FLNR JINR EXPERIMENTS ON SYNTHESIS OF SUPERHEAVY NUCLEI WITH ⁴⁸Ca BEAM*

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The investigations of the decay properties and formation cross sections of the heaviest isotopes of the 110 and 112 elements with atomic numbers 276, 277 and 282, 283, respectively, were performed at the FLNR JINR with the use of the intense internal and extracted ⁴⁸Ca beams. ²³²Th and ²³⁸U targets were used for the experiments. With the use of ²³⁸U target at the beam energy $E_{\text{targ.}} = 231 \pm 3 \text{ MeV}$ (E^{*} = 31 MeV) two spontaneous fission events were detected, that corresponds to the cross section — 5.0 ± 2 pb. The result could be explained as spontaneous fission of the even-odd isotope ²⁸³112 with half life $T_{1/2} = 117^{+283}_{-48}$ seconds. The probability of α decay of ²⁸³112 is not excluded and the possible branching ratio could be $b_{\alpha} \approx b_{\text{sf}} \approx 50 \%$.

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1. Introduction

For several reasons the projectile ⁴⁸Ca is of special interest for production of heaviest elements. Due to its neutron excess, it gives access to compound nuclei which are more close to the predicted magic neutron numbers 178– 184. Its neutron richness and doubly magic structure allows one to synthesize relatively cold compound nuclei at energies close to the fusion barrier. Also the fusion probability of ⁴⁸Ca with heavy target nuclei of the ²³⁸U, ²⁴⁴Pu or ²⁴⁸Cm type may be enhanced compared with more symmetric systems presently used for the synthesis of heaviest elements [1].

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2. Experiment

An extracted 48 Ca beam was obtained with the use of new ion source ECR–4M, installed on U400 cyclotron. Enriched (70%) 48 Ca material was used in the experiments. The material for ion source was metallic Ca prepared from the Ca oxide. The source material was situated into the Al₂O₃ oven heated to ≈ 1100 K. The consumption of material in the ECR source was less than 0.4 mg/h. The continuous time structure of the beam was used.

We performed two kinds of the experiments. First one was with the use of 232 Th target and with internal beam of Ca ions, because the intensity of the internal beam is factor of 3 higher than the external one. Thickness of 232 Th target installed on the internal beam probe was 8 mg/cm², more than the range of evaporation residues of 110 element but less than ranges of transfer reaction products. The original internal beam probe was constructed to allow the use of the satellite beams after the extraction foil and to make the possibility to irradiate Th target on internal beam probe in parallel with the separator experiment, which is using the main extracted ion charge state.

The separator targets were produced by electrodeposition of U_2O_3 or Th material onto a 1.6 mg/cm² Al backing disks 125 mm in diameter. They were used as a self supporting rotating targets. The thickness of ²³⁸U (99.999%) and Th layers were $\approx 300 \ \mu g/cm^2$.

The beam intensity on the separator target was typically $(1.7-2.4) \times 10^{12} \text{s}^{-1}$. The beam energy was controlled by measuring the energy of the ions scattered at 30° in a thin (200 $\mu g/\text{cm}^2$) gold foil. The energies of ⁴⁸Ca beams extracted from the U400 cyclotron were 255 and 264 MeV and at the middle of the target 231±3 and 238±3 MeV respectively.

In the case of ²³⁸U target the evaporation residues were separated inflight by the electrostatic separator VASSILISSA [2]. The detailed description of the determination of the transportation efficiency of recoil nuclei, background suppression and test experiments with ⁴⁸Ca beam is presented in [3].

The detector system consisting of two (start and stop) time-of-flight detectors and an array of silicon detectors has been developed and installed in the separator focal plane. After passing the time-of-flight detectors, recoil nuclei are implanted in an array of detectors. The detector array consists of five identical 16-strip silicon wafers. The active area of each of the silicon strip detectors is $60 \times 60 \text{ mm}^2$. In the case of the focal detector each strip is position sensitive in the vertical direction with a resolution of 0.6 mm between α decays of the correlated α decay chains, 1.0 mm in the case of recoil — α correlations and 1.5 mm in the case of recoil — fission fragment correlations. The typical energy resolution of 20 keV was obtained for α -decays

in the energy range from 6 to 9 MeV. The absolute error of the α energy is determined also by systematic uncertainties and amounts to ± 25 keV. The time resolution for the recorded events was 1 μ sec.

