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Yu.Ts.Oganessian, A.V.Yeremin, A.G.Popeko,
S.L.Bogomolov, G.V.Buklanov, M.L.Chelnokov,
V.I.Chepigin, B.N.Gikal, V.A.Gorshkov, G.G.Gulbekian,
M.G.Itkis, A.P.Kabachenko, A.Yu.Lavrentev,
O.N.Malyshev, J.Rohac, R.N.Sagaidak, S.Hofmann[1],
G.Münzenberg[1], S.Saro[2], G.Giardina[3], K.Morita[4]

THE SYNTHESIS OF AN ISOTOPE
OF THE NEW ELEMENT 114
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[1]Gesellschaft für Schwerionenforschung, D-64220 Darmstadt, Germany
[2]Department of Physics, Comenius University, SK-84215, Bratislava,
 Slovakia
[3]Dipartimento di Fisica dell'Universita di Messina, 98166 Messina,
 Italy
[4]Institute of Physical and Chemical Research (RIKEN), Wako-shi,
 Saitama, Japan

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Introduction

According to macro-microscopic nuclear theory the stability of heavy nuclides, undergoing mainly α-decay and spontaneous fission, is determined by the structural properties of nuclear matter. Their binding energy and lifetime increase most prominently in the vicinity of closed shells, corresponding to the spherical nuclear shape in the ground state. Following the well-known shells with Z=82 and N=126 in the stable spherical nucleus ^{208}Pb, the next closed neutron shell is expected at N=184. Calculations by R.Smolańczuk et al. [1,2] who successfully reproduced the decay properties of the known heavy nuclei, and P.Möller et al. [3] have shown that the stabilizing effect of the spherical shell starts already at N>168 for a broad region of isotopes with Z=104÷120. Such neutron-rich nuclei can be synthesized, as we have shown earlier (see for example, [4]), in fusion reactions using the heaviest isotopes of U, Pu and Cm as targets and a ^{48}Ca ion beam.

As a result of the significant mass excess of the doubly magic ^{48}Ca nucleus, the excitation energy of the compound nucleus at the Coulomb barrier only amounts to about 30 MeV. The deexcitation of this nucleus should proceed by the emission of 3 neutrons and γ-rays [5]. This circumstance should increase the survival probability of the evaporation residues (EVRs) as compared with the case of hot fusion reactions (E_x~50 MeV), which we have used for the synthesis of heavy isotopes of elements with atomic numbers Z=106, 108 and 110 [6]. On the other hand, the high asymmetry of the interacting nuclei in the entrance channel (A_p/A_T=0.2; Z_pZ_T=1880) should decrease possible dynamical limitations [7] on the fusion of massive nuclei as compared with more symmetrical cold fusion reactions.

In spite of these obvious advantages, previous attempts to synthesize new elements in ^{48}Ca-induced reactions gave only the upper limits of the production cross sections of superheavy elements [8,9,10].

The first positive result was obtained in spring 1998 in the ^{48}Ca+^{238}U reaction with a total beam dose of $3.5 \cdot 10^{18}$ ions. In this experiment two spontaneous fission events were observed, which were assigned to the decay of a new isotope of element 112 produced in the reaction ^{238}U(^{48}Ca,3n)^{283}112 with a cross section of $\sigma_{3n} = 5^{+6}_{-3}$ pb [11]. The half-life of the new nuclide, determined on the basis of the two events, was about 1.5 min. Thus it turned out to be about $3 \cdot 10^5$ times longer than the α-decay half-life of the known lighter isotope of element 112, synthesized in the cold fusion reaction ^{208}Pb(^{70}Zn,n)^{277}112 by S. Hofmann et al. [12].

The next experiment, carried out at the end of 1998, was aimed at the synthesis of element 114 in the reaction ^{48}Ca+^{244}Pu. In a 40-day irradiation with a beam dose of

$5.3 \cdot 10^{18}$ ions, a decay chain, consisting of three sequential α-decays and spontaneous fission, all takes about 34 min, was observed after the implantation of a heavy atom in the detector [13]. This decay chain is a good candidate for originating from the α-decay of the parent nucleus $^{289}114$ ($T_\alpha \sim 20$ s), produced in the 3n-evaporation channel with a cross section of about 1 pb.

