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Experimental studies of the ^{249}Bk + ^{48}Ca reaction including decay properties and excitation function for isotopes of element 117, and discovery of the new isotope ^{277}Mt

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Studies of superheavy nuclei produced in the ^{249}Bk + ^{48}Ca reaction were performed using the Dubna Gas Filled Recoil Separator. The cross section for the production of $^{293}117$ and $^{294}117$ isotopes was measured at five excitation energies of the $^{297}117$ compound nucleus ranging from 30 to 48 MeV and yielding maximum values of $1.1^{+1.2}_{-0.6}$ pb for the $3n$ and $2.4^{+3.3}_{-1.4}$ pb for the $4n$ reaction channels. Alpha emission from ^{281}Rg competing with spontaneous fission (α/SF decay probability 1:9) was observed for the first time leading to the identification of the new isotope ^{277}Mt ($T_{\mathrm{SF}} \approx 5$ ms). The measured decay properties are in good agreement with those expected based on the properties of neighboring even-Z and odd-Z nuclei. The α energies and half-lives of odd-Z isotopes observed in the $^{293}117$ and $^{294}117$ decay chains together with results obtained for lower-Z superheavy nuclei demonstrate enhanced stability with increasing neutron number toward the predicted new magic number $N = 184$.

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I. INTRODUCTION

The understanding of decay properties of the heaviest atomic nuclei produced at picobarn cross sections requires long duration experiments in order to detect competing decay channels and possibly new nuclei. Such experiments should be performed at optimum beam energies in order to maximize the discovery potential. These motivations guided a new six-month experimental campaign of the ^{249}Bk + ^{48}Ca reaction at the Dubna Gas Filled Recoil Separator (DGFRS). These new studies of superheavy nuclei were performed between April 23 and October 23, 2012, at the Joint Institute for Nuclear Research (JINR) in Russia, and used relatively short-lived ^{249}Bk target material produced at Oak Ridge National Laboratory (ORNL) in the United States.

Initial results of the irradiations of ^{249}Bk target with ^{48}Ca beams, at beam energies of 247 and 252 MeV were presented recently [1]. Here, in more detail, we present and discuss the complete data set obtained for the synthesis of $^{293,294}117$ isotopes in the ^{249}Bk + ^{48}Ca reaction at the 244, 247, 252, 256, and 260 MeV beam energies. The results include the excitation function for the $3n$ and $4n$ emission channels, identification of the new isotope ^{277}Mt, and improved decay data for the $^{293}117$ and $^{294}117$ decay chains. The α energies and half-lives of $Z = 105$ to $Z = 117$ isotopes observed in the $^{293}117$ and $^{294}117$ decay chains together with the results obtained for lower-Z superheavy nuclei provide experimental evidence for the existence of the "island of stability" associated with the predicted new magic neutron number $N = 184$.

II. EXPERIMENT

The experiments were performed at the DGFRS using the ^{48}Ca beam accelerated at the U400 cyclotron of the Flerov Laboratory of Nuclear Reactions, JINR. The ^{48}Ca ion beam intensity of about 1 particle μA was delivered to the target. The beam energy was determined with a systematic uncertainty of 1 MeV by a time-of-flight system placed in front of the DGFRS.

As in the previous experiments from 2009 and 2010 [2,3], the ^{249}Bk was produced at ORNL through the intense neutron irradiation of Cm and Am feed materials. In 2011, two irradiations were performed at the High Flux Isotope Reactor (HFIR). The first irradiation of four targets lasted for approximately 250 days (including reactor downtime). The second irradiation of five targets lasted for only one month (August 2011) but created about half of the total ^{249}Bk material. The targets for both irradiations consisted of the same initial concentration of heavier isotopes of curium (^{246}Cm and ^{248}Cm). The longer first irradiation was aimed at the production of ^{252}Cf and thus there was no optimization of the production of ^{249}Bk. However, the short irradiation was solely for the production of ^{249}Bk. Since the feed material of the targets was rich with the heavier isotopes of curium, this allowed for efficient production of ^{249}Bk; requiring one to three

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neutron captures. The ^{249}Bk production reaches equilibrium approximately five days after irradiation in the HFIR. The transmutation of ^{249}Bk to ^{250}Bk impacts the amount of ^{249}Bk that can be produced. Longer irradiations of this material allow for the production of the heavier elements and their isotopes, but do not significantly increase the overall production of the berkelium. The Bk fraction was chemically separated from all irradiated targets and was purified at the Radiochemical Engineering Development Center at ORNL; see Ref. [3] for a description of irradiations and chemical procedures. Half of the obtained ^{249}Bk material, 12.7 mg, was shipped to the JINR. The ^{249}Bk contained 0.51 mg of ^{249}Cf (β^- decay product of ^{249}Bk), less than 0.45 ng of ^{252}Cf, and no other detectable impurities. Six arc-shaped targets, each with an area of 5.4 cm^2, were made at the Research Institute of Atomic Reactors (Dimitrovgrad, Russian Federation) by depositing BkO$_2$ onto 0.72 mg/cm^2 Ti foils to a thickness of 0.33 mg/cm^2 of ^{249}Bk. The targets were mounted on the perimeter of a disk 12 cm in diameter that was rotated at 1700 rpm perpendicular to the beam direction.

The evaporation residues (ERs) were separated from projectiles, scattered particles, and transfer-reaction products by the DGFRS with an estimated transmission efficiency for $Z = 117$ nuclei of 35%. The magnetic rigidity of the separator of 2.395 T m was used in these experiments. Based on 14 observed decay chains of ^{293,294}117 in this experiment (see Table I), the ion charge of element 117 ERs was measured to be 6.54 ± 0.22, which agrees within uncertainties with the value ($6.1^{+0.3}_{-0.2}$) estimated from results of previous runs from 2009 and 2010 [2,3].

The detection system was modified in comparison with earlier experiments (see review [4] and references therein) to increase the position granularity of the detectors, which reduces the probability of observing sequences of random events that could imitate decay chains of synthesized nuclei. The new focal-plane detectors consisted of two 6×6 cm^2 silicon detectors each having 16 strips with position sensitivity in the vertical direction. These detectors were surrounded by six similar 6×6 cm^2 side detectors without position sensitivity. Behind the focal-plane detectors, which had a thickness of 0.3 mm, a pair of veto detectors similar to the side detectors was mounted for the detection and rejection of signals from high-energy long-range charged particles (α's, protons, etc., produced in direct reactions of projectiles with the DGFRS media) which can pass through the separator without being detected by the time-of-flight system placed in front of the detectors.