Four wafers are mounted in the backward hemisphere facing the stop detector. They measure escaping α 's or fission fragments with a total geometry efficiency of 85% of 4 π . Backward detectors have not the position resolution and each four neighbouring strips are connected galvanically so that 16 energy sensitive segments with resolution 100-120 keV are formed. 3 μ m degrader foil (Mylar) in front of the silicon detectors was placed.

3. Results

3.1. $^{48}Ca + ^{232}Th \ reaction$

Theory predicts [5,6] for the ²⁶⁸106, the granddaughter of ²⁷⁶110 — result of the 4n evaporation channel after fusion of 48 Ca and 232 Th, the half-life against α and SF decay of more than 2 hours. In this case it is possible to use fast chemistry and to perform after that off line measurements at the geometry close to 4π . The experiments with internal beam probe include the use of fast chemistry for extraction of the fraction containing 106 element nuclei from ²³²Th target material and the off line measurements of the correlation chains from α decay of ²⁶⁸106 and SF of ²⁶⁴104. The fraction extracted after chemistry was deposited onto 30 $\mu g/cm^2$ thick carbon foil and was 10 $\mu g/cm^2$ thick and 20 mm in diameter source. Selectivity of the chemical methods allowed to suppress the fraction of U isotopes, probable candidates for long lived background products, by factor of more than 10^5 . Carbon foils with these sources were placed between two silicon detectors (30 mm in diameter each) at special vacuum chambers. Total registration efficiency for α particles and SF fragments was close to 90 %. Altogether 8 vacuum chambers with pairs of silicon detectors were used in these experiments. In each chamber the measuring time was 3-6 days. The suppression factors for long lived transfer reactions products was more than 10^6 . Altogether 10 sources were prepared during these experiments, each after collecting of a number of projectiles $\approx 6 \times 10^{16}$. Total number of collected projectiles was $\approx 6 \times 10^{17}$. Two spontaneous fission events were detected during off line measurements, first after 40 hours from the end of irradiation and second after 92 hours from the end of irradiation. The preceding α particles detected at the energy range 7.5–9 MeV had a time distance of 1 hour in the case of first event and three hours for second events. It is at the level of the random counting rate of α particles at this energy region. One of the possible explanation of the obtained result could be that we detected the fission decay of even-odd isotopes ²⁶⁵104 or ²⁶⁹106, the descendants of ²⁷⁷110 formed after the evaporation of 3 neutrons from compound nucleus ²⁸⁰110. The estimated cross section value for two events is 6 ± 3 pb.

3.2. $^{48}Ca + ^{238}U$ reaction

The beam energies for the production of $^{282,283}112$ were calculated using the evaporation code HIVAP [4]. The set of parameters used was taken from [7]. It was tested for the 48 Ca + 206,208 Pb reactions and calculation results were in agreement with experimental data within errors.

At the beam energy $E_{\text{targ.}} = 231 \text{ MeV} (3n \text{ evaporation channel})$ calculated for reaction at half of the target thickness the excitation energy of compound nucleus was 31 MeV. The total number of collected projectiles was 3.2×10^{18} . The detailed data analysis of recoil- α -...- α -fission genetically correlated chains, using the obtained position and energy resolution, was performed. Two spontaneous fission events were detected, for both events the coincident high energy pulses were detected in focal plane and backward detectors. From these data, together with the target thickness and a calculated total efficiency of 25%, a cross section value of 5.0 ± 2 pb can be deduced. The given error represents statistical uncertainties only. The values are correct relative to our previously measured cross sections of the reactions ${}^{48}Ca + {}^{206,208}Pb$, but are uncertain within a factor of two on absolute scale due to estimated systematic deviations. The theoretical predictions [5] for T_{α} , Q_{α} for even-odd isotope ²⁸³112 and it's descendants and for neighbouring even-even isotopes ^{282,284}112, taking into account possible suppression factors for even-odd ²⁸³112, from [6] were used at the analysis (see Fig. 1). At the α -energy range from 8 to 13 MeV and time interval until 10000 seconds no correlations were found. In the position window ± 0.8 mm only recoil-fission correlations were found, for the first event (strip number 12) in time interval 182 seconds and for the second event (strip number 15) - 52 seconds. The following data analysis showed that for the position window ± 0.8 mm the time of the random recoil-fission correlations at the strip number 12 was approximately 1000 seconds and for the strip number 15 it was approximately 500 seconds.