In a third experiment, which is described in the present paper, an attempt was made to synthesize a new isotope, viz $^{287}114$, in the reaction $^{48}Ca + ^{242}Pu$.

If the identification performed in the first two experiments is correct, it is not difficult to predict the properties of the new isotope of element 114. According to the calculations and the above mentioned data, the isotope $^{287}114$ (N=173) should undergo predominantly α-decay to the daughter nucleus $^{283}112$, obtained formerly in the $^{48}Ca + ^{238}U$ reaction. Thus one could expect a short decay chain, α - SF, with a relatively short half-life with respect to α-decay (about several sec.) and a subsequent spontaneous fission with a considerably longer half-life (about several min.).

Experiment

The expected cross section for the production of the new isotope $^{287}114$ at the maximum of the excitation function for the 3n-evaporation channel is very small (~ 1 pb). This requires high sensitivity experiment. From this point of view, the production of an intense ion beam of the rare and extremely expensive isotope ^{48}Ca is the corner-stone of our attempts to synthesize superheavy elements.

The neutral atoms of calcium were injected into the plasma of the ECR-4M ion source [14] by a process of controlled heating of a metallic sample of ^{48}Ca (~ 50 mg) enriched to 70%. The sample was made from the calcium oxide right before being placed in the oven of the ion source. The whole preparation procedure of metallic Ca as well as the recuperation of the material from the ion source chamber was controlled by measuring the yield of γ-rays from the isotope ^{47}Ca (4.5 d), produced in the $^{48}Ca(\gamma,n)^{47}Ca$ reaction.

The $^{48}Ca^{+5}$ beam was extracted from the ECR ion source and injected into the Dubna U400 heavy ion cyclotron. The average intensity of the ion beam on the target was $4 \cdot 10^{12}$ pps at the material consumption rate of about 0.3 mg/h. The beam energy was determined with a precision of ~ 1 MeV, by measuring the energies of scattered ions and by a time-of-flight technique.

The targets consisted of the enriched isotope ^{242}Pu (97%) in the form of PuO_2, deposited onto 0.75 mg/cm^2 Ti foils of a thickness of 0.2 mg/cm^2. Each target had an area of 5 cm^2 in the shape of an arc segment with an angular extension of 60° and an average radius of 55 mm. Six targets were mounted on a disk that was rotated at 2000 rpm perpendicularly to the beam direction. A "chopper" system switched out the beam to make a pause (0.6 ms) at the moment when the target disk frames passed the beam.

The evaporation residues (EVRs) were separated in-flight from the beam particles and other reaction products by the electrostatic recoil separator VASSILISSA [15]. The values of the electric and magnetic fields were calculated according to a mean charge state of $<q> = 18$ of the EVRs and a mean energy of 36 MeV.

The transmission of the EVRs from the reaction $^{48}Ca + ^{242}Pu$ was estimated from test experiments. Cross sections of various xn-evaporation channels were measured in irradiations of ^{159}Tb, ^{174}Yb and $^{206,208}Pb$ targets with ^{48}Ca projectiles of different energies. The measurements showed that about 30% of the $^{290-x}114$ EVRs, produced with a ^{242}Pu target, would be implanted into the detector located at the focal plane of separator at a distance of 12 m from the target (Fig. 1).
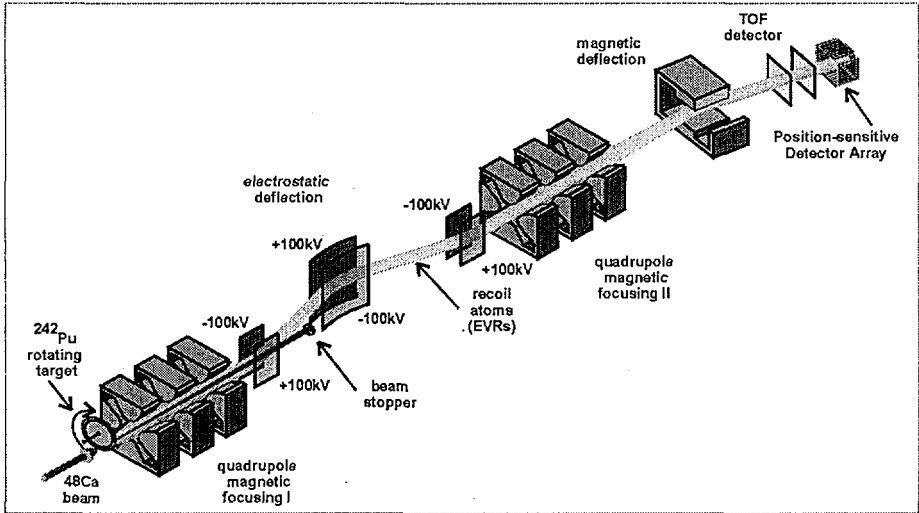

