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Isotopic trends in the production of superheavy nuclei in cold fusion reactions

G. G. Adamian,[1,2] N. V. Antonenko,[1] and W. Scheid[3]
[1]*Joint Institute for Nuclear Research, 141980 Dubna, Russia*
[2]*Institute of Nuclear Physics, 702132 Tashkent, Uzbekistan*
[3]*Institut für Theoretische Physik der Justus-Liebig-Universität, D-35392 Giessen, Germany*
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In the ^{208}Pb-based cold fusion-evaporation reactions the dependence of the yield of heaviest nuclei on the isotopic composition of the projectile nucleus is studied within the dinuclear system model for compound nucleus formation. Projectiles with a larger number of neutrons are not expected to increase always the production cross section of superheavy nuclei. New optimal reactions for the synthesis of superheavy nuclei with $Z=112$ and 114 are suggested.

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The study of the dependence of the evaporation residue cross section σ_{ER} on the isotopic composition of colliding nuclei is significantly important because the cross section for the synthesis of superheavy elements (SHE) continuously decreases with increasing charge number Z and the present experimental limit for the registration of heaviest elements has been reached in the recent experiments [1–4]. The production cross sections for $Z=111$ and 112 nuclei in the cold fusion-evaporation reactions with ^{208}Pb and ^{209}Bi targets [1,2], and for $Z=114$ and 116 nuclei in the ^{48}Ca-induced hot fusion-evaporation reactions with actinide targets [3] are of the order of picobarns. An important result in the synthesis of the element with $Z=110$ in cold fusion reactions was the enhancement of the cross section from 3.5 pb to 15 pb by changing the projectile from ^{62}Ni to ^{64}Ni [1,2]. This gave hope that the cross sections could decrease less steeply with more neutron-rich projectiles. But the later experiments ^{70}Zn+^{208}Pb,^{209}Bi showed that the production of elements $Z=112$ and 113 does not profit from the higher isospin value of the ^{70}Zn beam [1]. From the theoretical point of view, it was shown in Ref. [5] that in the Pb-based reactions the use of neutron-rich radioactive projectiles leads to a value of σ_{ER} comparable with evaporation residue cross sections with stable projectiles. The reason for this is the following. While the value of the survival probability of the compound nucleus increases, the value of the fusion probability can decrease with an increasing number of neutrons in the projectile. This conclusion is in agreement with the experimental observation that the probability of fusion of two heavy nuclei is diminished when the neutron numbers in the projectile or target deviate from the magic numbers or the nearest closed sub shells [6]. Therefore, the general opinion for providing a large neutron excess in the target as well as in the projectile nucleus in order to have a larger cross section of the production of the SHE with $Z>111$ should be reconsidered. Indeed, this opinion arises if only the survival probability of the compound nucleus against fission is considered as the crucial factor for the production of the SHE and the formation probability of the compound nucleus is disregarded.

The aim of the present paper is to investigate the evaporation residue cross sections σ_{ER} for the synthesis of SHE with $Z=110$, 112, and 114 in the Pb-based fusion reactions as functions of the isospin of the stable projectiles by means of the dinuclear system (DNS) model developed in Refs. [5,7–10]. In the DNS model, the compound nucleus is reached by a series of transfers of nucleons from the light nucleus to the heavy one. The dynamics of the DNS is considered as a combined diffusion in the degrees of freedom of the mass asymmetry $\eta=(A_1-A_2)/(A_1+A_2)$ (A_1 and A_2 are the mass numbers of the DNS nuclei) and of the internuclear distance R. The fusion barrier B^*_{fus} in η supplies a hindrance for fusion [5,7,8]. As a rule, B^*_{fus} for the initial DNS, $\eta=\eta_i$, decreases with increasing $|\eta_i|$. The basic assumptions of the DNS model, like the structure forbiddenness for the melting of the nuclei along R, were microscopically proved in Refs. [11]. The previous DNS model calculations of σ_{ER} for the cold and hot fusion reactions leading to heavy and superheavy nuclei are in good agreement with available experimental data [5,7,8]. The correct description of the mass (charge) distribution and kinetic energy of the products of quasifission, which accompanies the fusion process, is an additional justification of the DNS model [10].

