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Synthesis of superheavy nuclei in the reactions of ^{244}Pu and ^{248}Cm with ^{48}Ca

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Abstract. This paper presents results of the experiments aimed at producing long-lived superheavy elements located near the spherical-shell closures with $Z \geqslant 114$ and $N \geqslant 172$ in the ^{244}Pu + ^{48}Ca and ^{248}Cm + ^{48}Ca reactions. The large measured α-particle energies of the newly observed nuclei, together with the long decay times and spontaneous fission terminating the chains, offer evidence of the decay of nuclei with high atomic numbers. The decay properties of the synthesized nuclei are consistent with the consecutive α-decays originating from the parent nuclides ^{288,289}114 and ^{292}116, produced in the $3n$ and $4n$ evaporation channels with cross-sections of about a picobarn. The present observations can be considered as experimental evidence of the existence of the "island of stability" of superheavy elements.

PACS. 23.60.+e Alpha decay – 25.70.-z Low and intermediate energy heavy-ion reactions – 27.90.+b $A \geq 220$

1 Introduction

Beyond the domain of the heaviest known nuclei located near deformed shell closures with $Z = 108$ and $N = 162$, a substantial enhancement in the stability of heavy nuclei is predicted when approaching the next spherical shells, with $Z \geqslant 114$ and $N \geqslant 184$, above $Z = 82$ and $N = 126$. The most neutron-rich isotopes of superheavy elements and, consequently, the most stable, are expected to be produced in the fusion reactions of actinide targets with the doubly magic ^{48}Ca projectile. The resulting compound nuclei should have an excitation energy of about 30–33 MeV at the Coulomb barrier and should de-excite with the greatest probability by the evaporation of three or four neutrons. The investigation of the heaviest even-even nuclei produced in the $4n$ evaporation channel allows a more clear comparison with theoretical predictions due to their unhindered α-decays and spontaneous fission (SF). Our experiments with the ^{244}Pu + ^{48}Ca and ^{248}Cm + ^{48}Ca reactions were designed to produce elements 114 and 116 at the picobarn cross-section level, thus exceeding the sensitivity of previous attempts to synthesize new elements in ^{48}Ca-induced reactions with actinide targets by more than two orders of magnitude.

2 Experiment and results

A beam of ^{48}Ca^{+5} ions was delivered by the U400 cyclotron at FLNR, JINR. The average beam intensity at the target was 0.7 pμA at the consumption rate of ^{48}Ca material of ~ 0.3 mg h^{-1}. The 32 cm^2 rotating targets consisted of the enriched isotopes ^{244}Pu (98.6%) and ^{248}Cm (96.3% or 97.4%) in the form of dioxide deposited onto 1.5 μm Ti foils to a thickness of 0.32–0.37 mg cm^{-2}. We chose the bombarding energies for ^{48}Ca ions of 236 MeV and 240 MeV in the middle of the ^{244}Pu and ^{248}Cm layers, respectively. Taking into account the energy losses in the targets and the overall beam energy and target thickness variations, we expected the resulting compound nuclei ^{292}114 and ^{296}116 to have excitation energy in ranges of 31.5–39.0 MeV and 28.9–37.2 MeV, respectively. Thus, the compound nuclei which survive SF should de-excite most probably by the evaporation of 3 or 4 neutrons and γ-rays.

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 Dubna Gas-filled Recoil Separator [1]. The transmission efficiency of the separator for $Z = 114$ and 116 nuclei was estimated to be about 35–40%.

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The detection module mounted in the separator's focal plane consisted of a time-of-flight system (TOF) followed by a 4×12 cm^2 semiconductor detector array with 12 vertical position-sensitive strips, in which the recoils were implanted. 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 produced in the reactions $^{204,206\text{-}208}$Pb(^{48}Ca, xn) [1,2].

The energy resolution for α-particles absorbed in the focal-plane detector was about 55 keV. For α's escaping the focal-plane detector at different angles and registered by side detectors, the energy resolution of the summed signals was $\sim$190 keV. The FWHM position resolutions of the signals of correlated decays of nuclei implanted in the detectors were 1.4 mm and 1.2 mm for EVR-α and EVR-SF signals, respectively, in the experiments of 1998. Values of 0.8 mm and 0.5 mm, respectively, were obtained in subsequent experiments due to the improvement of the detection system.

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, for energy loss in the detectors' entrance windows, dead layers, and the pentane gas filling the detection system. The mean sum energy loss of fission fragments for ^{252}No was about 20 MeV.

The ^{244}Pu + ^{48}Ca bombardments were performed in November-December 1998, and June-October 1999. A total of 1.5×10^{19} ^{48}Ca projectiles was delivered to the target.

