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SYNTHESIS OF SUPERHEAVY NUCLEI: NEAREST AND DISTANT OPPORTUNITIES*

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There are only 3 methods for the production of heavy and superheavy (SH) nuclei, namely, a sequence of neutron capture and beta(−) decay, fusion reactions, and multinucleon transfer reactions. A macroscopic amount of the long-living SH nuclei located at the island of stability may be produced by using the pulsed nuclear reactors of the next generation only if the neutron fluence per pulse will be increased by about three orders of magnitude. Low values of the fusion cross sections and very short half-lives of nuclei with $Z > 120$ also put obstacles in synthesis of new elements. At the same time, an important area of SH isotopes located between those produced in the cold and hot fusion reactions remains unstudied yet. This gap could be filled in fusion reactions of ^{48}Ca with available lighter isotopes of Pu, Am, and Cm. New neutron-enriched isotopes of SH elements may be produced with the use of a ^{48}Ca beam if a ^{250}Cm target would be prepared. In this case, we get a real chance to reach the island of stability owing to a possible beta(+) decay of ^{291}114 and ^{287}112 nuclei formed in this reaction with a cross section of about 0.8 pb. Multinucleon transfer processes look quite promising for the production and study of neutron-rich heavy nuclei located in the upper part of the nuclear map not reachable by other reaction mechanisms. Reactions with actinide beams and targets are of special interest for synthesis of new neutron-enriched transfermium nuclei and not-yet-known nuclei with closed neutron shell $N = 126$ having the largest impact on the astrophysical r-process. The estimated cross sections for the production of these nuclei allows one to plan such experiments at currently available accelerators.

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1. Motivation

Due to the bending of the stability line toward the neutron axis, in fusion reactions of stable nuclei one may produce only proton rich isotopes of heavy elements. For elements with $Z > 100$ only neutron deficient isotopes (located to the left of the stability line) have been synthesized so far (see the left panel of Fig. 1). That is the main reason for the impossibility to reach the center of the "island of stability" ($Z \sim 110 \div 120$ and $N \sim 184$) in fusion reactions with stable projectiles.

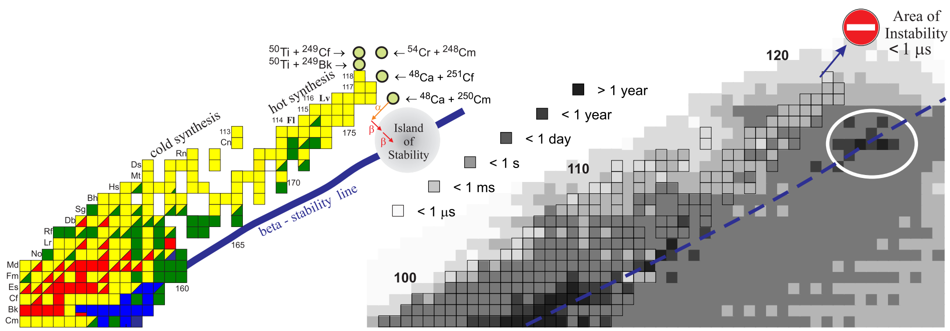

Fig. 1. Upper part of the nuclear map. Current and possible experiments on synthesis of SH elements are shown. (Right panel) Predicted half-lives of SH nuclei and the "area of instability". Known nuclei are shown by the outlined rectangles.

Further progress in the synthesis of new elements with $Z > 118$ is not quite evident. Cross sections of the "cold" fusion reactions decrease very fast with increasing charge of the projectile (they become less than 1 pb already for $Z \geq 112$ [1, 2]). For the more asymmetric ^{48}Ca induced fusion reactions, rather constant values (of a few picobarns) of the cross sections for the production of SH elements up to $Z = 118$ were found [3]. This unusual (at first sight) behavior of the cross sections has been predicted and explained in [4, 5] by the relatively slow decrease of the fusion probability (in contrast to the more symmetric "cold" fusion reactions), and by the increasing survival probability of compound nuclei (CN) owing to increasing values of their fission barriers caused by the larger shell corrections as the CN approach the neutron and proton closed shells in the region of the island of stability. These predictions have been fully confirmed by the experiments performed in Dubna and later in Berkeley [6], and at GSI [7, 8].