TABLE I. The summary of the 2012 DGFRS campaign focused on the production and studies of element 117 isotopes. The laboratory-frame beam energies in the middle of the target layer, resulting excitation energy intervals, total beam doses, and numbers of observed decay chains assigned to the parent nuclei ^{294}117 ($3n$) and ^{293}117 ($4n$) characterizing the studies in 2012 presented in Ref. [1] and this work, are listed.

E_{lab} (MeV)	E^* (MeV)	Beam dose $\times 10^{18}$	No. of chains $4n$ / $3n$	Ref.
243.7	30.4–34.7	9.4	0 / 1	this work
246.8	32.8–37.5	3.4	0 / 2	[1]
251.7	37.0–41.9	11.7	5 / 0	[1]
255.7	40.3–44.8	9.2	3 / 0	this work
259.8	43.8–48.3	11.9	3 / 0	this work

The detection system was calibrated by registering the recoil nuclei and α decays or spontaneous fission (SF) fragments of known isotopes of No and Th and their descendants produced in the reactions ^{206}Pb(^{48}Ca,$2n$) and ^{nat}Yb(^{48}Ca,$3n$–$5n$), respectively. The low-energy scale, used for registration of α particles and ERs, was calibrated by nine α lines of isotopes from ^{209}Rn ($E_\alpha = 6.04$ MeV) to ^{217}Th ($E_\alpha = 9.26$ MeV) [5]. The high-energy scale for detection of fission fragments was calibrated by the α lines of ^{216,217}Th and ^{217}Ra ($E_\alpha = 7.92$–9.26 MeV) and energies of ^{48}Ca scattered ions measured before DGFRS and then corrected for energy losses in the separator's solid [6] and gas [7] media. Fission fragments from the decay of ^{252}No implants produced in the ^{206}Pb + ^{48}Ca reaction were used for the total kinetic energy (TKE) calibration. The measured fragment energies presented in this work were not corrected for the pulse-height defect of the detectors or for energy losses of escaping fragments in the detectors and the pentane gas filling the detection system. From comparison of the measured average energy of fission fragments of ^{252}No registered by both the focal-plane and side detectors with TKE value determined in Ref. [8], the sum energy loss of fission fragments was estimated to be about 13 MeV. The position signals from the top and bottom of the strips were calibrated separately by signals registered in the low- and high-energy scales in calibration experiments.

The full width at half maximum (FWHM) energy resolutions of α particles completely absorbed in the focal-plane detector was 34 to 73 keV, while the summed signals recorded by the side and focal-plane detectors had an energy resolution of 83 to 120 keV. If α particles were detected only by a side detector (without a focal-plane energy and position signals), the energy of such events was estimated as the sum of the energy measured by the side detector and half of the threshold energy (0.73–0.98 MeV) with an uncertainty determined from the threshold energy and energy resolution of the side detector (68% confidence limit). 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 [9]. The FWHM position resolutions of the events registered in the focal-plane detector were 1.1–1.8 mm for ER-α and 0.5–1.2 mm for ER-SF signals. For α particles detected by both the focal-plane and side detectors, the ER-α position resolution depends on the energy deposited in the focal-plane detector and was, on average, 4.0–7.4 mm for energies lower than 3 MeV and 1.3–3.6 mm for energies larger than 3 MeV. In order to reduce the background rate in the detector, the beam was switched off for at least 3 min after a recoil signal was detected with $E_{\text{ER}} = 7$–18 MeV, followed by an α-like signal in the focal-plane detector within energy intervals of 10.7–11.3 and 9.6–10.7 MeV and time intervals of 0.4 or 2.0 s, respectively, in the same strip, within a 3.2 mm wide position window.

The experimental conditions and numbers of registered decay chains of ^{294}117 and ^{293}117 are summarized in Table I. Excitation energies of the compound nucleus at given projectile energies are calculated using the masses of Refs. [10,11], taking into account the thickness of the targets and the energy spread of the incident cyclotron beam during long experiments. The beam energy losses in the separator's entrance window (0.71 mg/cm^2 Ti foil), target backing, and target layer were calculated using the nuclear data tables [6,7].

III. EXPERIMENTAL RESULTS

The results of the first experiment reporting the study of production and radioactive decay of nuclei produced in the ^{249}Bk + ^{48}Ca at two projectile energies of 247 and 252 MeV (see Table I) were presented concisely in an earlier publication [1]. The decay chains of ^{294}117 and ^{293}117 observed at three other energies of ^{48}Ca ($E_{\text{lab}} = 244$, 256, and 260 MeV) are shown in Fig. 1. One can see that position deviations between ER signals and all α particles and five of seven SF events are in agreement with the position resolutions of detectors for consecutive ER-α and ER-SF signals. In two cases (chains No. 3 and 7 in Fig. 1) positions of SF events differ from ER signals by 2.8/2.9 and 3.5/3.2 mm (top/bottom signals), respectively. However, both of these decay chains were registered rather close to the edge of the strips (52.7 and 51.0 mm). Such deviations are permissible for events detected at the strip border and in different energy scales (different analog-to-digital converters). Moreover, in both chains the SF fragments were detected during beam-off intervals at very low counting rate of random SF events. Note, only six beam-off SF events were observed in experiments at $E_{\text{lab}} = 256$ MeV [Fig. 1(b)] and 260 MeV [Fig. 1(c)] and all of them were found at the end of decay chains of ^{293}117. The probability of the random appearance of the beam-off SF signals within a 10-mm position window and a 5-s time interval of triggering events in strips 15 and 16 was less than 10^{-5} [9].

