

Reaction dynamics of synthesis of superheavy elements

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Abstract. Based on the theory of the compound nucleus reaction, a brief review is given on the special aspects of the reaction dynamics in the synthesis of the superheavy elements (SHE), where the fusion probability is the most unknown factor. A new viewpoint of the fusion reaction is proposed that it consists of two processes; the first process up to the contact of two nuclei of the incident channel and the second one of a dynamical evolution to the spherical compound nucleus from the contact configuration. The fusion probability is, thus, given as a product of a contact probability and a formation probability. Analytic studies of the latter probability are discussed in the one-dimensional model, where a simple expression is given to the so-called extra-push energy in terms of the reduced friction, the curvature parameter of the conditional saddle point and the nuclear temperature. Preliminary results of numerical analyses of the contact probability are given, using the surface friction model (SFM). Remarks are given on the present status of our knowledge and for future developments.

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1 Brief reminder of the special aspects of SHE

Following the theory of the compound-nucleus reaction [1], residue cross-sections are given by the formula

$$\sigma_{\text{res}} = \pi\lambda^2 \Sigma_J(2J+1) \cdot P_{\text{fus}}^J(E_{\text{c.m.}}) \cdot P_{\text{surv}}^J(E_{\text{ex}}), \quad (1)$$

where P_{fus}^J and P_{surv}^J denote the fusion and survival probabilities for the total angular momentum J , respectively and are independent of each other except conserved quantities, such as the total angular momentum, etc. The excitation energy of the compound nucleus E_{ex} is equal to the sum of the incident c.m. energy $E_{\text{c.m.}}$ and the Q-value of the fusion reaction. Since the decay modes available to the compound nuclei of SHE are mainly fission and neutron emission, the probability P_{surv}^J is given as

$$P_{\text{surv}}^J(E_{\text{ex}}) = \frac{\Gamma_{\text{n}}^J(E_{\text{ex}})}{\Gamma_{\text{n}}^J(E_{\text{ex}}) + \Gamma_{\text{f}}^J(E_{\text{ex}})}, \quad (2)$$

where Γ_{f} and Γ_{n} are the decay widths of fission and neutron emission and are given by the statistical theory [2] *i.e.*, by Bohr-Wheeler formula or Kramers formula, and Weisskopf formula, respectively. Then, dependences on the excitation energy can be understood by approximate expressions with nuclear temperature, $\Gamma_{\text{n}} \sim \exp[-B_{\text{n}}/T]$, and $\Gamma_{\text{f}} \sim \exp[-B_{\text{f}}/T]$, where B_{f} and B_{n} are fission barrier height and neutron separation energy, respectively. (Explicit specifications of angular momentum dependences

are suppressed in physical quantities, and should be properly taken into account.) The temperature T is related to the excitation energy by the expression $E_{\text{ex}} = a \cdot T^2$ with the so-called level density parameter a . The special aspect here in SHE is that the barrier B_{f} is approximately given by the minus of the shell correction energy, ΔE_{shell} , because the Liquid Drop Model (LDM) fission barrier is almost equal to zero in accord with the fissility parameter x being nearly equal to 1. Thus, $\Gamma_{\text{f}} \gg \Gamma_{\text{n}}$ in most superheavy compound nuclei, and then, $P_{\text{surv}} \simeq \Gamma_{\text{n}}/\Gamma_{\text{f}} \sim \exp[-(B_{\text{n}} - B_{\text{f}})/T]$. And the energy ΔE_{shell} naturally depends on the excitation energy, which is readily understood by considering the situation in high excitation where many particle-hole excitations diminish the shell effect. Its dependence is parameterized with the shell-damping energy E_{d} , by $\Delta E_{\text{shell}}(E_{\text{ex}}) = \Delta E_{\text{shell}}(0) \cdot \exp[-E_{\text{ex}}/E_{\text{d}}]$ [3], where $\Delta E_{\text{shell}}(0)$ denotes the shell correction energy of the ground state and E_{d} is about 18 MeV theoretically. This means that compound nuclei with rather high excitation comparable to E_{d} have very small fission barriers and decay mostly through fission. Thus, the probabilities calculated with eq. (2) are very small. If the excitation energy is large enough for multiple emissions of neutrons, the expression of the r.h.s. of eq. (2) has to be used repeatedly and then, the final survival probability would be extremely small. This is the reason why experiments have been done in such a way that the formed compound nuclei have as low as possible excitation, especially in the so-called cold-fusion path. One remedy that remains is to