The evaporation residue cross section can be factorized as follows [5]:

$$\sigma_{ER}(E_{\text{c.m.}}) = \sigma_c(E_{\text{c.m.}})P_{CN}(E_{\text{c.m.}})W_{sur}(E_{\text{c.m.}}). \qquad (1)$$

The calculations of the σ_{ER} demand an analysis of all three factors in Eq. (1). The value of $\sigma_c=(\lambda^2/4\pi)(J_{max}+1)^2T(E_{\text{c.m.}})$ is the effective capture cross section for the transition of the colliding nuclei over the entrance Coulomb barrier with the transmission probability $T=0.5$ [8]. The contributing angular momenta in the evaporation residue cross section are limited by the survival probability W_{sur} and $J_{max}\approx 10$, when highly fissile superheavy nuclei are produced at bombarding energies $E_{\text{c.m.}}$ near the Coulomb barrier [5,8,12,13].

Dissipative large-amplitude collective nuclear motion, which occurs in fusion, can be analyzed within the transport theory [14,15]. The probability of complete fusion P_{CN} in Eq. (1) depends on the competition between complete fusion in η and quasifission in R (decay of the DNS after the capture stage). For cold fusion reactions, the decay of DNS takes place mainly from the initial configuration in contrast to the case of hot fusion reactions [5,10]. The probability is given by [5]

$$P_{CN} = \lambda_\eta^{Kr}/(\lambda_R^{Kr} + \lambda_\eta^{Kr}). \quad (2)$$

We use a two-dimensional Kramers-type expression [5] for the quasistationary rates of fusion λ_η^{Kr} and quasifission λ_R^{Kr} depending on the fusion barrier ($B_{\rm fus}^*$) in η and quasifission barrier (B_{qf}) in R, respectively [5]. The local thermodynamic temperature Θ of the DNS is calculated with the expression $\Theta=\sqrt{E^*/a}$, where the level density parameter $a=A/12$ MeV^{-1} ($A=A_1+A_2$) and E^* is the excitation energy of the initial DNS. The reduced friction coefficient and the frequencies in the harmonic oscillators approximation of the potential in R and η are given in Refs. [5,8].

The fusion ($B_{\rm fus}^*$) and quasifission ($B_{\rm qf}$) barriers are given by the potential energy $U(R,\eta,J)=B_1+B_2+V(R,\eta,J)$ of the DNS which is calculated as the sum of binding energies B_i of the nuclei ($i=1,2$) and of the nucleus-nucleus potential $V(R,\eta,J)$ [5,7,8]. The isotopic composition of the nuclei forming the DNS is obtained with the condition of a N/Z equilibrium in the system. The variations of the potential in η are caused by both shell and odd-even effects included into the calculations through realistic binding energies B_i [16,17]. The potential $V(R,\eta,J)$ was calculated with the double-folding procedure with a nuclear radius parameter $r_0=1.15$ fm and a diffuseness $a_0=0.54-0.56$ fm depending on the mass number of the nuclei [5,7,8]. Due to the large moments of inertia of the massive DNS considered and due to the restricted set of angular momenta ($J\leqslant 10\hbar$), we set $U(R,\eta,J)\approx U(R,\eta)$ and $V(R,\eta,J)\approx V(R,\eta)$. For a given η, the DNS is localized in the minimum of the pocket of the nucleus-nucleus potential $V(R,\eta)$ [5,8]. The deformation effects are taken into account in the calculation of the potential energy surface [5,7,8]. For the heavy nuclei in the DNS, which are deformed in the ground state, the parameters of deformation are taken from Ref. [18]. As in Ref. [5] the light nuclei of the DNS are assumed to be deformed only if the energies of their 2^+ states are smaller than 1.5 MeV. For the incident energies considered, the relative orientation of the deformed nuclei in the DNS follows the minimum of the potential energy during the evolution in η.

