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EXPERIMENTS ON THE SYNTHESIS OF SUPERHEAVY ELEMENTS WITH Z=110,112 VIA ^{232}Th,^{238}U+^{48}Ca REACTIONS.*

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The investigations of the decay properties and formation cross sections of the heaviest isotopes of the elements 110 ($^{276-277}$110) and 112 ($^{282-283}$112) were performed at the Flerov Laboratory of Nuclear Reactions at Dubna with the use of the internal and extracted high intensity ^{48}Ca beams. ^{232}Th and ^{238}U targets were used in the experiments. The experiments with internal beam probe include the use of fast chemistry for the extraction of the fraction containing 106 element nuclei from the ^{232}Th target material and the off line measurements of the possible correlation chains from the ^{268,269}106 α decay and ^{264,265}104 spontaneous fission. The ^{48}Ca+^{238}U reaction was investigated at the VASSILISSA recoil separator. The experiments were performed at two beam energies leading to excitation energies of 33 and 39 MeV. The collected beam doses were 3.5×10^{18} and 2.2×10^{18}, respectively. Two spontaneous fission events were observed at the lower beam energy, which tentatively were assigned to the new neutron rich isotope ^{283}112 produced by the reaction ^{238}U(^{48}Ca,3n)^{283}112. The measured cross-section is $(5.0\ ^{+6.3}_{-3.2})$ pb and the half-life is $(81\ ^{+147}_{-32})$ s. No event was observed at the higher beam energy resulting in an upper cross-section limit of 7.3 pb for the 4n evaporation channel.

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1 Introduction

The stability of heavy nuclei depends strongly on shell structure effects. These stabilizing effects increase significantly at closed proton and neutron shells. Beyond uranium the stability of nuclei diminishes rapidly with increasing element number Z. This trend is altered at $Z = 100$ and $N = 152$ due to the existence of shell gaps in the single-particle level spectra, which appear there for deformed shapes and which are the reason for the unusual high stability of ^{252}Fm against spontaneous fission (SF).

According to the macroscopic-microscopic theory, the next spherical shell closure for the neutrons beyond $N = 126$ is predicted at $N = 184$. The stability of the superheavy nuclei increases sharply when their neutron number approaches this spherical shell closure. Due to the spherical ground-state and strong ground-state shell-correction energy, the fission barrier is wider and higher than for deformed nuclei, which is the reason for the expected increased stability against SF. For the synthesis of spherical superheavy nuclides it is of advantage, to select reaction partners with the highest possible number of neutrons in order to approach the shell $N = 184$ as close as possible [1, 2].

2 Experiment

The production of an intense ion beam from the rare and extremely expensive isotope ^{48}Ca was the key problem of our aim to synthesize isotopes of elements 110 - 112 by "hot" fusion reactions.

Neutral atoms of ^{48}Ca were injected into the plasma of the ECR-4M ion source [3] by controlled heating of a 50 mg sample of metallic calcium. The ^{48}Ca enrichment was 70 %. The amount of calcium reaching the ECR resonance cavity as well as the recovery of the material was controlled and optimized by (γ,n)-activation of ^{48}Ca and measuring the γ rays emitted from ^{47}Ca ($T_{1/2}$ = 4.5 d).

The U-400 cyclotron was modified for an axial injection of the beam from the ECR source. The high charge state of the ions (5^+) extracted from the source allowed for a continuous operation of the cyclotron. This resulted in an increase of the beam intensity by a factor of 2–3 compared with the previously used pulsed mode. As a result of these improvements we reached an internal beam intensity of up to 2 pμA of ^{48}Ca at a material consumption rate of about 0.3 mg/h.

Two sets of experiments with ^{48}Ca (^{232}Th and ^{238}U targets) and a number of test experiments (^{174}Yb, ^{206,208}Pb targets) were performed from September 1997 to October 1998.