Fig. 1. Electrostatic recoil separator "VASSILISSA"

For the registration of the EVRs and their radioactive decay a system of time-of-flight (TOF) detectors and a silicon position-sensitive strip-detector array was installed in the focal plane of the separator. After registration by the TOF detectors, consisting of two secondary-electron foil converters with microchannel plates, the recoil atoms were implanted into a 16-strip silicon detector, which had an active area of 60x60 mm². Each strip had a longitudinal position sensitivity. A 3 μm thick degrader foil (Mylar) was placed in front of the silicon detectors for reduction of the number of low energy projectiles reaching the focal plane of the separator. Special measurements of the position resolution along each strip were performed. For sequential [α - α] decays it was 0.6 mm (FWHM), for correlated [EVR - α] - about 1 mm, and 1.5 mm for [EVR – SF]. The measurements were performed for recoil nuclei with energies of 4÷15 MeV, which corresponds to the expected energy interval for the signals originating from the implanted Z =114 nuclei.

The energy resolution for α-particles was 20 keV within the energy range from 6 to 9 MeV. The accuracy of the time registration for recording the events was about 1 μs. In order to increase the detection efficiency for α-particles, the front detector was surrounded by four silicon detectors of the same size as the stop detector. The entire array had the shape of a cube with a 60 mm edge length. In the present series of experiments

3

four neighbouring strips of the backward detectors were connected galvanicly so that a total of 16 energy sensitive segments were formed, each with an energy resolution of 120 to 150 keV. The efficiency of the silicon array for the detection of α-particles with full energy was 85% of 4π. Thus, the probability that the α-particle energy (E_α) was determined directly by the front detector was 50%; it was 35% when E_α was the sum of two signals $E_\alpha = E_{\alpha 1} + E_{\alpha 2}$ from the front and side detectors; and finally, it was 15% when the α-particle could be emitted backwards leaving only an escape signal ($E_{\alpha 1}$) in the front detector if $E_{\alpha 1} \geq 1$ MeV.

The signals from the TOF detectors were used to measure the velocity of the recoils and to distinguish the radioactive decays of previously implanted nuclei. The high efficiency of the TOF detectors made it possible to obtain very clean decay spectra. The time window for measuring decay chains could be considerably widened (up to several hours). The latter was particularly important for the correlation of decays with long half-lives in the presence of a continuously running beam.

At a beam intensity of $\sim 4 \cdot 10^{12}$ pps the total counting rate of all events at the focal plane detector was only 25-30 Hz. The counting rate at a single strip in a position interval of ± 1 mm (a 95% registration of correlated α - α decays) amounted: for the α-like signals (in the absence of signals from the TOF detector) with an energy higher than 7.5 MeV - to less than 1 h^{-1}; for the α-like signals with an energy from 1 to 4 MeV (signals corresponding to escaped α's) - to about 3 h^{-1}; for recoil-like signals (with a TOF signal) with an energy higher than 4 MeV - to about 4 h^{-1}.

Results

The experiment was performed during March 3 - April 5, 1999. The beam energy in the middle of target was $E_L = 235 \pm 2$ MeV ($E_x \sim 33.5$ MeV). Over a period of 32-days a total of $7.5 \cdot 10^{18}$ projectiles was passed through the target.

