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Fusion-fission of Superheavy Nuclei

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The process of fusion-fission of superheavy nuclei with Z = 102-122 formed in the reactions with ²²Ne, ²⁶Mg, ⁴⁸Ca, ⁵⁸Fe, and ⁸⁶Kr ions at energies near and below the Coulomb barrier has been studied. The experiments were carried out at the U-400 accelerator of the Flerov Laboratory of Nuclear Reactions (JINR) using a time-of-flight spectrometer of fission fragments CORSET and a neutron multi-detector DEMON. As a result of the experiments, mass and energy distributions of fission fragments, fission and quasi-fission cross sections, multiplicities of neutrons and γ -quanta, and their dependence on the mechanism of formation and decay of compound superheavy systems have been studied.

1. Introduction

Interest in the study of the fission process of superheavy nuclei interactions with heavy ions is connected first of all with the possibility of obtaining information, the most important for the problem of synthesis, on the production cross section of compound nuclei at excitation energies of $\approx 15-30$ MeV (i.e. when the influence of shell effects on the fusion and characteristics of the decay of the composite system is considerable), which makes possible prediction on its basis of the probability of their survival after evaporating 1, 2, or 3 neutrons, i.e. in "cold" or "warm" fusion reactions. However, for this problem to be solved, there is a need for a much more penetrating insight into the fission mechanism of superheavy nuclei and for a knowledge of such fission characteristics as the fission - quasi-fission cross section ratio in relation to the ion-target entrance channel mass asymmetry and excitation energy, the multiplicity of the pre- and postfission neutrons, the kinetic energy of the fragments and the peculiarities of the mass distributions of the fission and quasifission fragments, etc. Undoubtedly all these points are of great independent interest to nuclear fission physics.

In this connection this work presents the results of the experiments on the fission of superheavy nuclei in the reactions ${}^{48}\text{Ca} + {}^{208}\text{Pb} \rightarrow {}^{256}\text{No}, {}^{22}\text{Ne} + {}^{248}\text{Cm} \rightarrow {}^{270}\text{Sg}, {}^{26}\text{Mg} + {}^{248}\text{Cm} \rightarrow {}^{274}\text{Hs}, {}^{48}\text{Ca} + {}^{238}\text{U} \rightarrow {}^{286}\text{112}, {}^{48}\text{Ca} + {}^{244}\text{Pu} \rightarrow {}^{292}\text{114}, {}^{48}\text{Ca} + {}^{248}\text{Cm} \rightarrow {}^{296}\text{116}, {}^{58}\text{Fe} + {}^{208}\text{Pb} \rightarrow {}^{266}\text{Hs}, {}^{58}\text{Fe} + {}^{244}\text{Pu} \rightarrow {}^{302}\text{120}, {}^{58}\text{Fe} + {}^{248}\text{Cm} \rightarrow {}^{306}\text{122}, \text{and} {}^{86}\text{Kr} + {}^{208}\text{Pb} \rightarrow {}^{294}\text{118}$ carried out at FLNR JINR in the last years. The choice of the indicated reactions has undoubtedly been inspired by the results of the recent experiments on producing the nuclides ${}^{283}\text{112}, {}^{287}\text{114}, {}^{289}\text{114}, {}^{283}\text{116}$ at Dubna^{1,2} and ${}^{293}\text{118}$ at Berkeley³ in the same reactions.

2. Characteristics of Mass and Energy Distributions of Superheavy Element (SHE) Fission Fragments

Figure 1 shows the data on mass and total kinetic energy (TKE) distributions of fission fragments of ²⁵⁶102, ²⁸⁶112, ²⁹²114, and ²⁹⁶116 nuclei produced in the reactions with ⁴⁸Ca at one and the same excitation energy $E^* \approx 33$ MeV. The main peculiarity of the data is the sharp transition from the predominant compound nucleus fission in the case of ²⁵⁶102 to the quasi-

fission mechanism of decay in the case of the ²⁸⁶112 nucleus and more heavy nuclei. It is very important to note that despite a dominating contribution of the quasi-fission process in the case of nuclei with Z = 112-116, in the symmetric region of fission fragment masses ($A/2 \pm 20$) the process of fusion-fission of compound nuclei, in our opinion, prevails. It is demonstrated in the framings (see the right-hand panels of Figure 1) from



Figure 1. Two-dimensional TKE-Mass matrices (left-hand side panels) and mass yields (right-hand side panels) of fission fragments of ²⁵⁶No, ²⁸⁶112, ²⁹²114, and ²⁹⁶116 nuclei produced in the reactions with ⁴⁸Ca at the excitation energy $E^* \approx 33$ MeV.

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which it is also very well seen that the mass distribution of fission fragments of compound nuclei is asymmetric in shape with the light fission fragment mass = 132-134.