In the first search experiment a ^{232}Th target was used. It was performed in three runs at November 1997, May and October 1998. In the experiment we used an internal beam of Ca ions, because the intensity of the internal beam is a factor of 3 higher than the external one. This experiment included the use of fast chemistry for the extraction of the fraction containing 106 element nuclei from the ^{232}Th target material and the off line measurements of the correlation chains from the ^{268,269}106 α decay and ^{264,265}104 SF. Theory [4, 5] predicts for the ^{268}106 nucleus, the granddaughter of ^{276}110 - result of the 4n evaporation channel after fusion of ^{48}Ca and ^{232}Th, the half-life against α and SF decay more than 2 hours. The isotope ^{269}106, the granddaughter of ^{277}110 - result of the 3n evaporation channel after fusion of ^{48}Ca and ^{232}Th could have the half-life against α and SF decay even longer. In this case it could be possible to use fast chemistry and to perform after that off line measurements at the geometry close to 4π. The fraction extracted

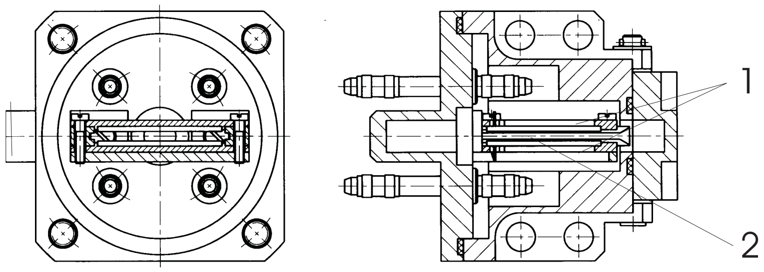

Fig. 1. The schematic view of the chamber for off-line measurements of the sources with fractions of 106 element. 1 - semiconductor detectors, 28.5 mm in diameter, distance between detectors 2 mm; 2 - supporting frame with source onto carbon foil (40 μg/cm^2 thick).

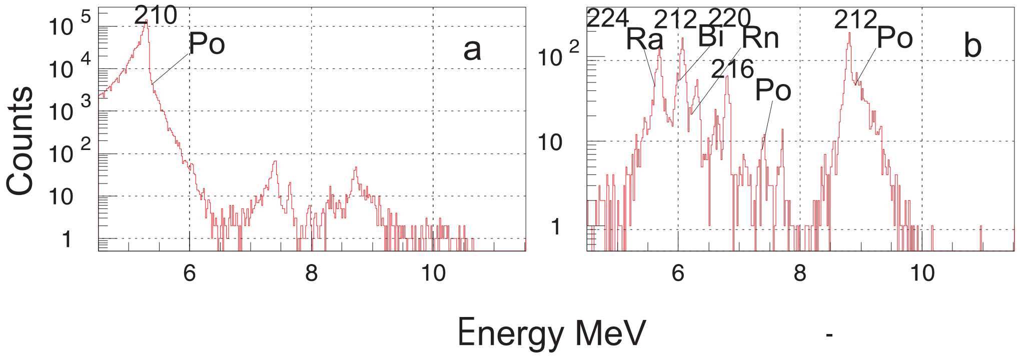

Fig. 2. An example of α spectra recorded during off line measurements. The beam dose collected - 4.5×10^{17}. Measuring time - 97 hours. a) Spectrum for the source with 106 element fraction, measurement starts 2 hours after stop of irradiation; b) Spectrum for 104 element fraction, measurement starts 12 hours after stop of irradiation.

after chemistry was deposited onto a 30 μg/cm^2 thick carbon foil and performed the 10 μg/cm^2 thick and 20 mm in diameter source. Carbon foils with those sources were placed between two silicon detectors (28.5 mm in diameter each) at special vacuum chambers (see Fig. 1). The 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 this experiment. The thickness of ^{232}Th targets was about 8 mg/cm^2, more than the range of the evaporation residues of the 110 element but less than the ranges of transfer reaction products. The selectivity of the chemical methods allowed us to suppress the fraction of U isotopes, probable candidates for long lived background products, by a factor of more than 10^5. Typical α spectra recorded during off line measurements are presented in Fig. 2.