The survival probability under the evaporation of 1 neutrons is considered according to Refs. [5,12,13] as

$$W_{sur}(E_{CN}^*) = P_{1n}(E_{CN}^*)\frac{\Gamma_n(E_{CN}^*)}{\Gamma_n(E_{CN}^*)+\Gamma_f(E_{CN}^*)}. \quad (3)$$

Here, P_{1n} is the probability of realization of the $1n$ channel at the excitation energy $E_{CN}^*=E_{\rm c.m.}+Q$ of the compound nucleus [12], Γ_n and Γ_f are the partial widths of neutron emission and fission [13], respectively. In the calculation of W_{sur} we used the microscopic corrections of Ref. [17] as fission barriers. The neutron binding energies B_n are also taken from Ref. [17]. In order to calculate $W_{sur}(E_{\rm c.m.},J=0)$ in Eq. (1), we used the following expression for Γ_n/Γ_f [15,19] in Eq. (3):

$$\frac{\Gamma_n(E_{CN}^*)}{\Gamma_f(E_{CN}^*)} = \frac{4A^{2/3}a(E_{CN}^*-B_n)}{k\{2[a(E_{CN}^*-B_n)]^{1/2}-1\}}\exp\{2a^{1/2}[(E_{CN}^*-B_n)^{1/2} - (E_{CN}^*-B_f)^{1/2}]\}, \quad (4)$$

where $k=9.8$ MeV and the ratio of the level density parameters in the fission and evaporation channels is equal to 1.0. Since the fission barrier B_f of the compound nucleus is defined by shell corrections, its value depends on the excitation energy E_{CN}^* as $B_f=B_f(E_{CN}^*=0)\exp[-E_{CN}^*/E_d]$, where $E_d=0.5A^{4/3}/a$ MeV is the shell-damping energy. For odd-even nuclei, the odd-even effects were taken into consideration by the following substitutions: $E_{CN}^*-B_f\to E_{CN}^* -[B_f(E_{CN}^*=0)+\delta]\exp[-E_{CN}^*/E_d]+\delta$ and $E_{CN}^*-B_n\to E_{CN}^*-B_n -\delta$ in the expression (6) with the pairing energy $\delta=11/A^{1/2}$ [12]. Here, the even-even nuclei are chosen as reference in energy. We take into consideration that the microscopic correction contains the pairing energy, but the fission barrier is related to shell correction.

Since the excitation energies of compound nucleus are about (10–15) MeV and (30–50) MeV for lead-based and actinide-based fusion reactions, respectively, the relative role of survival probability W_{sur} in the calculation of evaporation residue cross section σ_{ER} is smaller for cold fusion than for hot fusion. As a result of smaller mass asymmetry in the entrance channel, the fusion probabilities P_{CN} in the considered cold fusion reactions are between 10^{-9} and 10^{-5} and four to five orders of magnitude smaller than in the hot fusion reactions leading to superheavies with the same Z_0.

As was found in Ref. [5], in the Pb-based reactions with neutron-rich radioactive projectiles the increase of W_{sur} with neutron number is compensated by the decrease of P_{CN} and, thus, the isotopic dependence of σ_{ER} towards the increase of the number of neutrons is rather weak. Here, we show that there exist the isotopic compositions of colliding stable nuclei at which the product $P_{CN}W_{sur}$ gets larger by a slight decrease of the number of neutrons.

The calculated excitation functions $\sigma_{1n}(E_{CN}^*)$ are presented in Fig. 1 for the reactions ^{64}Ni, ^{67,68,70}Zn, ^{73,76}Ge $+^{208}$Pb. The agreement with available experimental data is quite good. The calculations for all reactions were performed with one set of parameters and with the same assumptions. The estimated inaccuracy of our calculations of σ_{ER} is within a factor of 2. However, it should be noted that the accuracy of relative values of cross sections is higher. The obtained σ_{1n} for the reactions ^{62,64}Ni, ^{68,70}Zn, ^{74,76}Ge$+^{208}$Pb are slightly different from the ones in Ref. [5] because in the present calculations the definition of the optimal excitation energy is more precise and for the calculation of E_d we used the factor 0.5 instead of 0.4 in Ref. [5] in order to have better agreement with the available latest experimental cold fusion data for the $Z=110$ and 112 elements.

