nucleus in this chain was detected with E_F=203 MeV and a position of 14.9 mm, 40.2 s after the last α-decay.

During this experiment, 4 other decay chains were observed that were very similar to the first one. In three events, as in the first case, the daughter nuclei in the de-

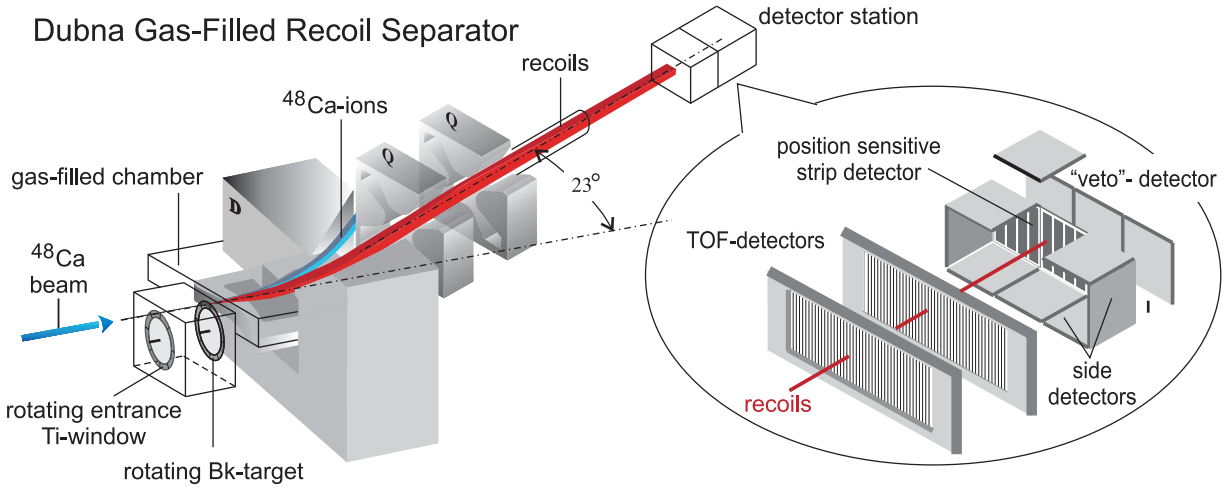

FIG. 2: Schematic of the Dubna Gas-Filled Recoil Separator (dipole magnet D followed by the quadrupole doublet Q_1 and Q_2). Also shown is a schematic of the detector station at the separator focal plane.

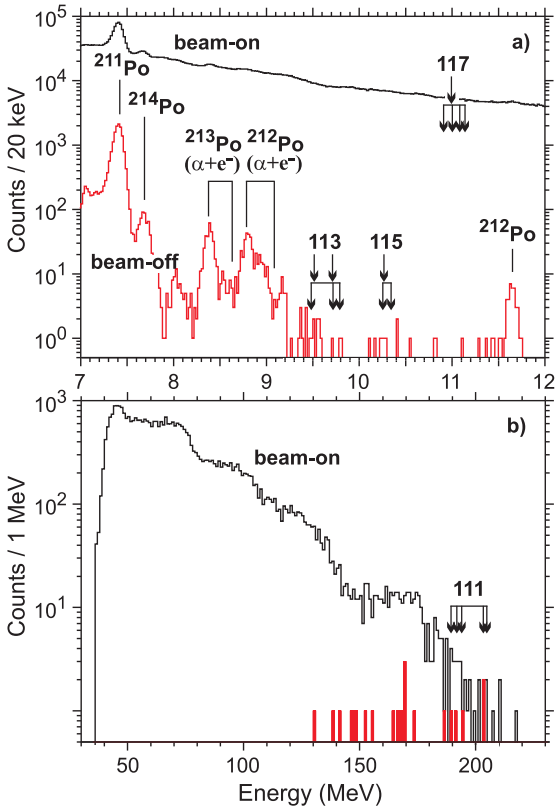

FIG. 3: Energy spectra [27] recorded during the 252 MeV ^{48}Ca+^{249}Bk run (E*=39 MeV). (a) Total energy spectra of beam-on α-like signals and beam-off α-particles. (b) Total fission-fragment energy spectra, both beam on and beam off. The arrows show the energies of events observed in the correlated decay chains; see Table I.

cay chains were detected with the beam turned off. These are shown in Table I enumerated 2, 4 and 5.

Event #2, which was similar to the first chain, consists of two sequential α-particles and spontaneous fission. After detecting the first α-particle with E$_\alpha$=11.14 MeV and switching off the beam, the side detector registered the second α-particle with E$_\alpha$=8.98 MeV in 2.2 s followed by SF in 4.3 s. The absence of the signal from the second α-particle in the front detector results in the position signal being lost and a subsequent energy determination of E$_\alpha$=9.52±0.43 MeV (see above). Attributing this particle to the given decay chain follows from the total counting rate of the side detectors with the beam switched off, $1.1\cdot10^{-3}$/s in the energy range E$_\alpha$=8.5-10.7 MeV. The probability of registering such an α-particle in a 6.5 s interval between the time- and position-correlated signals ER-α1-SF is less than 1%. In the spontaneous fission that terminates chain #2, two fission fragments were detected by the front and side detector that were coincident in time, with energy E$_{F1}$=185 MeV (this signal corresponds to the total energy of one SF fragment moving deeper into the detector plus part of the energy of the second fragment escaping the focal plane detector) and E$_{F2}$=7 MeV (part of the energy of the second fragment registered by the side detector); the sum energy was E$_{Ftot}$=E$_{F1}$+E$_{F2}$=192 MeV.

Chains #4 and #5 are also quite similar and give more complete information on the decay of the evaporation residues. In both cases after implantation of the recoil atoms in strips 8 and 10 with energies of 9.96 MeV and 9.36 MeV and emission in 53.0 ms and 20.2 ms of particles with energies of 10.91 MeV and 11.00 MeV, respectively, the beam was switched off. In further decays in strip 8 the sequential emission of two α-particles was observed: E$_{\alpha2}$ =10.25 MeV, t$_{\alpha2}$=0.51 s and E$_{\alpha3}$=9.79 MeV, t$_{\alpha3}$=0.24 s; then, in 31.7 s, a spontaneous fission with energy E$_{Ftot}$=E$_{F1}$+E$_{F2}$=153 MeV+36 MeV=189 MeV was observed. Similarly, in strip 10, after sequential emis-

TABLE I: Time sequences in the decay chains observed at two ^{48}Ca energies in the ^{249}Bk+^{48}Ca reaction.

