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Experiments on the synthesis of element 115 in the reaction ^{243}Am(^{48}Ca, xn)$^{291-x}$115

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The results of experiments designed to synthesize element 115 isotopes in the ^{243}Am+^{48}Ca reaction are presented. With a beam dose of 4.3×10^{18} 248-MeV ^{48}Ca projectiles, we observed three similar decay chains consisting of five consecutive α decays, all detected in time intervals of about 20 s and terminated at a later time by a spontaneous fission with a high-energy release (total kinetic energy ~220 MeV). At a higher bombarding energy of 253 MeV, with an equal ^{48}Ca beam dose, we registered a different decay chain of four consecutive α decays detected in a time interval of about 0.5 s, also terminated by spontaneous fission. The α decay energies and half-lives for nine new α-decaying nuclei are given. The decay properties of these synthesized nuclei are consistent with consecutive α decays originating from the parent isotopes of the new element 115, ^{288}115 and ^{287}115, produced in the 3n- and 4n-evaporation channels with cross sections of about 3 pb and 1 pb, respectively. The radioactive properties of the new odd-Z nuclei (105–115) are compared with the predictions of the macroscopic-microscopic theory. The experiments were carried out at the U400 cyclotron with the recoil separator DGFRS at FLNR, JINR.

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Our previous experiments were designed to synthesize even-Z superheavy elements (114−118) in the ^{48}Ca-induced reactions with actinide targets ^{244}Pu [1], ^{248}Cm [2], and ^{249}Cf [3]. The observed fusion-evaporation reaction products underwent two or three consecutive α decays terminated by spontaneous fission (SF).

For the neighboring odd-Z elements, especially their odd-odd isotopes, the probability of α decay with respect to SF should increase due to hindrance for SF. For such odd-Z nuclei one might expect longer consecutive α-decay chains terminated by the SF of relatively light descendant nuclides ($Z\leqslant105$).

The decay pattern of these superheavy nuclei is of interest for nuclear theory. In the course of α decays, the increased stability of nuclei caused by the predicted spherical neutron shell $N=184$ (or perhaps $N=172$) should gradually become weaker for descendant isotopes. However, the stability of these nuclei at the end of the decay chains should increase again due to the influence of the deformed shell at $N=162$.

The observation of nuclei passing from spherical to deformed shapes in the course of their consecutive α decays could provide valuable information about the influence of significant nuclear structure changes on the decay properties of these nuclei. For these investigations, we chose the fusion-evaporation reaction ^{243}Am+^{48}Ca, leading to isotopes of element 115. According to calculations based on the results of experiments on the synthesis of even-Z nuclei [1–4], the 3n- and 4n-evaporation channels leading to isotopes ^{288}115($N=173$) and ^{287}115($N=172$) should be observed with the highest yields.

The experiments were performed between July 14 and August 10, 2003, at the U400 cyclotron with the Dubna gas-filled recoil separator (DGFRS). The average incident beam intensity was 1.3 pμA. Over this period, equal beam doses of 4.3×10^{18} ^{48}Ca projectiles were delivered to the target at two bombarding energies. In this experiment, we chose the laboratory energies for the ^{48}Ca ions of 248 MeV and 253 MeV in the middle of the target. The systematic uncertainty in the beam energy is ~1 MeV. With the beam energy resolution, small variation of the beam energy during irradiation, and energy losses in the target (~3.3 MeV), we expected the resulting compound nuclei ^{291}115 to have excitation energies 38.0–42.3 MeV and 42.4–46.5 MeV, respectively [5].

The 32-cm^2 rotating target consisted of the enriched isotope ^{243}Am(99.9%) in the form of AmO_2. The target material was deposited onto 1.5-μm Ti foils to a thickness corresponding to ~0.36 mg cm^{-2} of ^{243}Am.

The evaporation residues (EVRs) recoiling from the target were separated in flight from the ^{48}Ca beam ions, scattered particles, and transfer-reaction products by the DGFRS [6]. The transmission efficiency of the separator for $Z=115$ nuclei was estimated to be about 35% [6].