The calculated maximal evaporation residue cross sections σ_{1n} and the corresponding optimal excitation energies of the compound nuclei in the $1n$ evaporation channel are presented in Figs. 2–4 for the reactions ^{A}Ni, ^{A}Zn, ^{A}Ge $+^{208}$Pb as functions of the mass number A of the projectile, respectively. The yield of the $Z=110$ element is larger in the ^{64}Ni$+^{208}$Pb reaction than in the $^{58-62}$Ni$+^{208}$Pb reactions due

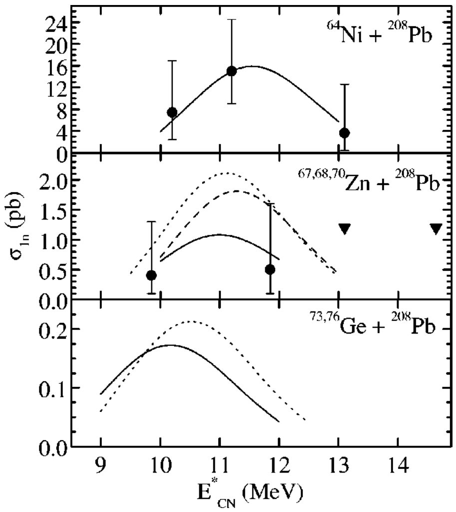

FIG. 1. The calculated excitation functions for the $1n$ channel of fusion reactions ^{64}Ni (solid line), ^{67,68,70}Zn (dotted, dashed, and solid lines, respectively), ^{73,76}Ge (dotted and solid lines, respectively) +^{208}Pb. The experimental data for the reactions ^{64}Ni, ^{70}Zn +^{208}Pb and upper limits for the reaction ^{68}Zn+^{208}Pb are shown by closed circles with error bars and triangles, respectively [2].

to the larger value of W_{sur}. The decrease of W_{sur} from ^{272}110 to ^{268}110 is not compensated by an increasing fusion probability P_{CN} and, therefore, σ_{1n} decreases. For the reactions with ^{58,59}Ni the increase of P_{CN} supplies a larger σ_{1n} than the ones in the reactions with ^{60,61}Ni. The cross sections in the reactions ^{64}Ni+^{207,208}Pb are comparable because the products $P_{CN}W_{sur}$ are practically the same in both cases.

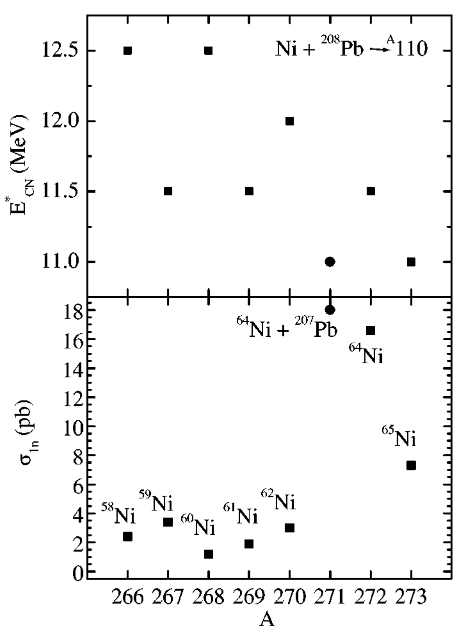

FIG. 2. The calculated maximal evaporation residue cross sections in the $1n$ channel (lower part) at the corresponding excitation energy of the compound nuclei (upper part) for the fusion reactions Ni+^{208}Pb$\rightarrow ^{A}$110 (closed squares) as functions of A. The projectiles are indicated. The results for the reaction ^{64}Ni+^{207}Pb$\rightarrow ^{271}$110 are shown by closed circle.