Location of the first moment of mass distribution in the region of $m \approx 132$ a.m.u. points to the decisive role of shell effects which is characteristic of heavy nuclear fission. Perhaps it is worthwhile to draw a phenomenological analogy between the heavy element fission and that of superheavy elements. Indeed, nuclear fission fragments belonging to the region of the spherical neutron shell N = 82 can be observed in the cases of all the synthesized SHE with a charge of higher than 110. It can be understood on the basis of the multi-modal fission concept. The fission fragments, belonging to the so-called first standard channel, make the light mass group in the case of superheavy nuclear fission, instead of the heavy mass group, as is the case with actinides. In the case of SHE, the number of nucleons in the heavy fission fragment may not be quite sufficient for the subsequent neutron shell N = 126 to manifest itself substantially. It looks as if the role of the valley (N = 82, Z = 50-52) is decisive in the formation of the structure of the potential barrier of heavy as well of superheavy nuclei. Sticking to such an assumption one can determine the reflected fission analogue of the ²⁹⁶116 nucleus. Simple calculations show that it should be ²⁴⁰U, the light fission fragment peak of which when reflected about the $m \approx 132-134$ a.m.u. yields the heavy fission fragments peak observed in the fission of the ²⁹⁶116 nucleus. Fortunately, there are detailed and reliable studies of the fission fragment properties made using the reaction ${}^{238}\text{U} + n$ in a wide neutron energies region. Direct comparison of the two data sets is presented in Figure 2. Note that in the case of uranium it was taken into account that we were dealing with the nuclei which undergo fission at different excitation energies due to a multi-chance character of the process. That is why from the whole bulk of the data, we chose a spectrum at $E_n = 50$ MeV characterized by the mean excitation energy of 30 MeV, according to theoretical prediction of the authors. As is seen from the figure, the mass spectra in the lapped region practically coincide within the limits of the errors. It is an indirect, but nonetheless weighty argument in favor of the above mentioned assumption stating that instant fission of the superheavy nucleus dominates over the rest channels of its decay. The rigidity q of the potential connected with the corresponding fission valley is therewith unknown. That is why any further consideration demands the use of new data. In the high temperature approximation, the variance of the fission fragment mass distribution is connected with the temperature Θ of the system in the scission point in the following manner: $\sigma_M^2 = \Theta/q$.

The temperature dependence was calculated up to $\Theta \sim 1$ MeV using numerous data on the fission of heavy nuclei from ²³³Th



Figure 2. Mass spectra of fission fragments of 238 U at the excitation energy $E^* \approx 30$ MeV (solid circles) and those of the superheavy nucleus 296 116 produced in the reaction 48 Ca + 248 Cm (open circles).

up to ²⁴⁰Pu. The dependences are shown in Figure 3. It is seen that rigidity of the generalized asymmetric valley in the case of actinides is approximately the same and the spread of the mass spectra widths, taken at the same temperatures in the scission point, does not exceed 10–20%. It reflects the fact that the fission mechanism for the actinides, which have low neutron excess above the shell N = 82, is unique and is determined exactly by that shell forming the valley structure of the fission barrier. This conclusion can definitely be spread to the class of superheavy nuclei which follows from Figure 3, demonstrating in fact ideal agreement between the mass spectra calculated for actinides and measured for the superheavy nucleus ²⁹⁶116 at $\Theta = 0.9$ MeV.

Figure 4 shows similar data for the reaction between ⁵⁸Fe projectiles and ²³²Th, ²⁴⁴Pu, and ²⁴⁸Cm targets, leading to the formation of the compound system ²⁹⁰116 and the heaviest compound systems ${}^{302}120$ and ${}^{306}122$ (where N = 182 - 184), i.e., to the formation of the spherical compound nucleus, which agrees well with theoretical predictions.⁴ As seen from Figure 4, in these cases we observe an even stronger manifestation of the asymmetric mass distributions of ³⁰⁶122 and ³⁰²120 fission fragments with the light fragment mass = 132. The corresponding structures are also well seen in the dependence of the TKE on the mass. Only for the reaction ${}^{58}\text{Fe} + {}^{232}\text{Th} \rightarrow {}^{290}116$ $(E^* = 53 \text{ MeV})$ the valley in the region of M = A/2 disappears in the mass distribution as well as in the average TKE distribution which is connected with a reducing of the influence of shell effects on these characteristics. Such a decrease in the role of shell effects with increasing the excitation energy is observed also in the induced fission of actinide nuclei.

Figure 5 shows mass and energy distributions of fission fragments for compound nuclei ²⁵⁶No, ²⁶⁶Hs, and ²⁹⁴118 formed in the interaction between a ²⁰⁸Pb target and ⁴⁸Ca, ⁵⁸Fe, and ⁸⁶Kr ions at the excitation energy of \approx 30 MeV. It is important to note that in the case of the reaction ⁸⁶Kr + ²⁰⁸Pb the ratio between the



Figure 3. Variances of mass spectra of the heavy nucleus fission fragments in a high temperature approximation.



