Attempt to produce the isotopes of element 108 in the fusion reaction ^{136}Xe + ^{136}Xe

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A setup of the experiment on the production of the isotopes with $Z = 108$ in the fusion reaction ^{136}Xe + ^{136}Xe and the obtained results are presented. At the excitation energy $0 \leqslant E_x \leqslant 30$ MeV of the ^{272}Hs* compound nucleus the upper limit of the cross section for evaporation residues $\sigma_{(1-3)n} \leqslant 4$ pb has been measured. The experimental results together with the data from asymmetric reactions point to a strong limitation of the Hs compound nucleus formation with increasing Coulomb forces in the entrance channel of the reaction.

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I. INTRODUCTION

Studies of most heavy (superheavy) nuclides in fusion reactions are connected, as known, by two key problems, i.e., the probability of formation and survival of a compound nucleus as well as its nucleon composition which determines the probability of different types of decay of the evaporation residues (EVRs).

Considering different types of fusion reactions [1], we point to advantages of the compound nucleus formation of the shell structure of interacting nuclei. These advantages were clearly manifested in key reactions leading to the synthesis of the heaviest nuclei, i.e., cold fusion between magic ^{208}Pb and projectiles of type ^{50}Ti, ^{54}Cr, . . . , ^{70}Zn [2,3] as well as hot fusion reactions between actinides target nuclei and magic ^{48}Ca projectiles [4].

It is also known that the shell structure strongly affects the character of collective motion in the direction opposite to fusion, i.e., in the process of fission into two fragments. The charge, mass, and energy distributions of the fission fragments are directly connected with the structure of the heavy nucleus in the process of fragment formation prior to separation at the scission point. Now considering fusion and fission processes at the landscape of the heavy nucleus potential energy surface we can expect an increase in the formation cross section (possibly quite a substantial one) in the cases where the nucleon composition of interacting nuclei close to the fission fragment of the synthesized nucleus are used.

As a rule, it is a type of cold fusion reaction in which magic nuclei with closed proton and/or neutron shells are involved. To demonstrate it, we present here the results obtained for the light isotope ^{222}Th. Fission fragment asymmetric mass distributions of ^{222,224}Th* with maximal fragment yields in the vicinity of magic nuclei ^{86}Kr($N = 50$) and ^{136}Xe($N = 82$) obtained in [5,6] were the basis for our further studies of fusion of these nuclei: ^{86}Kr + ^{136}Xe → ^{222}Th* [7].

It was shown that as compared with other target-ion combinations leading to the formation of ^{220,222}Th* nuclei [8–10] in this case the evaporation products $^{222-x}$Th ($x = 1, 2, 3 \ldots .7$) were formed with the maximal cross section. The maximal yield of the ^{136}Xe(^{86}Kr, xn)$^{222-x}$Th reaction products was observed at the compound nucleus excitation energy $E_x \sim 20$ MeV ($x = 2, 3$) and a cross section of about 3×10^{-29}cm^2 [7].

It is likely that in the transition to heavier and more neutron-rich nuclei (actinide isotopes) there will be a similar picture but with different closed shells $Z = 50$, $N = 82$. As is known, the effect of these shells leads to the asymmetry in the fission fragment mass distribution of uranium and transuranium elements. Thus, for the synthesis of heavier Th isotopes, for example, one has to choose the reaction ^{132}Sn + ^{96}Zr → ^{228}Th and use a beam of radioactive magic nuclei ^{132}Sn($T_{1/2} = 39$ s). In the fission of heavier nuclei situated beyond actinides and even in the region of superheavy nuclei with $Z = 112$–118 the effect of closed shells $Z = 50$, $N = 82$ is manifested in the formation of the light fragment. As a result the heaviest nuclei fission fragment distribution also becomes asymmetric [11,12].

The fusion of ^{132}Sn with stable nuclei, i.e., isotopes of Yb, Hf, and W, would lead to the formation of magic nuclei $Z = 120$, 122, 124 with large neutron excess ($N = 184$ and larger). In these nuclei according to the predictions of various microscopic models the maximal shell effect is expected [13,14]. Note that by their nucleon composition nuclides cannot be produced by any other way. At the same time, the compound nucleus formation in the fusion of such heavy nuclear systems looks quite problematic due to strong Coulomb repulsion forces in the entrance reaction channel. It is also quite difficult to estimate how strongly the ^{132}Sn shell structure can contribute to the formation of the heavy compound nucleus.

