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SEARCH OF THE DECAY OF THE HEAVIEST ISOTOPES OF ELEMENT 112

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INTRODUCTION

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According to the macro – microscopic theory predictions, the stability of the super heavy nuclei must increase sharply, while approaching the spherical neutron shell N = 184. The synthesis of the super heavy spherical nuclides, even if they are distant from the shell N = 184, takes place only at a significant neutron excess in the colliding nuclei. Thus, the isotopes of element 112, which have a spherical shape and are therefore, relatively stable, can be produced in the reaction ${}^{48}Ca + {}^{238}U$ [1].

The compound nucleus ²⁸⁶112 formed in this reaction appears to be weakly excited due to the significant mass excess of the double magic ⁴⁸Ca, the excitation energy at the Coulomb barrier being only $E_x \approx 35$ MeV. At such an excitation energy, the shell effects are still present in the heated nucleus. This may increase the survival probability of the evaporation residues (ER). The high mass asymmetry in the entrance channel (A₁/A₂ = 0.2 and Z₁Z₂ = 1840) seems to decrease the dynamic limitations for the fusion of the interacting nuclei, earlier observed for the more symmetric cold fusion reactions [2].

The expected decay properties of the isotopes with Z = 112 are quite specific. According to the calculations provided by Smolańczuk [3], the even – even isotopes $^{282}112$ and $^{284}112$ have partial half lives for α – decay respectively: $T_{\alpha} = 0.005$ s. and 1.0 s. (Fig. 1). Their spontaneous fission half life $(T_{s.f.})$ is slightly higher than T_{α} . At the same time, the isotope $^{280}110$, which is the α -decay daughter of the $^{284}112$ nucleus, will mainly decay by spontaneous fission. As for the chain of sequential decays of the other isotope $^{282}112$, all products with Z < 106 will have $T_{s.f.} \ll T_{\alpha}$ too. For the isotope $^{283}112$ the predictions are less clear, because the odd number of neutrons may lead to significant limitations for both α – decay and spontaneous fission. Here, a strong competition between the two modes of decay is also highly expected.

Note, that calculations performed by Möller et. al. [4], predict for the same nuclei the half life versus α - decay to be hundreds and thousands times longer than T_{α} from [3]. However, it seems less significant, since it does not change the main decay properties of the isotopes with Z = 112, produced in the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$. Either they will decay by spontaneous fission, or by the chains of their α - decay (one or several) will end by the spontaneous fission anyway. On the other hand, modern separation techniques allow synthesizing and identifying the ER, if their half lives are more than one microsecond. It covers the range of all the predictions and thus the experimental results obtained using these techniques can be used as a direct test of the predictive capabilities of the existing theoretical models.

EXPERIMENT

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Production of intensive ion beam from the rare and extremely expensive ⁴⁸Ca isotope presents itself as a key problem in our issue on the synthesis of the isotopes of element 112.

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FIGURE 1. The theoretical predictions for partial half lives of the isotopes with Z = 112. Open circles and open squares connected by solid lines are T_{α} and T_{sf} taken from [3] respectively. Open circles connected by dashed line are T_{α} taken from [4]. Black circle – T_{α} for the isotope ²⁷⁷112 obtained in the reaction ²⁰⁸Pb(⁷⁰Zn,1n); black square – T_{sf} for the isotope ²⁸³112 produced in the reaction ²³⁸U(⁴⁸Ca,3n) – present work

The neutral atoms of calcium were injected into the plasma of the ECR-4M ion source [5] by the process of controlled heating of the metallic sample of ⁴⁸Ca (50 mg) enriched to 70 %. The sample was made of calcium oxide right before being placed in the oven of the ECR source. The whole preparation procedure of metallic Ca as well as recuperation of the material from the ion source chamber was controlled by measuring the yield of γ - rays from the isotope ⁴⁷Ca (4.5 d), produced in the ⁴⁸Ca(γ ,n)⁴⁷Ca reaction.

The new injection system and the modified beam optics of the cyclotron allowed obtaining the internal beam of ${}^{48}\text{Ca}{}^{5+}$ ions with an intensity of up to 1 pµA at the material consumption rate of about 0.3 mg/h. The beam extraction from the U-400 cyclotron was provided by the stripping method. The variation of the energy of the extracted beam in the range from 200 MeV to 280 MeV was performed by a smooth variation of the magnetic field and by precise positioning of the stripping foil. The mean beam intensity of ${}^{48}\text{Ca}$ on the target was 2.2×10^{12} pps. The time structure of the beam was persistent.