For the moment, californium ($Z = 98$) is the heaviest available target that can be used in experiments. The half-life of the einsteinium isotope $^{254}_{99}$Es is 276 days, sufficient to be used as target material. However, it is impossible to accumulate the required amount of this matter (several milligrams) to prepare a target. To get SH elements with $Z > 118$ in a more realistic way, one should proceed to heavier than ^{48}Ca projectiles. ^{50}Ti is the most promising projectile for further synthesis of SH nuclei. Our calculations demonstrated that the use of the titanium beam instead of ^{48}Ca decreases the yield of the same SH element due to a worse fusion probability by about factor 20 [9]. Nevertheless, the elements 119 and 120 can be produced in the fusion reactions of ^{50}Ti with ^{249}Bk and ^{249}Cf targets (or in the ^{54}Cr+^{248}Cm fusion reaction) with the cross sections of about 0.04 pb [9] which are already at the limit of the experimental possibilities. The first attempts to perform these experiments have been already made at GSI [10, 11]. Only the upper limits of the cross sections have been obtained. For synthesis of element 119, this limit is very close to the predicted cross sections [9] (see Fig. 2).

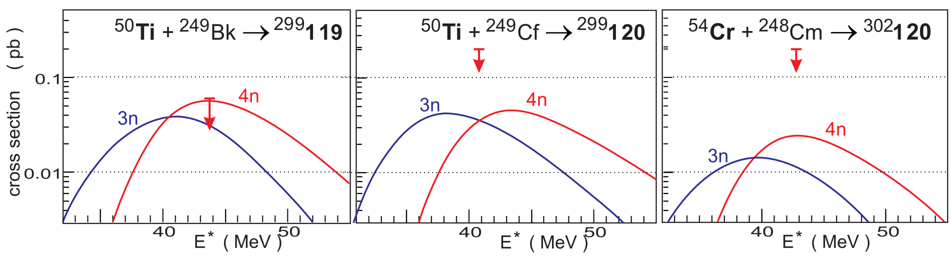

Fig. 2. Predicted cross sections for the production of new elements 119 and 120 in the Ti and Cr induced fusion reactions [9]. The arrows indicate the upper limits reached in the corresponding experiments performed at GSI [10, 11].

Synthesis of these nuclei may encounter also another important problem. The proton rich isotopes of SH elements produced in these reactions are rather short-living due to large values of Q_α. Their half-lives are very close to the critical value of one microsecond needed for the CN to pass through the separator up to the focal plane detector. The next elements (with $Z > 120$) being synthesized in such a way might be already beyond this natural time limit for their detection (see the right panel of Fig. 1).

2. Fusion reactions

There are only three methods for the production of transuranium elements, namely, fusion of heavy nuclei, multinucleon transfer reactions, and a sequence of neutron capture and β^- decay processes. Low values of the fusion cross sections and very short half-lives of nuclei with $Z > 120$ put obstacles in synthesis of new elements. At the same time, the extension of the area of known isotopes of SH elements is extremely important for better understanding of their properties and for developing the models which will be able to predict well the properties of SH nuclei located beyond this area (including those at the island of stability). An important region of SH isotopes located between those produced in the cold and hot fusion reactions remains unstudied yet (see the gap in the left panel of Fig. 1).