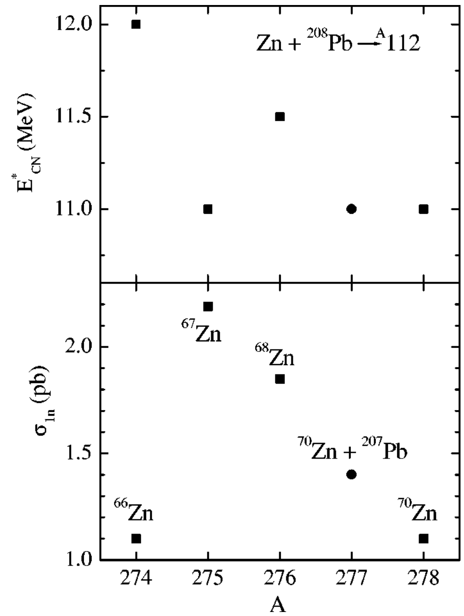

FIG. 3. The same as in Fig. 2, but for the fusion reactions Zn +^{208}Pb$\rightarrow ^{A}$112 and ^{68}Zn+^{207}Pb$\rightarrow ^{275}$112.

The calculations show that the production of the SHE with $Z=112$ and 114 in the cold fusion reactions ^{70}Zn,^{76}Ge +^{208}Pb does not profit from the higher isospin value of beams. One can expect quite large cross sections in the ^{208}Pb-based reactions with ^{67,68}Zn and ^{73}Ge projectiles. This effect is more pronounced for the Zn beam than for the Ge beam because the absolute value of the shell-correction energy and, respectively, the fission barrier, for isotopes of element 112 slightly increases with decreasing mass number from $A=278$ to $A=274$ due to a large level spacing at $N=162$ for deformed nuclei [17]. With the data of Ref. [17] the behavior of the shell-correction energy for the isotopes of

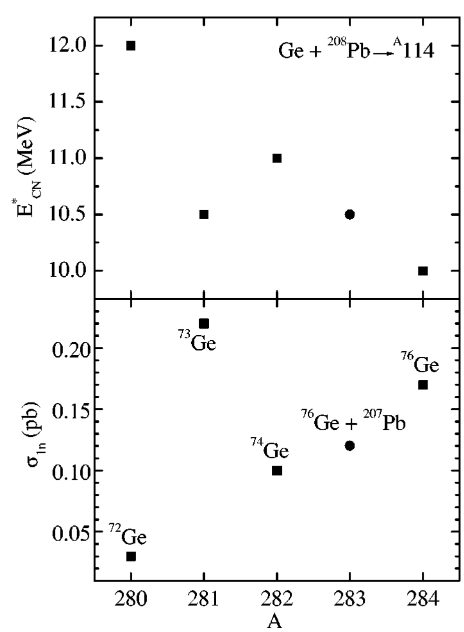

FIG. 4. The same as in Fig. 2, but for the fusion reactions Ge +^{208}Pb$\rightarrow ^{A}$114 and ^{76}Ge+^{207}Pb$\rightarrow ^{283}$114.

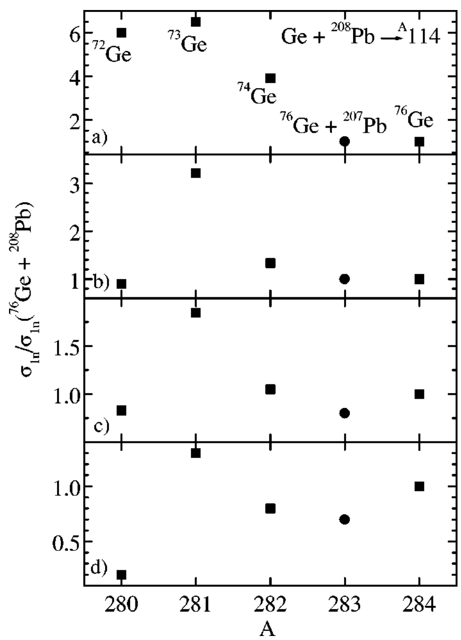

FIG. 5. The calculated ratios between the maximal production cross sections in the reactions Ge+Pb→^{A}114 and ^{76}Ge+^{208}Pb →^{284}114 as functions of A. The results obtained with data of Ref. [21], of finite range droplet model [20], of finite range liquid drop model [20], and of Ref. [17] are shown from upper part to lower part, respectively.