The evaporation residues were separated in-flight from the beam particles and other reaction products by the electrostatic recoil separator VASSILISSA [6]. The enriched isotope of ²³⁸U (99.999 %) with the thickness of 0.3 mg/cm² was used as a target material. It was deposited uniformly on a 1.6 mg/cm² thick Al backing disk 125 mm in diameter. The disk was rotating with a frequency $\omega = 2000$ rpm. The same target design using Tb and enriched isotopes of Yb and Pb was used in several test experiments.

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For registration and identification of the ER a system of time-of-flight detectors and a silicon position sensitive strip detector array was installed in the focal plane of the separator. After registration in the TOF detectors, the recoils were implanted into a 16 strip silicon detector with an active area of $60 \times 60 \text{ mm}^2$. Each strip had a longitudial position sensitivity. Special measurements of the position resolution along each strip were performed. For the sequential α - α decays it was 0.6 mm, for the correlated recoil and α - particle – 1.0 mm, and for the correlated recoil and spontaneous fission events it was 1.5 mm. The energy resolution for α - particles within the energy range from 6 to 9 MeV was 20 keV. The time accuracy for the recorded events was about 1 μ sec. The strip detector was surrounded with four other identical silicon detectors and the entire array had a shape of a cube with the dimensions $60 \times 60 \times 60 \text{ mm}^3$. The geometrical efficiency of the silicon array was 85 % of 4 π . In the present series of experiments each of the four neighboring strips of backward detectors were connected galvanicly and formed 16 energy sensitive segments with the resolution of 120 – 150 keV.

Scattered low energy projectiles were the main contribution to the background. An additional bending dipole magnet allowed placing the detectors at an angle of 8° with respect to the primary beam direction. After all, the counting rate of the frontal detector was only 25–30 Hz. In front of the silicon detectors the 3 μ m degrader foil (mylar) was placed to reduce the number of very low energy projectiles reaching the focal plane of the separator.

The separation efficiency of ER from the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$ was obtained in test experiments. The cross sections of the xn– evaporation channels were measured in the bombardment of targets of ${}^{159}\text{Tb}$, ${}^{174}\text{Yb}$ and ${}^{206,208}\text{Pb}$ by the ${}^{48}\text{Ca}$ projectiles in a wide energy range. The measurements showed that about 25 % of the ${}^{286-x}112$ ER, produced on the U target, would be implanted in the frontal detector. The signals from the time-of-flight detectors were used both for measurements of the velocity of the recoils and for distinguishing the decays of the previously implanted nuclei. The high efficiency of the TOF detectors allowed obtaining very clean decay spectra and significantly widening (up to several hours) the time window for measuring decay chains. The latter was particularly important for identification of the ER with a long half life and the continuous structure of the beam.

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RESULTS

To estimate the yield of evaporation residues in the reaction 238 U(⁴⁸Ca,xn) $^{286-x}112$ and to chose the optimal beam energy for 48 Ca, a set of additional experiments was performed. Using the recoil separator VASSILISSA, the excitation functions for the reactions 206,208 Pb(48 Ca,xn) $^{254-x}$ No, $^{256-x}$ No were measured. At the same time, on the time-of-flight fission fragment spectrometer CORSET [7] data were obtained on the cross sections and mass distribution of fission fragments in the reactions 48 Ca + 206,208 Pb and 48 Ca + 238 U. For all the reactions these compound systems were formed at low excitation energies, close to the Coulomb barrier.

These experiments will be described in full in separate articles (for example [8]), here we shall restrict our consideration to the main conclusions drawn from the data. As it follows from the analysis of the data observed above, the highest cross section for the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$ is expected for the 3n and 4n evaporation channels. The excitation functions reached their maxima at $E_x = 33$ and 39 MeV, which corresponds to the beam energy at the middle of the target $E_L = 231$ MeV and 238 MeV, respectively. The absolute value of the cross section for the xn evaporation channels can be estimated much less definitely. According to different calculations, the cross section at the maximum of the 3n evaporation channel varies from one pb to a few tens pb; although, it still remains by a factor of 3 - 5 higher than that for the 4n evaporation channel.