Figure 4. Two-dimensional TKE-Mass matrixes, the mass yields, average TKE, and the variances σ_{TKE}^2 as a function of the mass of fission fragments of ²⁹⁰116, ³⁰²120, and ³⁰⁶122 produced in the reactions with ⁵⁸Fe ions.

fragment yields in the region of asymmetric masses and that in the region of masses A/2 exceeds by about 30 times a similar ratio for the reactions with ⁴⁸Ca and ⁵⁸Fe ions. It signifies to that



Figure 5. Two-dimensional TKE-Mass matrices and mass yields of fission fragments for the reactions ${}^{48}Ca + {}^{208}Pb$, ${}^{58}Fe + {}^{208}Pb$, ${}^{86}Kr + {}^{208}Pb$ at an excitation energy of 28–34 MeV.

fact that in the case of the reaction $^{86}\text{Kr} + ^{208}\text{Pb} \rightarrow ^{294}118$ in the region of symmetric fragment masses the mechanism of quasi-fission prevails.

In analyzing the data presented in Figures 1, 2, and 4 one can notice two main regularities in the characteristics of mass and energy distributions of fission fragments of superheavy compound nuclei:



Figure 6. The average masses of the light (A_L) and heavy (A_H) fission fragments as a function of the compound nucleus mass. The asterisks show the data of this work, and the solid circles the data from Reference 5.



Figure 7. The dependence of TKE on the Coulomb parameter $Z^2/A^{1/3}$.

(*i*) Figure 6 shows the dependence of the average light and heavy fragment masses on the compound nucleus mass. It is very well seen that in the case of superheavy nuclei the light spherical fragment with mass 132–134 plays a stabilizing role, in contrast to the region of actinide nuclei.

(*ii*) Figure 7 shows the TKE dependence on the Coulomb parameter $Z^2/A^{1/3}$, from which it follows that for the nuclei with Z > 100 the TKE value is much smaller in the case of fission as compared with the quasi-fission process.

3. Capture and Fusion-fission Cross Sections

Figure 8 shows the results of measurements of the capture cross section σ_c and the fusion-fission cross section σ_{ff} ($\sigma_{A/2\pm 20}$) for the studied reactions as a function of the initial excitation energy of the compound systems.

Comparing the data on the cross sections $\sigma_{A/2\pm20}$ at $E^* \approx 14-15$ MeV (cold fusion) for the reactions ${}^{58}\text{Fe} + {}^{208}\text{Pb}$ and ${}^{86}\text{Kr} + {}^{208}\text{Pb}$, one can obtain the following ratio: $\sigma_{A/2\pm20}(108)/\sigma_{A/2\pm20}(118) \ge 10^2$. In the case of the reactions from ${}^{48}\text{Ca} + {}^{238}\text{U}$ to ${}^{58}\text{Fe} + {}^{248}\text{Cm}$ at $E^* \approx 33$ MeV (warm fusion) the value of *Z* changes by the same 10 units as in the first case, and the ratio $\sigma_{A/2\pm20}(112)/\sigma_{A/2\pm20}(122)$ is $\approx 4-5$ which makes the use of asymmetric reactions for the synthesis of spherical superheavy nuclei quite promising.

Another interesting result is connected with the fact that the values of σ_{ff} for ²⁵⁶102 and ²⁶⁶108 at $E^* = 14-15$ MeV are quite close to each other, whereas the evaporation residue cross sections $\sigma_{xn}{}^6$ differ by almost three orders of magnitude $(\sigma_{ff}/\sigma_{xn})$ which is evidently caused by a change in the Γ_f/Γ_n value for the above mentioned nuclei. At the same time, for the ²⁹⁴118 nucleus formed in the reaction ${}^{86}\text{Kr} + {}^{208}\text{Pb}$, the compound nucleus formation cross section is decreasing at an excitation energy of 14 MeV by more than two orders of magnitude according to our estimations ($\sigma_{ff} \approx 500$ nb is the upper limit) as compared with σ_{ff} for ${}^{256}102$ and ${}^{268}108$ produced in the reactions ${}^{48}\text{Ca} + {}^{208}\text{Pb}$ and ${}^{58}\text{Fe} + {}^{208}\text{Pb}$ at the same excitation energy. But when using the value of ≈ 2.2 pb for the cross section $\sigma_{ev}(1n)$ from the work in Reference 3, one obtains the ratio $\sigma_{xn}/\sigma_{ff} \approx 4 \times 10^{-6}$ for

²⁹³118, whereas for ²⁶⁶108 the ratio is $\sigma_{xn}/\sigma_{ff} \approx 10^{-6}$.