The detection system consisted of a multiwire proportional counter to measure time-of-flight (TOF) and a 4×12-cm^2 semiconductor focal-plane detector array with 12 vertical position-sensitive strips to measure the decay of the implanted recoils. This detector, in turn, was surrounded by eight 4×4-cm^2 side detectors without position sensitivity, forming a box of detectors open from the front side. The detection efficiency for α decays of implanted nuclei was 87% of 4π. The detection system was tested by registering the recoil nuclei and α and SF decays of the known isotopes of No and Th, as well as their descendants, produced in the

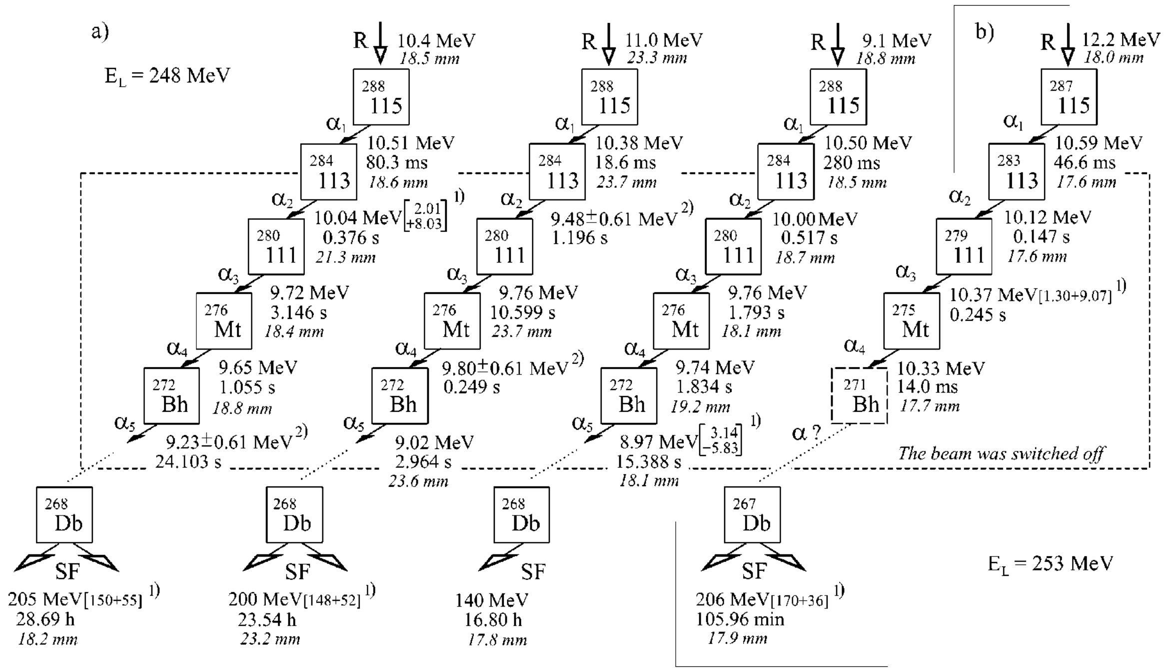

FIG. 1. Time sequences in the decay chains observed at two ^{48}Ca energies $E_L=248$ MeV (a) and $E_L=253$ MeV (b). Measured energies, time intervals, and vertical positions with respect to the top of the strips of the observed decay events are shown. (1) Energies of events detected by both the focal-plane detector and side detectors. (2) Energies of events detected by side detectors only. We have inserted an unobserved nuclide in the fourth decay chain.

reactions ^{206}Pb(^{48}Ca, $2n$) and $^{\mathrm{nat}}$Yb(^{48}Ca, $3-5n$). The full width at half maximum (FWHM) of the energy distribution for α particles absorbed in the focal-plane detector was 60–100 keV, depending on the strip and the position in the strip. For α's escaping the focal-plane detector at different angles and registered by side detectors, the energy resolution of the summed signals was 140–200 keV. The FWHM of the position distributions of correlated decays of nuclei implanted in the focal-plane detector were 0.9–1.9 mm for EVR-α correlations and 0.5–0.9 mm for EVR-SF correlations. If both the focal-plane detector and a side detector detected an α particle, the position resolution depended on the amplitude of the signal in the focal-plane detector (see, e.g., Fig. 4 in Ref. [3]), but was generally inferior to that obtained for the full-energy signal.