On the basis of above data, two long term irradiations of a 238 U target with 48 Ca at beam energies of 231 ± 3 MeV and 238 ± 3 MeV were performed. The dose of 3.5×10^{18} projectiles was collected in the first experiment in March 1998. Two spontaneous fission events were detected at that time. For the both events the fission fragments were registered as two coinciding signals with a high energy deposition both in the front and backward detectors.

The total kinetic energy (TKE) for the events has also been measured (Fig. 2). The detectors were calibrated by fragments from spontaneous fission of the implanted ER of ²⁵²No, produced in the reaction ²⁰⁶Pb(⁴⁸Ca,2n) using data from [9]. The determined values of the TKE for the two registered events are 190 and 212 MeV respectively. Their energies were calculated using the same procedure as for No and were not corrected taking into account higher masses of fission fragments.

The analysis of all the events collected in the experiment has been provided in order to find genetic decay links of the implants. For that purpose, the data of the position and energy resolutions of the correlated events, measured by the frontal detector for the various modes of decay were used. Thus, for the recoil $-\alpha - \alpha$ as well as recoil $-\alpha - \ldots$ -SF for an α - particle in the energy range from 8 to 13 MeV and in the time interval of up to 10000 s. no correlation was found. But at the same time, there were two correlated signals from the recoil and spontaneous fission found in strips 12 and 15 within the position window of \pm 0.8 mm. In the first case (strip 12) the position difference between the signals, "recoil" and "spontaneous



FIGURE 2. The spectra of TKE for spontaneous fission of 252 No. Open circles – data, dashed line – Gaussian fit to the data obtained with VASSILISSA in the reaction 206 Pb(48 Ca,2n) 252 No and normalized on the data taken from [9]. Black triangles correspond to the events from reaction 48 Ca + 238 U

fission", was 0.36 mm and the time interval was $\Delta t_1 = 182.39$ s., while in the second case (strip 15) the position difference was 0.75 mm and the time interval was $\Delta t_2 = 52.04$ s. As it became evident from the data of the long term detection, at the position window of \pm 0.8 mm the signals, similar to ER, were detected in strip 12 with a mean frequency of 0.001 Hz, while in strip 15 the frequency was 0.002 Hz. The detected time intervals were much less than that between recoil like events distributed randomly, thus we used Δt_1 and Δt_2 intervals for the half life estimation.

In the second experiment, the doze of 2.2×10^{18} projectiles was collected at the beam energy $E_L = 238$ MeV ($E_x=39$ MeV). No spontaneous fission events were detected and no $\alpha - \alpha$ correlations were observed in the whole α - particle energy range from 8 to 12 MeV in the time interval of up to 1000 sec. The upper limit for ER formation cross section at this excitation energy was 3.0 ± 1.5 pb.

It is very important to note that the two events observed in these experiments were collected practically under no background conditions. The fact that no SF – isomers as well as no other known SF – nuclei were observed in the two experiments

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with the total beam doze of 5.7×10^{18} makes evident the high selectivity of the device for the evaporation residues detection. That is why we also have to avoid explaining the effect in terms of a rare and unknown decay mode of known nuclei formed in the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$.

As mentioned above, no α - particles, preceding the spontaneous fission, were observed for either event despite the high efficiency of the detector array. The most reasonable explanation is that we observed the spontaneous fission of the heaviest new even – odd isotope ²⁸³112 with the half life $T_{1/2} = 81.25^{+200}_{-35}$ seconds. Despite the fact that only spontaneous fission was observed in the experiment, we cannot exclude also the α - decay with a branching ratio $b_{\alpha} \leq 50$ %. The cross section for the new isotope is (5.0 ± 2) pb (the statistical error is presented here, the absolute cross section accuracy value is within the factor of 2). The absence of any effect at the excitation energy $E_x = 39$ MeV is not surprising. The cross section of the 3n evaporation channel decreases with an increase in the excitation energy, while the cross section of the 4n evaporation channel is several times lower than that of the 3n channel, as it was already mentioned above.

CONCLUSION

The present article describes a new attempt to synthesize super heavy spherical nuclei in the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$. As compared with all previous experiments with ${}^{48}\text{Ca}$ ions, the sensitivity of the present experiment is more than 100 times higher. Two spontaneous fission events were detected at the beam energy below the Coulomb barrier. As it follows from the data analysis, the events were most probably triggered by the decay of the even – odd isotope ${}^{283}112$ formed in the 3n – evaporation channel. The half life of the new isotope is about 100 sec. It could also be that it undergoes α – decay with a branching ratio b_{α} \leq 50 %.