form neutron-rich compound nuclei, so that B_n is small and thereby P_{surv}^J is relatively large. This is especially crucial in hot fusion path [4].

However, as is seen in eq. (1), residue cross-sections depend not only on the survival probability but also on the fusion probability. In the former the cold-fusion path is surely favourable and the hot-fusion path is not, but in the latter the hot-fusion path is expected to be favourable and the cold-one is not, as is discussed below. Unfortunately, there is no reliable theory commonly accepted for the fusion reactions in the SHE region. Quantitative predictions, thus, are still difficult. So, let us start to understand special aspects of the fusion mechanism in SHE region.

In lighter mass region, P_{fus}^J is equal to the transmission coefficient $T^J(E_{\text{c.m.}})$. In massive systems, however, as is well known, fusion does not occur even if the incident energy is well above the barrier height, which means that P_{fus}^J is much smaller than T^J . This is called the fusion hindrance [5]. There are two possible interpretations for the hindrance. One is due to effects of frictions in the entrance channels, which cause a dissipation of the incident kinetic energy and thus reduce the probability for the system to overcome the barrier. An example is SFM [6] which was successful in explaining characteristic features of deep-inelastic collisions and was applied to the fusion hindrance with fair success in less massive systems, though it appears to hinder not enough in very massive systems. The other one is due to the effects of energy dissipation during shape evolutions towards the spherical compound nucleus, starting from the dumb-bell or pear-shaped configuration formed by the sticking of the two nuclei of the entrance channel. Important here is that there is a conditional saddle point (more precisely ridge line or ridge surface in a multi-dimensional deformation space) between the compound-nucleus configuration and the stucked di-nucleus configuration, which has to be overcome for fusion. The latter mechanism was proposed by Swiatecki [7] and was fairly successful in explaining experimental features of the hindrance observed in many massive systems, though not so good in less massive systems. It would be reasonable to consider both of them to exist, but which one is dominating is not clarified yet. That would depend on systems. Therefore, we propose that the fusion probability is given by two factors, the sticking and the formation probabilities,

$$P_{\text{fus}} = P_{\text{stick}} \cdot P_{\text{form}}, \quad (3)$$

which is reasonable from the dynamical viewpoint of the reaction process. The two factors on the r.h.s. of eq. (3) are not independent. As will be discussed below, mechanisms for passing the Coulomb barrier over not only determine the probability P_{stick} , but also provide initial conditions for the next stage of the dynamics of shape evolutions, *i.e.*, for calculations of P_{form} . In our previous predictions on $Z = 114$ [8] we employ the barrier penetration probability for P_{stick} and 3-dimensional Langevin calculations for P_{form} . More careful and detailed investigations are to be made. In the next section, the latter mechanism for P_{form} is discussed with the analytic solution for the one-dimensional overbarrier problem under

dissipation and fluctuation. In sect. 3, the former mechanism for P_{stick} , *i.e.*, the dynamics of the sticking of two nuclei from the encounter of the entrance ions are analysed. Preliminary results of the approaching phase with SFM are reported. Several remarks are also given in the last section.