Fission fragments from ^{252}No implants produced in the ^{206}Pb+^{48}Ca reaction were used for a fission-energy calibration. The measured fragment energies were not corrected for the pulse-height defect of the detectors, or for the energy loss in the detectors' entrance windows, dead layers, and low-pressure pentane gas filling the detection system. The mean sum energy loss of fission fragments from the SF-decay of ^{252}No was estimated to be about 20 MeV.

To greatly reduce the background, we switched off the beam after a recoil signal was detected with preset parameters of implantation energy and TOF expected for $Z=115$ evaporation residues, followed by an α-like signal with an energy of $9.6\ \mathrm{MeV}\leqslant E_\alpha\leqslant 11.0$ MeV in the same strip, within a position window of 1.4–1.9 mm and time interval of up to 8 s. The beam-off interval was initially set at 2 min. If, in this time interval, an α particle with $E_\alpha>8.6$ MeV was registered, the beam-off interval was extended to between 12 min and 2.5 h. Thus, all the expected sequential decays of the daughter nuclides with $Z\leqslant 113$ could be observed under very low background conditions. With the full beam intensity on target, the average counting rate of such "EVR$\rightarrow\alpha$" events by detector system was about 2 h^{-1}. The total counting rate for α particles with $E_\alpha>8$ MeV by the whole detector array during beam-off pauses was about 3 h^{-1}. The majority of the events with $E_\alpha>8$ MeV is caused by the α decays of the short-lived isotopes ^{212}Po ($T_{1/2}=0.3\ \mu$s, $E_\alpha=8.78$ MeV) and ^{213}Po ($T_{1/2}=4.2\ \mu$s, $E_\alpha=8.38$ MeV) detected in coincidence with β^- decays of the precursors ^{212}Bi and ^{213}Bi (see, e.g., Ref. [2]). Only four such EVR$\rightarrow\alpha$ events were followed by α particles with $E_\alpha>9$ MeV.

The three similar decay chains observed at 248 MeV are shown in Fig. 1(a). The implantations of recoils in strips 2, 3, and 4 of the focal-plane detector were followed by α particles with $E_\alpha=10.46\pm0.06$ MeV. These sequences switched the ion beam off, and four more α decays were detected in total time intervals of 29 s, 15 s, and 20 s, respectively, in the absence of a beam-associated background. The last α decay in the first chain and second and fourth α decays in the second chain were registered by the side detectors only. The energies deposited by these α particles in the focal-plane detector were not registered. However, with the actual α-counting rates, the probability that these α particles appeared in the detector ($\Delta t\sim 30$ s) as random events can be estimated to be approximately 1.5% [7], so we assign them to the decay of the same implanted nuclei. During the remainder of the 2.5 h beam-off period following the last position-correlated α particles, no α particles with $E_\alpha>7.6$ MeV were registered by the focal-plane detectors. The SF decay of the final nuclei in these chains was detected

28.7 h, 23.5 h, and 16.8 h, respectively, after the last α decay. A search was performed to identify α decays correlated closely in time (<60 s) and position to each of the three SF events in Fig. 1(a). No correlations were found.

In the first two decay chains, the focal-plane detector and side detectors simultaneously detected fission fragments with sum energies of 205 MeV and 200 MeV. In the third chain, only the focal-plane detector registered a fission fragment. All three SF events were registered in the corresponding strips and positions where the three EVR-α_1-α_5 decay chains were observed [see Fig. 1(a)]. Therefore, these SF events were assigned to the spontaneous fission of the descendant nuclei in these observed chains.