Date		8/20	9/10	9/18	10/17	10/23		11/22
Time		09:40	12:28	04:50	04:23	01:37		22:57
Event #		1	2	3	4	5		6
Bρ, Tm[a]		2.37	2.37	2.42	2.36	2.42		2.35
Isotope		Strip 10	Strip 10	Strip 10	Strip 8	Strip 10	Isotope	Strip 5
ER(117)	E_{ER}, MeV	11.89	13.87	13.51	9.96	9.36	ER(117)	8.67
	y_{ER}, mm	15.2	22.7	17.2	22.0	13.2		19.2
^{293}117	$\delta t_{ER-\alpha1}$, ms	17.01	7.89	4.60	53.0	20.24		112.4
	$E_{\alpha1}$, MeV	10.99	11.14	11.08[b]	10.91	11.00	^{294}117	10.81
	$y_{\alpha1}$, mm	15.0	23.2	17.2	22.2	13.3		19.0
^{289}115	$\delta\ t_{\alpha1-\alpha2}$, ms			17.50	511.8	424.4		22.6
	$E_{\alpha2}$, MeV	-	-	10.34	10.25	10.27[b]	^{290}115	9.95[c]
	$y_{\alpha2}$, mm			17.4	22.2	10.9		
^{285}113	$\delta t_{\alpha2-\alpha3}$, s	16.17	2.23	1.17	0.238	13.49		28.29
	$E_{\alpha3}$, MeV	9.72	9.52[c]	9.71[c]	9.79	9.48	^{286}113	9.63
	$y_{\alpha3}$, mm	15.4			22.1	13.6		19.5
^{281}Rg	$\delta t_{\alpha3-SF}$, s	40.19	4.25	12.03	31.66	76.56		0.74
	E_{SF}, MeV	203	192[d]	205[d]	189[d]	194	^{282}Rg	9.00
	y_{SF}, mm	14.9	22.7	16.5	22.0	13.2		19.0
	$\delta t_{\alpha4-\alpha5}$, s							11.05
	$E_{\alpha5}$, MeV						^{278}Mt	9.55[b]
	$y_{\alpha5}$, mm							
	$\delta t_{\alpha5-\alpha6}$, s							76.10
	$E_{\alpha6}$, MeV						^{274}Bh	8.80
	$y_{\alpha6}$, mm							19.1
	$\delta t_{\alpha6-SF}$, h							33.36
	E_{SF}, MeV						^{270}Db	196[d]
	y_{SF}, mm							19.9
N_{ran}		$6{\cdot}10^{-6}$	$1{\cdot}10^{-3}$	$1{\cdot}10^{-5}$	$3{\cdot}10^{-11}$	$3{\cdot}10^{-11}$		$6{\cdot}10^{-11}$

[a] Bρ values were calculated for corresponding strips assuming constant rigidity of transported atoms and using a separator dispersion of 7.5mm/1%Bρ and separator rigidities of 2.26 Tm, 2.31 Tm, and 2.39 Tm at which decay chains 1-2, 3-5, and 6 were observed, respectively.

[b] α-particles with E$_\alpha$=11.08 MeV, 10.27 MeV, and 9.55 MeV were detected by both the focal-plane and side detectors with energies of 6.64+4.44, 1.24+9.03, and 1.10+8.45 MeV, respectively.

[c] Energies of events detected by side detectors only.

[d] SF events with E$_{SF}$=192 MeV, 205 MeV, 189 MeV, and 196 MeV were detected by both the focal-plane and side detectors with energies E$_{tot}$=185+7, 180+25, 153+36, and 152+44 MeV, respectively.

sion of two particles: with E$_{\alpha2}$=10.27 MeV, t$_{\alpha2}$=0.42 s (this particle escaped into the back hemisphere and was detected by the front and side detectors) and E$_{\alpha3}$=9.48 MeV, t$_{\alpha3}$=13.5 s, spontaneous fission was detected after 76.6 s with E$_F$=194 MeV.

Switching off the beam occurs only in cases when the full energy of an α-particle from the decay of the parent nucleus is registered by the front detector. The efficiency

of registering α-particles from the decay of the implanted nuclei by the front detector is 52%. Therefore, some decay sequences may be detected in full or partially in the presence of the beam on the target. Such a decay was registered in strip 10, as shown in Table I (chain #3).

Here, the first α-particle was registered by the front (6.64 MeV) and side (4.44 MeV) detector ($E_{\alpha 1tot}$=11.08 MeV), 4.6 ms after a 13.5 MeV recoil nucleus registered in the same position ($\Delta_{yER-\alpha}$=0.02 mm). The beam was not switched off and the first decay was then quickly followed by the second decay ($E_{\alpha 2}$=10.34 MeV). Then, in 13.2 s two fission fragments were observed, with sum energy $E_{Ftot}=E_{F1}+E_{F2}$=180 MeV+25 MeV=205 MeV. Between the second particle and the spontaneous fission no α-signals with an amplitude E >8.4 MeV in the focal detector or signals detected by both the focal and side detectors were observed. In the side detector, 1.17 s after the second particle an "α-like" signal was registered with an energy of 9.17 MeV. Taking into account possible energy loss in the focal detector this signal could correspond to an α-particle with E_α =9.71± 0.41 MeV. The probability of such an α-particle occurring at random within this chain is rather high; therefore this event was not taken into account when determining the properties of the synthesized nuclei. However, the probability of observing the decay chain of the ER- α_1- α_2-SF type as a sequence of chance events, as shown below, is at the level of 10^{-5} for this chain (see Table I).

By using the approach offered in [42] and the calculation method [43] for all five events, the probabilities of chance correlations imitating each of the observed decay chains were determined. The total number of ER-like events with E_{ER}=7.5-17.5 MeV that were followed by α-like events in the focal-plane detector with $E_{\alpha 1}$=11.03± 0.25 MeV occurring in the same strip, within a position window corresponding to ±FWHM ER-α position resolutions (98% c.l.), within five half-lives of the parent nucleus (71.2 ms) was 1223 (N_{ER}-α_1) during the 70 days duration of the experiment. The total numbers of particles with E_α=9.2-10.7 MeV registered by the focal-plane detector or by both the focal-plane and side detectors and spontaneous-fission events with E_{SF} >135 MeV observed during the total 3.3 d beam-off pauses were 35, 23, and 21, respectively. The probabilities of observing random beam-off particles ($P_{\alpha i}$) and SF events (P_{SF}) were calculated separately for each strip assuming their equal distribution on the strip and applying position windows of 5 mm and 2 mm for αs and SFs, respectively. The time intervals were 1.5 s, 0.5 min and 2.5 min for detection of isotopes of elements 115, 113 and 111, respectively. The total number of random sequences ER-α_1-α_3-SF for chains 1, 2 (without applying a position factor for α_3 in chain 2) and ER-α_1- α_2- α_3-SF for chains 4, 5 were calculated as a sum of corresponding values $N_{ER-\alpha 1} \cdot \Pi(P_{\alpha i}) \cdot P_{SF}$ for each strip.