The second search experiment involved the use of the ^{238}U and ^{232}Th targets, an external beam of Ca ions and the VASSILISSA recoil separator [6]. The mean beam intensity of ^{48}Ca on the target was 2.2×10^{12} s^{-1}. The beam energy was controlled by measuring the energy of the ions scattered under an angle of 30^o from a thin gold foil ($200\,\mu g/cm^2$). The two beam energies used in the experiment were (255 ± 3) and (262 ± 3) MeV after extraction from the cyclotron.

As target we used ^{238}U with an enrichment of 99.999 % and a thickness of 0.3 mg/cm^2. The material was deposited uniformly by electroplating on a thin, 1.6 mg/cm^2, aluminium backing foil of 125 mm diameter, which was fixed between two supporting disks of 50 mm diameter. The disk was rotating with a frequency ω = 2000 rpm. The beam energies were reduced to (231 ± 3) and (238 ± 3) MeV, respectively, up to the middle of the target due to the energy loss mainly in the aluminium backing foil. The same target thickness and preparation method were used in the case of the ^{232}Th target in the June 1998 experiment. The target design was tested in several experiments using Tb and enriched isotopes of Yb and Pb before the main experiment.

For the registration of the evaporation residues (ER) 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 detectors with microchannel plates, the recoiled ions were implanted into the 16-strip silicon detector, which had an active area of 60×60 mm^2. Each strip was position sensitive in longitudinal direction. The position resolution along each strip was measured from test reactions. A value of 0.6 mm (FWHM) was obtained for sequential α-α decays, 1.0 mm for ER-α and 1.5 mm for ER-SF events. These values were obtained for ER energies ranging between 4 and 15 MeV.

3 Results

In the experiment with the ^{232}Th target and an internal beam probe the primary beam energy was $E_L \approx 260$ MeV. The energy losses of the ^{48}Ca beam at the ^{232}Th target were about 40 MeV, it covers the width of the excitation functions for 3 and 4 neutron evaporation channels. The experiment was performed as a number of irradiations followed by a chemistry procedure (≈ 1.5 – 2 hours) for the extraction of the 106 element fraction after irradiation stopped. The duration of one irradiation was approximately 8 – 9 hours, beam dose collected was $\approx 5\times10^{16}$. In November 1997 the total beam dose collected was 2.4×10^{17} (4 irradiations), 4 samples after the chemistry procedure were prepared and measured. 1 S.F. event (40 MeV + 70 MeV) was detected, the registered time of the event after irradiation stopped was 40 hours. In May 1998, the total beam dose collected was 2.5×10^{17} (5 irradiations), 5 samples were prepared and measured. One S.F. event (82 MeV + 54 MeV) with registered time after an irradiation of 92 hours and one SF event (50 MeV + 7 MeV) with registered time after an irradiation of 126 hours were detected. The background conditions were good enough, during two months of the measurements without samples we did not detect any high energy events, which could be explained as SF events. In October 1998 5 irradiations were performed. To accept higher beam intensity internal beam probe with rotating wheel were used (in the previous experiments self supporting standing Th foil was used). Thorium foils were fixed on the edge of the wheel. This construction allows to accept beam intensities up to 2 pμA. Altogether 5 irradiations were performed, the collected beam doses were 2.6×10^{17}, 9.6×10^{17}, 4.5×10^{17}, 4.0×10^{17} and 3.5×10^{17} respectively. After first and second irradiations more precise chemistry was used. The measurements started after 30 hours for the first irradiation and 17 hours after second one. After the second irradiation the source with the fraction of the 104 element was also prepared. Last three irradiations were followed by fast chemistry (≈ 2 hours) procedure for the extraction of the 106 element fraction, after the third irradiation fraction of the 104 element was also extracted. No SF events were detected.

During the measurements of the sample counting rate of the α- particles at the energy region from 7.5 to 9 MeV was approximately 1 event per 1 – 3 hours for each semiconductor detector facing the sample. No short α- SF correlations in the time interval up to 500 seconds were detected.