In the course of this experiment at 248 MeV, we observed only seven spontaneous-fission events. In addition to the three previously mentioned events, three other SF events, with measured energies of 147 MeV, 168 MeV, and 154 MeV, were detected 0.51 ms, 4.1 ms, and 2.07 ms after the implantation of corresponding recoil nuclei in strips 12, 10, and 11, respectively. For the second and third events, both the focal plane and side detectors registered fission fragments. Based on the apparent lifetime, we assign these events to the spontaneous fission of the 0.9-ms $^{244\text{mf}}$Am isomer, a product of transfer reactions with the ^{243}Am target. The DGFRS suppresses the yield of such products by a factor of 10^5 [2]. An additional long-lived SF event with $E_{\text{sum}}=146$ MeV was observed in strip 2. This SF event corresponds to the background level of long-lived SF Cf isotopes produced in incomplete fusion reactions in a previous experiment with a ^{249}Cf target [3] where the same set of detectors was used.

At 253 MeV, the aforementioned EVR-α_1-α_5 SF decay chains were not observed. However, a different decay chain, consisting of four α decays and a spontaneous fission, was registered [see Fig. 1(b)]. The beam was switched off after the detection of an EVR signal followed in 46.6 ms by an α particle with $E_\alpha=10.59$ MeV in the same position in strip 7. Three other α decays were detected in a time interval of about 0.4 s in the absence of beam-associated background. After 106 min, the terminal SF event was detected in-beam with a sum energy of 206 MeV in the same position in strip 7 [see Fig. 1(b)]. In addition to this SF event with $E_{\text{sum}}=206$ MeV, the decays of three other long-lived background SF nuclei, with measured fission-fragment energies of 168 MeV, 154 MeV, and 151 MeV, were also observed in strips 4, 3, and 1, respectively.

The radioactive properties of nuclei in this decay chain differ from those of the nuclei observed at the lower bombarding energy. The total decay time of this chain is about ten times shorter and the α decays are distinguished by higher α-particle energies and shorter lifetimes. Its production also required increasing the beam energy by about 5 MeV, so we assumed that the decay chain originates from another parent nucleus.

It is most reasonable that the different decay chains originate from neighboring parent isotopes of element 115, produced in the complete fusion reaction ^{243}Am$+^{48}$Ca followed by evaporation of three and four neutrons from the compound nucleus ^{291}115. Indeed, at the excitation energy $E^*=40$ MeV, close to the expected maximum for the

TABLE I. Decay properties of nuclei.

Isotope	Decay mode	Half-life	E_α (MeV)	Q_α (MeV)
^{288}115	α	87^{+105}_{-30} ms	10.46 ± 0.06	10.61 ± 0.06
^{284}113	α	$0.48^{+0.58}_{-0.17}$ s	10.00 ± 0.06	10.15 ± 0.06
^{280}111	α	$3.6^{+4.3}_{-1.3}$ s	9.75 ± 0.06	9.87 ± 0.06
^{276}Mt	α	$0.72^{+0.87}_{-0.25}$ s	9.71 ± 0.06	9.85 ± 0.06
^{272}Bh	α	$9.8^{+11.7}_{-3.5}$ s	9.02 ± 0.06	9.15 ± 0.06
^{268}Db	SF(α/EC)	16^{+19}_{-6} h		
^{287}115	α	32^{+155}_{-14} ms	10.59 ± 0.09	10.74 ± 0.09
^{283}113	α	100^{+490}_{-45} ms	10.12 ± 0.09	10.26 ± 0.09
^{279}111	α	170^{+810}_{-80} ms	10.37 ± 0.16	10.52 ± 0.16
^{275}Mt	α	$9.7^{+46}_{-4.4}$ ms	10.33 ± 0.09	10.48 ± 0.09
^{271}Bh	α			
^{267}Db	SF	73^{+350}_{-33} min		

3n-evaporation channel, we observed longer decay chains assigned to the odd-odd isotope ^{288}115. Increasing the beam energy by 5 MeV results in a reduced ^{288}115-isotope yield and, at the same time, an increased yield of the 4n-evaporation channel leading to the odd-even isotope ^{288}115. Corresponding cross sections for the 3n- and 4n-evaporation channels at the two projectile energies amount to $\sigma_{3n}=2.7^{+4.8}_{-1.6}$ pb and $\sigma_{4n}=0.9^{+3.2}_{-0.8}$ pb. Note that these values are comparable with results of recent experiments [4] where excitation functions for the reaction ^{244}Pu(^{48}Ca, xn) have been measured with maximum cross sections for the evaporation of three to five neutrons of $\sigma_{3n}=2$ pb, $\sigma_{4n}=5$ pb, and $\sigma_{5n}=1$ pb.