We found that this gap could be filled in ordinary fusion reactions of ^{48}Ca with available lighter isotopes of Pu, Am, and Cm. Several (rather cheap and available) isotopes of actinide elements can be used as the targets, for example, ^{233,235}U, ^{239,240}Pu, ^{241}Am, ^{243}Cm, and so on. It is much easier to fill the gap "from above" by the synthesis of new isotopes of SH elements with larger values of Z, their subsequent α decay chains just fill the gap. This unexpected finding is simply explained by greater values of survival probabilities of the corresponding nuclei with $Z = 114 \div 116$ as compared to those with $Z = 110 \div 112$. In the left panel of Fig. 3, the values of $B_{\rm f} - B_n$ are shown for the SH mass area, where $B_{\rm f}$ is the fission barrier and B_n is the neutron separation energy (an odd–even effect is smoothed here). As can be seen, the values of $B_{\rm f} - B_n$ are much higher for CN with $Z \sim 116$ as compared with CN of 112 element formed in fusion reactions of ^{48}Ca with neutron deficient isotope of uranium. As a result, the corresponding survival probability of lighter CN is smaller by more than one order of magnitude.

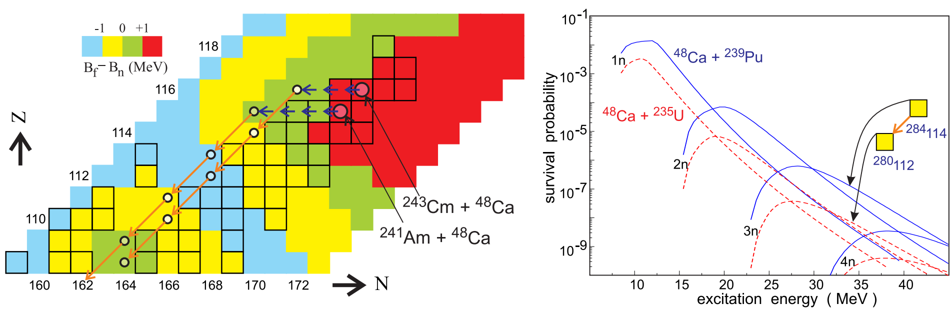

Fig. 3. The values of $B_{\rm f} - B_n$ as a function of proton and neutron numbers. Known isotopes of SH elements are marked by the bordered rectangles. The right panel shows the survival probability of CN ^{283}112 and ^{287}114 formed in the fusion reactions ^{48}Ca+^{235}U (dashed curves) and ^{48}Ca+^{239}Pu (solid curves).

The right panel of Fig. 3 shows survival probabilities of two CN, ^{283}112 and ^{287}114, formed in the fusion reactions ^{48}Ca+^{235}U and ^{48}Ca+^{239}Pu. The excitation energies of both CN (at collision energies equal to the corresponding Bass barriers, 195 and 198 MeV, correspondingly) are just the same for two reactions (they are about 30 MeV). In spite of the decrease of the fusion probability with increasing charge number of the target nucleus, we may conclude that the evaporation residue (EvR) cross sections for the ^{48}Ca+^{239}Pu reaction should be higher (by about one order of magnitude for the $3n$ evaporation channel) due to the larger survival probability of ^{287}114 compound nucleus as compared to $^{283}112$. Numerical calculations fully confirm this conclusion. This means that the new isotopes of element 112 (at least, $^{280,279}112$) could be easier synthesized and studied as α decay products of the heavier elements, 114 and/or 116.

In Fig. 4 the calculated EvR cross sections are shown for the production of new isotopes of elements 114 and 116 in the ^{48}Ca+^{239}Pu, ^{48}Ca+^{243}Cm and ^{40}Ar+^{251}Cf fusion reactions. High intensive beam of ^{40}Ar can be obtained quite easily. This material is also much cheaper than ^{48}Ca. However, as can be seen from Fig. 4, the use of an ^{40}Ar beam is less favorable as compared with ^{48}Ca. This is due to much "hotter" character of the ^{40}Ar+^{251}Cf fusion reaction (only the cross sections for the $5n$ evaporation channels are comparable for both reactions). More than ten new isotopes of even elements from $Z = 104$ to 116 could be produced in the ^{48}Ca+^{239}Pu and/or ^{48}Ca+^{243}Cm fusion reactions which just fill the gap in the superheavy mass area. Note that the production cross sections are high enough to perform such experiments at available facilities. All the decay chains, most probably, reach finally known nuclei (see Ref. [12]). This fact significantly facilitates the identification of the new SH isotopes.

