

Search for new isotopes of element 112 by irradiation of ^{238}U with $^{48}\text{Ca}^*$

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Abstract. The reaction $^{48}\text{Ca}+^{238}\text{U}$ was investigated at the recoil separator VASSILISSA at the Flerov Laboratory of Nuclear Reactions, JINR Dubna, in attempts to synthesize new isotopes of element 112. The experiments were performed at two beam energies resulting in excitation energies of the compound nucleus of 33 and 39 MeV. The collected beam dose was 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 in the reaction $^{238}\text{U}(^{48}\text{Ca},3n)^{283}112$. 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 the upper cross-section limit of 7.3 pb.

PACS. 21.10.Dr Binding energies, masses and excitation energy – 23.60.+e α -decay – 25.70.-z Heavy ion induced reactions and scattering – 25.85.Ca Spontaneous fission – 27.90.+b $220 \leq A$

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 the element number Z . This tendency changes 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 unusually 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 that

for deformed nuclei, which is the reason for the expected increased stability to SF. For the synthesis of spherical superheavy nuclides it is advantageous, to select reaction partners with the highest possible number of neutrons in order to approach the shell $N=184$ as close as possible [1].

During the last two decades the so called “cold” fusion reaction using lead and bismuth targets was successful in synthesizing the heaviest transfermium elements with $Z = 107-112$ [2]. The advantage of this reaction is that only slightly excited ($E^* \approx 10-20$ MeV) compound nuclei are produced at bombarding energies near the fusion barrier. This is a consequence of the double magic structure of the ^{208}Pb target and the strongly bound projectiles from ^{54}Cr to ^{70}Zn . Low excitation energies of fragile heavy compound nuclei are considered to be the reason for surviving the prompt fission.

The “hot” fusion reaction using targets of Th to Cf was also applied to synthesize isotopes of the elements $Z=102$ to 110 [3]. Due to the higher temperature of the compound nucleus, a number of neutrons (4–5) must be evaporated to reach the ground-state. These reactions allow the study of decay properties of more neutron rich isotopes of transfermium elements, which are impossible to synthesize directly by “cold” fusion reactions, e.g. long lived isotopes with neutron numbers close to $N=162$ and

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$Z \geq 106$. Very asymmetric systems fuse with higher probability at the Coulomb barrier, however simultaneously the excitation energies of the compound nuclei change from typically 10–20 MeV in the case of “cold” fusion reactions to 40–50 MeV. This increase of the excitation energy reduces the survival probability of heavy compound nuclei.

The minimum excitation energy in the case of the “hot” fusion reactions can be obtained with the use of a ^{48}Ca beam [1]. Therefore, the compound nucleus $^{286}112$ formed in the reaction $^{48}\text{Ca} + ^{238}\text{U}$ is less strongly excited. This is a result of the significant mass deficit of the double magic ^{48}Ca . The excitation energy at the fusion barrier [4] is only $E^* \approx 35$ MeV, which increases the survival probability of the compound nucleus considerably. The high mass asymmetry in the entrance channel ($A_1/A_2 = 0.2$ and $Z_1 Z_2 = 1840$) is also reason for a decrease of the dynamic hindrance of fusion, an effect, which was observed earlier for the more symmetric “cold” fusion reactions [5].

Using a modified version of the statistical code ALICE [6] we could rather well reproduce the measured cross-sections of reactions from $^{12}\text{C} + ^{248}\text{Cm}$ to $^{34}\text{S} + ^{244}\text{Pu}$. For these systems the effective fissility parameter x_{eff} varies from 0.68 to 0.81. Not so far from this range is the effective fissility parameter of the reaction $^{48}\text{Ca} + ^{238}\text{U}$, $x_{eff} = 0.85$, resulting in the compound nucleus $^{286}112$. Therefore, we used the same parameter set also for an estimate of the cross-section for production of element 112. The result was 5 to 20 pb for the 3n-evaporation channel and 1 to 5 pb for the 4n channel. These cross-sections would be high enough to be reached in a reasonable irradiation period of about 1 – 1.5 months. However, some improvements of the experimental set-up were necessary, concerning the preparation of the ^{48}Ca beam from the U-400 cyclotron and the detection system at the separator VASSILISSA.