The decay properties of the product nuclei are presented in Table I. In the decay chains shown in Fig. 1(a), we assigned SF events to the isotope ^{268}Db following five consecutive α decays. It is also possible that this isotope undergoes α decay or electron capture (EC). In the case of the α decay of ^{268}Db with $T_\alpha>2.5$ h (after the beam-off period), one would more reasonably expect SF or EC of ^{264}Lr, because the α decay of ^{264}Lr seems improbable due to the low expected α decay energy ($Q_\alpha=6.84$ MeV [8], $T_\alpha>100$ d). The electron capture of ^{268}Db or ^{264}Lr leads to the even-even isotopes ^{268}Rf or ^{264}No, for which rapid spontaneous fission can be expected (e.g., $T_{SF}=1.4$ s is predicted for ^{268}Rf [9]). Since both fission fragments of these terminal nuclei can have the $Z=50$ and $N=82$ closed shell configurations, one would expect their SF decays to result in narrowly symmetric mass distributions with rather high total kinetic energies, TKE$\approx230-240$ MeV (see, e.g., Ref. [10]). Indeed, the sum fission fragment energies observed for the terminal nuclei are close to this value. Note, the likelihood that EC decay occurs earlier in the observed decay chains is small because of the short T_α for the observed α decays of elements 115, 113, 111, Mt, and Bh.

In the single decay chain originating from ^{287}115 [see Fig. 1(b) and Table I], we propose that we have missed the α decay of ^{271}Bh. This assumption follows from the α-decay properties of nuclei located around $N=162$ as predicted by macroscopic-microscopic theory [8]. The expected T_α value for ^{271}Bh should be ~10 s ($Q_\alpha=9.07$ MeV [8], $T_\alpha\approx5$ s for

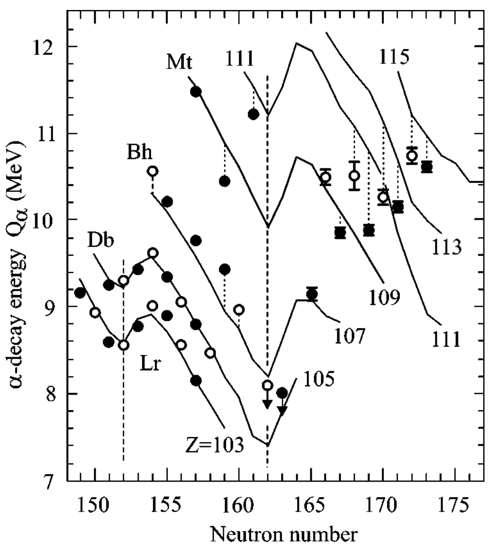

FIG. 2. α-decay energy vs neutron number for isotopes of odd-Z elements (solid circles, odd-odd isotopes; open circles, odd-even isotopes) [12–16]. Solid lines show theoretical Q_α values [8]. Data at $N \geqslant 162$ are from the present work. Upper Q_α limits for ^{267}Db and ^{268}Db were calculated assuming a hindrance factor of 3 (circles with arrows).

an allowed transition), which is much shorter than the interval between the last observed α particle and the terminating SF-event, but is much longer than the intervals between the observed correlated α particles. A search was performed to identify α particles with $E_\alpha > 7.0$ MeV correlated in position to the observed α decays. No correlated α particles were observed within an hour after the last α decay. In the correlated decay chains shown in Fig. 1, 19 α particles were registered using a detector with 87% efficiency, so the loss of one α particle seems rather probable. The experimental decay scheme for ^{287}115 is also supported by the agreement of the observed decay properties of the other nuclides in the decay chain with the expectations of theory (see Fig. 2). This means that the SF occurs directly in the decay of ^{267}Db since the calculated α-decay energies and EC-decay energies for this isotope are rather low ($Q_\alpha = 7.41$ MeV [8], $Q_{EC} = 1$ MeV [5]) and their expected partial half-lives significantly exceed the observed time interval of 106 min.