2 Analytic expression of the extra-push energy

Swiatecki studied shape evolutions by classical trajectory calculations in three-dimensional space and Swiatecki-Bjornholm [9] schematized the results to simple expressions for the extra-push and extra-extra-push energies, which well explain the experimental trends of fusion hindrance. In order to understand the mechanism, we take up a one-dimensional schematic model for dissipative overbarrier problem [10]. We approximate the shape of a conditional saddle by an inverted parabolic shape whose curvature is parameterized by a frequency ω , *i.e.*, $V(q) = -\frac{1}{2}\mu \cdot \omega^2 \cdot q^2$ with the inertia mass associated with the fusing coordinate q . Employing a Langevin equation for the collective coordinate q , the motion is described by

$$\begin{aligned} \mu \frac{d^2q}{dt^2} &= -\frac{\partial V}{\partial q} - \gamma \cdot \frac{dq}{dt} + R(t) \\ &= \mu\omega^2 q - \gamma \cdot \frac{dq}{dt} + R(t), \end{aligned} \quad (4)$$

where γ and $R(t)$ denote a friction coefficient and its corresponding fluctuating force, *i.e.*, Langevin force. The fluctuating force is assumed to be the Markovian of a Gaussian distribution and to satisfy the dissipation-fluctuation theorem

$$\langle R(t)R(t') \rangle = 2\gamma \cdot T \cdot \delta(t - t'), \quad (5)$$

where $\langle \rangle$ means an average over all the realizations of the random force and T denotes the temperature of the noise source, *i.e.*, of the nucleonic degrees of freedom. Equation (4) is rewritten as a linear equation in the phase space,

$$\frac{d}{dt} \begin{pmatrix} q \\ p \end{pmatrix} = \begin{pmatrix} 0 & \frac{1}{\mu} \\ \mu\omega^2 & -\beta \end{pmatrix} \begin{pmatrix} q \\ p \end{pmatrix} + \begin{pmatrix} 0 \\ R \end{pmatrix}, \quad (6)$$

where $\beta = \gamma/\mu$ is the so-called reduced friction. As has long been known [11] this is generally solved, giving $q(t)$ and $p(t)$ in terms of the initial value (q_0, p_0) and of the random force R , and the corresponding distribution function in the phase space at a time t is given by a Gaussian function of $(p - p(t))$ and $(q - q(t))$. In order to obtain the probability of the system being on the other side of the barrier at $t \rightarrow \infty$, we simply integrate the distribution over the relevant phase space. This corresponds to the fusion probability in Swiatecki *et al.* [7,9] but to the formation probability in the present new viewpoint. The result is expressed by an error function,

$$P_{\text{form}} = \frac{1}{2} \text{erfc} \left(-\frac{\langle q(t) \rangle}{\sqrt{2} \cdot \sigma(t)} \right)_{t \rightarrow \infty}, \quad (7)$$

where $\langle q(t) \rangle$ and $\sigma(t)$ denote an average of trajectories and their variance respectively, and are given as

$$\langle q(t) \rangle = q_0 e^{-\beta t/2} \left[\cosh \left(\frac{1}{2} \beta' \cdot t \right) + \frac{\beta}{\beta'} \cdot \sinh \left(\frac{1}{2} \beta' \cdot t \right) \right] + 2 \frac{p_0}{\beta'} e^{-\beta t/2} \sinh \left(\frac{1}{2} \beta' \cdot t \right), \quad (8)$$

$$\begin{aligned} \sigma^2(t) &= \langle q^2 \rangle - \langle q \rangle^2 \\ &= -\frac{T}{\mu\omega^2} \left\{ 1 - e^{-\beta t} \left[2 \left(\frac{\beta}{\beta'} \right)^2 \sinh^2 \left(\frac{1}{2} \beta' \cdot t \right) + \frac{\beta}{\beta'} \cdot \cosh(\beta' \cdot t) + 1 \right] \right\}, \end{aligned} \quad (9)$$

where $\beta' = \sqrt{\beta^2 + 4\omega^2}$. Replacing the initial values q_0 and p_0 with the barrier height $B = \frac{1}{2}\mu\omega^2 q_0^2$ and the initial kinetic energy $K = \frac{p_0^2}{2\mu}$, the argument of the r.h.s. of eq. (7) for a large time ($t \gg 1/\beta'$), becomes

$$-\frac{\langle q(t) \rangle}{\sqrt{2} \cdot \sigma(t)} \rightarrow \frac{\beta + \beta'}{\sqrt{2(\beta^2 + \beta \cdot \beta')}} \left[\sqrt{\frac{B}{T}} - \frac{2\omega}{\beta + \beta'} \sqrt{\frac{K}{T}} \right]. \quad (10)$$