The number of random ER-α_1-α_2-SF sequences for decay chain #3 observed with the beam on was calculated similarly. Here, the total number of ER- α_1 pairs was 438

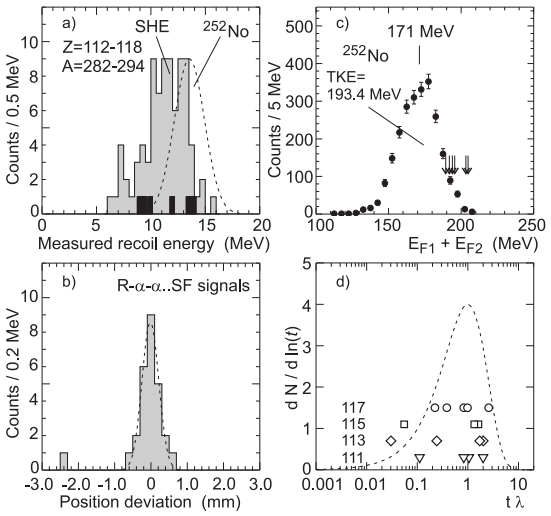

FIG. 4: (a) Energy spectrum of all ERs with Z = 112 - 118 produced in ^{48}Ca-induced reactions [5, 8–12, 20–22]. The energy distribution of ^{252}No implants measured in the calibration reaction ^{206}Pb+^{48}Ca is shown by the dashed line for comparison. Energies of Z = 117 ERs are shown by the black histogram. (b) Relative position deviations of all events in the decay chains observed in the reaction ^{48}Ca + ^{249}Bk (grey histogram) with a Gaussian fit (FWHM∼ 0.54 mm, dashed line). (c) The fission-fragment sum energy spectrum of ^{252}No. Energies of SF nuclei in the decay chains attributed to ^{293,294}117 are shown by arrows. (d) Time intervals t of all events in the observed decay chains originating from ^{293}117 multiplied by the average decay probability $\overline{\lambda}$ ($\overline{\lambda} = \ln 2/\overline{T}_{1/2}$) are compared to the average half-lives of respective nuclei ($t \cdot \overline{\lambda} = t \cdot \ln 2/\overline{T}_{1/2} = t/\overline{t}$). The dashed line is the expected time spectrum for correlated decays in $t \cdot \overline{\lambda}$ dimensionless units.

(α_1 is detected by both detectors), the number of beam-on α-particles with $E_{\alpha 2}$=10.31±0.27 MeV and SF events were $1.8 \cdot 10^5$ and 724, respectively. Applying a position window of 2.4 mm for α_2 and the same time intervals as for the other four chains we obtained $N_{ran}=1 \cdot 10^{-5}$. Results of the calculations of random probability for all five decay chains are given as the last line of Table I.

Let us consider now to what extent the measured characteristics defined by the experimental setup satisfy the criteria of observation of evaporation residues from ^{48}Ca-induced reactions and their subsequent decays. In this respect, we should note that the kinematics of the fusion reactions ^{249}Bk+^{48}Ca used in this work are practically the same as in all previous experiments where the mass of the target varied from ^{238}U to ^{249}Cf and the ^{48}Ca-beam energy changed by less than 30 MeV. This gives us an opportunity to compare all three kinds of registered signals: the energies of the recoiling nuclei and their decay by α-particle emission or spontaneous fission not only with the data from the reference reaction ^{206}Pb+^{48}Ca but also with the parameters of all 86 chains observed in

the synthesis of superheavy nuclei with Z = 112 - 118 [5, 8–12, 20–22].

As one can see from Fig.4a, the energies of all five recoiling nuclei (with their associated TOF signal) lie in the energy range 9.4 MeV $\leq E_{ER} \leq$ 13.9 MeV, in agreement with the energy values expected for the evaporation residues from the reaction ^{249}Bk+^{48}Ca. As seen in Fig. 4b, all the signals of the α and SF types subsequent to the signals of the ER implantation are in strict position correlation, which indicates that they belong to the decay of these implants. The average charge state determined for the five decay chains is $6.1^{+0.3}_{-0.2}$. This value is in agreement with data measured for Z = 112 - 118 nuclei produced in U-Cf+^{48}Ca reactions [5, 8–12, 20–22]. During the experiment the field in the dipole magnet was changed to shift the ER central trajectory to the middle of the focal plane detector array. The magnetic rigidities of recoils for each decay chain are shown in the fourth line of Table I.

When comparing the energy spectra of the signals that terminate the α chains with the sum energy spectrum of the fragments of spontaneous fission of ^{252}No (Fig. 4c) one can see that their sum energy $< E_{Ftot} >$=195 MeV is about 24 MeV higher than that characteristic of the SF decay of ^{252}No [35]. Hence, one can estimate the total kinetic energy of the observed spontaneous-fission fragments, $< TKE >\approx$218 MeV. Such a high sum energy of the fragments indicates the fission of a very heavy nucleus [11, 44].

In the spectra of α-transitions one can single out three groups of α particles differing in energy by 0.6-0.7 MeV. All five α particles emitted after the implantation of the recoils do not differ in energy from each other within the energy resolution of the focal plane detector. Their average energy is $E_{\alpha 1}$=11.03(8) MeV. The second group of particles, those of the daughter nuclei in chains #3, #4 and #5 (see Table I), $E_{\alpha 2}$=10.31(9) MeV, also do not differ in energy within the accuracy of the measurements. Finally, the third group includes all the decays preceding spontaneous fission. Four particles of this group lie in the energy range of 9.5-9.8 MeV. In chains #1 and #4 $E_{\alpha 3}$=9.74(8) MeV while in chain #5 $E_{\alpha 3}$=9.48(11) MeV. Here the difference in energy is greater than the detector's energy resolution, which might indicate a fine structure in α-decay of ^{285}113 isotope. From the time distributions of particles in each of the three energy groups $E_{\alpha 1}$, $E_{\alpha 2}$, $E_{\alpha 3}$ one can determine their half lives $T_{\alpha 1}$, $T_{\alpha 2}$, $T_{\alpha 3}$. Similarly, T_{SF} values can also be determined assuming that all the observed chains belong to the decay of one and the same nucleus (see Table II). Relative time intervals of all events in the observed decay chains (λ=ln2/$T_{1/2}$) compared with the average half-lives assigned to the appropriate nuclei are shown on Fig. 4d. In the figure, the decay time associated with each measurement is scaled to the above mentioned decay constant for the five measurements. This allows us to characterize each decay type by a single standard half-life value, bearing in mind the expected statistical distribution of values, indicated in the figure by the smooth curve. Therefore, one can conclude with high reliability, that in all five cases we observed decay chains originating from the same parent nucleus.

