Production of New Heavy Isotopes in Low-Energy Multinucleon Transfer Reactions

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It is shown that the multinucleon transfer reactions in low-energy collisions of heavy ions may be used for production of new neutron-rich nuclei at the "northeast" part of the nuclear map along the neutron closed shell $N = 126$ which plays an important role in the r process of nucleosynthesis. More than 50 unknown nuclei might be produced in such reactions (in particular, in collision of ^{136}Xe with ^{208}Pb) with cross sections of not less than 1 μb.

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During the last years, the progress in the investigation of nuclei far from stability has been impressive. Nowadays, nuclei far from stability are accessible for experimental study in almost any region of the chart of the nuclides. The only exception is the northeast part. The present limits of the chart of the nuclides in the region of heavy neutron-rich nuclei is still very close to stability although this region is extremely interesting for nuclear structure and nuclear astrophysics investigations. Study of the structural properties of nuclei along the neutron shell $N = 126$ would contribute to the present discussion of the quenching of shell effects in nuclei with large neutron excess. Moreover, this region is extremely interesting in astrophysics, in particular, for the production of heavy elements in stellar nucleosynthesis through the r process (the last "waiting point"). According to a recent report by the National Research Council of the National Academy of Sciences (USA), the origin of heavy elements from iron to uranium remains one of the 11 greatest unanswered questions of modern physics (see, for example, [1]) and it is likely to remain a hot research topic for the years to come.

Usually, new (neutron and proton rich) isotopes located far from the stability line are obtained in the fragmentation processes, fission of heavy nuclei and in low-energy fusion reactions. The first two methods are extensively used today for production of new isotopes in the light and medium mass region including those which are close to the drip lines. For example, in the fragmentation of ^{48}Ca beam with energy of about 140 MeV per nucleon, the neutron-rich nuclides ^{44}Si, ^{42}Al, and ^{40}Mg have been observed recently [2,3] in spite of extremely low cross section of their production at the level of 1 pb. In this region of the nuclear map, the neutron drip line may stretch up to very exotic nuclei like ^{40}O [4].

Because of the "curvature" of the stability line, in the fusion reactions of stable nuclei, we may produce only proton rich isotopes of heavy elements. For example, in fusion of ^{18}O with ^{186}W, one may get only the neutron deficient ^{204}Pb excited compound nucleus, which after evaporation of several neutrons shifts even more to the proton rich side. That is why we cannot reach the center of the "island of stability" ($Z \sim 114$, $N \sim 184$) in the superheavy mass region and have almost no information about neutron-rich isotopes of heavy elements (there are 18 known neutron-rich isotopes of cesium, for example, and only 4 of platinum).

In this Letter, we propose to fill this "blank spot" of the nuclear map by production of new heavy neutron-rich nuclei in the multinucleon transfer processes of *low-energy* damped collisions of heavy ions. It is well known that in the deep inelastic (or damped) collisions of heavy ions, the relative motion energy is transformed into the internal excitation of the projectile-like and target-like reaction fragments which are deexcited by evaporation of light particles (mostly neutrons). This does not seem to give us a chance for production of neutron-rich nuclei in such reactions at above-barrier energies. However, recently it was shown experimentally [5,6] that even at low collision energies of heavy ions (close to the Coulomb barrier), the cross sections for multinucleon transfer processes are still rather high. The excitation energy of the primary reaction fragments produced in these collisions should be not very high, and after evaporation of a few neutrons, one of the surviving residual nuclei might remain far from the stability line.

Here, we employ our model based on the Langevin-type dynamical equations of motion [7,8] to predict the cross sections for production of neutron-rich nuclei in the lead region of the nuclear map in the low-energy heavy ion collisions. We extended the model to consider the neutron and proton transfers separately (instead of using only one mass-asymmetry variable). This noticeably complicates the problem because of a necessity to deal with the multidimensional potential energy surface $V(R, \delta_1, \delta_2, \eta_N, \eta_Z)$ which depends on the distance between nuclear centers R, spheroidal type dynamic deformations of the fragments δ_1 and δ_2, and neutron and proton asymmetries, η_N and η_Z. All these degrees of freedom were found to be extremely important for evolution of low-excited heavy nuclear system, and no one of them can be ignored.