On the basis of the three detected events we could make the preliminary conclusion, that SF events could not be attributed to the spontaneous fission of the isotope $^{264}104$, the product of the α- decay chain of $^{276}110$ (the result of the 4 neutron evaporation channel). According to the theoretical predictions [4] the half-life of this isotope against spontaneous fission could be few seconds. The most reasonable explanation of the obtained results is that we detected the spontaneous fission events of isotopes $^{265}104$ or $^{269}106$, the descendants of $^{281}110$, the product of the 3 neutron evaporation channel. The cross-section evaluated for the production of the three fission events is approximately 3 - 5 pb.

Second experimental run was performed with the use of the separator VASSILISSA. Using the modified ALICE code [7], we calculated the excitation energies of $E^* = 33$ and 39 MeV for the maxima of the 3n and 4n channels, respectively. The corresponding beam energies for reactions in the middle of the target thickness are 231 and 238 MeV.

Using these two energies we investigated the reaction $^{48}Ca + ^{238}U$ at VASSILISSA. The irradiation started in March 1998. During a period of 25 days a beam dose of 3.5×10^{18} projectiles was collected at the lower energy and during a succeeding period of 15 days a dose of 2.2×10^{18} at the higher energy.

Whereas a number of registered α-particle-like signals was relatively high, only two signals from spontaneous fission were detected at the first irradiation at the lower beam energy.

Both SF events were characterized by a coincident event of a higher energy signal (161.8 and 192.9 MeV) in the stop detector and a lower energy signal (28.0 and 20.7 MeV) in the backward detectors. The first event was measured in the strip number 12 and the second in the strip number 15. The higher strip numbers are in the direction to the lower magnetic rigidity of ions after the deflection magnet. The shared energy results from the implantation close to the surface of the stop detector, in which one fission fragment is stopped and the stopping of the escaping fragment in the backward detectors after some energy loss in the inactive surface layers. The absence of signals from the TOF detectors identifies the coincidence events as radioactive decays and the high energy as fission of an implanted nucleus.

The analysis of events collected in the experiment has been provided to find generic decay links of the implants over a wide range of half-lives. For that purpose the data on the position and energy resolution of the focal plane detector for incoming particles and various decay modes were used.

Possible decay chains of the type $\mathrm{ER} \xrightarrow{E_{ER},t_{ER}} \alpha_1 \xrightarrow{E_{\alpha_1},t_1} \alpha_2 \xrightarrow{E_{\alpha_2},t_2} \cdots$ were searched for within time intervals $5\,\mu\mathrm{s} \leq t_i \leq 1000$ s and 8 MeV $\leq E_\alpha \leq$ 13 MeV. Signals from an α decay could be missed only in the case if an α particle escapes through the open front side of the detector array (15 % probability) or if the lifetime is smaller than ≈5 μs and the decay occurs in the dead time of the data acquisition system. The windows for the relative positions were twice the resolution at FWHM, as given before. No such decay chains were found for the two irradiation.

For decay chains of the type $\mathrm{ER} \xrightarrow{E_{ER},t_{ER}} \alpha_1 \xrightarrow{E_{\alpha_1},t_1} \cdots \mathrm{SF} \xrightarrow{E_{f_1,f_2},t_{SF}}$ the upper limit of the time intervals t_i was extended up to 10,000 s. Such decay chains were also not found for the two irradiation.

The most careful analysis needs the spontaneous fission of the implanted nucleus. It would result in a delayed coincidence event $\mathrm{ER} \stackrel{E_{ER},t_{ER}}{\longrightarrow} \mathrm{SF} \stackrel{E_{f_1,f_2},t_{SF}}{\longrightarrow}$. ER-like signals were randomly distributed in the time. The mean rate in the strip number 12 (first event) was $\theta_1 \approx 0.001$ Hz, in the strip number 15 (second event) it was $\theta_2 \approx 0.002$ Hz. No bursts of signals were found during the analysis. Both rates were determined for a position window of ± 0.8 mm.