TABLE II: Decay properties of nuclei produced in the reaction ^{249}Bk+^{48}Ca.

Isotope	Decay mode	Half-life[a,b]	E_α(MeV)	Q_α(MeV)
^{294}117	α	78^{+370}_{-36}ms (30ms)	10.81±0.10	10.96±0.10
^{290}115	α	16^{+75}_{-8}ms (1.7s)	9.95±0.40	10.09±0.40
^{286}113	α	20^{+94}_{-9}s (4s)	9.63±0.10	9.76±0.10
^{282}Rg	α	$0.51^{+2.5}_{-0.23}$s (70s)	9.00±0.10	9.13±0.10
^{278}Mt	α	$7.7^{+37}_{-3.5}$s (0.3s)	9.55±0.19	9.69±0.19
^{274}Bh	α	53^{+250}_{-24}s (14s)	8.80±0.10	8.93±0.08
^{270}Db[c]	SF(α/EC)	23^{+110}_{-10}h	-	<7.9
^{293}117	α	14^{+11}_{-4}ms (8ms)	11.03±0.08	11.18±0.08
^{289}115	α	220^{+260}_{-80}ms (160ms)	10.31±0.09	10.45±0.09
^{285}113	α	$5.5^{+5.0}_{-1.8}$s (1.7s)	9.74±0.08 9.48±0.11	9.88±0.08
^{281}Rg	SF	26^{+25}_{-8}s	-	<9.4

[a] Error bars correspond to 68% confidence level [42]. Expected half-lives for allowed transitions shown in parenthesis were calculated using the formula of Viola and Seaborg [45] with parameters from [11] and measured Q_α values.
[b] The half-lives given for the decay of ^{294}117 and its daughters, where only one decay chain was observed, are obtained by multiplying the measured time interval by $ln(2)$.
[c] The identity of the fissioning nucleus that terminates the ^{294}117 chain is uncertain. This nucleus cannot have an atomic number Z>105.

It is evident that the main difference between chains #1 and #2 from #4 and #5 consists of non-observation of the decays of first daughter nuclei in the first two chains. Indeed, when registering 15 α transitions in 5 decay chains with an efficiency of 87% (see above) missing two α particles is quite probable. The properties of the nuclei in the decay chains of element 117 are shown in Table II.

It is most reasonable that the observed decay chains

originate from the isotope ^{293}117, produced in the complete fusion reaction ^{249}Bk+^{48}Ca followed by evaporation of four neutrons from the compound nucleus ^{297}117. We can support this conclusion through the application of the systematic trends of the cross sections σ_{xn}(E*) measured previously for production of isotopes of superheavy nuclei with Z=108 [46], 112-116 and 118 [5] in ^{48}Ca-induced reactions. Despite the strong overlap in energy of the excitation functions of the major channels of these reactions, evaporation of three and four neutrons, the relationship between the maximum values of the cross sections depends considerably on the number of neutrons in the compound nucleus. In the process of de-excitation of the neutron-rich compound nuclei ^{292}114 and ^{296}116 ($N-Z$=64) with an excitation energy of E*≈40 MeV formed in the reactions ^{244}Pu, ^{248}Cm+^{48}Ca, the peak cross section σ_{4n} is about three times higher than that of σ_{3n} [5, 10, 11]. On the other hand, in the compound nuclei ^{291}115 and ^{293}116 ($N-Z$=61) formed in the reactions ^{243}Am, ^{245}Cm+^{48}Ca, this relationship is σ_{4n}/σ_{3n} ∼ 0.3 [12, 20, 21]. These data suggest that de-excitation of the neutron-rich nucleus ^{297}117 ($N-Z$=63) with E*≈39 MeV would occur by the emission of four neutrons with the highest probability.

In addition, we can use SF half-life systematic to support the mass assignment. Predicted SF half-lives of even-even nuclei with Z = 110 - 114 and N = 168 - 176 [47, 48] differ by less than one order of magnitude for each N; their logarithms increase from about -1.8 to -1.5 for N=168, 170, and become increasingly larger for N = 172, 174, 176 (log T$_{SF}$(s)≈ 0.4, 2.8 and 5.1, respectively). The experimental data for the three even-even isotopes ^{282}Cn (N = 170) and ^{284}Cn/^{286}114 (N = 172) [5] confirm such behavior (log T$_{SF}$(s) ≈ -3.1 and -1.0/-0.6, respectively). The SF half-lives for even-odd ^{279}Ds and ^{281}Ds are in agreement with this pattern (log T$_{SF}$(s)≈ -0.7 and 1.0) and would be in quantitative agreement with predictions [47] and measured T$_{SF}$ values for ^{282}Cn,^{284}Cn and ^{286}114, if one applied the same hindrance factor of three orders of magnitude for T$_{SF}$ caused by the unpaired neutron in both Ds isotopes. Thus, when going from ^{279}Ds (N = 169) to ^{282}Cn (N = 170), then to ^{281}Ds (N = 171), and ^{284}Cn/^{286}114 (N = 172) the unhindered SF half-lives would increase by about one order of magnitude for each neutron number from 0.2 ms to 1 ms and further to 10 ms and 100/260 ms, respectively. If the increase of T$_{SF}$ remains the same between the odd-odd isotopes ^{280}Rg and ^{282}Rg, the T$_{SF}$ lower limit for ^{282}Rg should exceed 10^3 s because the corresponding value for ^{280}Rg is about 10 s [20, 21]. The estimated T$_{SF}$ value for ^{282}Rg is about 40 times larger than the half-life of the terminal SF nucleus in the five observed decay chains. Following this consideration, the spontaneous fission with T$_{1/2}$=26 s should belong to the Rg isotope with a *smaller and even* number of neutrons, ^{281}Rg.