For the measured α decay energies of the newly produced isotopes, one can estimate half-lives for allowed transitions and compare them with experimental values under the Geiger-Nuttall treatment using the formula by Viola and Seaborg [11]. Parameters are obtained from fits to the T_α versus Q_α values of 65 known *even-even* nuclei with $Z > 82$ and $N > 126$. The ratios between experimental T_{exp} and calculated T_{calc} half-lives define the hindrance factors caused by odd numbers of protons and/or neutrons in the newly synthesized nuclei. The measured T_{exp} values closely reproduce the calculated ones for the first two nuclei of these chains; thus the element 115 and element 113 isotopes have rather low hindrance factors, if any, for α decay. For the isotopes of elements 111, Mt and Bh, the difference between measured and calculated T_α values results in hindrance factors of 3–10. These match the hindrance factors that can be extracted for the deformed odd-odd nuclei ^{272}111 and descendants produced in experiments at GSI [12] and RIKEN [13]. One can suppose that in this region of nuclei, a noticeable transition from spherical to deformed shapes occurs at $Z = 109-111$, resulting in the complication of the level structures of these nuclei and in an increased probability of α transitions going through excited states. Another sign of such a shape transition might be the significant increase in the difference of α-decay energies of the neighboring isotopes observed as the decay chains reach $Z = 111$, see Fig. 2. This assumption is in agreement with macroscopic-microscopic calculations [8]. The deformation parameter β_2 was calculated to be 0.072 and 0.138 for ^{288}115 and ^{284}113, respectively. As the decay chain recedes from the shell closure at $Z = 114$, the deformation parameter β_2 increases to 0.200, 0.211, and 0.247 for ^{280}111, ^{276}Mt, and ^{272}Bh, respectively.

The α-decay energies of these synthesized isotopes are plotted in Fig. 2, in which the α-decay energies of known odd-Z isotopes with $Z \geqslant 103$ together with theoretical Q_α values [8] for nuclides with $Z = 103-115$ are also shown. The α-decay energies attributed to the isotopes of Mt and Bh coincide well with theoretical values [8]. For the isotopes ^{279,280}111 and ^{283,284}113 the difference between theoretical and experimental Q_α values amounts to 0.6–0.9 MeV. Some part of this energy deficit can be explained by possible γ-ray emission from excited levels populated during α decay. Thus, low-lying level structure calculations for these nuclei are desirable for a more quantitative comparison with theory.

As a whole, the results of the present work suggest that in ^{48}Ca-induced reactions one can produce and study new nuclei over a wide range of Z and N. These investigations should result in significant new information on the influence of nuclear structure on the decay properties of superheavy nuclei.

Due to the hindrance to spontaneous fission caused by the odd proton, these odd-Z nuclei can undergo consecutive α decays leading to the lighter transactinide elements that have a large neutron excess. The number of neutrons in such nuclides can be varied downward by using other target nuclei, e.g., ^{241}Am, ^{237}Np, or ^{231}Pa. Therefore, the investigation of the region of nuclei located near deformed shell closures $Z = 108$ and $N = 162$ that has been explored from the $N < 162$ side in the cold fusion reactions [12,13] can be approached from the high-N side using ^{48}Ca-induced reactions. Note also that the rather long lifetimes of Db isotopes permit us to investigate in more detail the chemical properties of this element, previously studied only in on-line experiments with the short-lived isotopes ^{261,262,263}Db ($T_\alpha = 1.8$ s, 34 s, and 27 s) (see, e.g., Ref. [17] and references therein).

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