From our point of view, more definite conclusions could be drawn from the results of a model experiment on the production known isotopes of element 108 using the fusion reaction of two identical nuclei ^{136}Xe + ^{136}Xe → ^{272}Hs* [1].

Indeed, as is shown in the experiment ^{26}Mg + ^{248}Cm [15], the compound nucleus ^{274}Hs* mainly undergoes symmetric fission. Symmetric fission of a lighter isotope ^{272}Hs* into two ^{136}Xe fragments with the closed neutron shell $N = 82$ seems to be most likely. The isotopes$^{269-271}$Hs formed in the $1n$-$3n$ evaporation channels, according to data from [16], should undergo α-decay with $E_\alpha \sim 9$ MeV and partial half-lives $T_{1/2} \sim 4$–20 s. Their daughter products, i.e., the isotopes of element 106, $^{265-267}$Sg, undergo α-decay or SF; however their grand-daughter products, i.e., $^{261-263}$Rf, mainly undergo

spontaneous fission. That is why registration of EVRs in the reaction ^{136}Xe(^{136}Xe, xn)$^{272-x}$Hs by their characteristic decay properties (by the correlated α-decays with energies of 8–9 MeV and a few seconds later spontaneous fission) practically in background free conditions seems to be quite reliable.

As compared with the above mentioned reaction ^{136}Xe(^{86}Kr, 1–3n)$^{221-219}$Th [7], in the symmetric reaction ^{136}Xe(^{136}Xe, 1–3n) $^{271-269}$Hs the Coulomb repulsion forces grow by almost 40% which should lead to a strong decrease in the fusion probability for ^{136}Xe nuclei and, as a consequence, to a strong decrease in the Hs isotope formation cross section. But, as follows below, experiments on the synthesis of $^{271-269}$Hs should be conducted with a higher sensitivity, up to the cross section level of $\sim 10^{-36}$cm^2, which is 10^7 times lower than measured cross sections for ^{219,220}Th isotopes. Will the probability of fusion of 2 ^{136}Xe nuclei suppress by 7 orders of magnitude as compared with ^{86}Kr + ^{136}Xe?

Here are some calculation data obtained recently for the reaction ^{136}Xe(^{136}Xe, xn)$^{272-x}$Hs. In Fig. 1, solid lines show the calculated excitation functions [17] using a model earlier used for the description of experimental cross sections of cold fusion reactions [18]. Here, at $E_x \approx$ 15–25 MeV, the cross sections reach several tens of pb, but strongly decrease for a higher excitation energy of the compound nucleus. Note that proceeding from a method for the description of cold fusion reactions from an earlier work [19] a cross section of 170 pb was predicted for the reaction ^{136}Xe(^{136}Xe, 1n)^{271}Hs. Contrary to that, calculations from [20] made within another approach despite their larger uncertainty predict maximal cross sections (several pb) at the higher energy $Ex \approx$ 40–50 MeV (see dotted curves in Fig. 1).

In reality, despite the theoretical difficulty, the setup of the experiment on the synthesis of element 108 isotopes in the reaction ^{136}Xe(^{136}Xe, xn)$^{272-x}$Hs has a simple motivation. In the case of the observation of Hs isotopes one can obtain a qualitative estimation of the hindrance for fusion; in the case of absence of the effect—its lower limit.

In both cases, we will give more realistic ideas on the feasibility of the fusion reactions and use of the unique ^{132}Sn beam for the synthesis of superheavy elements, in particular.

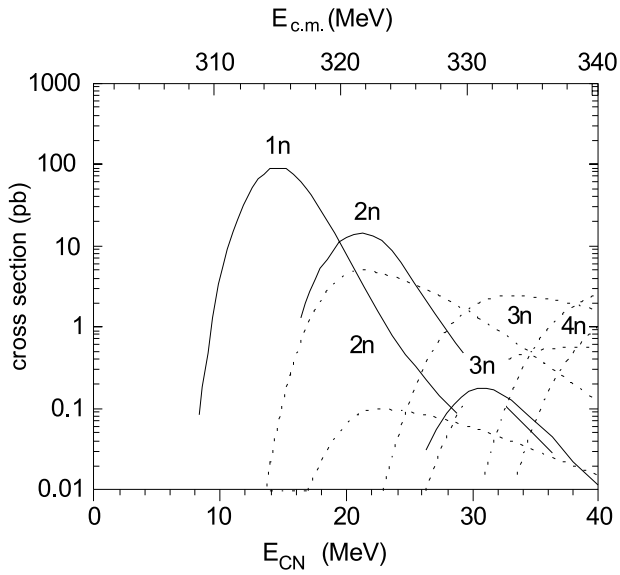

FIG. 1. The cross sections of xn-channels in the reaction ^{136}Xe + ^{136}Xe as a function of the excitation energy of the ^{272}Hs compound nucleus: solid lines—results of calculations within the diffusion model of the fusion process [18], the dotted line—data from work [20], with a variation of calculation parameters.