To search for the 3n-evaporation products of the reaction ^{249}Bk+^{48}Ca, the experiment was continued at the ^{48}Ca energy E$_{lab}$ = 247 MeV. This resulted in an excitation energy of the compound nucleus ^{297}117 spanning E*=33.2-37.5 MeV, the centroid about 4 MeV lower than in the previous case. For the evaporation residues of the compound nucleus ^{297}117 at E* ≈ 35 MeV, the σ_{4n} cross section will be several times lower in comparison to its maximum value at E* ≈ 39 MeV, while σ_{3n} will remain practically the same. This experiment was performed with some interruptions between October 23, 2009, and March 1, 2010, with an integrated beam dose of 2.0·10^{19}. A new chain was detected involving six consecutive α decays and ending in spontaneous fission (see chain 6 on the right side of Table I). After the beam switched off triggered by the implantation in strip #5 of a recoil with energy E$_{ER}$=8.67 MeV and the emission of an α-particle with E$_{\alpha 1}$=10.81 MeV (Δy$_{ER-\alpha}$ = 0.27 mm) 112.4 ms later, all five subsequent α transitions were detected in the 3-minute beam-off pause. Then 13 minutes after detection of the last α particle (E$_{\alpha 6}$=8.80 MeV), the beam was automatically switched on. After 2.5 hours it was manually switched off again in order to wait for the spontaneous fission of the terminal nucleus. Spontaneous fission was observed 33.4 hours after emission of the last α particle. A spontaneous fission with energy E$_{Ftot}$ = E$_{F1}$ + E$_{F2}$ = 152 MeV+44 MeV=196 MeV occurring in strip #5 had a position deviation $|\Delta y|$ ≤0.96 mm with respect to the preceding decays. In this experiment, the counting rate of the beam-off SF events with E$_F$ ≥ 135 MeV was 0.1/h. Thus, the probability to observe a random SF event within 33 h in the same strip and position window of 2 mm is about 1.5%. This indicates that the fission event belongs to the given decay chain with high confidence. This is also the only high-energy (>142 MeV) fission event detected during this time period in the entire detector array.

The total number of ER(7-17 MeV)- α_1(10.55-11.3 MeV) events registered in the focal-plane detector in the same strip, within a position window corresponding to ±FWHM ER- α position resolutions and within 3.5 lifetimes of the parent nucleus (390 ms) detected during the 70 days duration of this experiment was 2366. The total number of α particles with E$_\alpha$=8.6-10.5 MeV registered completely or partially by the focal-plane detector or by the side detectors observed during the total 57 hours beam-off pauses were 113 and 22, respectively. The probabilities of observing random beam-off α particles and SF events were calculated similarly to the first experiment. Because of large uncertainties in determining the half-lives of nuclei observed in one decay chain, we used extended time intervals of 5 s, 10 min and 100 h for detection of isotopes of elements 115, 113-107 and 105, respectively. The total number of expected random sequences ER- α_1-...- α_6-SF (without applying a position factor for α_2 and α_5) for this chain was N$_{ran}$=6·10^{-11}. The results of the experiment performed at E$_{lab}$ = 247 MeV (E* = 35 MeV) are also given in Table II with single-event elapsed times converted to half-lives by application of $ln(2)$ (T$_{1/2}$=$ln(2)\cdot\tau$).

IV. DISCUSSION

The six decay events of the nuclei produced in the reaction ^{249}Bk+^{48}Ca presented in Table I were observed in the energy range 33.2≤E* ≤ 41.4 MeV. At these excitation energies, as pointed out above, the excited nuclei ^{297}117 will decay with emission of three or four neutrons. From the systematic trends in the experimental data for the reactions between actinide targets and ^{48}Ca, it follows that in this relatively narrow energy interval evaporation residues of superheavy compound nuclei are produced with the highest probability. The excitation functions σ_{3n}(E*) and σ_{4n}(E*) overlap in energy and the relationship of their maxima depends on the neutron binding energy and the fission barriers (both values depend on Z and N of the compound nucleus). In even-Z nuclei the products of 3n and 4n evaporation are well distinguished by their decay properties. In odd isotopes, due to the strong hindrance of the spontaneous fission caused by an odd neutron, the probability of α-decay considerably increases. Accordingly, α-transitions in even-odd nuclei combine into longer chains and the overall decay time appears to be considerably higher than for the neighboring even-even isotopes.

In odd-Z evaporation residues, the difference in decay properties of the neighbors with even/odd neutron numbers becomes less significant. Decay chains of the two isotopes of element 115 synthesized in the reaction ^{243}Am(^{48}Ca,3-4n)^{288,287}115 look similar: five sequential α-decays that result in the spontaneous fission of the isotopes ^{268,267}Db [20, 21]. The energies and half-lives of the decays of the parent nuclei ^{287}115 and ^{288}115, and daughter activities of ^{283}113 and ^{284}113 do not differ significantly within the energy resolution of the detectors and statistical dispersion of the decay times. The differences in the experimental values of E$_\alpha$, T$_\alpha$ arise only in the decay-product nuclei with $Z \leq 111$ and in the T$_{SF}$ values of the chain-terminating nuclei.

The experimental results presented in Table I obtained in the reaction ^{249}Bk+^{48}Ca show that a similar pattern is observed for the isotopes of element 117. As pointed out above, all five decay chains observed at E* ≈ 39 MeV can be assigned to the decay of ^{293}117 - the product of the 4n evaporation channel in the reaction ^{249}Bk+^{48}Ca. The properties of the nuclei formed in the consecutive decays of this isotope are presented in the lower section of Table II. As shown in Table II, the nuclei ^{293}117, ^{289}115 and ^{285}113 are α-emitters; each of them is characterized by an α-transition with a definite energy: E$_{\alpha1}$=11.03 MeV, E$_{\alpha2}$=10.31 MeV and E$_{\alpha3}$=9.74 MeV/9.48 MeV, respectively. The nucleus ^{281}Rg in all five cases undergoes spontaneous fission with $T_{SF} = 26^{+25}_{-8}$ s. Since the α-decay of this nucleus was not observed only a lower half-life limit can be established T$_\alpha \geq$ 100 s (68% c.l.).

In the chain #6, which was observed at E* ≈ 35 MeV and close to the maximum of σ_{3n}(E*), the great-granddaughter nucleus with $Z = 111$ emitted an α-particle with E$_{\alpha4}$=9.00 MeV after 0.7 s, in contrast with the previous chains #1-5 produced at E* ≈ 39 MeV. This was followed by two more α-transitions and after 33 hours a spontaneous fission was observed.

