Superheavy elements created at the Joint Institute for Nuclear Research

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Abstract. This study is concerned with the search for the entrance channels suitable to synthesize of superheavy elements. The differences between "cold" and "warm" fusion reactions successfully used for superheavy element productions are discussed. A special attention is made on the quasifission process which competes with fusion process in the reactions with heavy ions and becomes dominant in the reactions leading to the formation of superheavy nuclei.

1. Introduction
The existence of the superheavy nuclei, as well as their decay properties, are mainly determined by fission barrier $B_f$. According to the Liquid Drop Model (LDM) the limit of existence of nuclei is located immediately after $Z \approx 100$ [1], but the presence of shell effects can lead to a considerable increase of stability of nuclei with respect to various decay modes: $\alpha$ decay or spontaneous fission. The island of stability in the region of nuclei with $Z = 114$ and $N = 184$ predicted theoretically [2] has induced an extensive experimental investigation in the field of superheavy element (SHE) synthesis. At present the SHE are synthesized in the complete fusion reactions of two heavy nuclei. More than 30 SHEs with $Z = 108 - 118$ have been synthesized in the reactions of "cold" fusion (reactions with $^{208}\text{Pb}$ or $^{209}\text{Bi}$ targets) and "warm" fusion (reactions with actinide targets).

In the "cold" fusion reactions the excitation energy of compound nucleus (CN) is 10-20 MeV near the reaction threshold. The excitation energy of a formed CN is about 30-40 MeV in "warm" fusion reactions and de-excitation of CN to the ground state is due to the emission of three or four neutrons and several $\gamma$-rays. Nevertheless in the "warm" fusion reactions the formed CN is more neutron rich and nearer to the closed neutron shell at $N = 184$ than in the case of "cold" fusion reactions. Consequently, the "warm" fusion reactions are more preferable for synthesis of the SHE. A big success was achieved in the reactions of actinides with double magic $^{48}\text{Ca}$ ions at FLNR [3]. The production cross sections of SHE in these reactions virtually do not change with increasing atomic number of CN and maintain the level of a few picobarn.

Due to the low production rate for SHE the experiments on synthesis of new elements lasted at least several months. The study of fusion-fission process also allows to obtain information about the fusion probability. Note that the most part of SHEs are strongly fissile nuclei and the counting rate for fusion-fission process is sufficiently larger. In heavy ion-induced reactions the CN formation cross section is suppressed strongly by competing quasifission process (QF) [4, 5]. Therefore, the measurement of binary reaction channel gives the information about the
competition between fusion-fission and QF processes and allows to estimate the probability of the CN formation.

2. Influence of entrance channel on the competition between fusion-fission and quasifission

The contribution of QF increases with increasing entrance channel mass asymmetry. As an example, to demonstrate this dependence the mass-energy distributions of binary fragments obtained in the reactions $^{22}$Ne+$^{249}$Cf, $^{26}$Mg+$^{248}$Cm, $^{36}$S+$^{238}$U and $^{58}$Fe+$^{208}$Pb leading to the formation of Hs* ($Z=108$) [6] are presented in Fig. 1.

![Mass-energy distributions of binary reaction fragments for the reactions $^{22}$Ne+$^{249}$Cf, $^{26}$Mg+$^{248}$Cm, $^{36}$S+$^{238}$U and $^{58}$Fe+$^{208}$Pb leading to the formation of elements with $Z=108$ at energies above (top panel) and below (bottom panel) the Coulomb barrier.](image)

**Figure 1.** Mass-energy distributions of binary reaction fragments for the reactions $^{22}$Ne+$^{249}$Cf, $^{26}$Mg+$^{248}$Cm, $^{36}$S+$^{238}$U, $^{58}$Fe+$^{208}$Pb leading to the formation of elements with $Z=108$ at energies above (top panel) and below (bottom panel) the Coulomb barrier.

