Diffusion mechanism for synthesis of superheavy elements

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The fusion-fission process in heavy systems is analyzed by the Smoluchowski equation with the finite-range droplet model potential of no pocket and the temperature-dependent shell correction energy which generates the pocket around the spherical shape. The evaporation residue cross sections of superheavy elements have been shown to have an optimum value at a certain initial temperature, due to the balance between the diffusibility for fusion at high temperature and the restoration of the shell correction energy against fission at low temperature. [S0556-2813(97)50303-7]

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In recent years, new heavy elements have been synthesized by the so-called *cold fusion* reaction [1], in which the target of Pb or Bi isotopes near doubly magic nucleus are bombarded by Ni or Fe isotopes, and the findings of the heaviest elements from 110 to 112 are reported with the cross section of the order of picobarn [2]. The cold fusion reaction is aimed at obtaining a high survival probability against fission, but it suffers a great loss of fusion probability into compound nuclei. On the other hand, a symmetric target-projectile combination can produce a rather cold heavy compound nucleus due to the interplay of the potential barrier and the Q value [3]. It turns out, though, that there is a fusion hindrance, i.e., a necessity of extra-push energy [4]. Therefore, also in those systems, there are the conflicting requirements of high fusion probability and high survival probability. The purpose of the present paper, thus, is to find out an optimum condition compromising the two requirements for synthesis of superheavy elements with massive target-projectile combinations.

Since there is no pocket around the spherical shape in the potential of the droplet model, and therefore no barrier, there is no formula for fusion probability, neither for fission decay probability of superheavy elements (we remind the reader that Bohr-Wheeler [5] as well as Kramers [6] formulas are not valid for cases without barrier). Therefore, there is no proper description for formation and decay of the superheavy compound nuclei before they cool down enough to restore the shell correction energy which generates the pocket. In other words, we have to employ a new dynamical description, at least for the early stage from the dinucleus complex with the fully dissipated incident kinetic energy to the spherical compound nucleus formation, and for its decay in the stage before the temperature becomes low enough for the restoring barrier to appear. If cooling due to neutron evaporation is much faster than the time scale of fission, which is now becoming well accepted from the analyses of γ-ray and neutron multiplicities [7–9], we can expect a certain probability for the system to remain around the spherical shape protected by the barrier.

In this paper, we describe the whole process by dissipative dynamics from the contact of two incident nuclei to the formation of the compound nucleus and further to the reseparation, namely, fission back into the symmetric fragments. From the analysis of prefission neutrons and fragment kinetic energies, a strong dissipation comparable to the one-body model is recommended [9], which permits us to use Smoluchowski equation for fusion-fission dynamics as an approximation of the Kramers or Langevin equation. As is well known and is readily seen in Eq. (1) below, the Smoluchowski equation describes diffusion over the potential energy surface with the diffusion coefficient $T/\mu\beta$, where T is the temperature of the compound nucleus, μ the inertia mass parameter, and β the reduced friction coefficient (the product of the latter two being simply a friction coefficient $\gamma = \mu\beta$). With this diffusion model, we can immediately expect that an optimum condition exists for residue cross sections. Qualitatively, in the formation process, higher temperature is favorable due to large diffusibility into the compact configuration from the dinucleus one at contact, while in the decaying process, lower temperature is favored for larger residue probability because of the higher fission barrier caused by the restored shell correction energy as well as the smaller diffusion coefficient. Therefore, a balance between the above two requirements gives rise to an optimum temperature or excitation energy of the compound system for the synthesis of superheavy elements. It should be noted here that, in the later stage of the decay process, the present treatment is more or less the same as the conventional one, but it is completely different from the latter in the formation stage and in the early stage of the decay. Thus, contrary to the conventional statistical analyses, the residue probability cannot be factored into the static fusion probability and the survival probability, but is given by the result of the dynamical evolution of the system.