Most probably, chain #6 belongs to the decay of the neighboring odd-odd nucleus ^{294}117 produced, as expected, in the 3n evaporation channel of the reaction ^{249}Bk+^{48}Ca at E* ≈ 35 MeV. The reasons for assigning the first five and the subsequent sixth decay chains to different isotopes (^{293}117 and ^{294}117, the 4n- and 3n-evaporation channels), are as follows: (1) for the isotope ^{293}117 with T$_{1/2}$=14 ms the probability of decaying after 112.4 ms is 0.4%; (2) the α-particle energies of parent isotopes differ by 0.2 MeV; (3) the five "short" and one "long" decay chains were observed at different ^{48}Ca energies; (4) the α-decay energies of the descendant nuclei ^{290}115 - ^{274}Bh correspond to the tendency observed in Q$_\alpha(N, Z)$ systematics for the lighter isotopes $^{287-289}$115 - $^{270-272}$Bh (see Fig. 5); (5) different decay modes were observed for the isotopes ^{281}Rg (SF) and ^{282}Rg (α). While each of these arguments considered independently cannot serve as strict evidence for the observation of different parent isotopes, observation of all five features *at once* strongly supports the assignment of the short and long chains to the decays of different parent isotopes ^{293}117 and ^{294}117. Properties of the nuclei in the decay chain of ^{294}117 are given in the upper section of Table II. In the above assignment the observed decay times of the isotopes ^{290}115 and ^{282}Rg appear to be lower than those expected from systematics of T$_{1/2}$ for their lighter neighbors (∼0.6 s and ∼1000 s, respectively, see Fig. 5). However, experimental observation of significantly scattered decay time values is a common occurrence (e.g., compare the 17.5-ms decay time for ^{289}115 in the 3rd decay chain, and 511.8 ms and 424.4 ms observed in the last two decay chains).

Comparing the α-decay energies and half-lives of the neighboring isotopes - the evaporation residues ^{293}117 and ^{294}117, their daughters ^{289}115 and ^{290}115, as well as granddaughters ^{285}113 and ^{286}113, one can see that they do not differ significantly. This situation changes in the great-granddaughter nuclei. Despite the strong hindrance, spontaneous fission is a principal decay mode of the odd-even nucleus ^{281}Rg. Note that all of the lighter isotopes of element $Z = 111$, the ^{280}Rg, ^{279}Rg [20, 21] and ^{278}Rg [22] produced in reactions with ^{48}Ca, as well as ^{274}Rg [49, 50] and ^{272}Rg [51–54] produced in cold fusion experiments are α-emitters. The heavier isotope ^{282}Rg registered in the decay chain at E* ≈ 35 MeV undergoes α-decay as well. A different decay mode of the isotope ^{281}Rg can be explained when comparing the results of the present experiment with the decay properties of the neighboring even-Z nuclei.

In the systematics of T$_{SF}(N)$ the decrease in the half-life of the even-Z nuclei with increasing neutron number in the region of nuclei with $N > 162$ gives way to a strong increase in stability associated with the effect of approaching the spherical shell $N = 184$ [47, 48]. Minimum values of T$_{SF}$ are characteristic of the transition

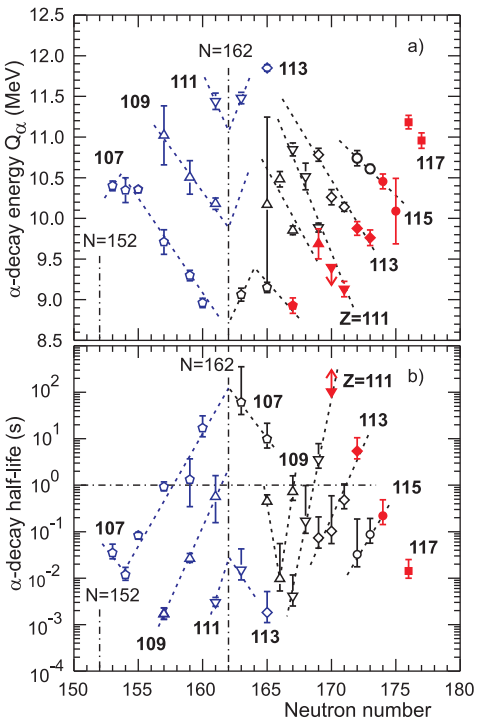

FIG. 5: (a) α-decay energy and (b) half-lives versus neutron number for the isotopes of odd-Z elements with Z = 107-117 (new results in red, results from ^{48}Ca-induced reactions [5, 20–22] in black). All the nuclides with $N > 165$ have been produced in ^{48}Ca induced reactions. T$_\alpha$(exp) values are given for nuclei belonging to the ^{293}117 decay chain (5 events). The limit for T$_\alpha$(^{281}Rg) was estimated from the measured half-life and number of observed nuclei.

region N = 168-170, where the effect of nuclear shells is at a minimum. Indeed, the partial half-lives of the even-even nuclei ^{282}Cn (N = 170) and ^{284}Cn (N = 172) with respect to spontaneous fission are only 0.8 ms and 0.1 s, respectively [5]. The two even-odd isotopes ^{279}Ds (N = 169) and ^{281}Ds (N = 171) also undergo spontaneous fission with half-lives of 0.2 s and 11 s [5]. Finally, the even-even isotope ^{286}114 (N = 172) undergoes spontaneous fission with a probability of 50% and T$_{SF}$=0.26 s [5]. Note that all the chains of sequential decays of the heavier nuclei - the isotopes of elements 112, 114, 116 and 118 produced in the reactions with ^{48}Ca terminate with spontaneously fissioning nuclei.

For the odd-Z nuclei produced in the reactions ^{237}Np+^{48}Ca and ^{243}Am+^{48}Ca a different decay pattern is observed. Because of the high hindrance of spontaneous fission in the decay of nuclei with an odd number of protons (neutrons) and their relatively low T$_\alpha$, the isotopes of elements 113 and 115 with N = 169-173 undergo α-decay [20–22]. In the products of the sequential decay of these nuclides, the T$_{SF}$ values increase upon approaching to the N = 162 shell. In the nuclei with $Z \geq$ 107, T$_\alpha <$ T$_{SF}$; and these nuclei are α-emitters. Spontaneous fission is observed only in the isotopes of element 105 whose α-decay half-lives reach T$_\alpha \geq 10^5$ s for ^{268}Db.