To search for the most probable candidate for the parent-recoil we have analyzed the distribution of time intervals between signals. The closest recoil-like signal was detected τ_1=182 s before the first SF-event. The probability to find 1 random correlated signal during the interval τ_1 at the mean frequency of signals θ_1 is described by the Poisson law. The probability that this correlation is a chance event is 0.15 and that it is the true one is 0.85. For the second SF-event corresponding probabilities are respectively 0.09 and 0.91. Thus we can conclude that the most probable candidates for signals from generic coupled recoils preceding the SF-events are the events coming 182 and 52 s before.

The position difference determined from the ER and SF signals was 0.36 mm for the first event and 0.75 mm for the second one. For the energies of the implantation signals we measured 3.84 and 4.69 MeV, respectively. These two values are within the range expected for ERs.

The mean value of the two measured time intervals results in a lifetime of $\tau = (117^{+213}_{-\ 46})$ s or a half-life of $T_{1/2} = (81^{+147}_{-\ 32})$ s. However, a longer lifetime cannot be excluded completely. This possibility must be taken into account for the case that the first ER like event preceding the SF event is not the true parent.

The total kinetic energy (TKE) of the two fission events was obtained by using the known TKE distribution of ^{252}No [8] for the calibration. The TKE distribution of ^{252}No was measured during the preparatory experiments using the ^{206}Pb(^{48}Ca,2n)^{252}No reaction. Fig. 3 shows a plot of the ^{252}No TKE distribution together with the energies of two events observed during the ^{48}Ca + ^{238}U irradiation. The TKE values for the two events are 190 and 212 MeV, respectively. A higher energy deficit for the signals of these two events due to a possible higher atomic number as nobelium was not taken into account.

The cross-section evaluated for the production of two fission events at a beam energy of 231 MeV in the middle of the target thickness is $(5.0^{+6.3}_{-3.2})$ pb. The upper cross-section limit obtained at 238 MeV energy is 7.3 pb, calculated at a probability level of 68 %. The error bars represent only the statistical uncertainties, the absolute values have an uncertainty of a factor of 2 due to uncertainties of the determination of the projectile dose and the separator efficiency.

4 Discussion

The assignment of SF events to a certain isotope is not a straightforward task as it is in the most cases of the observation of α-decay chains. Additional information is needed, like, e.g, the measurement of excitation functions or cross bombardment. However, these data are difficult to obtain, if the cross-sections are on a level of few picobarns. Nevertheless, we will present some arguments, which allow for a most plausible assignment of the two SF events observed in the irradiation of the ^{238}U target by ^{48}Ca.

Fissioning nuclei in the region of ^{238}U, which could be produced by transfer reactions, have either too long half-lives (ground-state SF) or too short half-lives (fission isomers) in order to

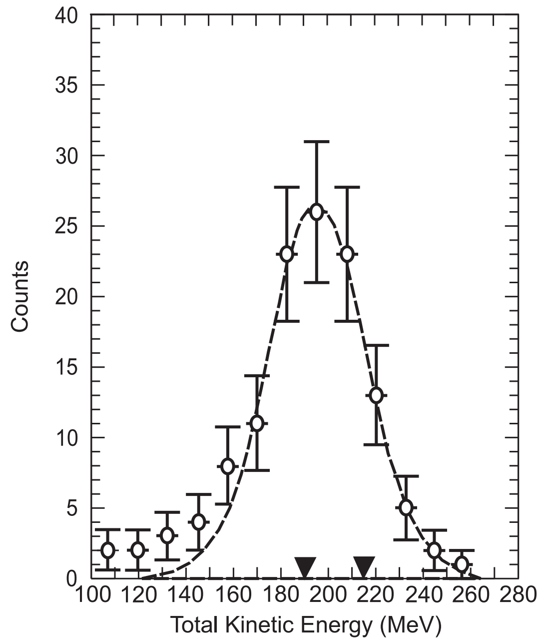

Fig. 3. The TKE spectrum for the SF of ^{252}No measured at VASSILISSA using the reaction ^{206}Pb(^{48}Ca,2n)^{252}No. The dashed line is a Gaussian curve fitted to the data. The error bars represent the statistical uncertainties only. The energy calibration was made using the results of Ref. [13]. The energies of the two fission events observed during the ^{48}Ca + ^{238}U irradiation are marked by the filled triangles.

