Synthesis and Properties of Even-even Isotopes with $Z = 110$–116 in ^{48}Ca Induced Reactions

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The paper presents results on the synthesis of superheavy nuclides with $Z = 114$ and 116 in the fusion reactions with ^{48}Ca ions. In the irradiation of targets made from enriched ^{244}Pu and ^{248}Cm isotopes with beam doses of 1.5×10^{19} and 2.3×10^{19}, respectively, the detector array situated in the focal plane of the gas-filled separator registered heavy atoms of new elements undergoing sequential α decays terminated by spontaneous fission. The time of the decay chains is approximately one minute. Decay properties of the synthesized nuclei are consistent the consecutive α decays originating from the parent nuclides ^{288}114 and ^{296}116 produced in the 4*n*-evaporation channels with the cross section of about a picobarn. Comparison of T_{SF} and T_α values for the nuclei with $Z = 110$ and 112 with those obtained earlier for lighter isotopes of these elements points to an enhanced stability of heavy nuclei with an increase in the neutron number. The α-decay energies Q_α measured experimentally in the chains 116-α_1–114-α_2–112-α_3–110 are compared with theoretical predictions of different nuclear models. This comparison shows that the difference between the experiment and theory is in the range of ±0.5 MeV. From this it follows that the theoretical models predicting the decisive influence of the nuclear structure on the stability of superheavy elements are well-founded not only qualitatively but in some sense also quantitatively. The prospects of further investigations in the field of superheavy nuclei are discussed briefly.

1. Introduction

It is known that one of the fundamental consequences of nuclear theory is the prediction of the "island of stability" of superheavy elements in the region of hypothetical superheavy elements. This intriguing hypothesis suggested 35 years ago has been developed lately and now seems to find its experimental confirmation in the currently conducted experiments.

A considerable increase in the nuclear stability at approaching closed spherical shells $Z = 114$ (and possibly 120–126) and $N = 184$ which follow the doubly magic nucleus ^{208}Pb ($Z = 82$, $N = 126$) is expected for the isotopes of superheavy elements with a high neutron excess (Figure 1). That is why for the synthesis of nuclei with $Z = 114$ and 116 we chose the fusion reactions ^{244}Pu, ^{248}Cm + ^{48}Ca in which the reaction products after the evaporation of neutrons have the maximal neutron excess.[1] In the fusion reactions with a doubly magic nucleus ^{48}Ca the compound nuclei ^{292}114 and ^{296}116 formed at the Coulomb barrier have the excitation energies of 33 and 31 MeV, respectively. It can be assumed that at these energies shell effects are still present in the excited nucleus which enhances the survival probability of evaporation residues as compared with typical "hot fusion" reactions ($E_x \geq 45$ MeV) used by us earlier for the synthesis of heavy isotopes with $Z = 106$, 108, and 110.[2]

Despite of these advantages, all previous attempts to synthesize new elements in reactions with ^{48}Ca ions and actinide targets only yielded the upper limit of their production cross section.[3] It was vital to increase the sensitivity of the experiment by three orders of magnitude to go down to the level of 0.5 pb where formation of superheavy nuclides was expected in the 3*n* and 4*n* evaporation channels. An increase in the sensitivity of the experiments could be achieved first of all due to increasing the intensity of the ^{48}Ca ion beam. For this purpose a new ion source ECR-4M operating on metallic Ca vapors was created. At a consumption of about 0.3 mg/h, a ^{48}Ca ion beam with the energy $E_L = 6$ MeV/*A* and intensity of about 0.5–0.8 pμA was produced.[4]

2. Experiment

Enriched ^{242,244}Pu and ^{248}Cm isotopes were used as a target matter. Rotating targets (about 0.3 mg/cm^2 in thickness) deposited on a Ti (1.5 μm) backing having a total area of about 30 cm^2 were used. Recoil nuclei knocked out of the target layer were separated in-flight from ^{48}Ca ions and other products of incomplete fusion reactions by kinematic separators. Two types of recoil separators were used in the experiments: VASSILISSA (energy selector) and a gas-filled separator DGFRS (Dubna Gas-Filled Recoil Separator).