In the reaction ^{249}Bk+^{48}Ca the daughter nuclei that originate from the evaporation residues ^{293}117 and ^{294}117 have one or two extra neutrons over those produced in the ^{243}Am+^{48}Ca reaction. Therefore, approaching the N = 184 shell should result in a decrease in their decay energy Q$_\alpha$ and an increase in T$_\alpha$ with respect to the neighboring lighter isotopes. This regularity is clearly observed experimentally for all the isotopes with $Z \geq$ 111 (see Fig. 5). Analogous to the neighboring even-Z isotopes all the nuclei in the decay chains of ^{293}117 and ^{294}117 with $Z >$ 111 and $N \geq$ 172 will undergo α decay. For these isotopes T$_\alpha < T_{SF}$. The nucleus ^{281}Rg (N = 170) belonging to the "critical" region between neutron shells might avoid spontaneous fission due to the hindrance resulting from an odd proton. The hindrance of the spontaneous fission of ^{281}Rg with respect to that of its even-even neighbor ^{282}Cn [5] is $\sim 3 \cdot 10^4$. Despite this, the isotope ^{281}Rg undergoes spontaneous fission with a probability $b_{SF} \geq$83%. Accordingly, even the high hindrance governed by single-particle oddness does not "save" the odd nucleus from spontaneous fission, caused by the weakening of the stabilizing effect of the neutron shells at N = 162 and N = 184.

Along with this, a doubly odd nucleus as well as an extra neutron in the neighboring isotope ^{282}Rg favor the α decay of this nucleus. It is also clear that in going further into the region with $N <$ 170, the lowering of T$_\alpha$ with an increase of neutron deficit and the increase of T$_{SF}$ upon approaching the N = 162 shell, will result in the lighter isotopes of Rg undergoing α decay in preference to SF.

The experimental values of Q$_\alpha$ and T$_\alpha$ for the isotopes with atomic numbers 107, 109, 111, 113, 115 and 117 produced in the reactions with ^{48}Ca are shown in Figs.5a and 5b. The heaviest isotopes of each element (marked red) belong to the decay chains of the nuclei of the new element 117 synthesized in the reaction ^{249}Bk+^{48}Ca. As seen in Fig. 5a, the effect of increasing the neutron number in odd-Z nuclei, is similar to that expressed by the even-Z nuclei, resulting in a decrease of the Q$_\alpha$ energy and a considerable increase of their half-life, T$_\alpha$. An especially strong growth of T$_\alpha(N)$ with increasing N is observed for the isotopes of elements 109, 111 and 113. All the nuclides presented in Figs.5a and 5b, except the above considered ^{281}Rg, are α emitters; for them T$_\alpha < T_{SF}$. This provides additional evidence of the high stability of the superheavy nuclei with respect to spontaneous fission.

In Fig. 6, the experimental Q$_\alpha$ results are compared with expectations based on the calculated masses and characteristics of nuclear decay in the macroscopic-microscopic nuclear model [55, 56]. The deviation between theory and experiment is given as the difference in decay energies $\Delta Q_\alpha = Q_\alpha^{exp} - Q_\alpha^{th}$ for all nuclei produced as evaporation residues in the ^{48}Ca-induced reactions and their daughter products as well as those syn-

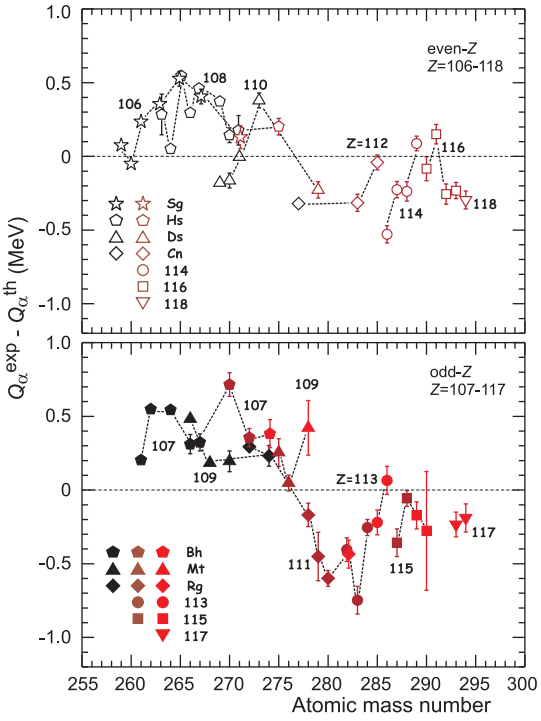

FIG. 6: Deviation between experimental and calculated α-decay energies for even-Z (top panel) and odd-Z (bottom panel) nuclei (new results in red for nuclei originating from ^{293}117 and ^{294}117; results from ^{48}Ca-induced reactions [5] in brown). Theoretical data [55, 56] were corrected by a method suggested in [26].

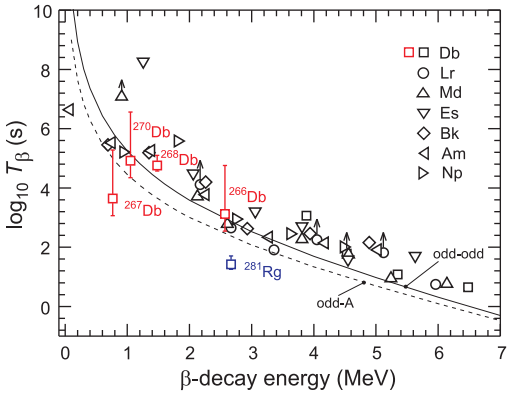

FIG. 7: Partial β^+/EC-decay half-lives vs. decay energy for known odd-odd isotopes of Np-Db (open symbols). The systematics [58] for odd-odd and odd-A isotopes are shown by lines. Experimental half-lives for terminal isotopes for which spontaneous fission was detected in the decay chains produced in ^{48}Ca induced reactions with ^{237}Np, ^{243}Am and ^{249}Bk targets are given by colored squares. Error bars correspond to 95% c.l. if a single event was detected and 68% c.l. in other cases. β^+/EC-decay energies for $^{266-268,270}$Db and ^{281}Rg were calculated from [26, 55].

thesized in other reactions. For the odd-Z nuclei - the isotopes ^{261,264}Bh, ^{266,268}Mt and ^{272}Rg with their multiline α-particle spectra, the value of Q_α^{exp} is determined from the maximum α-transition energy. Results of the present work for the 9 new α emitters obtained in the reaction ^{249}Bk+^{48}Ca are shown in red.