According to the concept of the "stability island" of superheavy elements, when an α-decay chain reaches the edge of the stability region, it should be terminated by SF [3,4]. In the course of this experiment, we observed five SF events. Two SF decays with total measured energies of 149 MeV and 153 MeV occurred *within milliseconds* following the implantation of the recoil. Based on the lifetime, we assigned these events to 0.9 ms $^{244\mathrm{mf}}$Am, a product of transfer reactions. Three other SF events terminated the α-decay sequences of relatively long-lived nuclei. Two such SF events, with energies of 221 MeV and 213 MeV, were observed in strips 2 and 8, respectively [5]. The full decay chains including these SF events are shown in fig. 1.

We calculated the probabilities that these decay sequences were caused by chance correlations of unrelated events at any position of the detector array and at the positions in which the events occurred [6]. The probability that both decay chains consist of random events is less than 5×10^{-13}.

The formation of the nuclei which initiated the observed decays resulted from "instant" ^{48}Ca beam energies of 237.6 and 237.0 MeV in the middle of the target. This would favor de-excitation of the compound nucleus by evaporation of 4 neutrons, which leads to the even-even nucleus ^{288}114. Indeed, the observed chains, including two α-decays and terminated by SF, match the decay scenario predicted for the even-even nuclide ^{288}114 [3,4]. The detected sequential decays have $T_{1/2}$ *vs.* E_α-values that correspond well to the decays of the even-even isotopes of elements 114 and 112 (see below). The measured total energies deposited in the detector array for both fission events exceed the average value measured for ^{252}No by about 40 MeV, which also indicates the fission of a rather heavy granddaughter nucleus. From the above considerations, we can conclude that the detected decay chains originate from the parent even-even nuclide ^{288}114, produced in the ^{244}Pu + ^{48}Ca reaction via the $4n$ evaporation channel.

The next SF event was observed in strip 8 with an energy of 172 MeV [7]. The entire position-correlated decay chain is shown in fig. 1. The probability that this decay sequence was caused by the chance correlation of unrelated events is 6×10^{-3}. This decay sequence evidently originates from a different parent nucleus than the chains that were assigned to the decay of ^{288}114. The best candidate for the parent nucleus is the even-odd isotope ^{289}114, produced in the $3n$ evaporation channel. Indeed, the α-decaying nuclides in this chain are characterized by lower decay energies than the corresponding members of the chain attributed to the decay of ^{288}114, while SF terminates the decay sequence at a later stage.

A priori, one cannot exclude that the investigated excitation energy range of 31.5–39 MeV was not optimal for the production of this isotope. To check this assumption, we performed an experiment in November-December 1999, using a lower projectile energy of 231 MeV ($E^* \approx 28.5$–34.5 MeV). A total beam dose of 4.6×10^{18} was accumulated. Only one fission event, the 0.9 ms $^{244\mathrm{mf}}$Am isomer with $E_{\mathrm{tot}} = 156$ MeV, was detected in this bombardment.

We estimate the cross-sections for producing both nuclides in this reaction to be about a picobarn. The bombardment performed at the lower projectile energy resulted in an upper production limit of 2 pb.

On June 14, 2000, we started an experiment aimed at the synthesis of superheavy nuclei with $Z = 116$ in the complete-fusion reaction ^{248}Cm + ^{48}Ca [8]. During June-July and November-December 2000, January and April-May 2001, we collected a beam dose of 2.3×10^{19} ^{48}Ca projectiles.

To improve background conditions for detecting long-time decay sequences, a special measurement mode was employed [1]. The beam was switched off after a recoil signal was detected with parameters of implantation energy and TOF expected for $Z = 116$ evaporation residues, followed by an α-like signal with an energy of $10.05\,\mathrm{MeV} \leqslant E_\alpha \leqslant 11.5\,\mathrm{MeV}$, in the same strip, within a position window $\Delta y = 2$ mm and time intervals of up to 5 s. The duration of the pause was determined from the observed pattern of out-of-beam α-decays and varied from 2 to 60 minutes. Thus, all the expected sequential decays of the daughter nuclides with $Z \leqslant 114$ could be observed in the absence of beam-associated background. The total counting rate for α-particles with $E_\alpha > 8$ MeV by the whole de-

