SYNTHESIS OF SUPERHEAVY ELEMENTS WITH ^{48}Ca AND LIGHTER ION BEAMS

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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. 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 can increase sharply, when their neutron number approaches this spherical shell closure [1,2]. Due to the spherical ground-state and strong ground-state shell-correction energy, the fission barrier is wider and higher that 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 if reaction partners with the highest possible number of neutrons are selected in order to approach the shell N = 184 as close as possible [3].

Two types of target–projectile combinations have been used for the synthesis of the transfermium elements: actinide targets from thorium to californium with corresponding beams from sulfur to carbon (the so called "hot" fusion reactions) or targets near lead and beams from zinc to titanium (the so called "cold" fusion reactions). More symmetric target–projectile combinations have been tried but didn't turn out to be promising in the heavy element synthesis.

Excitation energies at the Coulomb barrier for the actinide target based reactions are near 40 – 50 MeV whereas with lead, or bismuth targets typical excitation energies are 10 – 20 MeV. Correspondingly, the compound nuclei need four – five or one – two evaporation steps to dissipate the excitation energy. The production cross sections for the both types of reactions leading to the elements with Z $\geq$ 102 are of the same order, or "hot" fusion reactions have cross sections lower by not more than an order of magnitude. The explanation could be found when studying the neutron–to–total width ratios for highly excited heavy nuclei, and the ratio $< \Gamma_n/\Gamma_{tot} >= 0.4$–$0.6$ at the compound nucleus excitation energy E* > 40 MeV has been found recently [4]. One can conclude that the obtained values of $< \Gamma_n/\Gamma_{tot} >$ at high excitation energies signify the fact that the main losses in the yields of transfermium evaporation residues (ER), formed in heavy ion "hot" fusion reactions, arise at the final steps of deexcitation cascades. Partially this conclusion was confirmed by the recent experiments in which neutron rich isotopes of elements 108 and 110 were synthesised via the 5 neutron evaporation channel in the reactions ^{34}S+^{238}U [5] and ^{34}S + ^{244}Pu [6].

"Hot" fusion reactions were used for the synthesis and study of the decay properties of elements with atomic numbers 100 $\leq$ Z $\leq$ 106 until the mid–seventies [7]. During the last two decades the "cold" fusion reactions using lead and bismuth targets were successful in synthesizing the transfermium elements with Z = 107–112 [8]. At the same time the "hot" fusion reactions were also applied to the synthesis of neutron rich isotopes of the elements Z = 102 to 110 [6,9]. Using the gas–filled recoil separator [10] and an electrostatic recoil separator VASSILISSA [11] installed at the beam lines of the U–400 heavy ion cyclotron of the FLNR JINR we investigated the fusion reactions leading to No – Db and heaviest isotopes of Sg, Hs and element 110 (see fig. 1). The experiments performed at Dubna by employing the gas-filled recoil separator have resulted in the study of the new neutron rich nuclides ^{262}Rf, ^{265}Sg, ^{266}Sg, ^{267}Hs and ^{273}110.

The ground-state decay properties that were established for ^{266}106 and ^{262}104, by using the ^{248}Cm + ^{22}Ne reaction, revealed a large enhancement in their stability as compared to that of nuclides with lower Z or N values. The α decay energy measured for the neutron - rich ^{273}110, produced by the ^{244}Pu + ^{34}S reaction, provided indication that a neutron shell closure exists and is located at N = 162. The N = 162 shell closure appears much weaker than the spherical shell N = 126, but at least seems to be comparable in strength with the deformed shell N = 152 that is in good agreement with the recent theoretical predictions.

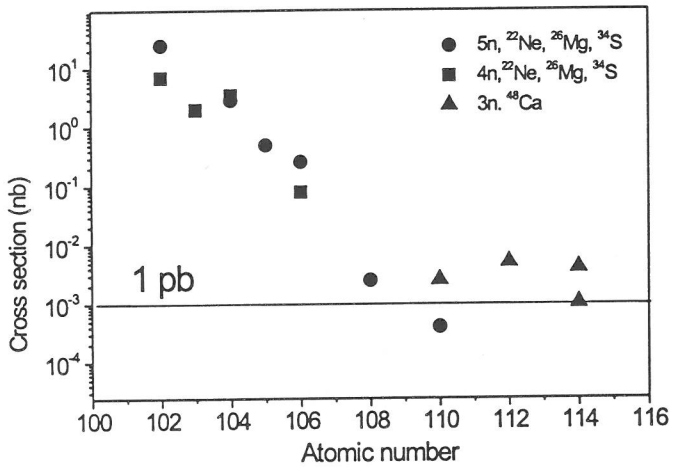

Figure 1: Experimental production cross sections for actinide based fusion evaporation reactions, leading to the transfermium region.