The half life of the new isotope is more than 5 orders of magnitude longer (Fig. 1) than that of the already known isotope $^{277}112$ (T_{1/2} ≈ 0.24 ms), obtained in the cold fusion reaction 208 Pb(70 Zn,n) $^{277}112$ [10]. The difference could indicate that the stability increases sharply at approaching the domain of the spherical shell corrections.

The present experiment is the first one in our long term research program with the 48 Ca beam, dedicated to the synthesis and study of the properties of super heavy elements. We strongly intend to increase the intensity of the beam, which will allow us to try to synthesize other isotopes with Z = 110 - 114.

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Поиск распада наиболее тяжелых изотопов элемента 112

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Для синтеза тяжелых изотопов элемента 112 в реакции ⁴⁸Ca + ²³⁸U были выполнены два эксперимента при разных энергиях бомбардирующих ионов. В первом эксперименте при энергии пучка в середине мишени 231 МэВ ожидалось образование составного ядра ²⁸⁶112 с энергией возбуждения $E_r = 33$ МэВ. В 20-дневном сеансе облучения мишени ²³⁸U при интегральной дозе ионов 3.5 × 10¹⁸ в фокальном детекторе сепаратора ВАСИЛИСА наблюдались два события спонтанного деления. Не были наблюдены ни α-частицы, предшествующие делению, ни коррелированные события испускания двух α-частиц в диапазоне энергий от 8 до 13 МэВ и временном интервале до 10000 с. Наиболее вероятным объяснением данных, полученных в этом эксперименте, является происхождение наблюдавшихся событий спонтанного деления в результате распада четно-нечетного изотопа (N = 171) элемента 112, образующегося в реакции ²³⁸U (⁴⁸Ca, 3n) ²⁸³112 с поперечным сечением ~5 пб. Период полураспада нового спонтанно делящегося изотопа составляет около 100 с. Во втором эксперименте энергия ионов пучка составляла 238 МэВ, что приводило к увеличению энергии возбуждения составного ядра до E_r = 39 МэВ. Интегральная доза ионов во втором случае составляла 2.2 × 10¹⁸. Не наблюдалось ни событий спонтаиного деления, ни событий последовательного испускания α-частиц в интервале энергий от 8 до 12 МэВ и временном интервале до 1000 с. Эти данные позволяют оценить верхнюю границу - 3 пб для сечения образования четно-четного изотопа ²⁸²112 в реакции ²³⁸U (⁴⁸Ca, 4n) ²⁸²112.

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Oganessian Yu.Ts. et al. Search of the Decay of the Heaviest Isotopes of Element 112

To produce heavy isotopes of element 112 in the reaction ${}^{48}\text{Ca} + {}^{238}\text{U}$ two experiments at different beam energies were performed. In the first experiment the beam energy at the middle of the target was 231 MeV which lead to the production of the compound nucleus ${}^{286}112$ with an excitation energy of $E_x = 33$ MeV. In a 20 — day irradiation of the ${}^{238}\text{U}$ target with a beam dose of 3.5×10^{18} on the focal plane of the recoil separator VASSILISSA two spontaneous fission events were detected. No α -particle emission preceding spontaneous fission nor $\alpha - \alpha$ correlation in the energy range from 8 to 13 MeV in the time interval of up to 10000 s. have been observed. The most probable explanation of the data obtained in this experiment is that the observed spontaneous fission corresponds to the decay of the even — odd isotope (N = 171) of element 112 produced in the reaction ${}^{238}\text{U}$ (${}^{48}\text{Ca}$, 3n) ${}^{283}112$ with a cross section of ~ 5 pb. The half life of the new spontaneous fission nuclide is about 100 s. In the second experiment the beam energy was 238 MeV which increases the excitation energy of compound nuclei up to $E_x = 39$ MeV. The total beam dose in that case was 2.2×10^{18} . No events due either to spontaneous fission or sequential $\alpha - \alpha$ decay in an energy range from 8 to 12 MeV and the time interval of 1000 s. were detected. These data give the upper limit of 3 pb for the production cross section of the even — even isotope ${}^{282}112$ in the reaction ${}^{238}\text{U}$ (${}^{48}\text{Ca}$, 4n) ${}^{282}112$.

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