From one to four nuclei in each decay chain shown in Fig. 1 were registered in the absence of beam-associated background. The expected maximum numbers of random sequences of the types given in Fig. 1 vary from 2×10^{-9} to 3×10^{-20} for all decay chains except for the last one for which this number was 2×10^{-4} [9]. Note that only 19 α particles with $E_\alpha = 8.5$–10.0 MeV were registered by the focal-plane detector only or simultaneously with the side detector during the beam-off time intervals with the sum length of 6.1×10^4 s at $E_{\text{lab}} = 244$ MeV, while in the experiments at $E_{\text{lab}} = 256$ and 260 MeV taken together the corresponding number is 25 α particles with $E_\alpha = 9.2$–10.6 MeV during 1.6×10^5 s. Among them three and seven α particles, respectively, belong to the decay chains of ^{294}117 and ^{293}117. We assume that, in the first and second chains detected at $E_{\text{lab}} = 256$ MeV that are shown in Fig. 1(b), the α decays of the parent isotope ^{293}117 did not switch the beam off because of a short 43-μs ER-α_1 time interval in the first case, and the difference in ER and α_1 position signals ($\Delta p_b = 1.5$ mm and $\Delta p_{t,b} = 1.7$ mm in the first and second cases, respectively) which could exceed the allowable ER-α_1 position deviation for the beam interruption [12]. But in both cases the next α particles registered by the focal-plane detector switched the beam off.

The loss of five α particles (marked "missing α" in Fig. 1) in decay chains which can consist of 26 α decays is in agreement with the 87% efficiency of the detector array for observing full-energy α particles. Two α particles [first and third decay chains in Fig. 1(b)] were registered by the side detector only. The probabilities of the random appearance of a beam-on and beam-off signals in one of the six side detectors with $E_\alpha = 9.6$–10.5 MeV and $E_\alpha = 8$–10 MeV and within time intervals of 0.02 and 10 s of a triggering events in strips 2 and 12 were about 0.25% and 0.6%, respectively [9]. We assigned these side-only signals to ^{289}115 and ^{281}Rg; their total energies were estimated to be the sum of the energy measured by the side detector and half of the threshold energy of the focal-plane detector; thus, the uncertainty in determining the total energy is increased to about 0.3 MeV.

The summary of decay properties of nuclei originating from the $4n$ evaporation reaction product, ^{293}117, measured in the five similar decay chains observed in 2009 and 2010 at $E_{\text{lab}} = 252$ MeV [2,3] and eleven chains observed in the second run at $E_{\text{lab}} = 252$, 256, and 260 MeV in 2012 ([1] and this work) as well as nuclei in the four decay chains originating from the $2n$ channel of the ^{243}Am + ^{48}Ca reaction [13,14], ^{289}115, are shown in Fig. 2. The α-particle energy spectra of the isotopes registered by the focal-plane detector only or together with the side one are shown in the left-hand side of Fig. 2. The decay-time distributions on a logarithmic scale for the same isotopes are given on the right-hand side of Fig. 2 except for events following an unobserved precursor(s). One can see that the results of the first experiment [2,3] are in good agreement with new data ([1] and this work). The radioactive decay properties of ^{293}117 and all descendant nuclei discovered in 2010 were completely confirmed by registration of eleven decay chains in the new series of experiments ([1] and this work). Moreover, the α-particle energies and decay times of the isotopes ^{289}115, ^{285}113, and ^{281}Rg registered after the α decay of the parent nucleus ^{293}117 in the reaction ^{249}Bk + ^{48}Ca and synthesized directly in the reaction ^{243}Am + ^{48}Ca [13,14] are comparable. Therefore, the isotope ^{289}115 was produced in two reactions with target nuclei ^{243}Am(^{48}Ca,$2n$)^{289}115 and ^{249}Bk(^{48}Ca,$4n$)$^{293}117 \rightarrow ^{289}$115 that provides cross-bombardment evidence for the discovery of elements 115 and 117.

For the first time in recent experiments, we observed two decay chains (see Fig. 1) in which the isotope ^{281}Rg underwent α decay ($E_\alpha = 9.28$ MeV, estimated α-decay branch of $0.13^{+0.13}_{-0.04}$) instead of fissioning, which was then followed by SF of the new isotope ^{277}Mt with $T_{1/2} = 4.9^{+8.8}_{-2.0}$ ms [9], which is also the first observation of fission in any isotope of Mt.

In agreement with the well known behavior of excitation functions of the complete-fusion reactions, with reduction of excitation energy of the compound nucleus, the yield of the lower xn-evaporation channel should be detected (see, e.g., [4,13,14]). Indeed, in the excitation energy interval $E^* = 30.4$–37.5 MeV, we observed a total of four decay chains of the $3n$-evaporation channel, ^{294}117 (see [1–3] and