element 114 is opposite to element 112 due to the stronger effect of spherical shell closure at $N=184$ than of subshell closure at $N=162$. Within the considered intervals of A the value of P_{CN} becomes larger with decreasing A in the most cases. In spite of the odd-even effect in Eq. (4), the larger P_{1n} and larger fission barriers and smaller neutron binding energies lead to larger W_{sur} for odd nuclei like ^{275}112 and ^{281}114 in comparison to the neighboring even-even nuclei (see Figs. 3 and 4). This produces a higher cross section for the reaction with ^{67}Zn(^{73}Ge) beam than for the reactions with ^{66,68}Zn(^{74,72}Ge) beams in spite of their larger fusion probability at energies at the maximum of the 1n channel. The gain in fusion probability P_{CN} for the reaction ^{66}Zn+^{208}Pb(^{72}Ge +^{208}Pb) in comparison with the reaction ^{68}Zn+^{208}Pb(^{74}Ge +^{208}Pb) is smaller than the loss in the survival probability W_{sur}. A further decrease of A results in much smaller W_{sur} and, thus, smaller σ_{ER}.

Using other nuclear mass tables [20,21] for the calculations of P_{CN} and W_{sur}, we found for the production of element 114 (Fig. 5) that reactions with a smaller neutron excess are even more favorable than ones with the larger neutron excess. This effect is more pronounced with data of Ref. [21] because the calculated negative shell-correction energies slightly decrease with the mass number of element 114 like in the case of element 112 due to the shell closure at $N=162$. It should be noted that the experiments [22] have established the existence of deformed shell closures at $N=162$ and $Z=108$ predicted by the modern macroscopic-microscopic approach [23]. Using different mass tables and the same set of other parameters, we obtained practically the same cross sections for the elements 110 and 112. For the isotopes of element 114, the cross sections are more than ten times smaller with the data of Ref. [21], of the finite range liquid drop model [20] and finite range droplet model [20] than with the data of Ref. [17]. To obtain the same cross section $\sigma_{1n}\approx 0.2$ pb for element ^{283}114 as with the data of Ref. [17], we must take the level density parameter $a_n=A/14$ for the data of the finite range droplet model [20] and $a_n=A/21$ for the data of Ref. [21] and finite range liquid drop model [20]. The calculations in Fig. 5 were performed with these level density parameters. However, the ratio between the production cross sections of different isotopes is not sensitive to the change of a_n in all cases. Presently one cannot give a preference to any nuclear mass table because of lack of experimental information. Although, the dependence of the level density parameter for the superheavies is extrapolated from known systematics, it is the subject of a future study.

The increase of the production cross section with decreasing isospin in the case of element 112 was first suggested in Ref. [2]. There was the experimental attempt to prove this effect in the reaction ^{68}Zn+^{208}Pb [1,2]. The reason why the element 112 was not observed in the experiment at the cross section limit of 1.2 pb could be that the excitation function for the 1n channel is very narrow and the beam energy was not chosen in accordance with the maximum production cross section (Fig. 1). The lowest excitation energy of the compound nucleus in this experiment was about 1.5 MeV higher than the optimal one. We obtain $\sigma_{1n}=0.3$ pb at this bombarding energy with our model.

In conclusion, the probabilities of fusion and survival probabilities, ingredients of the evaporation residue cross sections, depend decisively on the neutron number of projectile. Certain ^{208}Pb-based cold fusion-evaporation reactions with smaller neutron excess are even more favorable than those with larger neutron excess for the production of the elements with $Z=112$ and 114. For the first time, the optimal excitation energies and the combinations of the colliding nuclei, such as ^{67,68}Zn, ^{73,74}Ge+^{208}Pb, are suggested for future experiments. The systematical experimental study of these reactions is needed to reveal the role of the subshell at $N=162$ for $Z>110$. Our results favor the use of ^{67,68}Zn and ^{73,74}Ge beams on ^{209}Bi target in the production of the 113 and 115 elements, respectively. Work on these reactions is in progress.

For hot fusion reactions, we found in Ref. [24] that the optimal reaction partners are the calcium ^{48}Ca projectile and actinide targets with smaller neutron excess. The ^{208}Pb and ^{209}Bi, and ^{48}Ca nuclei are most favorable targets and projectile in the cold and hot fusion reactions, respectively. For cold and hot fusion reactions, we must look for the optimal projectile and target, respectively.

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