II. EXPERIMENTAL SETUP

The setup of the experiment on the separation of EVRs produced in symmetric reactions from a huge amount of by-product nuclei and further registration of their decay has some specific features. Due to the high energy of recoils ($E_R \approx$ 300 MeV) it is practically impossible to use kinematic separators. Indeed, the separators' type of velocity or energy selector requires extremely strong electric fields. In the case of gas-filled separator the setup loses its separation qualities due to comparable magnetic rigidities of recoil nuclei and the beam. That is why we chose a variant of chemical separation of the sought products and their fast delivery to detectors, similar to the setup previously used for studies of the chemical properties of element 108 [21]. The essence of the method is in stopping EVRs, i.e., isotopes of element 108, in a gas volume, in oxidation of Hs atoms into the HsO_4 compound with high volatility and their fast transport to remote detectors. The detectors are under the conditions of low temperature since according to experimental data from [22] HsO_4 molecules are effectively adsorbed at the surface at a temperature of $\leqslant -50^\circ$C. The scheme of the experiment is shown in Fig. 2.

The ^{136}Xe ion beam produced at the U-400 cyclotron (FLNR, JINR) passed through a target chamber filled with the ^{136}Xe isotope enriched to 98% at a pressure of 1 bar. The target thickness was 5 mm (2.95 mg/sm^2). An entrance 4 μm Ti-foil separated the target chamber from the accelerator's vacuum volume. The second "stop" chamber, 50 mm in depth, filled with a transport gas (mixture of Ar–70%, He–20%, O–10%) at a pressure of 1.2 bars was placed directly behind the target chamber. The chambers were separated from each other by 13 μm Be or 6 μm Al foils. The recoil nuclei–Hs isotopes–stopped in the second chamber were oxidized up to the volatile

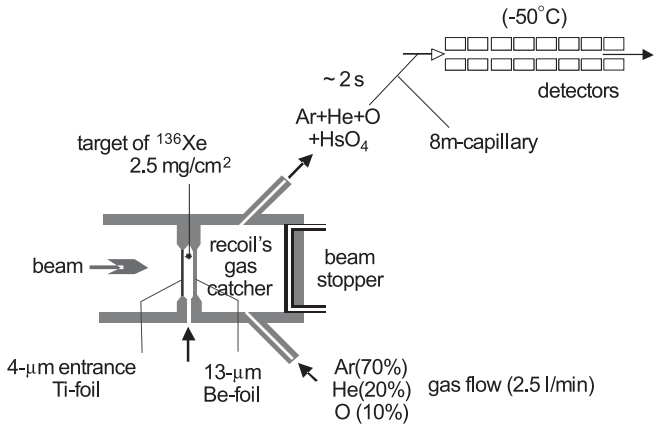

FIG. 2. Schematic view of the setup employed for the synthesis of element 108 isotopes in the fusion reaction ^{136}Xe + ^{136}Xe.

tetra-oxide chemical form HsO_4 and together with transport gas through a 8-m long Teflon capillary were delivered to the detectors. For additional purification of the volatile compounds from low-volatile and nonvolatile reaction products of a filter from quartz cotton at 700°C was placed at the entrance of capillary. At a gas flow of 1.5 l/min the average transport time of recoils from stopping chamber to the detectors was about 2 s.

For the registration of α-particles and spontaneous fission fragments in decay chains of Hs isotopes a detector array, 85 mm in length, was used. It consisted of eight pairs of semi-conductor detectors, each 1×1 cm^2 in area, situated in opposition to each other with a gap of 1 mm.

The temperature of detectors could be varied from room temperature to −65°C. The major part of the experiment at low temperature was carried out at −50°C. Volatile HsO_4 molecules in a gas flow through a 1 mm gap were diffused and adsorbed at the cold detector surface. According to the data from works [20,21] under these conditions the detectors could collect more than 50% of Hs recoil nuclei stopped in the second chamber.