In the chosen reactions the parameters of mass-asymmetry $\alpha$ and the Coulomb factor $Z_1Z_2$ in the entrance channel vary strongly: for the reaction $^{58}$Fe+$^{208}$Pb: $\alpha = 0.571$ and $Z_1Z_2 = 980$, for $^{36}$S+$^{238}$U: $\alpha = 0.737$ and $Z_1Z_2 = 1152$, for $^{26}$Mg+$^{248}$Cm: $\alpha = 0.810$ and $Z_1Z_2 = 1472$, and for $^{22}$Ne+$^{249}$Cf: $\alpha = 0.838$ and $Z_1Z_2 = 2132$. As demonstrated in Fig. 1 the TKE-mass distributions change with decreasing asymmetry in the entrance channel from symmetric for incoming $^{22}$Ne-ions to strongly asymmetric for incoming $^{58}$Fe-ions. These changes are understood as reflecting the relative contributions of CN-fission and QF to the fission process of excited Hs* isotopes as depending on the reaction studied.

It is clearly seen that even at similar CN excitation energies the TKE-mass distributions are vastly different for these reactions. In the case of the reactions $^{22}$Ne+$^{249}$Cf and $^{26}$Mg+$^{248}$Cm the mass distributions have a near Gaussian shape with no evidence for asymmetric fission. The reactions are considered to be mainly attributed to the CN-fission process. For the $^{36}$S+$^{238}$U reaction the mass distributions of the fission-like fragments change markedly. This difference in mass distribution for the $^{26}$Mg+$^{248}$Cm and $^{36}$S+$^{238}$U reactions is connected with an increasing contribution of the QF process for the $^{36}$S-induced reaction. At low excitation energies QF is the dominant process for the reaction $^{36}$S+$^{238}$U. At higher excitation energies the mass distribution
becomes symmetric and similar to the reaction $^{26}$Mg$+^{248}$Cm though - due to a remaining trace of QF - slightly wider.

The TKE-mass distribution for the $^{58}$Fe$+^{208}$Pb reaction has a wide two-humped shape even at excitation energy of 48 MeV (well above the Bass barrier). For this reaction the QF process dominates at energies below and above the Bass barrier. At the last decade the large efforts have been done in the investigation of suppressing of fusion process by QF for different "ion-target" combinations \cite{4, 5, 6, 7, 8, 9, 10, 11}. It was found that the balance between fusion and QF strongly depends on entrance channel properties, such as mass asymmetry, deformation of interacting nuclei, collision energy, and the Coulomb factor $Z_1Z_2$. The presence of QF has to be taken into account for experiments devoted to the production of SHEs in order to choose the optimal reaction and collision energy.

3. "Cold" and "warm" fusion reactions

![Figure 2](image1.png)

**Figure 2.** Mass-energy distributions of fragments obtained in the "cold" fusion reactions $^{208}$Pb$+^{50}$Ti, $^{58}$Fe, $^{86}$Kr, $^{136}$Xe at energies close to the Bass barrier.

![Figure 3](image2.png)

**Figure 3.** Mass-energy distributions of fragments obtained in the "warm" fusion reactions $^{48}$Ca$+^{232}$Th, $^{238}$U, $^{244}$Pu, $^{248}$Cm at energies close to the Bass barrier.

Coming back to "cold" and "warm" fusion reactions one should note that the entrance channel is strongly different. For SHEs with $Z \geq 108$ at the former reaction type the values of the Coulomb factor $Z_1Z_2 \geq 2000$, while at the latter reaction type the Coulomb factor $Z_1Z_2 = 1960$ even in the case of reaction $^{48}$Ca$+^{249}$Cf leading to the SHE with $Z = 118$. In Figs. 2 and 3 the
mass energy distributions of binary fragments obtained in the reactions of "cold" and "warm" fusion are shown.