2 Experimental set-up

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 element 112 by hot fusion reactions.

Neutral atoms of ^{48}Ca were injected into the plasma of the ECR-4M ion source [7] 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 $1 \mu\text{A}$ of ^{48}Ca at a material consumption rate of about 0.3 mg/h.

The beam was extracted from the cyclotron by the stripping method. The mean beam intensity of ^{48}Ca on the

target was $2.2 \times 10^{12} \text{ s}^{-1}$. The beam energy was controlled by measuring the energy of the ions scattered under an angle of 30° from a thin gold foil ($200 \mu\text{g}/\text{cm}^2$). 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 $0.3 \text{ mg}/\text{cm}^2$. The material was deposited uniformly by electroplating on a thin ($1.6 \text{ mg}/\text{cm}^2$) aluminium backing foil of 125 mm in diameter, which was fixed between two supporting disks of 50 mm in diameter. The disk was rotating with a frequency $\omega = 2000$ rpm. To the middle of the target the beam energies were reduced to (231 ± 3) and (238 ± 3) MeV respectively due to energy loss mainly in the aluminium backing foil. The target design was tested in several experiments using Tb and enriched isotopes of Yb and Pb before the main experiment.

The evaporation residues (ERs) were separated in-flight from the beam particles and other reaction products by the electrostatic recoil separator VASSILISSA [8]. Scattered low energy projectiles are the main contribution to the background at the focal plane. In order to reduce this background, we installed an additional 8° dipole magnet after the second quadrupole triplet of the separator. This magnet allowed for mounting the detectors out of the primary beam direction.

A 3 μm thick degrader foil (mylar) was placed in front of the silicon detectors for reduction of the number of elastically scattered low energy beam-like projectiles reaching the focal plane of the separator.

The field settings were calculated according to a mean charge state of $(19 \pm 1)^+$ of the ERs and the two mean energies of 38 and 40 MeV, respectively.

The separation efficiency of ERs from the reaction $^{48}\text{Ca} + ^{238}\text{U}$ was estimated from preparatory experiments. Cross sections of various xn-evaporation channels were measured for the reactions of ^{159}Tb , ^{174}Yb and $^{206,208}\text{Pb}$ targets irradiated by ^{48}Ca projectiles of different energies. The measurements showed that about 25 % of the $^{286-x}112$ ERs, produced with a ^{238}U target, would be implanted into the detector.

For the registration of the ERs 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 $60 \times 60 \text{ mm}^2$. 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 energies of the ERs in the range from 4 to 15 MeV.

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 strip 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 long edge. In the present series of experiments four neighboring strips of the backward detectors were connected galvanically so that a total of 16 energy sensitive segments was formed, each with a resolution within the range from 120 to 150 keV. The efficiency of the silicon array for detection of α particles with full energy was 85 % of 4π , however, the small energy loss from escaping α 's in the stop detector could be measured only for 50 % of events, because of the low implantation energy of the ERs.

The signals from the TOF detectors were used both for measurement of the velocity of the recoils and for distinguishing the radioactive decays of the previously implanted nuclei. The high efficiency of the TOF detectors allowed obtaining very clean decay spectra. The time window for measuring decay chains could be significantly widened (up to several hours). The latter was particularly important for the correlation of decays with a long half-life at the presence of a continuously running beam.

The counting rate of all background events at the focal plane detector was only 25–30 Hz. The counting rate on one strip of the detector of α -like events (background without TOF signal) with energies more than 7.5 MeV was 20–30 per hour, and of recoil-like events (background with TOF signal) with energies more than 4 MeV it was 80–120 per hour. The background rate depended on the horizontal position of the strip. From the results of the test reactions we estimated an energy range from 4 to 15 MeV for the signals originating from implanted $Z=112$ nuclei.