The evolution of the probability distribution $P(x,l;t)$ in the collective coordinate space is assumed to follow Smoluchowski equation,
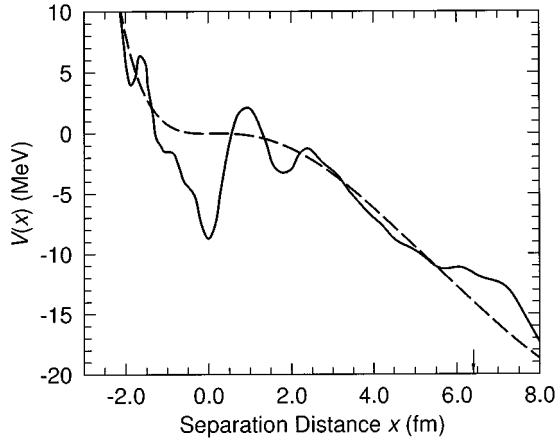
FIG. 1. The finite-range droplet model potential energy for the element 114 is drawn by the dashed line and the potential energy including the shell and paring corrections is drawn by the solid line. The initial probability density distribution is settled at x_0, which is marked by the arrow.

$$\frac{\partial}{\partial t}P(x,l;t) = \frac{1}{\mu\beta}\frac{\partial}{\partial x}\left\{\frac{\partial V(x,l;t)}{\partial x}P(x,l;t)\right\}$$
$$+ \frac{T}{\mu\beta}\frac{\partial^2}{\partial x^2}P(x,l;t). \tag{1}$$

The coordinate x is defined as $x=R_{\text{c.m.}}-\frac{3}{4}r_0A^{1/3}$ so that $x=0$ corresponds to the spherical shape, where $R_{\text{c.m.}}$ denotes the separation distance between the mass centers of the nascent fission fragments in the case of symmetric fission, A the mass number of the nucleus, and $r_0=1.16$ fm. The angular momentum of the system is expressed by l. Both the inertia mass μ and the reduced friction β are assumed to be independent of the shape of the nucleus in the present calculations. The parameter μ is taken to be the reduced mass for the symmetric separation and β is $5\times10^{21}\,\text{s}^{-1}$ corresponding to the weakest value of one-body dissipation in a series of shapes. Note that Eq. (1) actually does not depend on the inertia mass, but only on the friction $\gamma=\mu\beta$.

The time-dependent potential energy curve appearing in Eq. (1) is defined as follows:

$$V(x,l;t) = V_{\text{DM}}(x;t) + \frac{\hbar^2 l(l+1)}{2I(x)} + V_{\text{shell}}(x)\Phi(t),$$

$$V_{\text{DM}}(x;t) = (1-\xi T^2(t))E_S(x) + E_C(x), \tag{2}$$

where $I(x)$ is the moment of inertia of the rigid body at deformation x. V_{DM} and V_{shell} are the potential energy of the finite-range droplet model and the shell plus pairing correction energy at $T=0$, respectively. Both are calculated with the code developed by Möller [10]. E_S denotes the sum of the surface and the curvature energy and E_C is the Coulomb energy of the droplet model. The temperature dependence of E_S is introduced with $\xi=0.014$ MeV^{-2} [11]. The potential energy curve along the minimum valley is calculated with the ϵ parametrization [12] and is shown in Fig. 1 for the nucleus with $Z=114$ and $N=184$. The solid and dashed curves denote $V_{\text{shell}}+V_{\text{DM}}$ and V_{DM}, respectively. When the nucleus is in high temperature, the shell plus pairing correc-

tion energy disappears. It, however, is restored as the nucleus cools down and the potential energy curve changes gradually from the dashed curve to the solid one. Thus, one of the most important ingredients are the shell and the pairing correction energies, depending on the shape and temperature of the composite system.

The temperature dependence of the shell correction energy is extracted from the free energy [13] calculated with single particle energies [14]. We assume that both the shell and the pairing correction energies have the same dependence on temperature; hereafter, the term ''shell correction energy'' is used to refer to the shell plus pairing correction energy. The temperature dependent factor $\Phi(t)$ in Eq. (2) is parametrized as

$$\Phi(t) = \exp\left(-\frac{aT^2(t)}{E_d}\right), \tag{3}$$

following the work by Ignatyuk et al. [15], where a denotes the level density parameter of Töke and Swiatecki [16]. The shell-damping energy E_d is chosen as 20 MeV according to the above results. The cooling curve $T(t)$ is calculated by the statistical model code SIMDEC [14], which is confirmed to give similar results to the codes in the market for medium and heavy nuclei. The Smoluchowski equation is solved numerically with the finite difference method.