It is clearly seen that in the "cold" fusion reactions at the transition from $^{50}$Ti to $^{86}$Kr the shape of mass-energy distributions of binary reaction fragments changes strongly due to the QF process. For the $^{50}$Ti ion the contribution of symmetric fragments into the capture cross-section is around 60%; in the case of $^{86}$Kr the QF and deep-inelastic scattering are dominant processes. For the heaviest system $^{136}$Xe+$^{208}$Pb the main process is deep-inelastic scattering [14]. For the reactions with $^{48}$Ca-ions ("warm" fusion) the mass-energy distributions are similar for all the reactions: the clearly pronounced asymmetric QF with heavy fragments near the double magic lead is observed, the contribution of symmetric fragments into the capture cross section is about 10%.

Today the properties of fission of nuclei up to element with $Z = 106$ are well studied. It is known that at low excitation energy and spontaneous fission the spherical nuclear shells with $Z = 50$ and $N = 82$ and deformed neutron shell at $N = 88$ play a dominant role in the formation of fission fragments. The fission of nuclei with excitation larger than 40 MeV (when shell effects practically disappear) is well described by the LDM and characterized by symmetric mass distribution with the Gaussian shape with parameters depending on the temperature at saddle (or scission) point. Therefore the most probable kinetic energy of the fission fragments depends on the parameter $Z^2/A_{CN}^{1/3}$ and can be estimated by the Viola systematics based on experimental data [15]. Therefore as the first approximation the symmetric fragment region with mass $A_{CN}/2 \pm 20$ u may be attributed to CN-fission. The red contours in Figs. 2 and 3 show the regions of masses and TKE’s where the main part of CN fission fragments is expected.

However, this estimation corresponds to the upper limit for fusion-fission process due to the population of the mass region $A_{CN}/2 \pm 20$ u by both the CN-fission and QF fragments. The QF is characterized by large nucleon exchange and energy dissipation as well as CN-fission process and separation between CN-fission and QF fragments is a complicated problem, especially for symmetric mass split. A realistic description of deep inelastic scattering, QF and CN-fission processes in low energy heavy ion collisions shows [12, 13] that the potential energy surface for these systems is strongly modulated by shell effects and leads to the appearance of deep valleys corresponding to the formation of well bound magic nuclei. In accordance with these calculations, at least three paths leading to the formation of fission-like fragments can be distinguished: asymmetric QF caused by the influence of proton shells with $Z = 28, 82$ and neutron shells with $N = 50, 126$; symmetric QF determined by the shells with $Z = 50$ and $N = 82$; fusion-fission leading to the formation of symmetric fragments.

It is important to note that in the case of "warm" fusion reactions all target nuclei are well-deformed nuclei. In the reactions with deformed nuclei the potential energy surface strongly depends on the relative orientation of the reaction partners, which changes the Coulomb barrier and the distance between the centers of the colliding nuclei. When two interacting nuclei touch each other by their lateral surfaces (near-side collisions), a high probability of formation of a spherical CN is expected, whereas in the elongated configuration, when nuclei touch each other by their poles (near-tip collision), a high QF probability is expected. One should note that all the events observed in the production of isotopes of elements 112, 114, 116, 117 and 118 with the fusion reactions $^{238}$U, $^{242, 244}$Pu, $^{248}$Cm, $^{249}$Bk, $^{249}$Cf, $^{48}$Ca were detected at energies above the Coulomb barrier [16, 17, 18]. In contrast, in "cold" fusion reactions of the target nuclei of $^{208}$Pb and $^{209}$Bi with more massive projectiles, the maximum yield of evaporation residues is observed at subbarrier projectile energies [19, 20].