3 Experiment and results

In a first part of the experimental program we investigated the reactions $^{208}\text{Pb}(^{48}\text{Ca},\text{xn})^{256-x}\text{No}$ and $^{206}\text{Pb}(^{48}\text{Ca},\text{xn})^{254-x}\text{No}$, both at the recoil separator VASSILISSA and at the in-beam time-of-flight fission fragment spectrometer CORSET [9]. At CORSET fusion cross-sections of the compound nuclei were determined by observing the prompt fission events and by measurement of the fission fragment mass distribution. For the irradiation of the lead targets the beam energy was varied over a wide range covering the evaporation channels $1n$ to $3n$ so that a set of excitation functions was obtained from both experiments. These data allowed for a comparison of the ER cross-sections measured at VASSILISSA [10] and the total fusion cross-sections measured in-beam.

Then, the measurement was repeated at CORSET using a ^{238}U target. Compound-nucleus formation was observed and from the yield as function of the beam energy we concluded that the residue cross-section must be highest for the $3n$ and $4n$ channels. Detailed results of these experiments will be published elsewhere, see e.g. [9]. Using the modified ALICE code [6], we calculated beam energies for reactions in the middle of the target thickness $E_{lab}=231$ and 238 MeV for the maxima of the $3n$ and $4n$ channels, respectively. The corresponding excitation energies are $E^*=33$ and 39 MeV. The known masses of projec-

tile and target and the predicted mass of the compound nucleus were used [11,12].

Using these two energies we investigated the reaction $^{48}\text{Ca}(^{238}\text{U},\text{xn})^{286-x}112$ 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 190.9 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 performed to find generic decay links of the implants over a wide range of half-lives. For that purpose we used the data on the position and energy resolution of the focal plane detector for incoming particles and various decay modes.

Possible decay chains of the type $\text{ER} \xrightarrow{E_{ER}, t_{ER}} \alpha_1 \xrightarrow{E_{\alpha_1}, t_1} \alpha_2 \xrightarrow{E_{\alpha_2}, t_2} \dots$ were searched for within time intervals of $5 \mu\text{s} \leq t_i \leq 1000$ s and $8 \text{ MeV} \leq E_{\alpha} \leq 13 \text{ MeV}$. Signals from an α decay could be missed only in the case if an α particle escaped through the open front side of the detector array (15 % probability) or if the lifetime was smaller than $\approx 5 \mu\text{s}$ and the decay occurred 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 irradiations.

For decay chains of the type $\text{ER} \xrightarrow{E_{ER}, t_{ER}} \alpha_1 \xrightarrow{E_{\alpha_1}, t_1} \dots$ 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. Neither were such decay chains found for the two irradiations.

The spontaneous fission of the implanted nucleus needs the most careful analysis. It would result in a delayed coincidence event $\text{ER} \xrightarrow{E_{ER}, t_{ER}} \text{SF} \xrightarrow{E_{f_1}, f_2, t_{SF}}$. ER-like signals were randomly distributed in time. The mean rate in the strip number 12 (the first event) was $\theta_1 \approx 0.001$ Hz, in the strip number 15 (the 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 ± 1.5 mm.

To search for the most probable candidate for the parent-recoil we have analyzed the distribution of time in-

tervals between signals. The closest recoil-like signal was detected $\tau_1=182$ sec before the first SF-event. Before the second SF-event the closest recoil-like signal was detected $\tau_2=52$ sec. The probability to find one random correlated signal during the interval τ_i at the mean frequency of signals θ_i is described by the Poisson law. The probability that the first correlation is a chance event is 0.15, and that of its being the true one is 0.85. For the second SF-event the 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 sec 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 energy range expected for ERs. The values of the TOF signals of the ERs associated to the two SF events equal to the values expected for complete fusion reaction products.