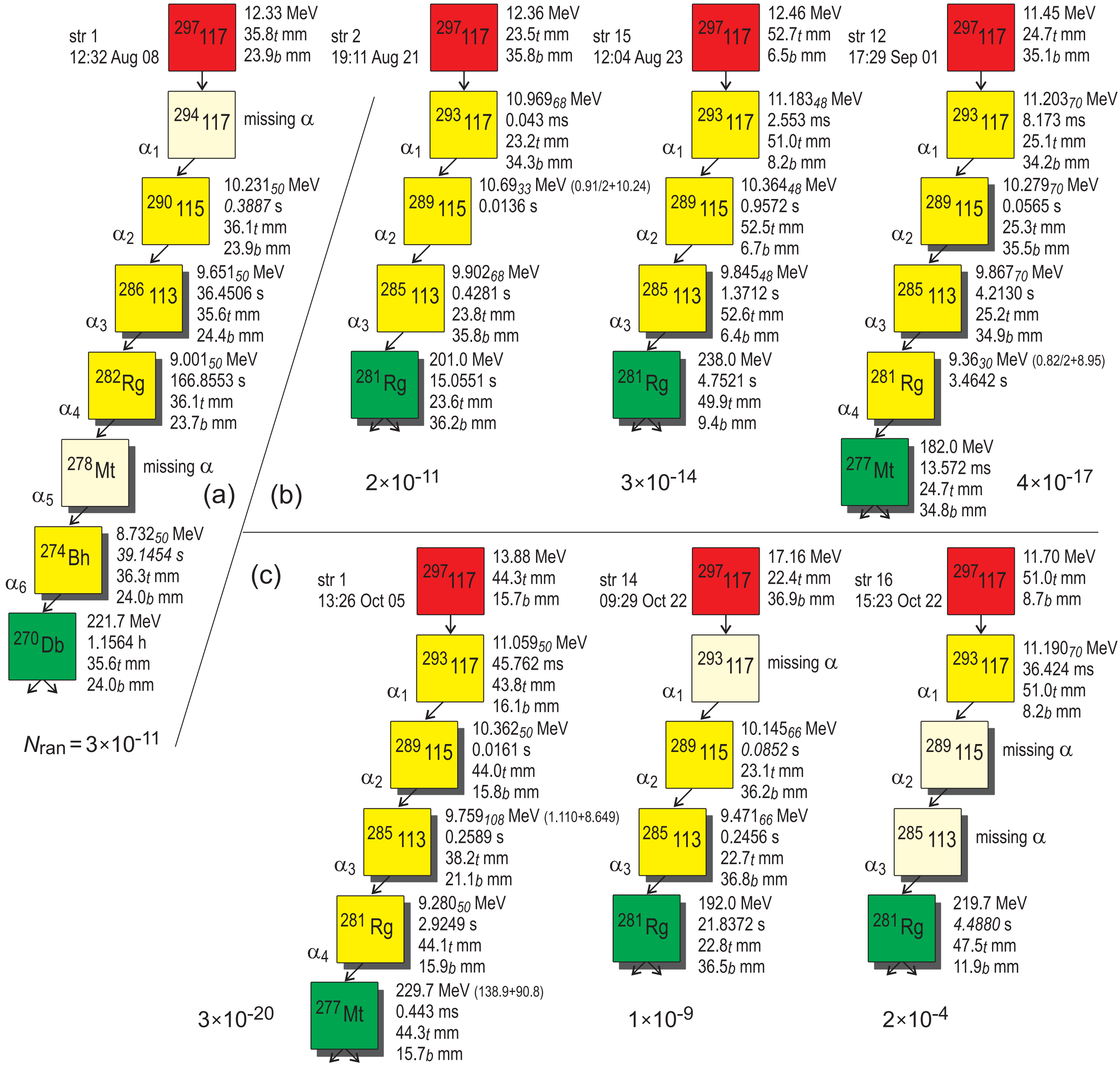

FIG. 1. (Color online) Decay properties of ^{294}117 (a) and ^{293}117 [(b) and (c)] and their decay products observed at ^{48}Ca energies of 244, 256, and 260 MeV, respectively. The upper rows for each chain show strip number (str) and time and date of registration (on the left side) as well as ER (in red) energies and positions from the top (t) and bottom (b) of the strip (on the right side). Subsequent rows provide α-particle (in yellow) and SF fragment (in green) energies, time intervals between events and their positions. Energies of summed signals are given in parentheses. Events marked with a shadow were registered during the beam-off periods. The α-particle energy errors are shown by smaller italic numbers. Time intervals for events following a "missing α" were measured from preceding registered events and are shown in italic. The calculated number of random sequences is given at the bottom of each chain [9].

Fig. 1(a)). The summary α-particle energy spectra of the isotopes observed in the decay chains originating from ^{294}117 and decay-time distributions on a logarithmic scale for the same isotopes are given in Fig 3. As we noted in [1,3], in the three new decay chains we observed longer lifetimes for ^{290}115 and ^{282}Rg compared with the values detected in the first experiment [2,3]. All other decay properties of all nuclei in the new decay chains are in good agreement with data measured in the first experiment [2,3] and point to the same activities arising from ^{294}117 detected in the two experiments using the ^{249}Bk target.

Experimental values of the cross section for the production of the isotopes of element 117 measured in the ^{249}Bk + ^{48}Ca reaction at five excitation energies of the compound nucleus, ^{297}117, are given in Fig. 4. As it was mentioned in [1], the cross sections for the 3n and 4n evaporation channels at

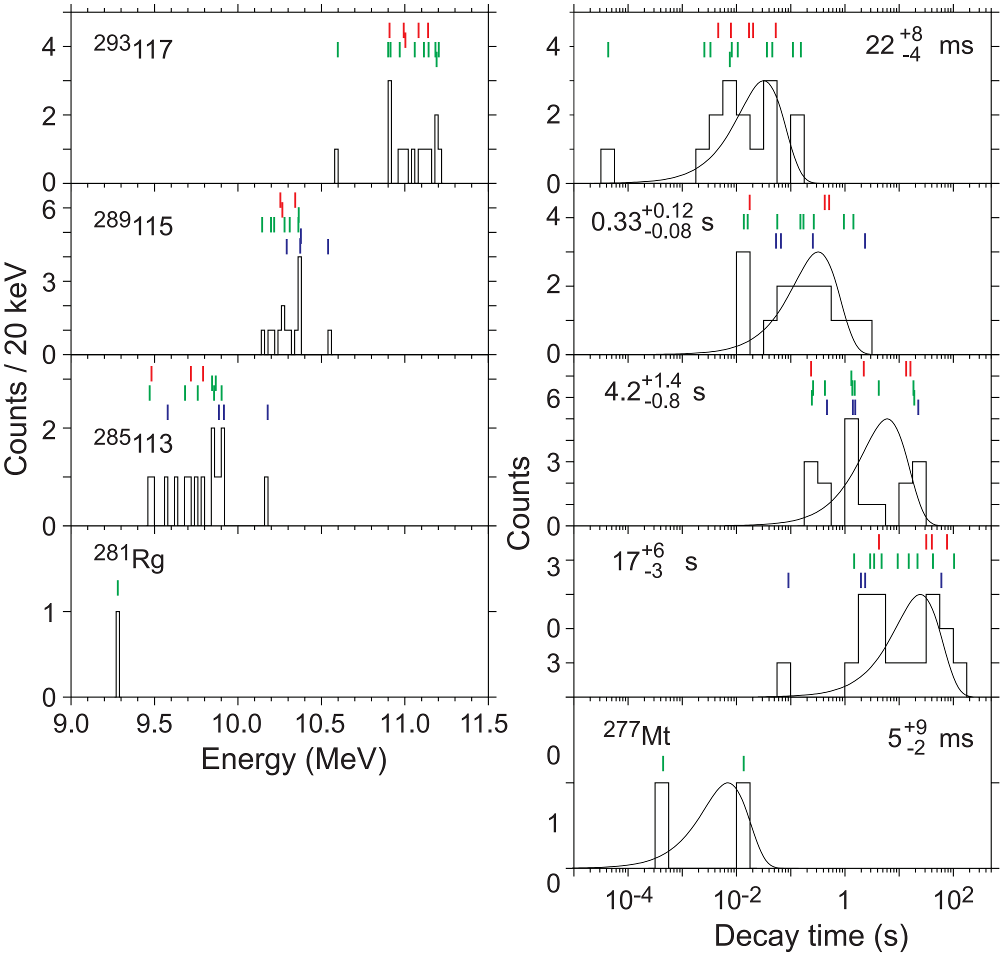

FIG. 2. (Color online) α-particle energy spectra (left-hand panel) and decay-time distributions on a logarithmic scale (right-hand panel) for ^{293}117 and descendant nuclei. The events originating from ^{293}117 observed in the reaction with ^{249}Bk in the first experiment [2,3], in the second experiment ([1] and this work), and those originating from ^{289}115 produced in the reaction with ^{243}Am [13,14] are shown in the histograms by red (top), green (middle), and blue (bottom) lines, respectively. The smooth curves are the time distributions for exponential decays calculated with the half-lives $T_{1/2}$ shown in the figures.