Since the extra-push energy is defined as $P_{\text{form}} = 1/2$, it is given by the condition that the argument of the error function be equal to zero. Then, the critical kinetic energy for 1/2 of the probability K_c is

$$K_c = \left(\frac{\beta + \beta'}{2\omega} \right)^2 \cdot B. \quad (11)$$

In the case of $\gamma = 0$, *i.e.*, $\beta = 0$, eq. (11) gives $K_c = B$, which is trivial. It is worth to emphasize here that the factor in front of B on the r.h.s. of eq. (11) is approximately equal to $(\beta/\omega)^2$ in cases of strong friction. In the one-body model of friction [12] this is about several to 10. That is, the additional energy required is not simply equal to the saddle point height, but equal to a much higher one, which is in agreement with fusion hindrance [5]. Since we have the analytic expression for the formation probability as a function of incident energy, we can discuss not only the extra-push energy (virtual shift of the barrier height), but also a slope of increase of the probability as incident energy increases. A kind of ‘‘variance of barrier distribution’’ can be defined as an energy difference between the cases of the probabilities of 0.2 and 0.8 for example. The square of the ‘‘variance’’ is found to be effectively linear in the extra-push energy, which appears to be consistent with the analyses of the experiments [13].

For quantitative predictions or comparisons with experiments, the one-dimensional model may not be accurate enough, because other degrees of freedom such as neck, mass asymmetry etc. come into play, especially in decaying back to reseparations. Effects of the neck degree of freedom in P_{form} are now being investigated [14]. Nevertheless, the result, eqs. (7) and (10) are extremely useful for qualitative understanding of fusion hindrance,

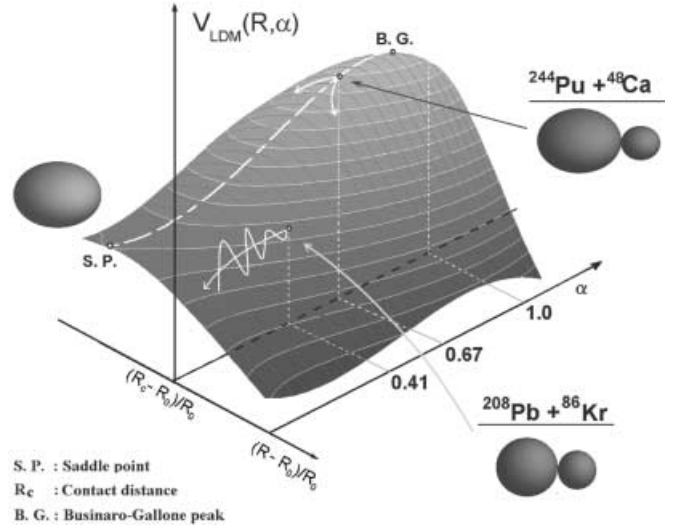


Fig. 1. LDM energy surface as a function of the distance between two fragments and the mass asymmetry $\alpha = (A_1 - A_2)/(A_1 + A_2)$. As for the former, R_c is the distance at the contact with R_0 being the c.m. distance of one-center limit, *i.e.*, the c.m. distance between two semi-spheres. The dashed line is connecting the conditional saddle points, *i.e.* denotes the ridge line. S.P. is the true saddle point for fission. Note that the figure is schematic, because the neck degrees of freedom etc. are frozen.