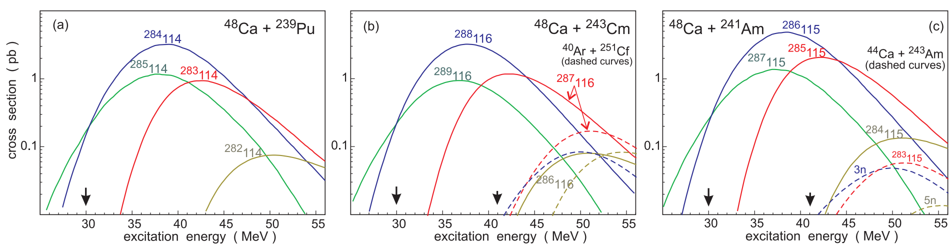

Fig. 4. Production cross sections for the new isotopes of elements 114 (a), 116 (b), and 115 (c) in the ^{48}Ca+^{239}Pu, ^{48}Ca+^{243}Cm, ^{40}Ar+^{251}Cf (dashed curves), ^{48}Ca+^{241}Am and ^{44}Ca+^{243}Am (dashed curves) fusion reactions. The arrows show positions of the corresponding Bass barriers.

The ^{48}Ca+^{241}Am fusion reaction is the best for the production of the new isotopes of odd SH elements filling the gap. The production cross sections for the new isotopes $^{284-286}115$ in this reaction are about 0.1 pb, 2 pb and 4 pb, respectively, *i.e.* high enough to be measured. The corresponding excitation functions are shown in Fig. 4. The more neutron deficient isotopes of element 115 could be produced in the ^{44}Ca+^{243}Am fusion reaction (note that ^{44}Ca is a more abundant and available material as compared to ^{48}Ca). However, in this reaction the excitation energy of the formed CN is 10 MeV higher than in the ^{48}Ca+^{241}Am fusion reaction. As a result, the corresponding excitation functions (see the dashed curves in Fig. 4 (c)) are shifted to higher energies at which the survival probability of the CN is much lower. Thus, the ^{48}Ca beam remains preferable also for the production of neutron deficient SH nuclei in fusion reactions with lighter isotopes of actinide targets as compared to the use of $^{42-44}$Ca or ^{40}Ar beams.

Recently, the synthesis of SH elements at the level of 1 pb became more or less a routine matter for several laboratories. The corresponding experiments require about 2-week irradiation time to detect several decay chains of SH element. This means that many new isotopes of SH elements could be synthesized now, and the gap between nuclei produced in the cold and hot fusion reactions could be closed at last.

It is well known that there are no combinations of available projectiles and targets, the fusion of which may lead to SH nuclei located at the island of stability. Only the proton-rich isotopes of SH elements have been produced so far in fusion reactions (see Fig. 1). The use of radioactive ion beams cannot solve this problem because of their low intensity. Two new neutron rich isotopes of elements 116 (^{294,295}116) may be synthesized in $3n$ and $4n$ evaporation channels of the ^{48}Ca+^{250}Cm fusion reactions with the cross sections of about 1 pb [12]. α-decay chains of these nuclei lead to absolutely new neutron enriched isotopes of SH elements ended by fission of seaborgium and/or rutherfordium isotopes located already at the beta-stability line.

Another interesting feature of the fusion reaction ^{48}Ca+^{250}Cm is an unexpected possibility to reach the middle of the island of stability just in ordinary fusion processes of "stable" nuclei. In this reaction, new neutron enriched isotopes ^{291}114 and ^{287}112 are formed as α-decay products of $3n$-evaporation residue of the corresponding CN. These isotopes should have rather long half-lives and, thus, they could be located already in the "red" area of the nuclear map, *i.e.*, they might be β^+-decaying nuclei. In Fig. 5 several possible decay chains are shown along with the corresponding values of Q_α and half-lives calculated with the use of nuclear masses predicted by Sobiczewski *et al.* [13] and by Möller *et al.* [14].