$E^* = 35$ and 39 MeV were measured to be $\sigma_{3n} = 3.6^{+6.1}_{-2.5}$ pb and $\sigma_{4n} = 2.0^{+2.2}_{-1.0}$ pb in this run, which are larger but within experimental uncertainties when compared with the previous results of $\sigma_{3n} = 0.5^{+1.1}_{-0.4}$ pb and $\sigma_{4n} = 1.3^{+1.5}_{-0.6}$ pb [2,3]. The average cross sections for the production of ^{294}117 and ^{293}117 nuclei at these energies determined from the observation of 3 and 10 events amount to $\sigma_{3n} = 1.1^{+1.2}_{-0.6}$ pb and $\sigma_{4n} = 1.5^{+1.1}_{-0.5}$ pb, respectively. The cross section of the 4n evaporation channel at $E^* = 43$ MeV was measured to be $\sigma_{4n} = 2.4^{+3.3}_{-1.4}$ pb. These cross-section values are consistent with the results of previous experiments where cross sections for the reactions of ^{238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, and ^{249}Cf targets with ^{48}Ca beams have been measured [4,13,14]. Also shown in Fig. 4 are calculated cross sections for the 3n to 5n evaporation channels of the ^{249}Bk + ^{48}Ca reaction within a model based on multidimensional Langevin-type dynamical equations [15] and the fusion-by-diffusion model [16]. The calculated cross sections are in good agreement with results of experiment. Some other models predict comparable cross-section values but the energies of the maxima of the xn evaporation channels are predicted to be somewhat lower (see, e.g., [17,18]) than those measured in this experiment.

IV. DISCUSSION

The systematics of α-decay energy (Q_α) vs neutron number for the isotopes of odd-Z elements 105–117 produced in the ^{48}Ca-induced reactions are shown in Fig. 5. For the most of the odd-Z nuclei, the multiline α-particle spectra were observed with the difference in α energies clearly exceeding the energy resolution of detectors (see Figs. 2 and 3). This observation most likely reflects fine structure in α decays. The α-decay energy for each isotope was determined from the largest measured α-particle energies detected within energy interval $\Delta E_\alpha \approx 0.1$ MeV. Only the two highest energies of 10.540 and 10.178 MeV observed for isotopes ^{289}115 and ^{285}113 (see Table III in [14]), respectively, were excluded from calculations because of their deviation by more than 0.15 MeV from the bulk of other α-particle energies. Both of these values were observed in two of four decay chains of

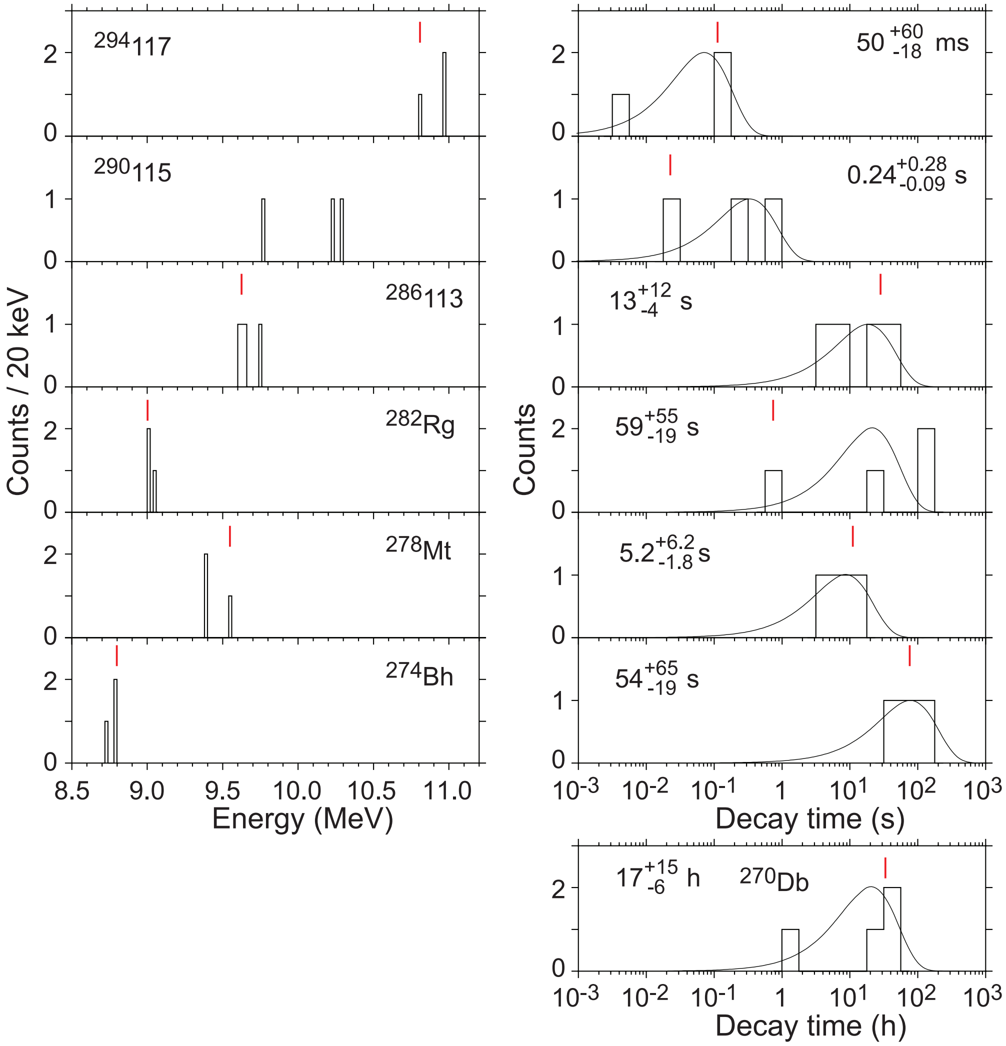

FIG. 3. (Color online) α-particle energy spectra (left-hand panel) and decay-time distributions on a logarithmic scale (right-hand panel) for $^{294}117$ and descendant nuclei. The events originating from $^{294}117$ observed in the reaction with ^{249}Bk in the first experiment [2,3] are shown in the histograms by red lines. The smooth curves are the time distributions for exponential decays calculated with the half-lives $T_{1/2}$ shown in the figures.