For calibrating the detectors α-radioactive nuclides in the decay chain ^{219}Rn (4c)-$\alpha \rightarrow {}^{215}$Po (1.8 MC)-$\alpha \rightarrow {}^{211}$Pb were used. The isotope ^{227}Th deposited onto a plate from porous tantalum served as a source of ^{219}Rn nuclei. The plate was located in the stop chamber of recoil nuclei, ^{219}Rn atoms were transported by a gas flow to the detectors which registered in-flight their α-decay as well as second fast decay of the daughter ^{215}Po nuclei.

The fusion reaction $^{20}\mathrm{Ne} + {}^{156}\mathrm{Dy}$ which produced Os isotopes, a chemical homologue of Hs, was also used for calibrating the detectors and testing the work of the setup. Despite the fact that the effective adsorption of osmium tetra-oxide (OsO_4) at a cold surface occurs at temperatures lower than −50°C, a considerable amount of light α-radioactive isotopes of this element was observed in the experiment. As seen from the α-particle spectrum shown in Fig. 3, the energy resolution of the detectors is about 80 keV. Note that the detector registers α-particles in a large solid angle (from the surface of all nearby detectors) and from nuclei decaying in-flight.

And finally, in a long-run irradiation with the ^{136}Xe ion beam, the working regime of the setup was also controlled by the yield of Os isotopes produced in the other reaction $^{\mathrm{nat}}\mathrm{Ti} + {}^{136}\mathrm{Xe}$. The 4 μm Ti foil, serving at the same time as an entrance window of the gaseous ^{136}Xe target, was the target matter.

The initial energy of the ^{136}Xe ion beam from the accelerator was $E_L = 750$ MeV; it was determined by the TOF-detectors with an accuracy of ±3 MeV and was monitored nonstop during all experiments. After the energy loss in the entrance Ti foil, the beam energy in the working part of the ^{136}Xe target was 650–600 MeV which corresponded to the excitation energy range of ^{272}Hs* compound nuclei $0 \leqslant E_x \leqslant 30$ MeV. Under these conditions evaporation products with the energy $\varepsilon_R \approx 1.14$ MeV/nuclear were to stop approximately in the middle of the chamber filled with gaseous Ar/He/O.

The ^{136}Xe ion beam with an intensity of about 2×10^{12}/s was produced at the U-400 accelerator. However, in the long-run experiment $^{136}\mathrm{Xe} + {}^{136}\mathrm{Xe}$ the beam intensity was limited to a level of 3×10^{11}/s due to the radiation damages of the Ti entrance foil.

III. RESULTS AND DISCUSSION

In a 21 day irradiation of a ^{136}Xe target with the ^{136}Xe ion beam a total beam dose of $3.7 \times 10^{17}$was collected. Figure 4 shows a summary spectrum of α-particles registered by eight detectors at a temperature of −50°C during the whole experiment. Figure 5 separately shows the spectrum of α-particles registered during 3 h by eight detectors at a temperature of −50°C. The spectrum of α-particles registered within the same time by the same detectors at room temperature is also shown in this figure.

It is seen that the major effect is connected with the registration by cold detectors of α-particles from the decay of light Os isotopes in the energy interval $E_\alpha = 4.5$–5.5 MeV. Despite the fact that Os-isotopes produced in the reaction $^{\mathrm{nat}}\mathrm{Ti} + {}^{136}\mathrm{Xe}$ mostly undergoing electron capture

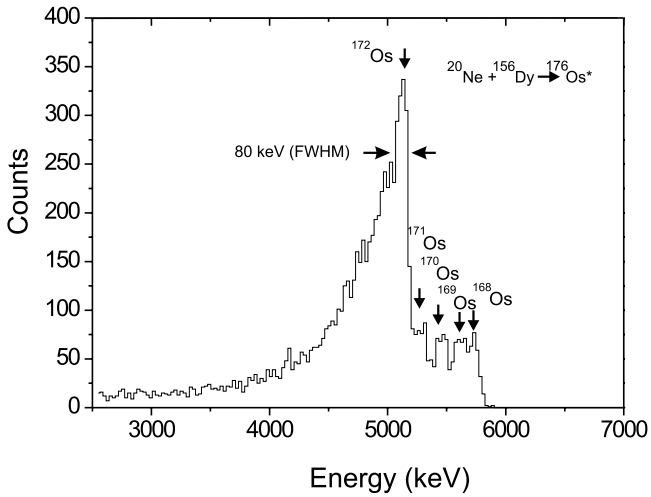

FIG. 3. Alpha-particle energy spectra from the decay of Os isotopes (shown in the figure) observed in the reaction $^{20}\mathrm{Ne} + {}^{156}\mathrm{Dy} \rightarrow {}^{176}\mathrm{Os}^*$ at the ^{20}Ne ion beam energy $E_L = 170$ MeV.