In accordance with our calculations of decay properties of SH nuclei, the isotopes ^{291}114 and ^{287}112 may experience not only α decay but also electron capture with half-life of several seconds. If it is correct, the narrow pathway to the middle of the island of stability is surprisingly opened by production of these isotopes in subsequent α decay of element 116 produced in the ^{48}Ca+^{250}Cm fusion reactions, see Fig. 5. The corresponding cross section is rather high, it is about 0.8 pb [12]. For the moment, this is the only method which is proposed for the production of SH nuclei located just in the middle of the island of stability. Further careful study of the decay properties of unknown SH nuclei located closer to the beta-stability line is needed to confirm the existence of such a possibility.

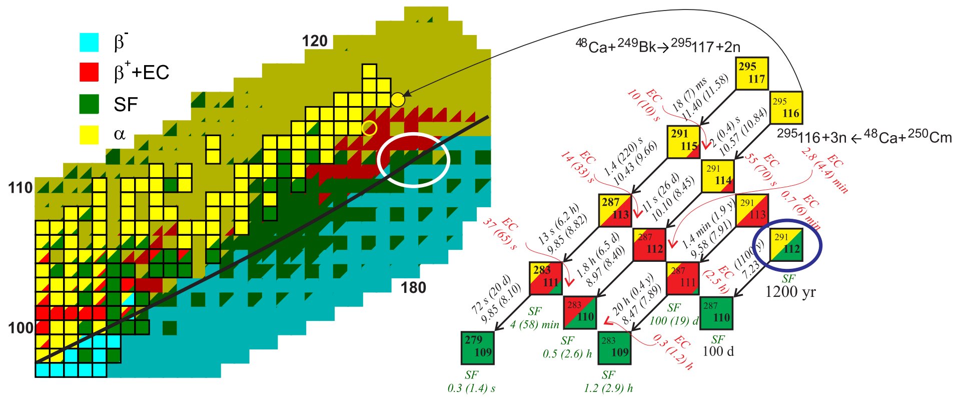

Fig. 5. The pathway to the middle of the island of stability via a possible β^+ decay of the isotopes ${}^{291}115$ and ${}^{291}114$. The first isotope may be formed after α decay of ${}^{295}117$ ($2n$ channel of the ^{48}Ca+^{249}Bk fusion reaction, cross section is 0.3 pb [9]). The second one, ${}^{291}114$, is formed after α decay of ${}^{295}116$ in the $3n$ evaporation channel of the ^{48}Ca+^{250}Cm fusion reaction with cross section of about 0.8 pb.

3. Transfer reactions

The multinucleon transfer processes in near barrier collisions of heavy ions, in principle, allow one to produce heavy neutron rich nuclei including those located at the island of stability. These reactions were studied extensively about thirty years ago. Among other topics, there had been great interest in the use of heavy-ion transfer reactions to produce new nuclear species in the transactinide region [15–20]. The cross sections were found to decrease very rapidly with increasing atomic number of surviving heavy fragments. However, several Fm and Md isotopes have been produced at the level of 0.1 μb [18].

Renewed interest in the multinucleon transfer reactions with heavy ions is caused by the limitations of other reaction mechanisms for the production of new neutron rich heavy and SH nuclei. Multinucleon transfer processes in near barrier collisions of heavy (and very heavy, U-like) ions seem to be the only reaction mechanism (besides the multiple neutron capture process [21]) allowing us to produce and explore neutron rich heavy nuclei including those located at the SH island of stability.

Appropriate description of proton and neutron transfers in damped collisions of heavy ions meets several fundamental and technical difficulties. Calculations performed within the microscopic time-dependent Schrödinger equation [22] clearly demonstrated that, at low collision energies of heavy ions, nucleons do not “suddenly jump” from one nucleus to another. Instead, the wave functions of valence nucleons occupy two-center molecular states, spreading gradually over volumes of both nuclei. The same adiabatic low-energy collision dynamics of heavy ions was found also within the time-dependent Hartree–Fock (TDHF) calculations [23]. This means that the perturbation models based on a calculation of the sudden overlapping of single-particle wave functions of transferred nucleons (in donor and acceptor nuclei, respectively) cannot be used for description of multinucleon transfer and quasi-fission processes in low-energy damped collisions of heavy ions. Indeed, the two-center shell model and the adiabatic potential energy look most appropriate for the quantitative description of such reactions.