Briefly about the conditions of the experiment.

Separated heavy atoms are implanted into a strip position-sensitive detector (∼50 cm^2 in area) situated in the focal plane. The front detector is surrounded with side detectors so that the whole assembling looks like a box with an open front wall. It increases the efficiency of detection of α particles from the decay of implanted nuclei up to 87% of 4π. For every implanted atom velocity is measured (by TOF detectors) as well as energy and location of implants on a sensitive surface of the front detector. In the case the nuclei of implanted atoms emit α particles or fission fragments, the last-mentioned will be registered in a strict correlation with the implant.

Experiments have been carried out in such setting since late 1998 and up to date. Without mentioning the results of the first

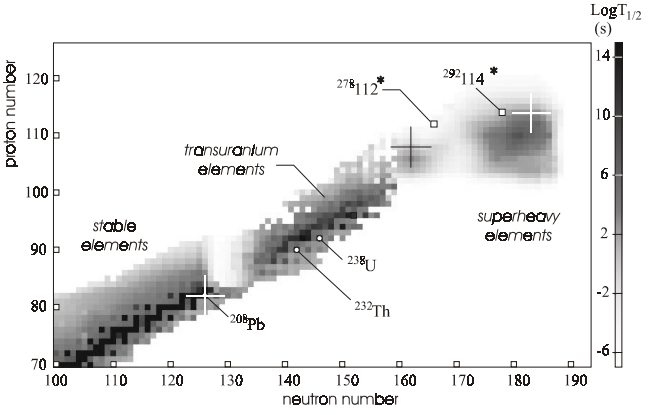

Figure 1. The map of nuclides in the region of heavy elements. The intensity of the color reflects the half-lives of the nuclei (the right-hand scale). The crosses indicate the location of the doubly magic nuclei with closed spherical shells $Z = 82$, $N = 126$ and $Z = 114$, $N = 184$ and with closed deformed shells $Z = 108$, $N = 162$. The white squares indicate the compound nuclei with $Z = 112$ and 114 formed in the reactions of cold fusion ^{70}Zn + ^{208}Pb and hot fusion ^{48}Ca + ^{244}Pu, respectively.

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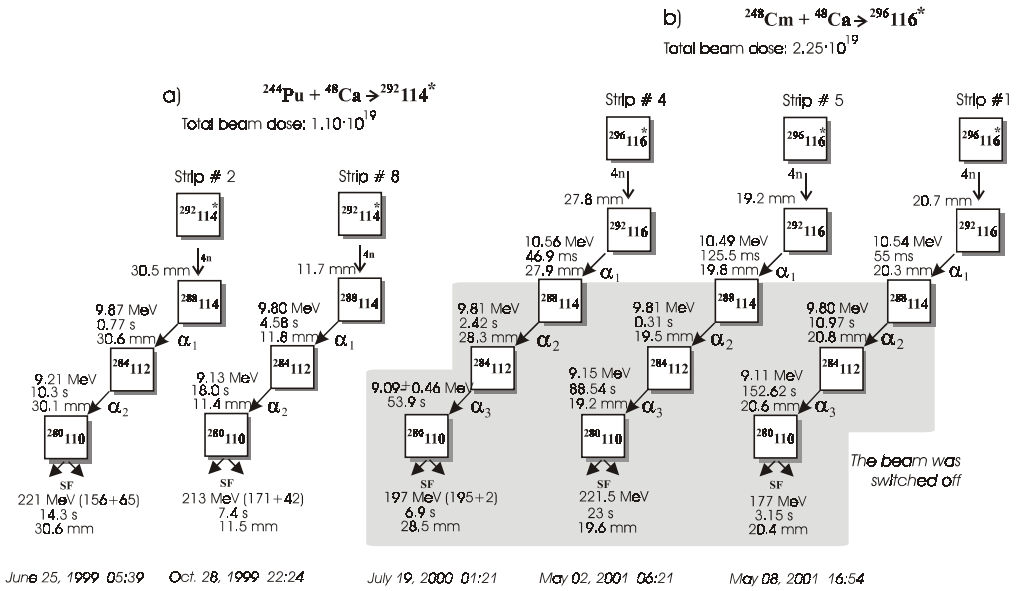