In the analysis of the experimental data we proceeded from the expected short decay chain of $^{287}114$ terminated by spontaneous fission of the daughter nucleus (or its grand-daughters). So, first of all, the decay events with a large energy deposit in the front detector (E>100 MeV) were selected. Four such events were observed in the reaction $^{48}Ca + ^{242}Pu$.

Two events with energies E=144 MeV and 175 MeV were registered 59 μs and 20 μs after the implantation of the corresponding position-correlated recoil atoms, respectively. We assigned these events to the SF - isomers, tentatively to the 24-μs $^{241mf}Pu$, produced in a transfer reaction, and being a neighbor of the ^{242}Pu-target nucleus.

For the other two events spontaneous fission was observed as two coincident signals (two fission fragments) with an energy deposited in the front detector $E_{F1} = 130$ MeV and in the side detector $E_{F2} = 65$ MeV, with $E_{tot} = 195$ MeV for the first event; and $E_{F1} = 110$ MeV, $E_{F2} = 55$ MeV, with $E_{tot} = 165$ MeV for the second event.

We searched for the recorded data backwards in time from each one of these two events for a preceding α-particles correlated in position. For the first case only one α-particle was detected in the front detector 1.32 s after the implantation in the middle of

4

the strip of a recoil nucleus with a measured energy E_{EVR}=10 MeV. This value fits the energy expected for the element-114 recoils, and the TOF signal is consistent with that expected for a complete-fusion EVR, as determined in the calibration reactions. The energy of the α-particle was E_α=10.29 MeV. The SF event was observed 559.6 s later. All the three signals (EVR, α and SF) appeared within a position interval of 0.82 mm, which indicates that there is a correlation between the observed decays. The probability for the observed correlation to be a random coincidence of signals imitating the decay chain (EVR, α and SF) at a given position window amounts to $\sim 2 \cdot 10^{-5}$.

In the second case, spontaneous fission was observed 243 s after the registration of the implanted recoil nucleus with an energy E_{EVR}=13.5 MeV. In the search for an α-particle in the time interval EVR-SF only a single escape signal with $E_{\alpha 1}$=2.31 MeV was found 14.45 s after the implantation of the recoil nucleus. All the three signals (EVR, α and SF) appeared within a position interval of 1.0 mm, which again indicates a correlation between the observed decays. The probability for the observed correlation to be a random coincidence of signals of the EVR, α and SF type in this case is $3 \cdot 10^{-4}$. The entire position correlated decay chains are shown in Fig. 2a. In both sequences the parent nucleus undergoes α-decay. It should be noted that the time intervals preceding the α-particle emission differ by about factor of 10. This is not surprising at the low statistics we have. As the total energy E_α for the second event is not determined, we assume that the α-decay in both cases proceeds from one and the same state. The half-life of the parent nucleus, derived on the basis of the two events , amounts to T_α= 5.5^{+10}_{-2} s.

The detected daughter nuclei undergo spontaneous fission. Their decay properties are comparable with the ones for the spontaneously fissioning emitter synthesized earlier in the reaction ^{48}Ca+^{238}U [11]. The spontaneous fission observed in these reactions is characterized by a considerable fission fragment energy deposit $<E_{tot}> \sim 190$ MeV. It is determined from comparison with the total kinetic energy (TKE) distribution of the fission fragments from ^{252}No, that was used for the calibration of the detectors. The spectra of the ^{252}No fission fragments were measured during preparatory experiments and were normalized to the literature data [16]. This procedure was described in Ref. [11] in detail .

The time intervals measured for all four spontaneous fission events (this work and Ref. [11]), as could be seen from Figs. 2a and 2b, correspond to the same half-life within the limits of probability errors. This makes possible the conclusion that in both reactions we observe the spontaneous fission of one and the same nuclide with T_{SF}~3min. In the ^{48}Ca + ^{238}U reaction it was produced directly as an EVR in the 3n-evaporation channel, while in the ^{48}Ca + ^{242}Pu reaction it is the daughter from the α-decay of the parent ^{287}114 nucleus. The half-life of the nucleus ^{283}112 determined on the basis of four events amounts to T_{SF}= 180^{+170}_{-60} sec.