The potential energy is calculated within the double-folding procedure (sum of the nucleon-nucleon forces averaged over "frozen" densities of colliding nuclei) at initial (fast or diabatic) reaction stage. For slow collisions (near-barrier incident energies), when nucleons have enough time to reach equilibrium distribution after overlapping of the nuclear surfaces (adiabatic conditions), the potential energy of the nuclear system should be calculated within the two-center shell model. Thus, for the nucleus-nucleus collisions at energies near the Coulomb barrier, we need to use a time-dependent potential energy, which after contact gradually transforms from a diabatic potential energy into an adiabatic one (see Refs. [8,9]).

For all the variables, with the exception of the neutron and proton asymmetries, we use the usual Langevin equations of motion with the inertia parameters, μ_R and μ_δ, calculated within the Werner-Wheeler approach [10]. For the mass and charge asymmetries, the inertialess Langevin-type equations are used (derived from the master equations for the corresponding distribution functions)

$$\begin{aligned}\frac{d\eta_N}{dt} &= \frac{2}{N_{\mathrm{CN}}} D_N^{(1)} + \frac{2}{N_{\mathrm{CN}}} \sqrt{D_N^{(2)}}\Gamma_N(t),\\ \frac{d\eta_Z}{dt} &= \frac{2}{Z_{\mathrm{CN}}} D_Z^{(1)} + \frac{2}{Z_{\mathrm{CN}}} \sqrt{D_Z^{(2)}}\Gamma_Z(t),\end{aligned} \qquad (1)$$

where $\Gamma(t)$ is the normalized random variable with Gaussian distribution, $D^{(1)}$, $D^{(2)}$ are the transport coefficients, $\eta_N = (2N - N_{\mathrm{CN}})/N_{\mathrm{CN}}$, $\eta_Z = (2Z - Z_{\mathrm{CN}})/Z_{\mathrm{CN}}$, N, and Z are the neutron and proton numbers in one of the fragments, whereas N_{CN} and Z_{CN} are referred to compound nucleus.

Assuming that sequential nucleon transfers play a main role in mass rearrangement, i.e., $A' = A \pm 1$, we have

$$\begin{aligned}D_{N,Z}^{(1)} &= \lambda_{N,Z}(A \to A + 1) - \lambda_{N,Z}(A \to A - 1),\\ D_{N,Z}^{(2)} &= \frac{1}{2}[\lambda_{N,Z}(A \to A + 1) + \lambda_{N,Z}(A \to A - 1)],\end{aligned} \qquad (2)$$

where the macroscopic transition probability $\lambda_{N,Z}^{(\pm)}(A \to A' = A \pm 1)$ is defined by nuclear level density [11,12], $\lambda_{N,Z}^{(\pm)} = \lambda_{N,Z}^0 \sqrt{\rho(A \pm 1)/\rho(A)}$ and $\lambda_{N,Z}^0$ are the neutron and proton transfer rates [note that the nuclear level density $\rho \sim \exp(2\sqrt{aE^*})$ depends on the excitation energy $E^* = E_{\mathrm{c.m.}} - V(R, \delta_1, \delta_2, \eta_N, \eta_Z) - E_{\mathrm{rot}}$ and, thus, on all the degrees of freedom used in the model]. There is no information in literature on a difference between neutron and proton transfer rates, and for simplicity, we assume here that $\lambda_N^0 = \lambda_Z^0 = \lambda^0/2$, where λ^0 is the nucleon transfer rate which was estimated to be $\sim 10^{22}$ s^{-1} [11,12]. We treat λ^0 as a parameter which should be chosen from appropriate description of available experimental data on deep inelastic scattering [7,8].