The mean value of the two measured time intervals results in a lifetime (117_{-46}^{+213}) s or a half-life of $T_{1/2} = (81_{-32}^{+147})$ 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 [13] for calibration. The TKE distribution of ^{252}No was measured during the preparatory experiments using the reaction $^{206}\text{Pb}(^{48}\text{Ca},2n)^{252}\text{No}$. Figure 1 shows a plot of the ^{252}No TKE distribution together with the energies of two events observed during the $^{48}\text{Ca}+^{238}\text{U}$ 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 $(5.0_{-3.2}^{+6.3})$ 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 as straightforward a task as it is in 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 at a level of a few picobarns. Nevertheless, we will present some arguments, which allow a most plausible assignment of the two SF events observed.

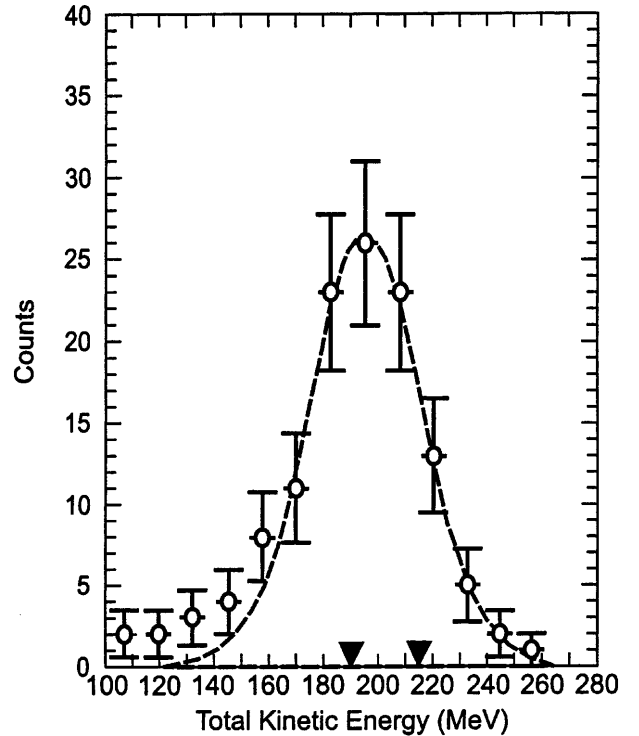


Fig. 1. The TKE spectrum for the SF of ^{252}No measured at VASSILISSA using the reaction $^{206}\text{Pb}(^{48}\text{Ca},2n)^{252}\text{No}$. The dashed line is a Gaussian curve fitted to the data. The error bars represent the statistical uncertainties only. The energy calibration was made using the results of [13]. The energies of the two fission events observed during the $^{48}\text{Ca} + ^{238}\text{U}$ irradiation are marked by the filled triangles

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

SF 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 would have the velocity and corresponding TOFs significantly differing from that of complete fusion products.

ERs resulting from fast “pre-compound” alpha emission could have TOFs and implantation energies close to that of complete fusion ERs. Such processes have a very low probability, especially at that low bombarding energy, and have not been measured until now in the region of transfermium nuclei. Additionally we can mention that calculations [15] resulted the VASSILISSA suppression factors of more than 100 for products of such processes.