The maximal cross sections for evaporation residues obtained in the "cold" and "warm" fusion reactions are presented in Fig. 4. The evaporation residue cross section drops dramatically with increasing atom number of CN in the case of "cold" fusion reaction. In the "warm" fusion reactions their cross sections virtually do not change. This behavior may be explained by the
Figure 4. Maximal cross sections of 1n- evaporation channel in the "cold" fusion reactions of \(^{208}\)Pb and \(^{209}\)Bi target nuclei with different projectiles (indicated in the figure) and cross sections of 3n and 4n channels in the "warm" fusion reactions of \(^{48}\)Ca as a function of compound nucleus atomic number. The lines are drawn as guide the eye. Data were taken from [3].

fact that the contribution of QF increases strongly with increasing atom number of CN in the case of "cold" fusion reaction. For the "warm" fusion reaction the contribution of QF process nearly the same due to small difference in the reaction entrance channel. While the survival probability against fission is higher in the "cold" fusion reactions due to lower excitation energy of formed CN, the evaporation residues are some 10-15 u shifted from \(\beta\) stability line, that leads to a considerable decrease in their half-lives. The isotopes of SHE formed in the \(^{48}\)Ca induced reactions cannot reach the neutron closed shell with \(N = 184\) due to the lack of 7-9 neutrons, but nevertheless the additional neutrons increase strongly the half-lives of these nuclei up to several seconds.

The analysis of the available experimental data on the fusion and fission of nuclei of \(^{286}\)Cn, \(^{292}\)Fl, and \(^{296}\)Lv, produced in the reactions \(^{48}\)Ca+\(^{238}\)U, \(^{48}\)Ca+\(^{244}\)Pu, and \(^{48}\)Ca+\(^{248}\)Cm, as well as experimental data on the survival probability of those nuclei in evaporation channels of three- and four-neutron emission, enables us to reach the quite reliable conclusion that the fission barriers of those nuclei are really quite high [21], which results in their relatively high stability. The lower limits that we obtained for the fission barriers of nuclei of \(^{283-286}\)112, \(^{288-292}\)114, and \(^{292-296}\)116 are 5.5, 6.7, and 6.4 MeV, respectively.

4. Alternative ways for SHE

While the relative contribution of QF to the capture cross section mainly depends on the reaction entrance channel properties, the features of asymmetric QF are determined essentially by the driving potential of composite system. Generally, in heavy-ion-induced reactions the formation of QF fragments is connected with the strong influence of the nuclear shell at \(Z = 82\) and \(N = 126\) (doubly magic lead) and \(Z = 28\) and \(N = 50\) (double magic nickel). The experimental mass distribution of the fission-like fragments formed in the reaction \(^{48}\)Ca+\(^{248}\)Cm at energy close to the Coulomb barrier is presented in the top panel of Fig. 5. The symmetric fragment mass distribution (the open circles in Fig. 5) has been extracted from the experimental mass distribution of the all fission-like fragments using the Gaussian fitting procedure of asymmetric
peaks. The driving potential for this composite system at scission point calculated in the frame of the proximity model is also shown in the same figure. As it was mentioned above the maximum fragment yields correspond to the positions of the local minima of the driving potential as in the case of formation of asymmetric fragments, as well as symmetric ones.

![Driving potentials for different reactions](image)

**Figure 5.** The driving potentials as a function of mass at scission point for the reactions $^{48}\text{Ca}+^{248}\text{Cm}$, $^{238}\text{U}+^{248}\text{Cm}$, and $^{136}\text{Xe}+^{248}\text{Cm}$.

In the middle and bottom panels of Fig. 5 the driving potentials for the composite systems formed in the reactions $^{238}\text{U}+^{248}\text{Cm}$ and $^{136}\text{Xe}+^{248}\text{Cm}$ calculated in the same approximation...
are presented, respectively. The dashed arrows indicate the position of the entrance channels, while the solid arrows show the position of the proton and neutron closed shells. It is clearly seen that the local minima in the driving potential exist for all reactions, though in the latest two reactions these minima are located from the outside the entrance channel. Thus, we may expect an increase of the fragment yields in the mass region around these minima. W. Greiner and V. Zagrebaev in [22] proposed to call this process inverse QF. Notice, that in the case of the reaction $^{238}\text{U}+^{248}\text{Cm}$ one of the minima corresponds to doubly magic lead valley and the complementary fragment is a SHE around $Z=106$. In the reaction $^{136}\text{Xe}+^{248}\text{Cm}$ the both fragments have closed shells: the light fragment is near $N=50$, the heavy one is close to $Z=114$ and $N=184$ (predicted "island of the stability").