We use the model based on the Langevin-type dynamical equations of motion which was proposed recently [24, 25] for simultaneous description of strongly coupled multinucleon transfer, quasi-fission, and fusion–fission reaction channels (which are difficult to distinguish experimentally in many cases). The diabatic potential energy is calculated within the double-folding procedure at the initial reaction stage, whereas in the adiabatic reaction stage we use the extended version of the two-center shell model [26].

Within this model, we found that the shell effects (clearly visible in fission and quasi-fission processes) also play a noticeable role in near barrier multinucleon transfer reactions [27, 28]. These effects may significantly enhance the yield of searched-for neutron rich heavy nuclei for appropriate projectile–target combinations. In particular, the predicted process of anti-symmetrising ("inverse") quasi-fission may significantly enhance the yields of long-living neutron rich SH isotopes in collisions of actinide nuclei (such as U+Cm). However, the role of the shell effects in damped collisions of heavy nuclei is still not absolutely clear and was not carefully studied experimentally. Very optimistic experimental results were obtained recently [29] confirming such effects in the surrogate ^{160}Gd+^{186}W reaction, for which the similar "inverse quasi-fission" process (^{160}Gd$\rightarrow^{138}$Ba while ^{186}W$\rightarrow^{208}$Pb) was also predicted [28].

In multinucleon transfer reactions, the yields of heavier-than-target (trans-target) nuclei strongly depend on the reaction combination. The cross sections for the production of *neutron rich* transfermium isotopes in reactions with ^{248}Cm target change sharply if one changes from medium mass projectiles to the uranium beam. Even for rather heavy projectiles (such as ^{136}Xe) the nuclear system has a dominating symmetrizing trend of formation of reaction fragments with intermediate (heavier than projectile and lighter than target) masses (see Fig. 6).

Of course, the yield of survived SH elements produced in the low-energy collisions of actinide nuclei is rather low, though the shell effects give us a definite gain as compared to a monotonous exponential decrease of the cross sections with increasing number of transferred nucleons. In Fig. 7, the calculated cross sections for the production of primary and survived

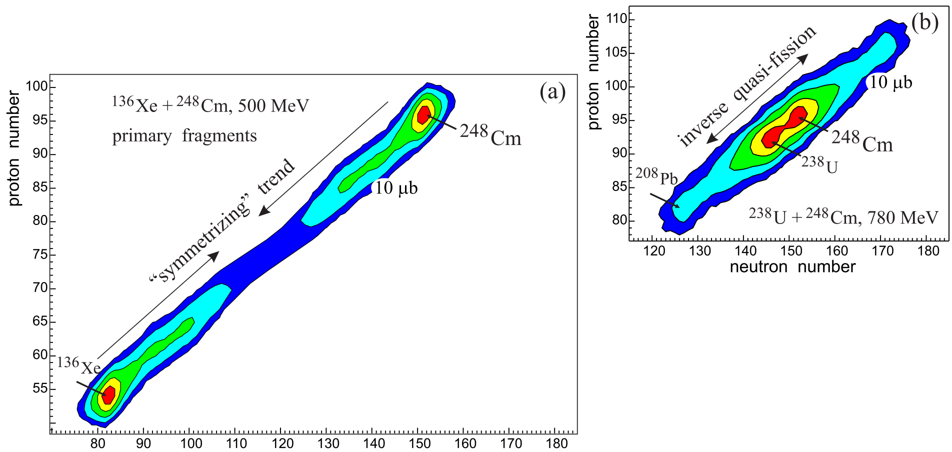

Fig. 6. Landscapes of the calculated cross sections for the production of primary reaction fragments in collisions of ^{136}Xe (a) and ^{238}U (b) with ^{248}Cm target (contour lines are drawn over one order of magnitude).