$^{289}115$ produced in the $^{243}\text{Am} + {}^{48}\text{Ca}$ reaction and could be explained by transitions from excited levels of these isotopes. Otherwise, the α-decay energies of $^{289}115$ and $^{285}113$ would be larger by 0.18 and 0.31 MeV, respectively, than those given in Fig. 5 (these larger values correspond to the top edge of vertical blue lines and deviate from linear behavior $Q_\alpha(N)$ for neighboring isotopes). In the same figure, we show Q_α values resulting from the new atomic mass evaluation [10] for nuclei with $Z = 105$–118.

As a whole, the results of the calculations demonstrate good agreement with the new experimental data in Fig. 5. One can see small deviations for the lightest isotopes of Rg and a few other nuclides which could reflect not only the inaccuracy of the model calculations but also could be due to decays going through the excited levels of nuclei. Nevertheless, the α-decay energies of the neighboring isotopes of even-Z and odd-Z elements with the same number of neutrons clearly show a difference in Fig. 5. The measured α-particle energies for isotopes of odd-Z elements have intermediate values between neighboring even-Z nuclei.

The data obtained in this work helps to analyze a potential α decay of Db isotopes. If the model of [10] reproduces the α-decay energies of Db isotopes, such as ^{270}Db or ^{268}Db, with similar accuracy as those of the isotopes of Bh and heavier elements, the Q_α values should be 8.31 and 8.26 MeV for these Db isotopes, respectively, with the corresponding energies of α particles being 8.19 and 8.14 MeV, respectively, or somewhat lower. Applying the Viola-Seaborg formula [19] with parameters, e.g., from [20], one would expect their unhindered partial α-decay half-lives to be about 0.1 h. The hindrance factors for α decay of all odd-Z nuclei shown in Fig. 5 do not exceed 25. If so, the upper limits of half-lives of both ^{270}Db and ^{268}Db would be about of 2–3 h. This is much lower than half-lives measured for the terminal SF nuclei in the decay chains originating from $^{294}117$ [1–3] and $^{288}115$ [13,14,21] and assigned to decays of ^{270}Db and ^{268}Db.

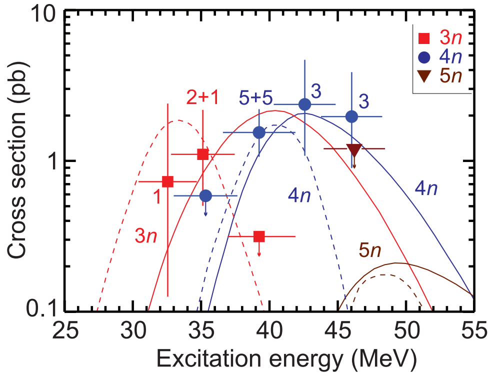

FIG. 4. (Color online) Measured cross sections for the 3n and 4n evaporation channels for the ^{249}Bk + ^{48}Ca reaction. Vertical error bars correspond to statistical uncertainties [9]. Horizontal error bars represent the range of excitation energies populated at given beam energy. Symbols with arrows show upper cross-section limits. Numbers of detected decay chains are given for each ^{48}Ca energy. The results of theoretical calculations are shown by solid [15] and dashed [16] lines.

In the ^{249}Bk + ^{48}Ca experiment, the beam-off intervals were relatively short for detection of ∼1-h activity. In one case [see Fig. 1(a)], the time interval between decay of ^{274}Bh and the SF event was about 1.2 h. Here no α particles with $E_\alpha = 7$–10 MeV were registered solely by the focal-plane detector or together with the side detectors in the same strip and within ±FWHM position deviation after observed chain of isotopes $^{294}117$–^{274}Bh when the beam was switched on. In each of three other chains [1–3] a few α-like events with $E_\alpha = 7.7$–8.2 MeV were found during the beam-on intervals.

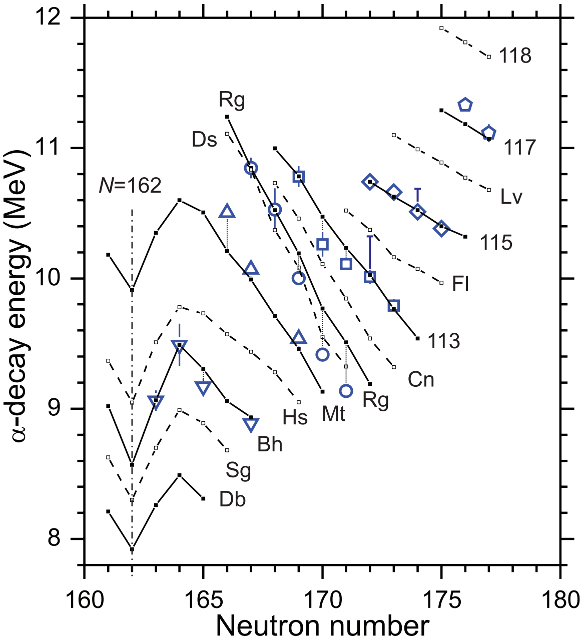

FIG. 5. (Color online) Measured α-decay energy vs neutron number for the isotopes of odd-Z elements 105–117 produced in the ^{48}Ca-induced reactions (open blue symbols). Predicted Q_α values for odd-Z and even-Z nuclei [10] are shown by solid and dashed lines, respectively.