Compared with the calculated values of T_{SF} for the neighbouring even-even isotopes the hindrance factor for the decay of ^{283}112 amounts to about 10^3. This value seems reasonable as far as spontaneous fission is concerned. In contrast, the absence of the α-decay of this nucleus leads to an anomalously large hindrance ($>2 \cdot 10^3$) for this kind of decay. According to the calculations by S.Cwiok, W.Nazarewicz and P.Heenen

[17] considerable hindrance on α-decay of even-odd nuclei may arise as a result of the properties of low-lying one-quasiparticle states in the parent and daughter nuclei. Note that the calculations carried out by the same authors for the [289]114 nucleus are in good agreement with the experimental data [13] (see Fig. 2c.).

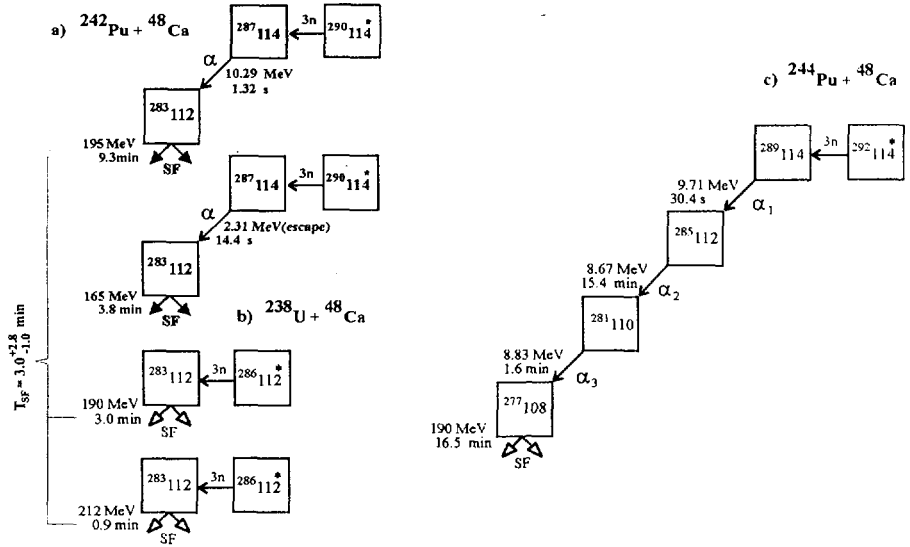Fig. 2. Position-correlated decay chains: **a)** of [287]114, produced in reaction [48]Ca+[242]Pu (present work); **b)** of [283]112, produced in reaction [48]Ca+[238]U [11] and **c)** of [289]114, produced in reaction [48]Ca+[244]Pu [13].

Conclusions

In the reaction [48]Ca + [242]Pu at the beam energy close to the Coulomb barrier, two events of radioactive decay were observed as decay sequences, consisting of α-decay and subsequent spontaneous fission. The decay properties of the parent nucleus amounts to $T_\alpha = 5.5^{+10}_{-2}$ s and $E_\alpha = 10.29 \pm 0.02$ MeV. The daughter nucleus undergoes spontaneous fission with $T_{SF} = 180^{+170}_{-60}$ sec. From the analysis of the data we draw the conclusion that the observed α-SF chains arise from the decay of superheavy nucleus, which is the isotope of element 114 with mass number 287 ($Q_\alpha = 10.44 \pm 0.02$ MeV). This nucleus is produced in the 3n-evaporation channel with a cross section of $\sigma = 2.5^{+15}_{-1}$ pb.

The half-life of the new isotope ^{287}114 is several times shorter than that of the previously observed heavier isotope ^{289}114, formed in the reaction ^{48}Ca+^{244}Pu (Fig. 2c). Such a trend is expected with a decrease of the neutron numbers of the superheavy nuclei.

The observed radioactive properties of the new nucleus ^{287}114, together with the data obtained earlier for the isotope ^{289}114 and the products of its α–decay, viz the isotopes ^{283}112 [12] and ^{289}114 [14], is considered as a direct proof of the existence of the "island of stability" of superheavy elements.

Acknowledgements

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