combined with the height of conditional saddle point as a function of mass asymmetry. Figure 1 shows a LDM potential landscape in the two-dimensional space. As is readily seen, entrance channels with very large mass asymmetry such as $^{48}\text{Ca} + ^{244}\text{Pu}$ [15] have contact configurations (on the line with R_c/R_0) around their conditional saddle points, which means that once a system reaches its contact point after overcoming the Coulomb barrier, it goes down to the spherical compound configuration as well as to the reseparations with similar probabilities. Therefore, fusion hindrance due to dynamical shape evolutions under the dissipation is inferred not to exist at all or to exist very weakly. On the other hand, in the mass-symmetric side (smaller α), even such as in $^{86}\text{Kr} + ^{208}\text{Pb}$ [16] contact configurations locate much outside of the conditional saddle points (the ridge line) and much lower in energy than the saddle points, which means that systems have to climb up to the condition saddle point and the necessary energy for oversaddle, *i.e.*, the extra-push energy is very large, as is given in eq. (11). According to ref. [9], the extra energies necessary for fusion are calculated to be 121 MeV for $^{86}\text{Kr} + ^{208}\text{Pb}$ and 0.0 MeV for $^{48}\text{Ca} + ^{244}\text{Pu}$, which is qualitatively consistent with eq. (11) and fig. 1. A probability for oversaddle at any incident energy is again given by eq. (7) with the argument given by eq. (10). How much hindered is the formation probability depends, of course, on the reduced friction β whose precise value is not yet known, especially at low temperatures. Therefore, we should not take the numerical numbers seriously, but consider them as indications of the qualitative features. This point is discussed in the last section.

3 Dynamics of sticking

It is also important to know how much flux reaches the contact point from where the dynamical evolutions discussed in the previous section start. In order to calculate probabilities of overpassing the Coulomb barrier to the contact and to analyse how in contact di-nucleus systems are, or more precisely how much the sticking limit is reached among them, we employ the SFM, *i.e.*, a classical trajectory model in the approaching phase of colliding ions [17]. Here, we simply recapitulate the equations, together with the Langevin forces added consistently with the dissipation fluctuation theorem,

$$\begin{aligned} \frac{dr}{dt} &= \frac{1}{\mu} p, \\ \frac{dp}{dt} &= -\frac{dV}{dr} - K_r \frac{p}{\mu} + R_r(t), \\ \frac{d\varphi}{dt} &= \frac{l}{\mu r^2}, \\ \frac{dl}{dt} &= -K_\varphi \frac{(l - l_s)}{\mu} + R_\varphi(t), \end{aligned} \quad (12)$$

where the notations are standard, and Langevin forces are assumed to be Gaussian and to satisfy the relation

$$\langle R_i(t) R_j(t') \rangle = \delta_{ij} \cdot \delta(t - t') \cdot 2 \cdot K_i \cdot T \quad (13)$$

with i specifying the coordinate r or φ .

The potential is a sum of the Coulomb potential V_C and the folding potential V_N employed in ref. [6], and the friction form factors are

$$K_i = K_i^0 \cdot \left(\frac{dV_N}{dr} \right)^2. \quad (14)$$

The values of K_i^0 are those of ref. [6],

$$\begin{aligned} K_r^0 &= 3.5 \times 10^{-23} (\text{s/MeV}), \\ K_\varphi^0 &= 0.01 \times 10^{-23} (\text{s/MeV}). \end{aligned} \quad (15)$$

The sticking limit angular momentum denoted by l_s is given by

$$l_s = l_0 \times \frac{\mu r_c^2}{\mu r_c^2 + I_1 + I_2}, \quad (16)$$

where l_0 is the incident angular momentum, r_c is the contact point and I_1, I_2 are the rigid-body moments of inertia of the incident ions 1 and 2, respectively.

Preliminary results on $^{86}\text{Kr} + ^{208}\text{Pb}$ collisions are given in fig. 2 and 3. Figure 2 shows the contact probability for $L = 0$ as a function of the incident energy relative to the potential barrier top, where the contact means the contact of two incident droplets, *i.e.*, the contact distance is defined as $r_c = R_1 + R_2$ with R_i being the radius of the i -th ion, *i.e.*, $1.25 \cdot A_i^{1/3}$.