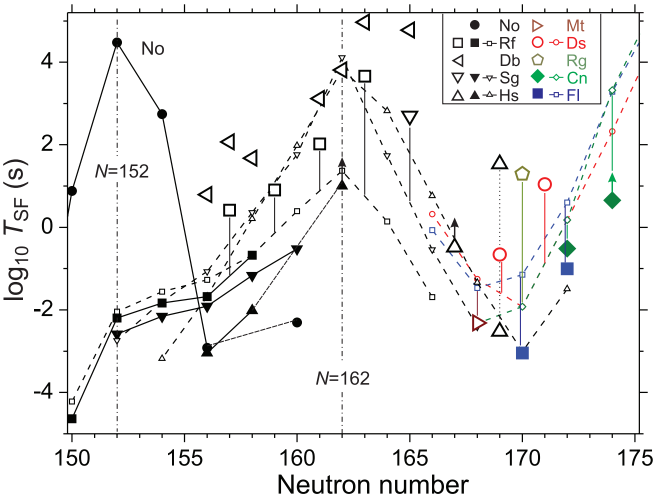

FIG. 6. (Color online) Common logarithm of partial spontaneous fission half-life vs neutron number for isotopes of elements with $Z = 102$–114 ([1–4,13,14,21,26–32] and this work). Data for even-even and odd-N or Z isotopes are given by solid and open symbols, respectively. Dashed lines show the theoretical $T_{\rm SF}$ values [24,25] for even-even $Z = 104$–114 isotopes. Vertical lines connect experimental and predicted values.

Similarly, most decays of $^{288}115$ produced in the ^{243}Am + ^{48}Ca reaction were registered within short beam-off periods [13,14,21]. But four of all 31 decay chains (first event in [21] and chains No. 7, 26, and 28 in [14]) were followed by long beam-off intervals (2.7 h and about 1 d, 1 d, and 3 d, respectively). In all these cases, no α particles with $E_\alpha = 7.7$–8.2 MeV following decay of ^{272}Bh were found. The lack of α decays of ^{268}Db is in agreement with results of chemical experiments where SF activity, with the same half-life as was measured in the DGFRS experiments [21], was observed in fraction of transactinide elements [22,23] and attributed to SF of ^{268}Db. If we assume that ^{268}Db and ^{270}Db undergo predominantly SF (or electron capture leading to relatively short SF of even-even ^{268}Rf and ^{270}Rf [24], as was discussed in [3]), then upper limits for their α-decay energies would be 7.7 and 8.0 MeV, respectively, if one assumes a hindrance factor of 25. Such a difference between predicted and estimated Q_α values could be explained by lower α-decay energies than those given in [10] with uncertainty of 0.3 MeV and/or relatively high hindrance factors for α decay of these nuclei. Further investigations would be desirable for understanding the origin of SF activities observed in the decay chains of $^{294}117$ and $^{288}115$.

The systematics of SF half-life vs neutron number for isotopes of elements with $Z = 102$–114 are shown in Fig. 6. The results of calculations within a macroscopic-microscopic model [24,25] reproduce rather closely the experimental values for the given even-even isotopes of elements from Rf to Fl, deviating from them by less than three orders of magnitude. Reasonable agreement between experiment and theory can be seen for light even-even isotopes of Rf–Hs as well as for

^{282,284}Cn and ^{286}Fl; here the measured partial SF half-lives are lower than calculated values [24,25] by less than two orders of magnitude. The SF half-lives for even-Z nuclei with an odd number of neutrons reasonably exceed values predicted for even-even neighboring isotopes by about 1–3 orders of magnitude. The T_{SF} values for Db isotopes are larger than those for Rf or Sg isotopes with the same number of neutrons by factor of about 10–100. Such an additional hindrance factor caused by an odd number of protons cannot be ruled out also. Thus, occurrence of SF in ^{268,270}Db together with partial α or EC decays cannot be excluded.

The SF half-life of ^{277}Mt is rather close to that for ^{277}Hs [29,30]; see Fig. 6. Here we also show a much longer T_{SF} value for ^{277}Hs observed in [31] and presumably assigned to the decay from a high-spin isomer. In the ^{243}Am + ^{48}Ca or ^{249}Bk + ^{48}Ca reactions, ^{277}Hs could be produced after electron capture/β^+ decay of ^{277}Mt (that could occur, if so, within a few milliseconds between the last observed α decay and SF) or its precursors, ^{281}Rg, ^{285}113, etc. But the α-particle energy of ^{281}Rg (9.28 MeV) is evidently larger than that for ^{281}Ds (8.73 MeV [29,30]) while EC/β^+-decay energy of ^{277}Mt (1.28 MeV [10]) seems to be too low for observing EC decay with a half-life of about 1 ms (compare with ^{268,270}Db for which similar Q_{EC/β^+} values are predicted [10,33]). Thus, the close half-lives that were observed for ^{277}Mt ($N = 168$) and ^{277}Hs ($N = 169$) could indicate comparable hindrance factors for these odd-even and even-odd isotopes. The SF half-life value for ^{277}Mt is even lower than those for the neighboring even-even $N = 168$ isotopes, ^{276}Hs and ^{278}Ds, predicted in [24] (46 and 56 ms, respectively). Bearing in mind that SF half-lives of odd-Z nuclei could be hindered by several orders of magnitude, one can expect considerable weakening of the shell stabilization effect and, therefore, a decrease of T_{SF} values for $N = 168$ nuclei over those predicted in [24]. In accordance with this, the SF half-life for ^{276}Hs is calculated to be about 10^{-5} s within another macroscopic-microscopic model [34]. However, in this approach T_{SF} values for ^{282,284}Cn and ^{286}Fl are underestimated by about one order of magnitude and those for the known SF decaying even-even Rf and Hs isotopes by about three orders. In contrast, a self-consistent Hartree-Fock-Bogoliubov approach [35] results in overestimated T_{SF} values by 3–4 orders of magnitude for even-even nuclei shown in Fig. 6. But, $T_{\text{SF}} \approx 40$ ms is predicted for ^{276}Hs, similarly to [24], which allows one to assume lower SF half-lives for nuclei at $N = 168$ than are predicted in [35]. Thus, experimental observations of SF for isotopes ^{282,284}Cn [20,27,28,36,37], ^{277}Hs [29,30], and ^{277}Mt indicate a gap in stability against SF for nuclei with $N = 168$–170 that is in line with all the above mentioned theoretical predictions.