The theoretical predictions [55, 56] were corrected by a method suggested in [26], based on the assumption that nuclei with odd numbers of protons or neutrons undergo α decays not from ground-state to ground-state but to an excited level of the daughter nuclei with the average excitation energies determined in [57]. One can see that the model approach based on the shell corrections to the macroscopic potential energy of the nucleus provides a good description of decay energies of the deformed nuclei in the region of neutron shells at $N = 152$ and $N = 162$. With increasing neutron number (in the model this means approaching the spherical nuclei at $N = 184$) the measured decay energy Q_α^{exp} becomes less than Q_α^{th}, but this difference varies weakly up to the heaviest nuclei being within $\Delta Q_\alpha = \pm 0.6$ MeV for the even-Z nuclei and $\Delta Q_\alpha = \pm 0.8$ MeV for nuclei with odd-Z. As seen in Fig. 6, the decay energies of the isotopes of element 117, as well as those of their decay daughters also lie within these limits and are consistent with the decay properties of the neighboring nuclei. The data from the reaction ^{249}Bk+^{48}Ca corroborate in full the theoretical predictions and provide an extra and direct proof of the existence of the nuclear shells that form zones (islands) of stability in the domain of superheavy elements.

Partial β^+/EC-decay half-lives vs. decay energy for known odd-odd isotopes of Np-Db are shown in Fig. 7. Experimental half-lives together with predicted β^+/EC-decay energies for the isotopes ^{266,268,270}Db are in agreement with T_β vs. Q_β systematics for Np-Db isotopes. This indicates that the heaviest odd-odd isotopes have significant probability of decaying by β^+/EC decay, leading to the spontaneous fission of even-even Rf isotopes for which T_{SF}=23 s, 1.4 s, and 20 ms are predicted [47]. Values of the observed half-lives leading to fission for ^{267}Db as well as ^{281}Rg are somewhat lower than the bulk of β^+/EC data for lighter isotopes. This could indicate a larger probability for decay by spontaneous fission in preference to β^+/EC decay leading to 1 h ^{267}Rf and 11 s ^{281}Ds [5]. However, large uncertainties in half-life and/or the calculated β^+/EC-decay energy [26, 38, 39, 55, 59–62] for these isotopes prevent us from drawing definite conclusions.

Another possible indication of the observation of β^+/EC decay leading to spontaneous fission of daughter Rf isotopes instead of direct fission of Db isotopes follows from the pattern of their observed half-lives with increasing neutron number. The measured half-lives for Db isotopes gradually become larger with increase in neutron number on approach to the neutron shell $N =$

TABLE III: Experimental and calculated cross sections for the xn-evaporation channels in the ^{249}Bk+^{48}Ca reaction.

E* (MeV)	xn	Experiment	[23]^a	[24]^a	[25]^a
39.3±2.1	$2n$	-	0.3	-	-
	$3n$	≤0.7	2	0.3	0.6
	$4n$	$1.3^{+1.5}_{-0.6}$	1	0.3	0.8
35.3±2.1	$2n$	-	0.2	0.01	-
	$3n$	$0.5^{+1.1}_{-0.4}$	0.9	0.8	1.2
	$4n$	≤0.8	0.1	0.1	0.7

^{a} Cross sections represent average values within given excitation-energy intervals, compare refs.[23],[24] and [25].

162 (^{267}Db) and continue to increase beyond that point ($T_{1/2}$=22 min, 1.2 h, 29 h, and 23 h for isotopes with masses 266, 267, 268, and 270, respectively). However, crossing the neutron magic number is expected to result in a decrease of T_{SF} for nuclei with $N > 162$, e.g., for ^{270}Db in comparison with ^{268}Db. Conversely the half-life behavior of ^{266,268,270}Db isotopes is similar to that observed for, e.g., Md, Lr or Db isotopes located near magic number $N = 152$.

The cross sections for producing the isotopes for element 117 in the reaction ^{249}Bk+^{48}Ca are $\sigma = 0.5^{+1.1}_{-0.4}$pb and $\sigma = 1.3^{+1.5}_{-0.6}$ pb at E*=35 MeV and E*=39 MeV, respectively (see Table III). Error bars correspond to statistical [42] and experimental uncertainties.

The cross section values for the ^{249}Bk(^{48}Ca,3-4n)^{294,293}117 reaction are comparable with the results of previous experiments [5] where cross sections for the reactions of ^{233,238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm and ^{249}Cf+^{48}Ca have been measured. As one can see in Table III experimental cross sections of the 3n and 4n reaction channels within experimental uncertainties do not contradict the calculated values of [23–25].

V. CONCLUSIONS

From the data obtained in this work and based on the above discussion of the experimental results several conclusions can be derived.

1. In the fusion reaction of the radioactive target nuclei ^{249}Bk and the doubly-magic ^{48}Ca ions, a new chemical element with the atomic number 117 was synthesized.

2. In the experiments at E*=35 MeV and 39 MeV that correspond to the expected maximum cross sections of the channels associated with the evaporation of 3 and 4 neutrons the production of two isotopes of element 117 with similar properties was observed. These correspond to ^{294}117 and ^{293}117.

3. These isotopes of element 117 undergo α-decay with $Q_\alpha = 11.18 \pm 0.08$ MeV and 10.96±0.10 MeV and half-lives $T_\alpha = 14^{+11}_{-4}$ ms and 78^{+370}_{-36} ms, for ^{293}117 and ^{294}117, respectively. These parent isotopes initiate long chains of sequential decays that end up in the spontaneous fission of ^{281}Rg ($T_{SF} = 26^{+25}_{-8}$ s) and ^{270}Db ($T_{SF} = 23^{+110}_{-10}$ h).

4. Decay characteristics of the eleven new isotopes produced in the reaction ^{249}Bk+^{48}Ca are in a good agreement with those expected based on the properties of the neighboring even-Z and odd-Z nuclei. They substantially expand the systematics of decay properties of odd-Z nuclides in the region of the most neutron-rich isotopes of elements 105-117 and display considerable increase in the stability of the superheavy elements associated with an increase in neutron number.

5. The new isotopes, together with superheavy nuclides previously synthesized in reactions with ^{48}Ca, present a consistent picture of nuclear properties in the area of heaviest nuclei. They demonstrate the critical role of nuclear shells and represent an experimental verification of the existence of the predicted island of stability of superheavy elements.

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