Nucleon transfer for slightly separated nuclei is also rather probable. This intermediate nucleon exchange plays an important role in sub-barrier fusion processes and has to be taken into account in Eq. (1). It can be treated by using the following final expression for the transition probability

$$\lambda_{N,Z}^{(\pm)} = \lambda_{N,Z}^0 \sqrt{\frac{\rho(A \pm 1)}{\rho(A)}} P_{N,Z}^{\mathrm{tr}}(R, \beta, A \to A \pm 1). \qquad (3)$$

Here, $P_{N,Z}^{\mathrm{tr}}(R, \beta, A \to A \pm 1)$ is the probability of one nucleon transfer, which depends on the distance between the nuclear surfaces. This probability goes exponentially to zero at $R \to \infty$, and it is equal to unity for overlapping nuclei. Here, we used the semiclassical approximation for $P_{N,Z}^{\mathrm{tr}}$ proposed in Ref. [13]. Equation (1) along with (3) defines a continuous change of mass asymmetry in the whole space (obviously, $d\eta_{N,Z}/dt \to 0$ for far separated nuclei).

The double differential cross sections of all the processes are calculated as follows:

$$\frac{d^2\sigma_{N,Z}}{d\Omega dE}(E, \theta) = \int_0^\infty bdb \frac{\Delta N_{N,Z}(b, E, \theta)}{N_{\mathrm{tot}}(b)} \frac{1}{\sin(\theta)\Delta\theta\Delta E}. \qquad (4)$$

Here, $\Delta N_{N,Z}(b, E, \theta)$ is the number of events at a given impact parameter b in which a nucleus (N, Z) is formed with kinetic energy in the region $(E, E + \Delta E)$ and center-of-mass outgoing angle in the region $(\theta, \theta + \Delta\theta)$, $N_{\mathrm{tot}}(b)$ is the total number of simulated events for a given value of impact parameter. Expression (4) describes the mass, charge, energy and angular distributions of the *primary* fragments formed in the binary reaction. Subsequent deexcitation cascades of these fragments via fission and emission of light particles and gamma-rays were taken into account explicitly for each event within the statistical model leading to the *final* distributions of the reaction products. The sharing of the excitation energy between the primary fragments is assumed to be proportional to their masses. For both excited fragments, the multistep decay cascade was analyzed taking into account a competition between evaporation of neutrons and/or protons and fission.

The model was already applied successfully for description of the angular, energy, and mass distributions of reaction products observed in the deep inelastic scattering of heavy ions at above-barrier energies [7,8]. Here, we test our model for a possibility to apply it also for description of multinucleon transfer reactions at energies close to the Coulomb barrier. We used the same values of the model parameters as in Ref. [8], and, in particular, the nucleon transfer rate was fixed at $\lambda^0 = 0.05 \times 10^{22}$ s^{-1}. This rather small value was found to be sufficient to reproduce the mass distributions of reaction products in low-energy deep inelastic scattering of heavy ions.

In Ref. [5], light and heavy reaction products in collisions of ^{58}Ni with ^{208}Pb were measured at $E_{\mathrm{lab}} =$ 328.4 MeV. Potential energy of the nuclear system formed in this reaction is shown in Fig. 1 for configuration of two

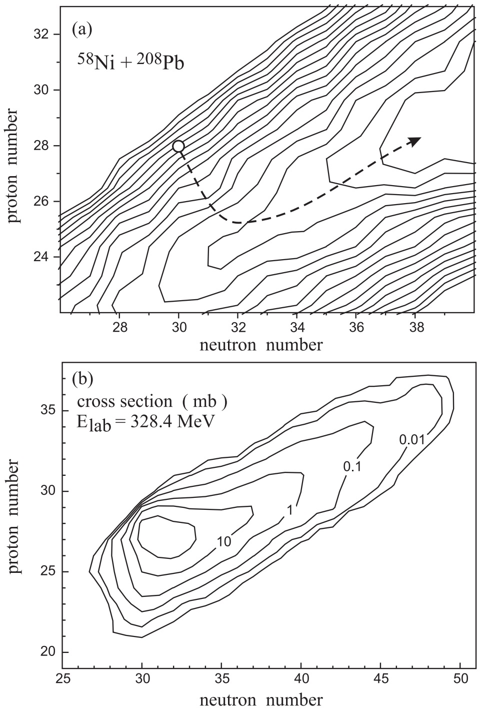

FIG. 1. (a) Landscape of potential energy surface of the nuclear system formed in collision of ^{58}Ni with ^{208}Pb. Potential energy is calculated for the contact configuration, $R = R_1(Z_1, N_1) + R_2(Z_2, N_2)$, at zero deformations $\delta_1 = \delta_2 = 0$. Contour lines are drawn over 1 MeV energy interval. Dashed curve with arrow indicates the most probable evolution of the system during nucleon exchange. (b) Landscape of the cross section (mb) for production of projectile-like fragments (logarithmic scale, the contour lines are drawn over 1 order of magnitude).