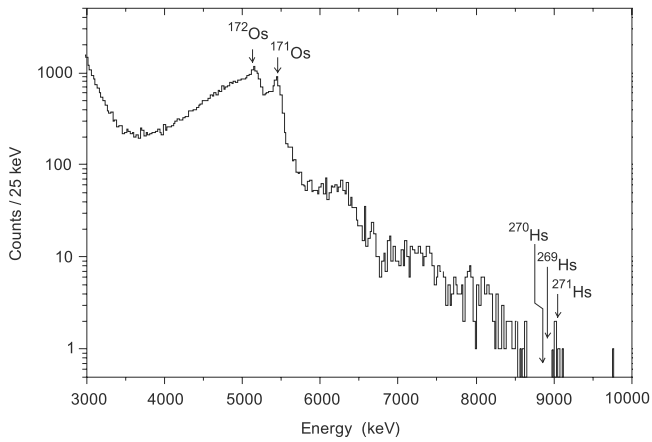

FIG. 4. Energy spectra of α-particles registered by eight detectors ($T = -50$°C) in the reaction $^{136}\mathrm{Xe} + {}^{136}\mathrm{Xe}$ during the whole experiment.

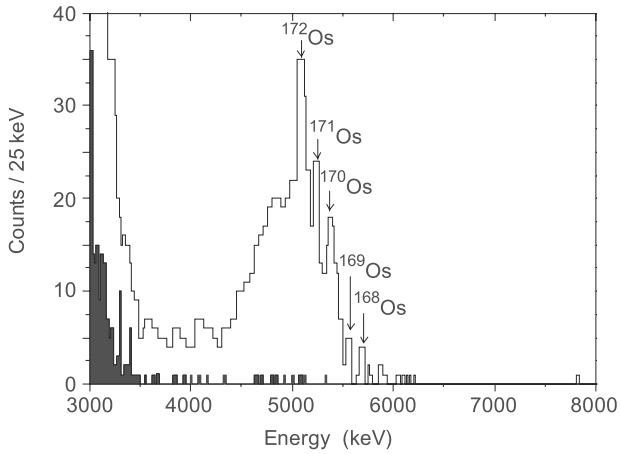

FIG. 5. The spectrum of α-particles from the reaction ^{136}Xe + ^{136}Xe registered during 3 h by eight detectors at a temperature of -50°C. The spectrum of α-particles (filled area) registered by the same detectors at room temperature is shown for comparison.

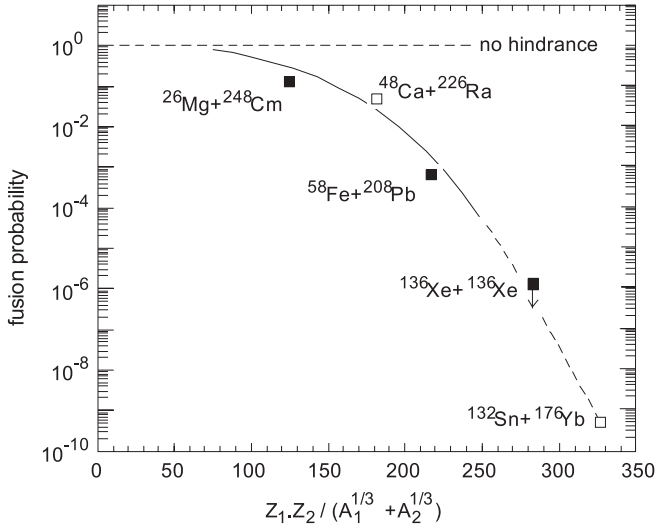

FIG. 6. Fusion probability leading to the formation of the compound nucleus $Z = 108$. Filled squares represent calculated values obtained from experimental evaporation residue cross sections of the reactions ^{26}Mg + ^{248}Cm [21], ^{58}Fe + ^{208}Pb [23], and ^{136}Xe + ^{136}Xe (this work). The open square shows predictions for the reaction ^{48}Ca + ^{226}Ra [24]. Explanations for the solid and dotted curves are in the text.