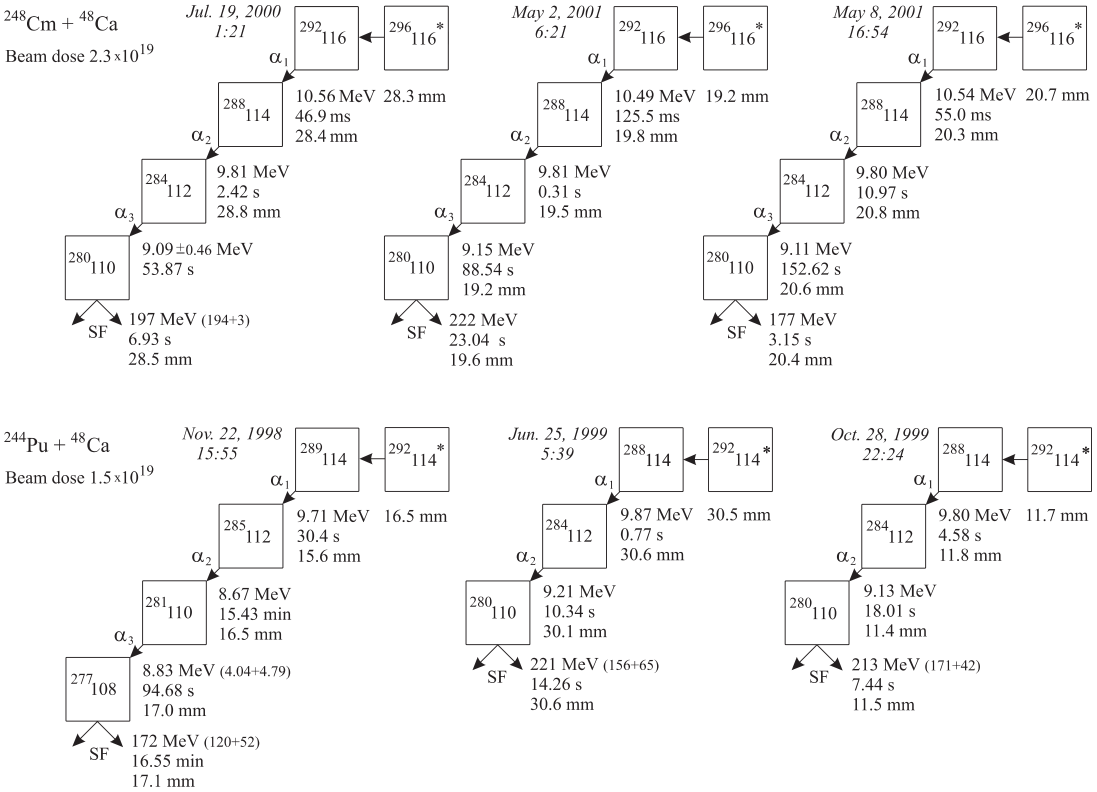

Fig. 1. Time sequences in the observed decay chains. Vertical positions of the observed events are given with respect to the top of the strip. Values in parentheses show α-particle and fission energies measured by the focal-plane and side detectors, respectively.

tector array during beam-off pauses was about 2 h^{-1}, the counting rate of the focal-plane detector alone was about 6×10^{-5} min^{-1} per pixel $\Delta y = 1.4$ mm. The majority of these events is caused by the α-decays of the short-lived isotopes ^{212}Po and ^{213}Po detected in coincidence with β^--decays of their precursors ^{212}Bi and ^{213}Bi produced in the transfer reactions with ^{244}Pu [5,7] and ^{248}Cm [8] targets. During the bombardments of the ^{248}Cm target and subsequent off-line measurements we observed SF events that could be attributed to the spontaneous fission of ^{252,254}Cf and ^{256}Fm, long-lived products of transfer reactions with the ^{248}Cm target. For a pixel $\Delta y = 1.4$ mm, the signals from SF events were observed with an average frequency of 7×10^{-7} min^{-1}.

During these irradiations three similar decay sequences were observed that can be assigned to the implantation and decay of the isotope of element 116 with mass number 292 (fig. 1). The implantation events of heavy recoils in strips 4, 5, and 1 of the focal-plane detector were followed by α-particles with $E_\alpha = 10.53 \pm 0.06$ MeV. These sequences switched the ion beam off, and further decays were detected under lower-background conditions. All events in the three decay chains appeared within position intervals less than 0.6 mm and are consistent with one another taking into account the energy resolution of the detectors and statistical uncertainty in lifetimes determined from a few events. The third α-decay with the energy of 8.63 MeV in the first decay chain was registered by a side detector only. The energy deposited by this α-particle in the focal-plane detector was not registered because it was lower than the detection threshold of 0.92 MeV. However, the probability that the third α-particle appeared in the chain ($\Delta t \sim 1$ min) as a random event can be estimated to be only $\sim 1\%$, so we assign it to the decay of the same implanted nucleus. Thus its total energy is determined with a larger uncertainty to be $E_\alpha = 9.09 \pm 0.46$ MeV. The probability of the observed event chain being totally of random origin is negligible [6].