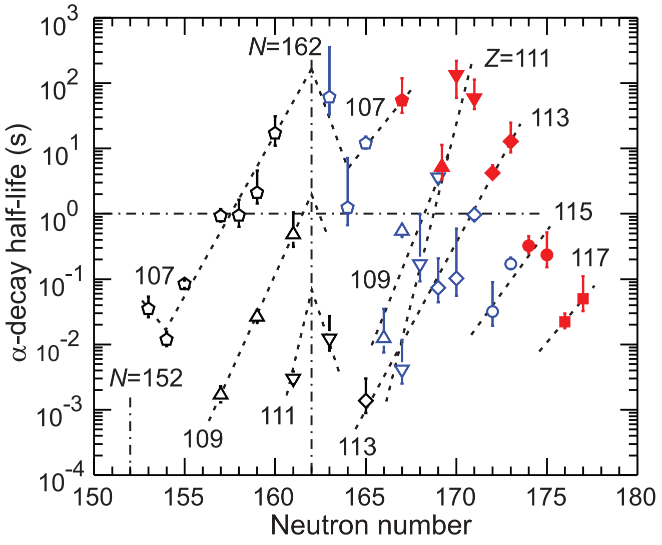

FIG. 7. (Color online) Half-lives vs neutron number for the isotopes of odd-Z elements with $Z = 107$–117 (results from ^{249}Bk + ^{48}Ca reaction are shown by full red symbols and results from ^{48}Ca-induced reactions [1–3,13,14,21,38] by open blue symbols).

Variations of stability against α decay vs neutron number are shown in Fig. 7. Here the experimental half-lives, T_α, for the isotopes of odd-Z elements with atomic numbers from 107 to 115 produced in the reactions with ^{48}Ca are shown by open blue symbols [13,14,21,38]. The value $T_\alpha = 0.44$ s [38] for ^{274}Mt was not included in this figure because the energy $E_\alpha = 9.76$ MeV measured by the focal-plane detector is far below the value following from the expected Q_α energy (by about 0.7 MeV, see Fig. 5) and could originate from a transition to an excited level of the daughter nucleus. The heaviest isotopes of each element (full red symbols) belong to the decay chains of the nuclei originating from ^{293,294}117 synthesized in the ^{249}Bk + ^{48}Ca reaction ([1–3] and this work). The increase of neutron number in odd-Z nuclei with $N \geqslant 165$ results in a decrease of the Q_α energy (see Fig. 5) and a considerable increase of their half-lives (see Fig. 7). 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 Fig. 7 and produced in ^{48}Ca-induced reactions,

TABLE II. Decay properties of nuclei originating from ^{294}117 and ^{293}117.

Z	N	A	No. observed[a]	Decay mode, branch (%)[b]	Half-life[c]	E_α (MeV)[d]
117	177	294	4(3/3)	α	50^{+60}_{-18} ms	10.81 – 10.97
	176	293	16(15/15)	α	22^{+8}_{-4} ms	10.60 – 11.20
115	175	290	4(3/4)	α	$0.24^{+0.28}_{-0.09}$ s	9.78 – 10.28
	174	289	20(15/16)	α	$0.33^{+0.12}_{-0.08}$ s	10.15 – 10.54
113	173	286	4(4/4)	α	13^{+12}_{-4} s	9.61 – 9.75
	172	285	20(17/17)	α	$4.2^{+1.4}_{-0.8}$ s	9.47 – 10.18
111	171	282	4(4/4)	α	59^{+55}_{-19} s	9.01 ± 0.05
	170	281	20(17/2)	α/SF:10/90	17^{+6}_{-3} s	9.28 ± 0.05
109	169	278	4(3/3)	α	$5.2^{+6.2}_{-1.8}$ s	9.38 – 9.55
	168	277	2(2/-)	SF	5^{+9}_{-2} ms	
107	167	274	4(3/4)	α	54^{+65}_{-19} s	8.76 ± 0.05
105	165	270	4(4/-)	SF	17^{+15}_{-6} h	

[a] Number of observed decays and number of events used for calculations of half-lives and α-particle energies, respectively.

[b] Branching ratio is not shown if only one decay mode was observed.

[c] Error bars correspond to 68% confidence level.

[d] Energy range of α particles detected by the focal-plane or both the focal-plane and side detectors is shown if it exceeds the energy resolution of the detector.

except for ^{281}Rg, are α emitters; their stability is determined by α decay. This provides additional evidence of the high stability of the superheavy nuclei with respect to spontaneous fission.

The radioactive properties of nuclei observed in the ^{249}Bk + ^{48}Ca reaction in 2010 [2,3] and in 2012 ([1] and this work), as well as in the reaction ^{243}Am(^{48}Ca, $2n$)^{289}115 [13,14], are shown in Table II.

V. CONCLUSION

The discovery of element 117 that was reported for the first time in 2010 [2,3] has now been corroborated through the observation in total of 11 additional decay chains originating from the $4n$ evaporation product, ^{293}117, and three decay chains of the $3n$ channel, ^{294}117, of the reaction ^{249}Bk + ^{48}Ca ([1] and this work). The radioactive decay properties of the twelve ^{293,294}117, ^{289,290}115, ^{285,286}113, ^{281,282}Rg, ^{277,278}Mt, ^{274}Bh, and ^{270}Db isotopes measured in this work are in full agreement with the results of the first experiment [2,3] as well as with the decay data determined for ^{289}115, ^{285}113, and ^{281}Rg measured in the cross-bombardment ^{243}Am(^{48}Ca,$2n$)^{289}115 reaction [13,14]. An α-decay branch of ^{281}Rg leading to the new SF nucleus, ^{277}Mt, was observed for the first time. The excitation functions of the production of the ^{293,294}117 isotopes in the ^{249}Bk + ^{48}Ca reaction have been measured in the excitation energy range of 30.4–48.3 MeV providing additional evidence of the identification of the nuclei of element 117. The maximum cross sections for the production of ^{294}117 and ^{293}117 nuclei at $E^* = 35$ and 43 MeV were found to be $\sigma_{3n} = 1.1^{+1.2}_{-0.6}$ pb and $\sigma_{4n} = 2.4^{+3.3}_{-1.4}$ pb, respectively.

The decay properties of odd-Z nuclei produced in the ^{249}Bk + ^{48}Ca reaction are in good agreement with those expected based on the properties of the neighboring even-Z and odd-Z nuclei and semiempirical systematics. They demonstrate a considerable increase in the stability of the superheavy elements associated with an increase in neutron number when approaching the magic neutron number $N = 184$. The observation of a SF isotope, ^{277}Mt, together with the previously synthesized even-even nucleus ^{282}Cn [20,27,28,36,37] and even-odd nucleus ^{277}Hs [29,30] indicate a weakening of the shell stabilization effect for nuclei at $N = 168$–170, in the intermediate region between $N = 162$ and $N = 184$. The presence of such a “fission corridor” was recently analyzed theoretically for even-even superheavy nuclei [39].

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