Concerning the initial condition, we assume that the kinetic energy of the relative motion in the entrance channel dissipates completely just inside the contact distance. The initial probability distribution $P(x,l;t=0)$ has a Gaussian shape with a very small width and is imposed at $x_0=x_{\text{cont}}-0.5$ fm, where x_{cont} is the contact distance evaluated as $x_{\text{cont}}=2r_0(A/2)^{1/3}-\frac{3}{4}r_0A^{1/3}$. The position of x_0 is marked by the arrow in Fig. 1 for $A=298$. (The effect of the approaching phase before contact such as the barrier penetration, and coherent or incoherent interactions should be taken into account [17]. But in the present calculation, they are neglected while the penetration will be discussed later.) Obviously, the present initial condition is crude, but should be allowed for the first preliminary calculations. As the fusion process is supposed to be diabatic, the potential would be a little different from that for the decay process (supposedly adiabatic). The difference surely changes the results quantitatively, but not qualitatively, so in the present calculation we use the one-dimensional potential. An extension to multidimensional coordinate space including the neck degree of freedom from the contact stage with velocity distributions, etc. is straightforward and will be made in the near future for realistic calculations with various mass asymmetric combinations of projectiles and targets.

The evaporation residue cross section is defined as the probability which is left inside the fission barrier in the final stage of the cooling process and is proportional to the quantity $d(T_0,l;t)$ at $t=\infty$:

$$d(T_0,l;t) = \int_{-\infty}^{x_{\text{sad}}}P(x,l;t)dx. \tag{4}$$

Here, T_0 is the initial temperature and x_{sad} stands for the first saddle point in Fig. 1. The evaporation residue cross section $\sigma_{\text{EV}}\equiv\Sigma_y\sigma(\text{HI},yn)$ is calculated as
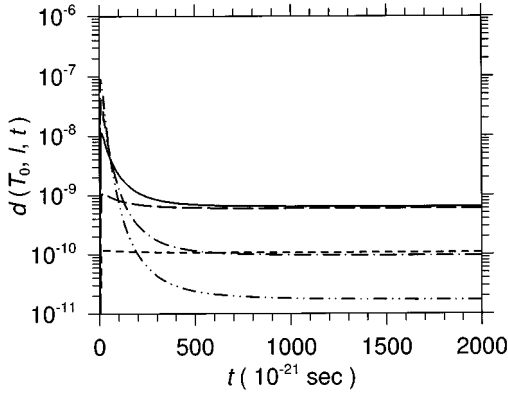
FIG. 2. The time evolution of the probability density in the compact configuration region $d(T_0, l = 10; t)$. The curves for five initial temperatures are plotted: $T_0 = 0.68$ (short-dashed), 0.79 (long-dashed), 0.96 (solid), 1.11 (dot-dashed), and 1.24 MeV (dot-dot-dashed).

$$\sigma_{EV} = \frac{\pi \hbar^2}{2 \mu_0 E_{c.m.}} \sum_l (2l+1) d(T_0, l; t = \infty), \qquad (5)$$

where μ_0 denotes the reduced mass in the entrance channel and $E_{c.m.}$ the incident energy in the center-of-mass frame.

As an example of reactions forming the doubly closed superheavy nucleus, we consider the reaction $^{149}_{57}\text{La} + ^{149}_{57}\text{La} \rightarrow ^{298}114$. The time-dependent feature of the probability $d(T_0, l = 10; t)$ is plotted in Fig. 2 for five different incident energies which correspond to $T_0 = 0.68$, 0.79, 0.96, 1.11, and 1.24 MeV. Up to the time of around 30×10^{-21} s, the probability density in the region of the compact configuration is supplied by diffusion from the contact region and its yield increases rapidly. But during that time, the main part of the probability initially at x_0 has descended down the slope of the potential and thus the supply ceases. After $t \sim 30 \times 10^{-21}$ s, the probability density accumulated in the compact configuration area diffuses back over the fission barrier arising from the restoration of the shell correction energy. At low temperatures such as $T_0 = 0.68$ MeV, 60% of the correction energy is restored and the fission barrier is about 6 MeV. Therefore, the fission width is very small and $d(T_0, l; t)$ becomes flat quickly. On the contrary, in the case of $T_0 = 1.24$ MeV, restoration sufficient to prevent the system from fissioning takes time, during which the yield accumulated in the compact configuration area diffuses out rapidly as shown in Fig. 2.