To check a hypothetical existence of nuclear shell closures at Z = 114 and N = 178 - 184, which is one of the fundamental predictions of modern nuclear theory, a series of experiments is now being performed at the FLNR JINR, aiming at the production of neutron rich nuclides with Z = 110, 112 and 114 (N = 166-167, 170-171 and 174-175, respectively,) via the complete fusion reactions between ^{232}Th, ^{238}U and ^{242,244}Pu targets and ^{48}Ca projectiles delivered by the U400 cyclotron. With the use of ^{48}Ca as a beam the minimum excitation energy for the asymmetric fusion reactions ("warm" fusion) could be obtained [3]. Therefore, the compound nuclei ^{280}110, ^{286}112 and ^{290,292}114 formed in the reactions with ^{48}Ca were less excited. This is the result of a significant mass deficit of the doubly magic ^{48}Ca. The excitation energy at the Coulomb barrier is only $E^* \approx 31 - 35$ MeV, which decreases the number of evaporated neutrons from 5 to 3. According to the calculations, the de–excitation channel with evaporation of 3 neutrons will have the maximum cross section. This could considerably increase the survival probability of the heavy ER at the exit channel. The high mass asymmetry in the entrance channel ($A_1/A_2 = 0.2$ and $Z_1Z_2 = 1840$) is also a reason for a decrease in the dynamic hindrance of fusion, the effect, which was observed earlier for the more symmetric "cold" fusion reactions [12]. Theory [1,2] predicts for the neutron rich nuclides with Z = 110, 112 and 114 (N = 166-167, 170-171 and 174-175, respectively,) a strong increase in the half-lives against α and SF decay of more than 5 orders of magnitude in comparison with known neutron deficient isotopes of elements 110 - 112. That fact allows one, on the one hand, to plan experiments with the use of rather slow chemistry methods and off – line measurements, but, on the other hand, this fact raises the requirements for the beam quality and background

conditions while using the recoil separators. Production of an intense ion beam from the rare and extremely expensive isotope ^{48}Ca was the key problem in our atempts to synthesize isotopes of elements 110 – 114 by "warm" fusion reactions. Neutral atoms of ^{48}Ca were injected into the plasma of the ECR–4M ion source by a controlled heating of a $\approx$ 50 - 60 mg sample of metallic calcium. The ^{48}Ca enrichment was 70 %. The amount of calcium reaching the ECR resonance cavity and 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 a continuous operation of the cyclotron. This resulted in an increase in 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 pμA 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. As targets we used ^{232}Th, ^{238}U (99.999 %), ^{242}Pu (97 %) and ^{244}Pu (98.6 %) with thicknesses from 0.2 to 0.4 mg/cm^2. The material was deposited uniformly by electroplating on a thin, 1.6 mg/cm^2, aluminium or a 0.7 mg/cm^2 titanium backing foils, which were fixed on disks with windows.

The experiments with ^{48}Ca beams started at the end of 1997. Two series of experiments with ^{48}Ca were performed.

In the first series of experiments 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 higher that external one by a factor of 3. This experiment included the use of the chemistry procedure for the extraction of a fraction containing 106 element nuclei from the ^{232}Th target material and off line measurements of the correlation chains from α decay of ^{268,269}Sg and SF of ^{264,265}Rf. In the experiment with the ^{232}Th target and the internal beam probe the primary beam energy was $E_{Lab} \approx$ 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 the 3 and 4 neutron evaporation channels. The experiment was peformed as a number of irradiations with a following chemistry procedure ($\approx$ 1.5 - 2 hours) for the extraction of the 106 element fraction after the end of irradiation. The duration of an irradiation was approximately 8 – 9 hours, the collected beam dose was $\approx 5\times10^{16}$. Three SF events were altogether detected during this run. 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 [13]. The cross-section evaluated for the production of the three fission events is approximately 2 - 3 pb.

In the second series of experiments the evaporation residues were separated in–flight from the beam particles and other reaction products by the electrostatic recoil separator VASSILISSA and the gas filled recoil separator. The reaction ^{48}Ca + ^{238}U was investigated at the recoil separator VASSILISSA. The experiments were performed at two beam energies resulting in 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 were tentatively assigned to the new neutron rich isotope ^{283}112 produced by the reaction ^{238}U(^{48}Ca,3n)^{283}112 [14]. The measured cross-section was (5.0 $^{+6.3}_{-3.2}$) pb and the half–life was (81 $^{+147}_{-32}$) s. Not a single event was observed at the higher beam energy resulting in an upper cross–section limit of 7 pb for the 4n evaporation channel. The reaction ^{48}Ca + ^{232}Th was also investigated at the recoil separator VASSILISSA. The experiments were performed at two beam energies resulting in excitation energies of 31 and 38 MeV. The collected beam doses were

4.8×10^{18} and 1.8×10^{18}, respectively. A single decay chain was observed at the lower beam energy, which could be assigned to the new neutron rich isotope $^{277}110$ and its descendants, produced by the reaction ^{232}Th(^{48}Ca,3n)$^{277}110$. The cross–section corresponding to a possible event was about 2 pb. Not a single event was observed at the higher beam energy resulting in an upper cross–section limit of 9 pb for the 4n evaporation channel. The reaction ^{48}Ca + ^{244}Pu was investigated at the gas filled recoil separator. The experiment was performed at the beam energy resulting in an excitation energy of 35 MeV. The collected beam dose was 5.2×10^{18}. A single decay chain was observed, which could be assigned to an isotope of the new 114 element with the mass number 289 and its descendants, produced by the reaction ^{244}Pu(^{48}Ca,3n)$^{289}114$. The measured cross-section was about 1 pb [15]. Now, in March 1999 the reaction ^{48}Ca + ^{242}Pu is being investigated at the recoil separator VASSILISSA. The beam energy corresponds to the compound nucleus excitation energy of 33.5 MeV, which coincides with the calculated maximum of the cross section for the 3n evaporation channel. For the beam dose of 2.4×10^{18} a decay chain ER $\rightarrow \alpha \rightarrow$ SF was observed, which could be assigned to an isotope of the new 114 element with the mass number 287 and its descendant, $^{283}112$, produced by the reaction ^{242}Pu(^{48}Ca,3n)$^{287}114$. The cross-section corresponding to the already collected beam dose was about 4 pb. The experiment and data analysis are in progress.

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