Figure 2. Chains of radioactive decay of nuclei synthesized in the reactions (a) $^{48}Ca + ^{244}Pu$ and (b) $^{48}Ca + ^{248}Cm$. The shaded area corresponds to the switched-off beam.

experiments with ^{238}U, ^{242}Pu, and ^{244}Pu targets which were published in Reference 5, we will only present the data obtained within the past year.

In May–November 2000, the second irradiation of the ^{244}Pu target was performed with ^{48}Ca ions, the beam dose was 1×10^{19} ions.[6]

The highly enriched (98.5%) target matter was provided by our colleagues and collaborators from the Livermore National Laboratory. In that experiment two more identical decay chains were observed. Each of them consisted of two sequential α decays and was terminated with spontaneous fission accompanied by a high energy release in the detector array (Figure 2a). Only the above mentioned events of spontaneous fission were registered in this experiment. The lifetime of the new nuclei was about 0.5 min. The probability that the observed decays were the result of incidental coincidences of the signals in the front detector was less than 5×10^{-13}. Note that both events were observed at an energy of the ^{48}Ca ion beam corresponding to the excitation energy of the compound nucleus $E_x = 36 \pm 2$ MeV. At this energy the most probable channel of the $^{292}114$ nucleus deexcitation corresponds to the emission of 4 neutrons and γ rays. Proceeding from this, new decay chains may be attributed to the decay of the even-even isotope of element 114 with mass 288.

The experiment that followed was devoted to the study of the reaction $^{48}Ca + ^{248}Cm$ and was aimed at testing this conclusion.

The target matter — enriched isotope of ^{248}Cm ($Z = 96$) — was produced at the high-flux reactor in Dimitrovgrad (Russia) in the quantity of 10 mg. The other target matter of enriched isotope ^{248}Cm was provided by the Livermore National Laboratory (USA).

Changing the ^{244}Pu target to the ^{248}Cm one, all other conditions of the experiment being kept, should lead to the formation of the new heavy nucleus with $Z = 116$ and mass 292 in the $4n$ evaporation channel of the fusion reaction $^{48}Ca + ^{248}Cm$. As a result of the α decay of this nucleus, expected with a high probability, we should obtain the daughter nucleus-isotope $^{288}114$ earlier produced in the reaction $^{48}Ca + ^{244}Pu$. That is why after the decay of the $^{292}116$ nucleus the whole chain of the daughter nucleus decay $^{288}114 \rightarrow ^{284}112 \rightarrow ^{280}110$ (SF) should also be observed in the experiment.

Usually the separator operates with a continuous ^{48}Ca beam. In the experiment on the synthesis of element 116 this regime was changed.

After the implantation into the focal plane detector of the heavy nucleus with the expected parameters (energy and velocity) and its decay with emission of an α particle with $E_\alpha \geq 10$ MeV (two signals are strictly position-correlated) the beam was switched off.

Measurements made straight after switching off the beam showed that the rate of the α particles ($E_\alpha \geq 9$ MeV) and spontaneous fission fragments in any strip within $\Delta x = 0.8$ mm, which is determined by the position resolution of the detector, is 0.45/year and 0.1/year, correspondingly. Incidental coincidences of the signals simulating the 3-step 1-min chain of the nucleus decay $^{288}114$ (α-α-SF) are practically excluded even for a single event.[7]

In these conditions, at a beam dose of 2.25×10^{19} ions three decay chains of element 116 were registered (Figure 2b). After the emission of the first α particle ($E_\alpha = 10.53 \pm 0.06$ MeV) the following decay occurred in the absence of the beam (see the gray area). As is seen from Figure 2, two decay chains of the $^{288}114$ nucleus produced in the reaction $^{48}Ca + ^{244}Pu$ and three new decay chains observed in the reaction $^{48}Ca + ^{248}Cm$