The height of the peak around 30×10^{-21} s is essentially determined by the diffusibility into the compact configuration area, while the decrease from the peak value to the final yield at $t_\infty = 2000 \times 10^{-21}$ s is determined by how fast the shell correction energy is restored to give rise to a sufficient barrier height. Thus, the final yield surviving in the compact configuration area is determined by two factors: the diffusibility depending on the temperature and the restoration of the shell correction energy.

In terms of the obtained values of $d(T_0, l; t_\infty)$, we can calculate the evaporation residue cross section σ_{EV} with Eq. (5). The excitation function of σ_{EV} for the $^{149}\text{La} + ^{149}\text{La}$
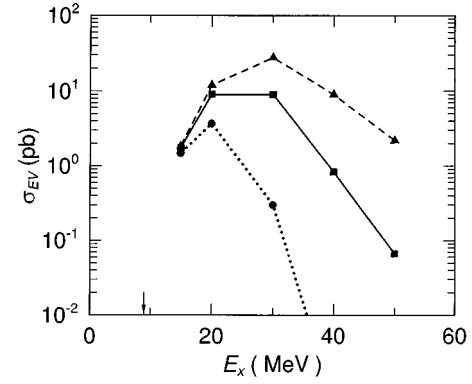

FIG. 3. The excitation function of the evaporation residue cross section for $^{149}_{57}\text{La} + ^{149}_{57}\text{La} \rightarrow ^{298}114$ reaction calculated from $d(T_0, l; t_\infty)$. Results for three values of reduced friction parameter β are plotted: $\beta = 2.5 \times 10^{21}\,\text{s}^{-1}$ (circles), $5.0 \times 10^{21}\,\text{s}^{-1}$ (squares), and $7.5 \times 10^{21}\,\text{s}^{-1}$ (triangles). The corresponding Bass potential barrier is indicated by the arrow.

$\rightarrow ^{298}114$ reaction is shown in Fig. 3 by squares. The results with $\beta = 2.5 \times 10^{21}\,\text{s}^{-1}$ and $7.5 \times 10^{21}\,\text{s}^{-1}$ are also plotted by circles and triangles, respectively. It is seen that the characteristic feature in the excitation function exists over a wide range of the friction strength. In this reaction system, the Bass potential barrier height [18] is 320 MeV in the center-of-mass frame and corresponds to $E_x = 9$ MeV in the compound nucleus as shown in Fig. 3. Thus, above $E_x = 9$ MeV, the penetration factor for the Bass barrier does not drastically change the energy dependence of $d(T_0, l; t_\infty)$ obtained by the diffusion model. The excitation function shows a bell shape having a maximum around $E_x \sim 25$ MeV which is produced by the two competing factors discussed above, not by the usual origin, i.e., accumulation of partial waves and disappearance of the fission barrier in high angular momenta. It should be emphasized that the optimum cross section can be realized above the Bass barrier in this reaction system and thereby can be observed experimentally.

In summary, a diffusion model which takes into account dynamical evolution of a distribution including statistical fluctuations in the deformation parameter space is shown to be a necessary and appropriate way to describe fusion-fission process for systems without, as well as with, pocket. With the model, it is shown for the synthesis of superheavy elements that there exists the optimum initial temperature or the excitation energy of compound system due to the balance between the diffusibility for fusion and the restoration of the shell correction energy against fission. Roughly speaking, the optimum temperature is around the restoration temperature of the shell correction energy. In the present symmetric system, the maximum cross section of about ten picobarns is obtained around $E_x = 20 \sim 30$ MeV. The absolute value of the cross section, of course, depends on the friction coefficient $\gamma = \mu \beta$ as well as the initial condition, etc. as stated above, which should be treated in more realistic ways, but the proposed mechanism will not essentially be affected by them. For example, the qualitative feature does not change with β as shown in Fig. 3. We can also take into account the temperature dependence of the level density parameter a used in the statistical calculation. It was confirmed that the

qualitative feature does not change with the modification. The authors believe that this novel mechanism can inspire new experimental studies for the synthesis of superheavy elements. Details of the model and the results will be given in a full paper, which is now in preparation. A more realistic model of two-dimensional dynamics including the neck degree of freedom has been prepared, in which effects of the difference between the fusion and the fission paths will be investigated in detail in comparison with the present one-dimensional model.

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