The idea of the production of SHE in the multi-neutron transfer reactions in the collision of U+U nuclei (or similar reactions) was already proposed in [23]. In this work it was found that at an incident energy of 7.42 MeV/u (about 22% above the Coulomb barrier) a direct search for $\alpha$-decay or fission of SHE being produced in a deep inelastic collision resulted in an upper cross section limit of 2nb. Although the stronger penetration of nuclei leads to enhanced mass transfer, the higher excitation energies involved drastically reduce the survival probability of the nuclei produced. The decrease in collision energy to the Coulomb barrier energy leads to the lower total excitation and consequently to larger cross section of survived SHE. According to the calculation of the cross section of survived SHE formed in the reaction $^{232}\text{Th}+^{250}\text{Cf}$ at 800 MeV center-of-mass energy (near the Coulomb barrier) from [22] there is a real chance for production of the long-lived neutron-rich SHE in such reactions.

Even though the "warm" fusion reaction leads to the formation of more neutron-rich nuclei than in the case of cold fusion even after the de-excitation process, the isotopes of SHE formed in these $^{48}\text{Ca}$ induced reactions cannot reach the neutron closed shells with $N=184$ due to the lack of 7-9 neutrons. From the investigation of the mass-energy distributions of binary reaction fragments obtained in the reactions $^{48}\text{Ca}+^{238}\text{U}$, $^{58}\text{Fe}+^{244}\text{Pu}$ and $^{64}\text{Ni}+^{238}\text{U}$ [9] it was found that the cross section drops three order of magnitude for the formation of the compound nucleus with $Z=120$ obtained in the reaction $^{64}\text{Ni}+^{238}\text{U}$ compared to the formation of the compound nucleus with $Z=112$ obtained in the reaction $^{48}\text{Ca}+^{238}\text{U}$ at an excitation energy of the compound nucleus of about 45 MeV. This is unfortunately a limiting factor. Furthermore, the relative contribution of the CN-fission fromed in $^{64}\text{Ni}+^{238}\text{U}$ is much lower than in the case of $^{58}\text{Fe}+^{244}\text{Pu}$, leading to the formation of the same composite system.

Recently the experiments aimed at the synthesis of isotopes of element $Z=120$ have been performed using the $^{244}\text{Pu}$ ($^{58}\text{Fe}, xn)^{302-}\text{Xe}$120 reaction [24] and $^{238}\text{U}(^{64}\text{Ni}, xn)^{302-}\text{Xe}$120 reaction [25]. A cross section limit of 0.4 pb at $E^*=44.7$ MeV for the former reaction and 0.09 pb at $E^*=36.4$ MeV for the latter reaction were obtained. In the case of $^{48}\text{Ca}$ induced reactions the evaporation residue cross section for 3n, 4n channels is about a few picobarns even for the heaviest nucleus with $Z=118$.

A possible alternative pathway is represented by the "inverse" QF or deep-inelastic reactions in the collision of $^{136}\text{Xe}$, $^{232}\text{Th}$ and $^{238}\text{U}$ with actinide targets. According to the theoretical expectations the cross section for the survived nuclei formed in such processes is higher than in the reaction of complete fusion. The reaction $^{136}\text{Xe}+^{248}\text{Cm}$ can be more interesting due to the fact that the heavy valley of the driving potential for this system corresponds to the nuclei close to the "island of stability" while in the case of the collision of Th and U with actinide targets the heavy valley lies outside the stability line. To estimate the formation probabilities of superheavy elements in these reactions the additional investigations are needed.

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