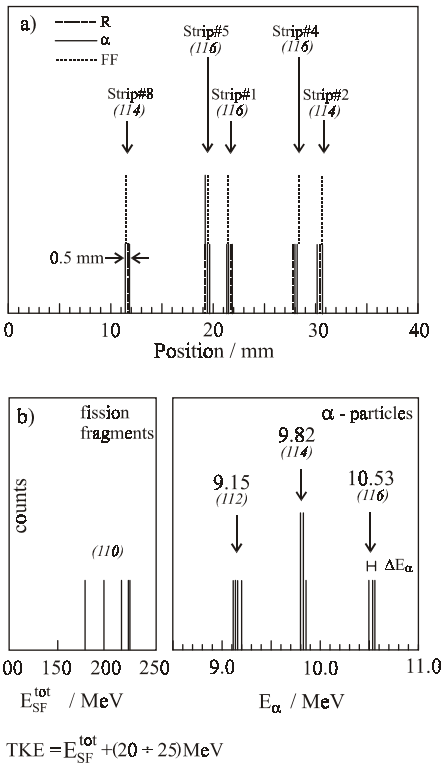

Figure 3. (a) Position and (b) energy correlations in the decay chains shown in Figure 2.

are strictly correlated: five signals arising in the front detector, i.e., the recoil nucleus, three α particles and fission fragments (in the case of the ^{288}114 nucleus decay: R-α-α-SF), differ in their position by no more than 0.6 mm (Figure 3a).

Energies of α particles and nuclear half-lives obtained in the reaction ^{48}Ca + ^{248}Cm coincide with each other within the limits of the detector energy resolution ($\Delta E_\alpha \sim 60$ keV) and statistical fluctuations determined by the nuclear decay in the chains. Figure 3b shows α-particle energy spectra for the three events corresponding to the ^{292}116 α-decay and the five events corresponding to the ^{288}114 and ^{284}112 decay, as well as the spectrum of combined energies of fission fragments from the five events of the ^{280}110 spontaneous fission obtained in the experiments with ^{244}Pu and ^{248}Cm targets.

Isotopes of elements 114 and 116 produced in the reactions ^{244}Pu, ^{248}Cm + ^{48}Ca are most likely formed in the 4*n* evaporation channels. This conclusion follows from the excitation energy values for compound nuclei which after the emission of neutrons and γ rays led to the observed evaporation products.

As it had been expected, in the case of even-even nuclei the experimentally observed α radiation is characterized by the strictly definite decay energy which corresponds to the difference between the masses of mother and daughter nuclei in the ground states (Figure 3b). For the allowed α transitions (even-even nuclei) the decay energy Q_α and the decay probability (or the half-life T_α) are connected by the well-known relation of Geiger-Nuttall. This relation is strictly fulfilled for all the known by now 60 even-even nuclei heavier than Pb, for which Q_α and T_α have already been measured. Figure 4 shows experimental and calculation data for the region of heavy nuclei with $Z \geq 100$. The results obtained for the new even-even superheavy nuclei with $Z = 112$, 114, and 116 are also presented there. On the other hand from the quantities Q_α and T_α it is possible to determine the atomic numbers of the α-emitters. With a probability of ≥96% the observed α-decays can be attributed to the chain with $Z = 116$–114–112–110.

Finally, in spontaneous fission of ^{280}110 nuclei the total energy release from the fission fragments in the detectors is about $E = 206$ MeV. This value with a correction to the energy losses in the dead layers of the focal plane pentane gas, and side detectors corresponds to the mean total kinetic energy of the fission fragments TKE ∼ 230 MeV. Such a high value of the fission fragment energy points to the spontaneous fission of a rather heavy nucleus. Note that in the fission of ^{235}U induced by thermal neutrons TKE = 168 MeV.

TABLE 1: Radioactive properties of even-even isotopes of the superheavy elements with *Z* = 110, 112, 114, and 116.