In order to estimate the possible background sources of spontaneous fission we have three possibilities. The first is the spontaneous fissioning isomers at Pu–Am region. But the most long lived one has a half life of 14 ms, that is in contradiction with obtained recoil–fission correlation times. And we do not detect any alpha peaks in the region of the energies, corresponding to these Pu–Am isotopes, which should be present, taking into account isomeric ratio about $10^{-3}-10^{-4}$. The second possibility is the spontaneous fission of 252 No, the reaction product of 48 Ca with Pb admixture in the 238 U target. However, the half life of the 252 No (2.3 s) is in contradiction with obtained results. In addition, in order to estimate the amount of lead in 238 U we studied the 20,22 Ne + 238 U reaction and on the basis of well known cross section data [8] for the 20,22 Ne + Pb reactions, we estimated the admixture



Fig. 1. The theoretical predictions for half lives of the isotopes of 112 and 110 elements. $\Box - T_{\alpha}$ from [6]. • $- T_{\alpha}$ from [7]. • $- T_{\text{sf}}$ from [7]. The investigated isotopes of element 112 and it's descendants are indicated by arrows.

of lead in ²³⁸U target is less than 10^{-5} . For the beam energy 231 MeV at the middle of the target and known cross section values for the reactions ⁴⁸Ca + Pb we could estimate the apparent cross section for the reaction of Ca with lead admixture as 0.2–0.4 pb. In addition at the end of our experiment with ²³⁸U target we irradiated the natural lead target at the same conditions (beam energy and separator's settings for 112 element). Collected number of fission events confirms our estimation of 0.3 pb for the reaction with lead admixture. The third possibility is presently unknown exotic isomers with long half life. It is an open question, but seems to be not very probable. During the following experiments with ⁴⁸Ca and actinide targets it will be possible to collect more information.



Fig. 2. The spontaneous fission spectra (TKE), collected during test experiments using ${}^{48}\text{Ca} + {}^{206}\text{Pb}$ reaction. Two events from ${}^{48}\text{Ca} + {}^{238}\text{U}$ reaction are indicated by arrows.

On the basis of above mentioned estimations the more probable explanation of the obtained results is, that we detected the spontaneous fission of the even-odd isotope ²⁸³112 with half life $T_{1/2} = 117^{+283}_{-48}$ seconds. We do not exclude the probability of α decay of ²⁸³112 and estimate the possible branching ratio as $b_{\alpha} \approx b_{sf} \approx 50$ %. In Fig. 2 the spontaneous fission spectrum, collected during test experiments using ⁴⁸Ca + ²⁰⁶Pb reaction, is presented. The energy calibration of spectrum was performed using data from Ref. [9]. In this spectra two events from ⁴⁸Ca + ²³⁸U reaction are indicated by arrows.

At the beam energy $E_{\text{targ.}} = 238 \text{ MeV} (4n \text{ evaporation channel})$ calculated for reactions at half of the target thickness the excitation energy of compound nucleus was 39 MeV. The total number of collected projectiles was 2.2×10^{18} . No events were detected, including spontaneous fission or $\alpha - \alpha$ correlations in a range of α energies from 8 to 12 MeV and time interval up to 1000 seconds. From these data a cross section limit for non observation of any events 3.0 ± 1.5 pb can be deduced. This fact is not surprising, because the calculations predict the maximum cross section value for the 4nevaporation channel less than for 3n evaporation channel by a factor of 3-5.

4. Conclusion

Among all the target-projectile combinations available at present, complete fusion reactions induced by ⁴⁸Ca ions on heavy actinide targets, such as ²³²Th, ²³⁸U and ^{242,244}Pu, provide the closest approach to the neutron number expected for the spherical superheavy elements. These reactions result in about 30 MeV excitation energy of the compound nuclei. This is 20 MeV lower than the corresponding values for compound nuclei produced in the ³⁴S + ²³⁸U, ²⁴⁴Pu reactions, in which the isotopes ²⁶⁷108 and ²⁷³110 have been synthesized after evaporation of 5 neutrons [10]. In the ⁴⁸Ca case, this can mean a higher probability of 3n and 4n evaporation channels, which can increase production cross sections of corresponding nuclei compared to 5n channel, due to lower fission losses in deexcitation process.

We plan to continue the experiments aimed to the synthesis of even–even isotope $^{282}112$, to start the experiments leading to the synthesis of $^{276}110$ with 232 Th target and $^{286}114$ with 242 Pu target, which is very important for the comparison with modern theory predictions.

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