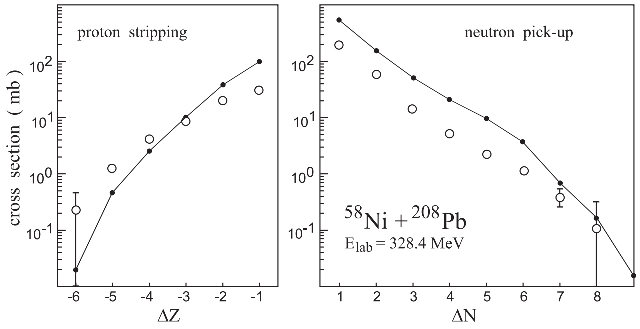

FIG. 2. Total cross sections for pure proton stripping (left side) and pure neutron pickup (right side) in collision of ^{58}Ni with ^{208}Pb at 328.4 MeV laboratory energy. Experimental data (open circles) are from Ref. [5].

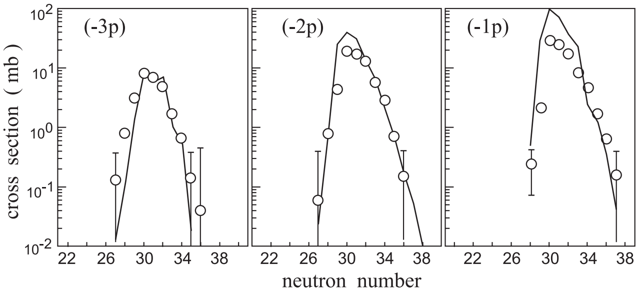

FIG. 3. Angular and energy integrated cross sections for proton and neutron transfer in collision of ^{58}Ni with ^{208}Pb at 328.4 MeV laboratory energy. Experimental data (open circles) are from Ref. [5].

touching nuclei depending on neutron and proton rearrangement. As can be seen, neutron pickup and proton stripping from ^{58}Ni are the most favorable (downward descent along the potential energy surface). Indeed, the calculated cross section, $d^2\sigma/dZdN$, reveals such behavior [Fig. 1(b)] and agree well with experimental data shown in Fig. 2 and in Fig. 3.

The satisfactory agreement of our calculations with available experimental data gives us a definite confidence in receiving rather reliable estimations of the cross sections for low-energy multinucleon transfer reactions leading to formation of new heavy neutron-rich nuclei in the northeast part of the nuclear map. It is clear that a choice of appropriate combination of colliding nuclei for production of specific neutron-rich isotopes is quite important. At low incident energies (low excitations), just a relief of the potential energy surface and, in particular, the shell effects play a very important role in the mass rearrangement not only in the fusion-fission processes but also in the deep inelastic collisions [14,15].

For production of heavy neutron-rich nuclei located along the neutron closed shell $N = 126$ (probably it is the last "waiting point" in the r process of nucleosynthesis), we propose to explore the multinucleon transfer reactions in low-energy collisions of ^{136}Xe with ^{208}Pb. The idea is to use the stabilizing effect of the closed neutron shells in both nuclei, $N = 82$ and $N = 126$, correspondingly. Pure proton transfer from lead to xenon might be

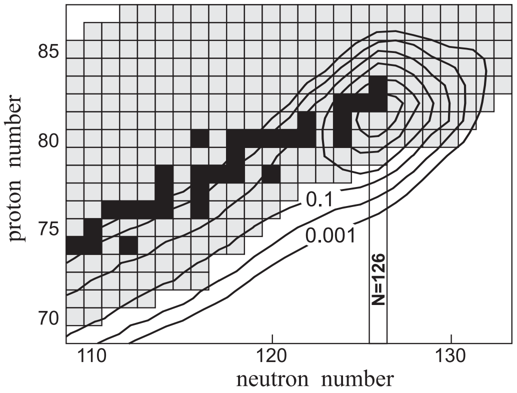

FIG. 4. Landscape of the total cross section $d^2\sigma/dZdN$ (mb, numbers near the curves) for production of heavy fragments in collisions of ^{136}Xe with ^{208}Pb at $E_{c.m.} = 450$ MeV. Contour lines are drawn over 1 order of magnitude.