Z	Decay mode	Q_α / MeV	T_α
116	α	10.68 ± 0.06	53^{+62}_{-19} ms
114	α	9.96 ± 0.06	$2.6^{+2.0}_{-0.8}$ s
112	α	9.28 ± 0.06	$0.75^{+0.57}_{-0.23}$ min
110	SF	TKE ∼ 230	$7.6^{+5.8}_{-2.3}$ s

3. Properties of Superheavy Nuclides

Radioactive properties of the new nuclides are presented in Table 1. Comparison of the partial half lives T_{SF} and T_α of the synthesized nuclei with $Z = 110$ and 112 with the data obtained earlier for lighter isotopes of these elements[9] point to a considerable increase in the stability of heavy nuclei with increasing the neutron number. Addition of 10 neutrons to the ^{270}110 nucleus ($T_\alpha \sim 100$ μs) leads to an increase in the half-life almost by 10^5 times. For the nuclei of element 112 with $A = 277$ and the isotopes with $A = 284$ and 285 formed in the synthesis of element 114 nuclei in the reaction ^{244}Pu(^{48}Ca; 3*n*,4*n*) this difference is 2×10^5 and about 10^6, respectively.

Comparison of the experimental Q_α values for even-even nuclei with the calculation carried out using different theoretical models[8,10–13] shows that the difference between the experiment and theory is in the range of ±0.5 MeV (Figure 5).

Without making so far a more detailed analysis, one can conclude that theoretical predictions about the decisive influence of the nuclear structure on the stability of superheavy nuclides has been confirmed not only qualitatively but also to some extent quantitatively.

An increase in the half-lives of the most heavy nuclei up to tens of seconds and minutes substantially extends the area for the investigation of superheaviers, including the study of their chemical properties, measuring atomic masses, etc. On the other hand, development of acceleration and experimental technique will allow us to make an advent into the region if even heavier nuclei in the future.

4. Prospects

(1) Experimental and calculated cross sections of evaporation products of the fusion reactions with ^{48}Ca ions obtained in the synthesis of superheavy nuclides with $Z = 112$–116 show that these reactions can also be in principle used for the synthesis of nuclides with $Z > 116$. In particular, the reaction ^{249}Cf + ^{48}Ca leading to the formation of the $Z = 118$ element nuclei in the *xn*-evaporation channels can be regarded as a next step. In this

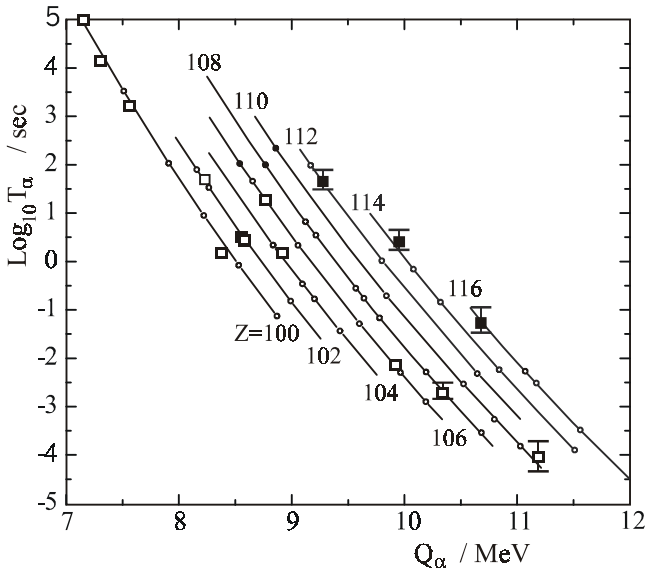

Figure 4. Calculated[8] and the experimental T_α vs. Q_α relationship for even-even isotopes of transfermium elements.