(evaporation residue) SH nuclei in damped collisions of ^{238}U with ^{248}Cm at 770 MeV center-of-mass energy are shown along with available experimental data. As can be seen, really many new neutron-rich isotopes of SH nuclei with $Z \geq 100$ might be produced in such reactions.

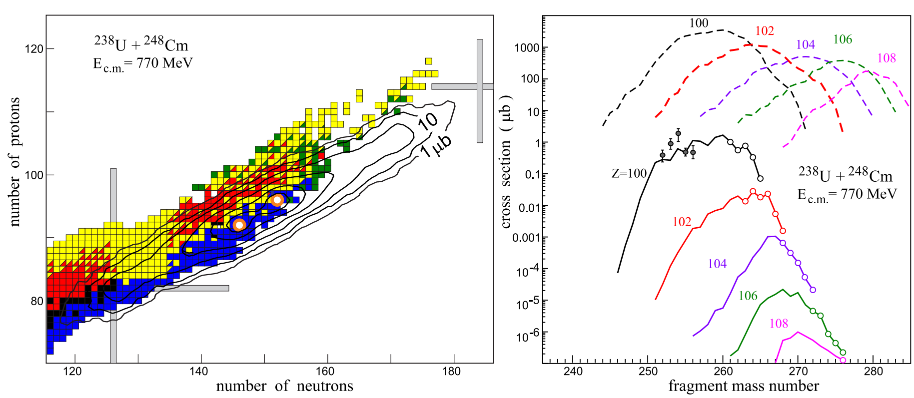

Fig. 7. Cross sections for the production of primary (left panel) and survived (right panel) transfermium nuclei in collisions of ^{238}U with ^{248}Cm target at $E_{\rm cm} = 770$ MeV. Open circles indicate new isotopes of transfermium elements. The dashed curves indicate the cross sections for the production of primary fragments. Experimental data are taken from [18] for the production of fermium isotopes in this reaction at beam energy $E_{\rm cm} = 862$ MeV before entering the thick ^{248}Cm target.

The choice of collision energy is very important for the production of desired neutron-rich SH nuclei. With increasing beam energy the yield of primary fragments increases. However, the excitation energy of these fragments also increases and thus decreases their survival probabilities. We found that the optimal beam energy for the production of neutron-rich isotopes of SH elements in multinucleon transfer reactions with heavy actinide nuclei (such as U+Cm) is very close to the energy needed for these nuclei to reach the contact configuration (there is no ordinary barrier: the potential energy of these nuclei is everywhere repulsive). For ^{238}U+^{248}Cm, it is about 770 MeV center-of-mass collision energy.

Actinide beams (as well as actinide targets) might be successfully used also for the production of new neutron-rich heavy nuclei around the closed neutron shell $N = 126$, the region having the largest impact on the astrophysical r-process. Near-barrier collisions of ^{136}Xe and ^{192}Os with a ^{208}Pb target were predicted to be quite promising for the production of new neutron rich nuclei with $N \sim 126$ [27, 30]. However, low-energy collisions of stable neutron rich isotopes of elements located below lead (such as ^{192}Os or ^{198}Pt) with available actinide targets look even more favorable.

Distribution of primary fragments formed in transfer reactions is concentrated around the line connecting projectile and target (just due to conservation of proton and neutron numbers). If one reaction partner has a neutron excess (such as ^{238}U), then this line will be inclined to the neutron axis. Distribution of primary fragments in (Z, N) plane is shown in the left panel of Fig. 8 for the case of transfer reaction products formed in low en-