(EC) the detector also registers rare α and β^+ radioactive evaporation products. With the warm detectors the effect sharply decreases which is connected with the extremely low probability of the OsO_4 compound adsorption at room temperature.

In the range of higher energies $E_\alpha \geqslant 8.7$ MeV, where α-particles from the decay of $^{269-271}$Hs isotopes and their daughter products $^{265-267}$Sg are expected, a total of six signals were registered. Among them no correlations by energy and time of α-particles, which could be attributed to the decay of $^{269-271}$Hs isotopes, were detected.

Meanwhile the observation of one decay of any isotopes of element 108 with mass 269–271 produced via 1n, 2n, or 3n evaporation channels would correspond to a total cross section of 2.5 pb. The absence of this event determines the limit of the formation cross section for ^{272}Hs* evaporation products in the excitation energy range $E_x = 0$–30 MeV (see Fig. 1) at the level of 4 pb.

Actually it is the first experiment aimed at the study of the reaction ^{136}Xe + ^{136}Xe with high sensitivity, in which the value (or the limit) of the formation cross section of evaporation products is comparable or even noticably lower than the expected (calculated) values. Note that from a technical point of view the rotational target could accept a more intense beam and would increase the sensitivity of the experiment by another order of magnitude.

But even from the obtained limit it is already seen that in the symmetric reaction the Hs isotope formation cross section in 1–3 neutron evaporation channels is considerably lower (by an order or more) than that in the more asymmetric cold fusion reaction ^{208}Pb(^{58}Fe, n)^{265}Hs [23]. This difference, from our point of view, is directly connected with increased hindrance for fusion of two colliding ^{136}Xe nuclei, as compared with the asymmetric reaction ^{208}Pb + ^{58}Fe leading to the ^{266}Hs* compound nucleus.

An even stronger limitation of fusion follows from a comparison of EVRs cross sections of the ^{222}Th* and ^{272}Hs* compound nuclei produced in fusion reactions ^{86}Kr + ^{136}Xe and ^{136}Xe + ^{136}Xe, respectively. For these compound nuclei in the considered excitation energy range $E_{x=}$10–30 MeV the probability of survival does not differ greatly. That is why the difference in the evaporation product cross sections of about seven orders observed in the experiment is mainly determined by dynamic limitations for the compound nucleus formation.

Figure 6 shows the probability of nuclear fusion in different target-ion combinations leading to the formation of element 108 isotopes as a function of the Coulomb parameter $z = Z_1 \cdot Z_2/(A_1^{1/3} + A_2^{1/3})$. Black squares denote the values obtained from calculations using experimental cross sections of evaporation residues. Such calculations do not have pretensions of high accuracy. At the same time, the limitation for the formation of Hs isotopes are well fitted into experimental data from the other 28 reactions in a wide parameter range $84 \leqslant z \leqslant 245$, for which using a similar procedure the limitation for fusion were obtained [25]. They are shown in Fig. 6 by the averaged solid curve.

Our experimental results point to a further increase in the hindrance with increasing mass and charge of the interacting nuclei. In the case of the symmetric reaction ^{136}Xa + ^{136}Xa ($z = 283.5$) the limitation for the formation of ^{272}Hs* compound nucleus already amounts to six or more orders.

We may expect that with an increase in masses of the synthesized nuclei such limitation will continue to grow. Now, taking into account results from the reaction ^{136}Xa + ^{136}Xa, extrapolation into the region of superheavy nuclei, element 120 isotopes, in particular, can be performed more accurately (the dotted line in Fig. 6).

As is seen from the figure, due to a strong hindrance of more than 10^9 for the formation of the ^{308}120* compound nucleus, the expected cross section for neutron-rich superheavy nuclides in the reaction ^{132}Sn + ^{176}Yb ($z = 327.2$) will

be extremely small. It is very unlikely that such a strong suppression of fusion can be overcome by the high intensity of the ^{132}Sn radioactive beam due to limited technical feasibility. However, this circumstance does not decrease general interest in the production of the unique ^{132}Sn radioactive ion beam for the purposes of extensive studies in the region of more light nuclei.

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