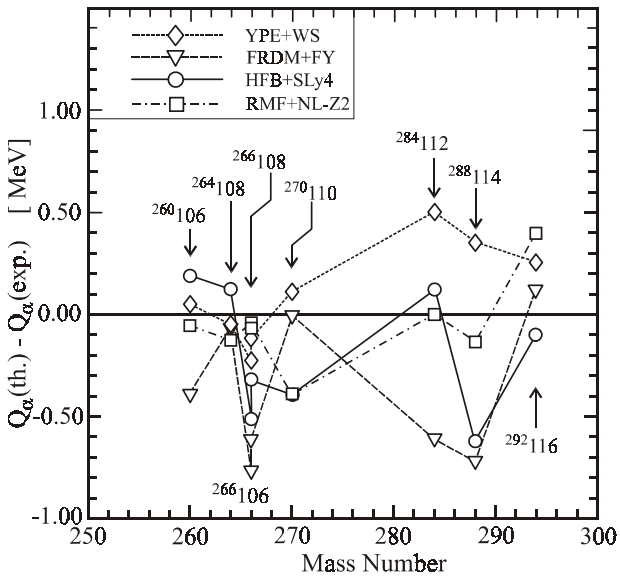

Figure 5. The difference between calculated and experimental values of Q_α for even-even isotopes with $Z = 106$–116 and $N = 154$–176.

reaction the excitation energy of the ^{297}118 compound nucleus at the Coulomb barrier is about 26 MeV. At this energy, the largest formation cross section of the evaporation products is expected for the 3*n* evaporation channel with the formation of the even-even isotope ^{294}118 ($N = 176$).

For the registration of sequential decays of this nuclide, the same setting of the experiment as earlier used in the synthesis of nuclei with $Z = 116$ can be employed. The energy of the allowed α transitions in the chains of the ^{294}118 nucleus decay together with earlier obtained data for $Z = 114$ and 116 can yield more information on the location and strength of the closed proton shell in the region of superheavy nuclei.

(2) Relatively long half-lives of the new nuclides amounting to several seconds in the case of $Z = 114$ and nearly one minute in the case of $Z = 112$ open up possibilities for the investigation of chemical properties of superheavy elements. In our opinion, the most suitable candidates are the isotopes of Eka Hg due to their relatively long half-lives and expected unique chemical properties of element 112. One of possible approaches to this task is to determine the probability of formation of intermetallic bonds between Eka Hg and Pd, Au, or Pt which are well-known for Hg. On-line experiments of this type will soon be launched using ^{48}Ca ion beams.

(3) An enhanced stability of superheavy nuclei changes the entire approach to their synthesis in the fusion reactions with heavy ions. The necessity of separating in-flight the reaction products for a speedy delivery of superheavy atoms to detector array is not a decisive factor in the synthesis of superheavy nuclei due to their long half-lives. The use of classical on-line separators is more preferable since the latter alongside their high efficiency, connected with a possibility of using thick targets and a higher selectivity, allow us to measure the superheavy nucleus mass.

This idea should be implemented in the design of the setup MASHA (Mass Analyzer of Superheavy Atoms), which is now being developed at the Flerov Laboratory.

(4) The use of fusion reactions with ^{48}Ca ions presently employed for the synthesis of superheavy elements is restricted by the availability of the target material. The isotope ^{249}Cf which is planned to be used as a target in the experiments on the synthesis of nuclei with $Z = 118$ is apparently the last one in the row of actinides. At the same time, according to predictions of microscopic models of the type of Hartree-Fock-Bogoliubov or Relativistic Mean Field, theory a change in the nuclear density (the so call "semi-bubble nuclei") also leading to the stability of hyperheavy nuclei is expected in the case of nuclei with $Z \geq 120$.[13]

For the synthesis of these nuclei it will be necessary to use fusion reactions with ions heavier than ^{48}Ca, i.e. of the ^{58}Fe, ^{64}Ni, etc., type. Two fusion reactions, ^{248}Cm + ^{54}Cr and ^{238}U + ^{64}Ni, lead to the formation of the weakly excited compound nucleus ^{302}120 with $N = 182$. It is planned to study the fusion and fission processes involving such massive nuclei.

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(1) As is seen from Figure 1, in the region of deformed shells $Z = 108$ and $N = 162$ an increase in the stability of heavy nuclei is also expected.

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