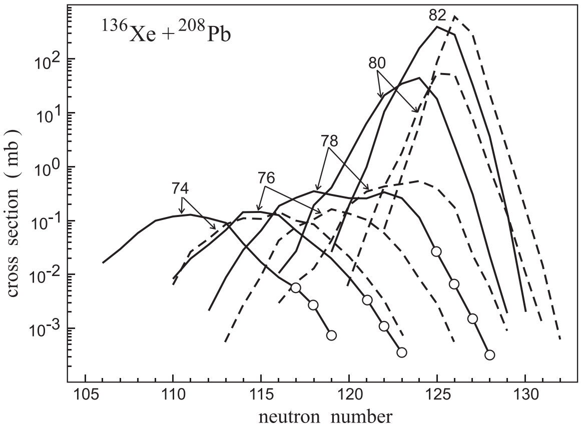

FIG. 5. Cross sections for production of heavy neutron-rich nuclei in collisions of ^{136}Xe with ^{208}Pb at $E_{\mathrm{c.m.}} = 450$ MeV. Dashed curves show the yield of primary fragments, whereas the solid ones correspond to survival nuclei. Open circles indicate unknown isotopes.

rather favorable here because the lighter fragments formed in such a process are well bound (stable nuclei) and the reaction Q-values are almost zero.

The landscape of the calculated cross sections for the yield of the different reaction fragments in low-energy collision of ^{136}Xe with ^{208}Pb is shown in Fig. 4. The cross sections for production of primary and survival heavy neutron-rich nuclei in this reaction at the energy $E_{\mathrm{c.m.}} =$ 450 MeV which is very close to the Coulomb barrier (Bass barrier for this combination is about 434 MeV) is shown in Fig. 5, whereas Fig. 6 demonstrates the yield of nuclei with neutron closed shell $N = 126$.

Thus, our calculations demonstrate that the production of unknown heavy neutron-rich nuclei is quite possible in the multinucleon transfer processes of low-energy heavy ion collisions. As can be seen from Fig. 4, about 50 new nuclides in this region can be produced with a cross section of 1 μb which is much higher than the level reached at available experimental facilities. These nuclei cannot be produced in fission reactions (they are too heavy). In principle, they can be produced in the quasifission decay of superheavy nuclear system (formed, for example, in collision of ^{48}Ca with ^{248}Cm). In this case, a lot of reaction fragments in the lead region really appear. However, all of them are highly excited (result of large positive reaction Q-values) and should evaporate many neutrons thus shifting to the stability line. This problem, nevertheless, needs additional study.

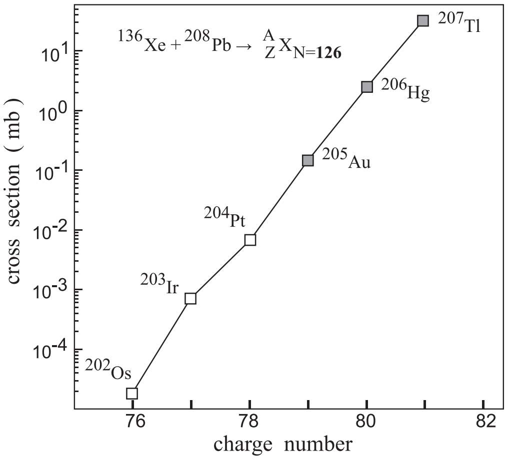

FIG. 6. Yield of nuclei with neutron closed shell $N = 126$ in collisions of ^{136}Xe with ^{208}Pb at $E_{\mathrm{c.m.}} = 450$ MeV. Open rectangles indicate unknown isotopes.

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