All the decays following the first 10.53 MeV α-particles agree well with the decay chains of ^{288}114, previously observed in the ^{244}Pu + ^{48}Ca reaction (see fig. 1). Thus, it is reasonable to assign the observed decays to the nuclide ^{292}116, produced via evaporation of four neutrons in the complete-fusion reaction ^{248}Cm + ^{48}Ca. All the decay chain members follow the Geiger-Nuttall Q_α *vs.* T_α relationship for even-even nuclei. Substituting the values $E_{\alpha 1} = 10.53$ MeV and $T_{\alpha 1} = 53^{+62}_{-19}$ ms, $E_{\alpha 2} = 9.82$ MeV and $T_{\alpha 2} = 2.6^{+2.0}_{-0.8}$ s, and $E_{\alpha 3} = 9.15$ MeV and $T_{\alpha 3} = 45^{+34}_{-14}$ s, measured in the ^{244}Pu + ^{48}Ca and ^{248}Cm + ^{48}Ca reactions into the formula by Viola and Seaborg, with parameters fitted to all the known even-even nuclides with $Z > 82$ and $N > 126$ [3], results in the atomic numbers $Z_1 = 115.8^{+1.3}_{-0.8}$, $Z_2 = 114.8^{+0.9}_{-0.8}$, and $Z_3 = 112.1^{+0.9}_{-0.7}$, respectively.

3 Discussion

The lifetimes of the new isotopes $^{285}112$ and $^{281}110$ appear to be approximately 10^6 times longer than those of the known nuclei $^{277}112$ and $^{273}110$ [9,10], which have eight fewer neutrons. Note that $^{289}114$, $^{285}112$ and $^{281}110$ are about 10^4–10^5 times more stable than $^{285}114$, $^{281}112$ and $^{277}110$, the α-decay products of $^{293}118$ that was recently produced in the bombardment of ^{208}Pb with ^{86}Kr ions [11].

The newly observed nuclides $^{284}112$, $^{288}114$, and $^{292}116$ are the heaviest known α-decaying even-even nuclides, following the production of ^{260,266}Sg [12], ^{264,266}Hs [12,13], and $^{270}110$ [13]. A comparison with theoretical calculations [3,4] of the measured decay properties of these new nuclides, including $^{280}110$ ($T_{1/2} = 7.6^{+5.8}_{-2.3}$ s), indicates that nuclei in the vicinity of spherical-shell closures with $Z = 114$ and $N = 184$ could be even more stable than predicted by theory. It can be seen in fig. 2 that α-decay energies of the heaviest new even-even nuclides with $Z = 112$, 114, and 116 are 0.35–0.5 MeV less than the corresponding predicted values. The heaviest even-odd nuclides follow this trend as well. Such a decrease in Q_α-values leads to an increase of partial α-decay lifetimes by an order of magnitude. Calculations are far less definite regarding spontaneous fission; however, we note that the observed SF half-life of $^{280}110$ exceeds the predicted value [3] by more than two orders of magnitude.

The principal result of the present work is the observation of the considerable increase in lifetimes of superheavy nuclei with $Z \geqslant 110$ with increasing neutron number. Comparison of the present data with predictions of macroscopic-microscopic models [3,4,14,15] and recent self-consistent models [16,17] shows that theoretical predictions agree with experimental results (see fig. 2). The observed chains, including three α-decays and terminated by SF, match the decay scenario predicted for the even-even nuclide $^{292}116$ [3,4]. In this respect, the decay properties of the new nuclides observed in present experiments confirm theoretical expectations and can be considered the proof of the existence of enhanced stability in the region of superheavy elements around $Z = 114$ and $N = 184$.

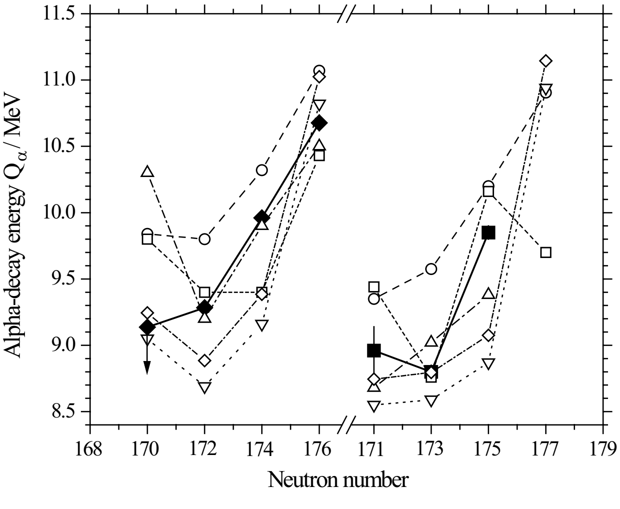

Fig. 2. Comparison of experimental (solid symbols) and calculated (open symbols) Q_α-values for the α-decay chains of $^{292}116$ and $^{289}114$. Circles show data from refs. [3,4] (mean values for neighbouring even-even nuclei are used for odd-N isotopes); squares, from ref. [16]; diamonds, from ref. [15]; triangles up, from ref. [17]; and triangles down, from ref. [14].

The ^{248}Cm + ^{48}Ca experiment is in progress.

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