In one of recent works⁷ it has been proposed that such unexpected increase in the survival probability for the ²⁹⁴118 nucleus is connected with the sinking of the Coulomb barrier below the level of the projectile's energy and, as a consequence, leads to an increase in the fusion cross section. However, our data do not confirm this assumption.

4. Neutron and γ-ray Multiplicities in the Fission of Superheavy Nuclei

Emission of neutrons and γ rays in correlation with fission fragments in the decay of superheavy compound systems at excitation energies of near or below the Coulomb barrier had not been properly studied before this publication. At the same time such investigations may be extremely useful for an additional identification of fusion-fission and quasi-fission processes and thus a more precise determination of the cross sections of the above mentioned processes in the total yield of fragments. On the other hand, the knowledge of the value of the fission fragment neutron multiplicity may be used in the identification of SHE in the experiments on their synthesis.

The results of such investigations are presented in Figure 9 for the reactions ${}^{48}\text{Ca} + {}^{244}\text{Pu} \rightarrow {}^{292}114$ and ${}^{48}\text{Ca} + {}^{248}\text{Cm} \rightarrow {}^{296}116$ at energies near the Coulomb barrier. As seen from the figures, in all the cases the total neutron multiplicity $\langle v_n^{tot} \rangle$ is considerably lower (by more than twice) for the region of fragment masses where the mechanism of quasi-fission predominates as compared with the region of fragment masses where, in our opinion, the process of fusion-fission prevails (in the symmetric region of fragment masses).

Another important peculiarity of the obtained data is the large values of $\langle v_n^{tot} \rangle \approx 9.2$ and 9.9 for the fission of $^{292}114$ and $^{296}116$ compound nuclei, respectively. As well as for $\langle v_n^{tot} \rangle$ noticeable differences have been observed in the values of γ -ray multiplicities for different mechanisms of superheavy compound nucleus decay.

5. Conclusion

As a result of the experiments carried out, for the first time the properties were studied of the fission of the compound nuclei ²⁵⁶No, ²⁷⁰Sg, ²⁶⁶Hs, ²⁷¹Hs, ²⁷⁴Hs, ²⁸⁶112, ²⁹²114, ²⁹⁶116, ²⁹⁴118, ³⁰²120, and ³⁰⁶122 produced in reactions with ions ²²Ne, ²⁶Mg, ⁴⁸Ca, ⁵⁸Fe, and ⁸⁶Kr at energies close to and below the Coulomb barrier.

On the basis of those data a number of novel important physics results were received:

(*i*) It was found, that the mass distribution of fission fragments for compound nuclei ²⁸⁶112, ²⁹²114, ²⁹⁶116, ³⁰²120, and ³⁰⁶122 is asymmetric one, whose nature, in contrast to the asymmetric fission of actinides, is determined by the shell structure of the light fragment with the average mass 132–134. It was established that TKE, neutron and γ -ray multiplicities for fission and quasi-fission of superheavy nuclei are significant different.

(*ii*) The dependence of the capture (σ_c) and fusion-fission (σ_{ff}) cross sections for nuclei ²⁵⁶No, ²⁶⁶Hs, ²⁷⁴Hs, ²⁸⁶112, ²⁹²114, ²⁹⁶116, ²⁹⁴118, and ³⁰⁶122 on the excitation energy in the range 15–60 MeV has been studied. It should be emphasized that the fusion-fission cross section for the compound nuclei produced in reaction with ⁴⁸Ca and ⁵⁸Fe ions at excitation energy of \approx 30 MeV depends only slightly on reaction partners, that is, as one goes from ²⁸⁶112 to ³⁰⁶122, the σ_{ff} changes no more than by the factor 4–5. This property seems to be of considerable importance in planning and carrying out experiments on the synthesis of superheavy nuclei with Z > 114 in reaction with ⁴⁸Ca and ⁵⁸Fe ions. In the case of the reaction ⁸⁶Kr + ²⁰⁸Pb, leading to the production of the composite system ²⁹⁴118, contrary to reactions with ⁴⁸Ca and ⁵⁸Fe, the contribution of quasi-fission is dominant in the region of the fragment masses close to A/2.



Figure 8. The capture cross section σ_c and the fusion-fission cross section σ_{ff} or $\sigma_{A/2\pm 20}$ for the measured reactions as a function of the excitation energy.



Figure 9. Two-dimensional TKE-Mass matrices (top panels) and the mass yields (the solid circles), neutron (the stars) and γ -ray multiplicities (the open circles) in dependence on the fission fragment mass (bottom panels) for the reactions ${}^{48}Ca + {}^{248}Cm \rightarrow {}^{296}116$ and ${}^{48}Ca + {}^{244}Pu \rightarrow {}^{292}114$.

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