be candidates for an assignment of the two SF events observed in the ^{48}Ca+^{238}U reaction. In addition, as measured in the case of the ^{48}Ca+^{208}Pb reaction, the transfer channels are strongly suppressed by VASSILISSA.

Spontaneous fission half-lives of the order of 100 s or longer, as measured here, are known from the region of neutron rich isotopes of elements from californium to dubnium. These known isotopes or possibly still heavier unknown isotopes could be reached by a ^{48}Ca break-up reaction. However, in this case the ERs produced by fusion of a ^{48}Ca fragment with ^{238}U have a much smaller velocity in comparison to the ERs from a complete fusion and are, therefore, suppressed by VASSILISSA.

The exclusion of transfer products and incomplete fusion products makes the assignment of the two observed SF events to the complete fusion of ^{48}Ca + ^{238}U likely. However, a definite assignment to a specific isotope is made difficult by the lack of any data from previous experiments leading into the region of these relatively neutron rich nuclei. The only one known isotope of element 112 so far is ^{277}112 synthesized in the cold fusion reaction [9]. This isotope is an α emitter with a half-life of $T_{1/2}$ = 0.24 ms.

The non-observation of α decay, the unprobable evaporation of protons or α particles from the compound nucleus and the relatively low excitation energy of 33 MeV let us tentatively assign the fission events measured in this work to the isotope ^{283}112 produced via a 3n evaporation

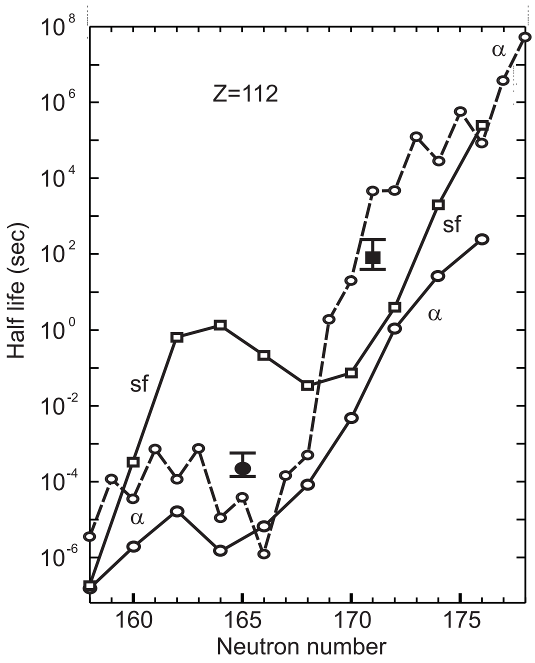

Fig. 4. Theoretical predictions for partial α and SF half-lives for isotopes of element $Z = 112$. Open circles and open squares connected by solid lines: $T_{1/2,\alpha}$ and $T_{1/2,SF}$ from Ref. [8]. Open circles connected by a dashed line: $T_{1/2,\alpha}$ from Ref. [9]. Filled circle: $T_{1/2,\alpha,exp}$ for $^{277}112$ from Ref. [14]. Filled square: $T_{1/2,SF,exp}$ from this work and tentatively assigned to $^{283}112$.

channel in the $^{48}Ca+^{238}U\rightarrow^{286}112^{*}$ reaction.

Results of calculations from Smolańczuk [4] and Möller et al. [5] describing the decay properties of isotopes of element 112 are shown in Fig. 4. The theoretical data were obtained from using the macroscopic-microscopic method. Both calculations were able to reproduce the decay properties of the so far known nuclei of the heavy elements rather well within an accuracy of about 1 to 2 orders of magnitude. This can be seen by comparing the measured α half-life of $^{277}112$ (neutron number $N = 165$) with the calculations (Fig. 4). For this isotope SF is expected to have a much longer half-life than α decay.