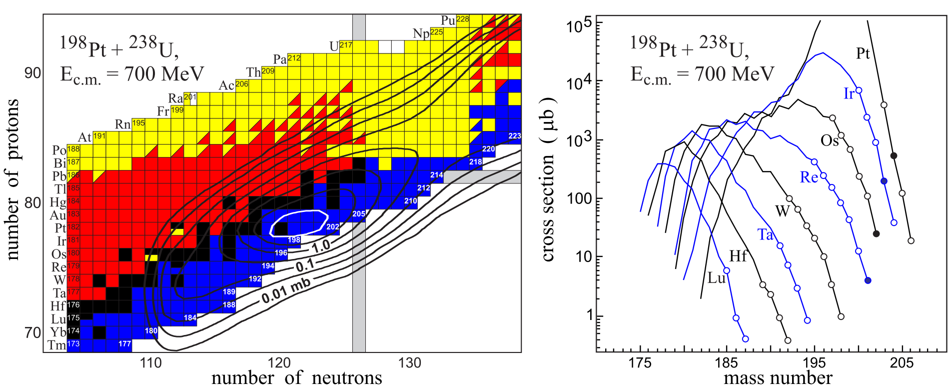

Fig. 8. Contour plot of the cross sections for the formation of *primary* reaction fragments in collisions of ^{198}Pt with ^{238}U at $E_{\rm cm} = 700$ MeV. Contour lines are drawn over half order of magnitude and units of measurement are millibarns (numbers on lines). Right panel: Isotopic yields of elements below lead (from Lu to Pt) in collisions of ^{198}Pt with ^{238}U at $E_{\rm cm} = 700$ MeV. Circles denote not-yet-known isotopes (the solid ones show isotopes with closed neutron shell $N = 126$).

ergy collisions of ^{198}Pt with ^{238}U at $E_{\rm cm} = 700$ MeV. As can be seen, a lot of new isotopes in the region of the closed neutron shell $N = 126$ can be synthesized in this reaction.

Estimated cross sections for the production of the final (survived) isotopes of heavy elements with $Z = 71 \div 78$ in low energy collisions of ^{198}Pt with ^{238}U are shown in the right panel of Fig. 8. On average, the cross sections for the production of new neutron rich heavy nuclei (including those located along the closed neutron shell $N = 126$) in this reaction are higher than in collisions of ^{136}Xe or ^{192}Os with ^{208}Pb target [30].

4. Summary

First, we hope that new SH elements 119 and 120 will be successfully synthesized within one or two nearest years with the use of Ti and/or Cr beams. Synthesis of SH elements with $Z > 120$ is rather problematic in near future due to extremely low cross sections and short half-lives of these elements. One might think that the epoch of ^{48}Ca in the production of SH nuclei was finished by the synthesis of element 118 in the ^{48}Ca+^{249}Cf fusion reaction [3]. However this projectile still could be successfully used for the production of new isotopes of SH elements. The extension of the area of known isotopes of SH elements is extremely important for better understanding of their properties and for developing the models which will be able to predict well the properties of SH nuclei located beyond this area (including those at the island of stability). We found that the ordinary fusion reactions could be used for the production of new isotopes of SH elements. The gap of unknown SH nuclei, located between the isotopes which were produced earlier in the cold and hot fusion reactions, could be filled in fusion reactions of ^{48}Ca with available lighter isotopes of Pu, Am, and Cm.

Then we must redirect our interests onto the production of longer living neutron enriched SH nuclei. New neutron-enriched isotopes of SH elements might be produced with the use of ^{48}Ca beam if a ^{250}Cm target would be prepared. In this case, we get a real chance to reach also the island of stability due to a possible β^+ decay of ^{291}114 and ^{287}112 nuclei formed in this reaction. The same path to the island of stability is opened also in the $2n$ evaporation channel of the ^{48}Ca+^{249}Bk fusion reaction leading to the isotope ^{291}115 having a chance for β^+ decay.

Low-energy multinucleon transfer reactions look quite appropriate for the production of new neutron-enriched heavy nuclei. Reactions with actinide beams and targets are of special interest for synthesis of new neutron-enriched transfermium nuclei and not-yet-known nuclei with the closed neutron shell $N = 126$ having the largest impact on the astrophysical r-process. However, it is rather difficult to perform these experiments because of the low beam intensities of the massive projectiles and problems with separating and detecting the heavy reaction products. The available experimental data on the production of heavy nuclei in low-energy multinucleon transfer reactions are still insufficient and fragmentary to make accurate predictions. Urgently needed are new experiments, including those in which the role of shell effects in reaction dynamics can be clarified much better.

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