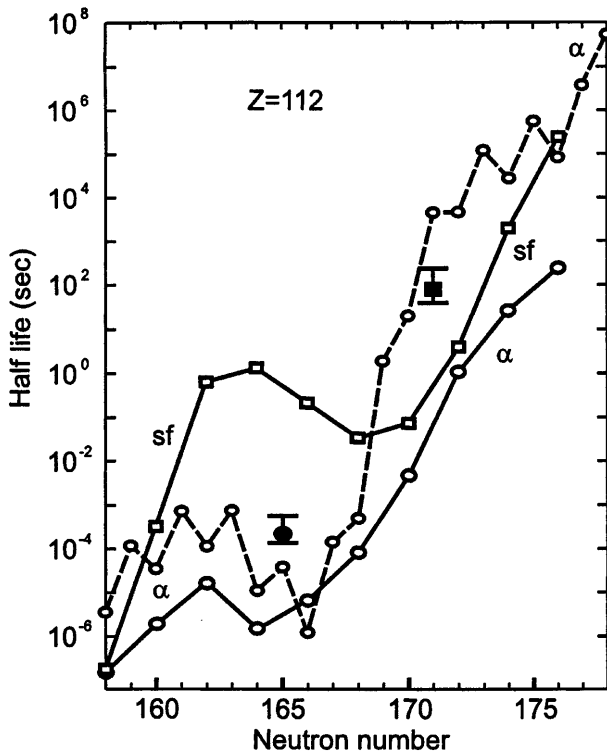


Fig. 2. Theoretical predictions for partial α and SF half-lives for isotopes of element $Z = 112$. Open circles and open squares connected by solid lines: $T_{1/2,\alpha}$ and $T_{1/2,SF}$ from [16]. Open circles connected by a dashed line: $T_{1/2,\alpha}$ from [12]. Filled circle: $T_{1/2,\alpha,exp}$ for $^{277}\text{112}$ from [14]. Filled square: $T_{1/2,SF,exp}$ from this work and tentatively assigned to $^{283}\text{112}$

The exclusion of transfer products and incomplete fusion products makes the assignment of the two observed SF events to the complete fusion of $^{48}\text{Ca}+^{238}\text{U}$ 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 $^{277}\text{112}$ synthesized in cold fusion reaction [14]. This isotope is an α emitter with a half-life of $T_{1/2}=0.24$ ms.

The nonobservation of α decay, the improbable 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 $^{283}\text{112}$ produced via a 3n evaporation channel in the reaction $^{48}\text{Ca}+^{238}\text{U}\rightarrow^{286}\text{112}^*$.

Results of calculations from Smolańczuk [16] and Möller et al. [12] describing the decay properties of isotopes of element 112 are shown in Fig. 2. The theoretical data were obtained 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 $^{277}\text{112}$ (neutron number $N=165$) with the calculations (Fig. 2). 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 with the SF half-life calculations performed for the even-even nuclei as shown in Fig. 2, 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 $^{283}\text{112}$ is in the range from 100 to 1,000 s, which agrees with the experimental half-life of (81_{-32}^{+147}) s.

The calculated partial α half-lives also increase steeply with increasing neutron number. They start to become comparable with [16] or even exceed [12] the SF half-lives at $N=168-170$. For almost all isotopes shown in Fig. 2 the calculations of [12] result in the longer half-lives, and still, in the case of the known α emitter $^{277}\text{112}$, the prediction is a factor of ten smaller than the experimental value. Concerning $^{283}\text{112}$ only a lower limit of $T_{1/2,\alpha} \geq 190$ s can be deduced from the experiment. This value is in agreement with the data by Möller et al. and does not completely contradict the data by Smolańczuk, taking into account that his values were calculated only for the even isotopes.

The cross-section for the production of element 112 is higher (5.0 pb) using the hot fusion reaction and a ^{48}Ca beam as the value obtained using the cold fusion reaction and a ^{70}Zn beam (1.0 pb) [14]. The relatively high cross-section is probably a result of the relatively low excitation energy of the compound nucleus that can be achieved at the fusion barrier using ^{48}Ca . The negative result of the irradiation at the higher beam energy and the obtained upper limit of 7.3 pb show 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 setup at VASSILISSA, which allows investigation of the hot fusion reaction on a cross-section level of 1 pb. The investigated reaction $^{48}\text{Ca}+^{238}\text{U}$ results in 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 plan to continue our investigation of the reaction $^{48}\text{Ca}+^{232}\text{Th}\rightarrow^{280}\text{110}^*$. Using this reaction, ERs should be produced halfway the known isotopes in the region of deformed heavy elements and the isotope $^{283}\text{112}$. The results will hopefully provide more data for establishing 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.

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