The calculations predict a steep rise of the half-lives with increasing neutron number. This is a result from the decreasing negative shell-correction energy and a transition of the nuclear shape from well deformed quadrupole to spherical, which both increase the stability and the fission barrier and thus the partial α and SF half-lives. Compared to the SF half-life calculations performed for the even-even nuclei as shown in Fig. 4, hindrance factors due to a specialization energy for the odd neutron isotopes of up to about three orders of magnitude may be expected. Therefore, the calculated SF half-life for $^{283}112$ is in the range from 100 to 1,000 s, which agrees with the experimental half-life of (81^{+147}_{-32}) s.

The cross-section for the production of element 112 is higher (5.0 pb) using the hot fusion reaction ($E^* = 33$ MeV) and a ^{48}Ca beam respect to the value obtained by using the cold fusion reaction ($E^* = 10$ MeV) and a ^{70}Zn beam (1.0 pb) [9]. The relatively high cross-section of the hot fusion reaction, despite the higher excitation energy, may be caused by the higher ground state shell correction of the evaporation residues or by the lower fusion hindrance in the hot fusion reaction. The negative result of the irradiation at the higher beam energy and the obtained upper limit of 7.3 pb shows that the 4n channel does not have a significantly higher cross-section as the 3n channel.

The irradiation of ^{238}U by ^{48}Ca was a first experiment at our Laboratory using an improved experimental set-up at VASSILISSA, which allows for investigation of the hot fusion reaction on a cross-section level of 1 pb. The investigated ^{48}Ca+^{238}U reaction leads to ERs, which are far from the region of known isotopes of the heavy elements. Moreover, no α decay chains were observed, whose properties would provide additional spectroscopic information in order to establish a final assignment of the observed reaction product. Therefore, we continue our experiments with an investigation of the ^{48}Ca+^{232}Th$\rightarrow^{280}$110* reaction. Using this reaction, ERs should be produced halfway between the known isotopes in the region of deformed heavy elements and the ^{283}112 isotope. The results could hopefully provide more data in order to establish a trend of the decay properties and cross-sections for the synthesis of isotopes by hot fusion on the way to the region of spherical superheavy elements. Using the energy $E_{targ.}$ = 238 $\pm$ 2 MeV (E* = 39 MeV) we investigated the reaction ^{48}Ca(^{232}Th,4n)^{276}110 at VASSILISSA. The irradiation started in June 1998. During a period of 15 days a beam dose of 1.8×10^{18} projectiles was collected. No spontaneous fission events were detected and no α-α correlations were observed in the whole α-particle energy range from 8 to 12 MeV in the time interval of up to 1000 s. The upper cross-section limit obtained at 238 MeV energy is 9.1 pb, calculated at a probability level of 68 %.

5 Conclusion

We report a new attempt to synthesize super heavy spherical nuclei by the ^{48}Ca+^{232}Th and ^{48}Ca+^{238}U reactions. As compared with all previous experiments with ^{48}Ca ions, the sensitivity of the present experiments is more than 100 times higher. Three spontaneous fission events were detected in the ^{48}Ca+^{232}Th reaction and two spontaneous fission events were detected in the ^{48}Ca+^{238}U reaction 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 isotopes ^{277}110 and ^{283}112, respectively, formed in the 3n – evaporation channel. The half-life of the new isotope ^{283}112 is about 100 s. It could also be that it undergoes α-decay with a branching ratio $b_\alpha \leq 50$ %.

The present experiments are the first ones 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 continue these experiments in order to collect more statistics and to try to synthesize other isotopes with $Z = 110-114$. It is also planned to improve chemical methods in order to obtain more clean and detailed information, when chemical extraction of the long-lived fractions of the 104 and 106 elements and off-line α and SF spectrometry are used in the experiments.

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