It is readily seen that an extra energy necessary for the probability to be 1/2 is about 50 MeV, which could be compared with an experimental extra-push energy if the

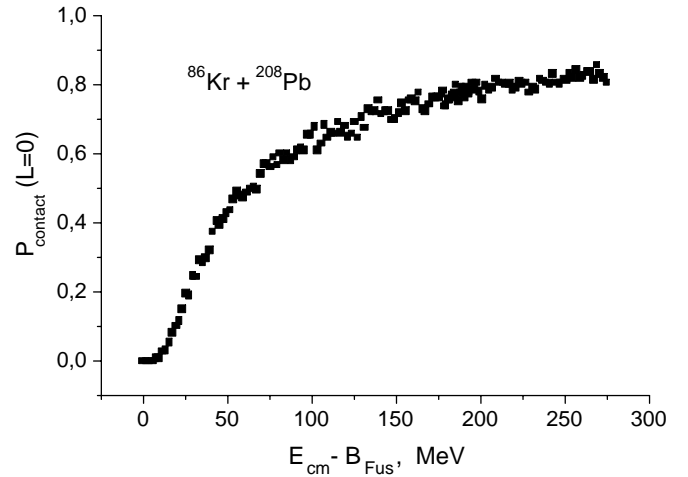


Fig. 2. The contact probabilities of two incident nuclear droplets for the $^{86}\text{Kr} + ^{208}\text{Pb}$ system are calculated with SFM, and shown as a function of incident c.m. energy relative to the Coulomb barrier. Note that quantum tunneling effects are neglected.

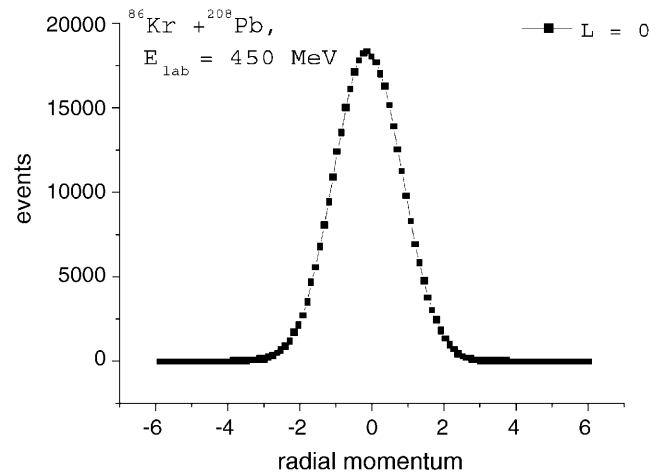


Fig. 3. The distribution of the radial relative momentum at the contact relative distance. The initial energy is taken to be about 10 MeV above the Coulomb barrier. The unit of the abscissa is $10^{-21} \text{ s} \cdot \text{MeV}/\text{fm}$. The initial value of the incoming momentum at the top of the barrier is about -3.5 in this unit. Therefore, radial momenta are completely dissipated at the contact.

subsequent process is neglected, *i.e.* P_{form} is assumed to be equal to 1. In $^{48}\text{Ca} + ^{244}\text{Pu}$ system, it is about 15 MeV (not shown). Again, in fusion processes, the $^{48}\text{Ca} + ^{244}\text{Pu}$ system is much more favourable than the $^{86}\text{Kr} + ^{208}\text{Pb}$ system. It is worth to mention here that if we apply the same model to light-ion combinations, there is no additional energy necessary, which is consistent with the well-known systematics of heavy-ion fusion reactions. This is due to the form factors of the friction forces given in eq. (14). In lighter systems, the frictions are not effective around the Coulomb barrier top, while in massive systems they are appreciable even outside the barrier top.

As for the angular-momentum dissipation, we have calculated orbital angular momenta of the trajectories as a function of the radial distance with $E_{c.m.}$ being 10 MeV above the barrier, and found that the average angular momenta reach the sticking limit at the contact point, up to the initial angular momentum of 30 which is the maximum of the calculated examples. This means that the systems averagely form the sticking configuration once they reach the contact point.

Next, we analyse the intrinsic excitation of the sticking configuration or the dissipation of the radial relative momenta initially carried in. Figure 3 shows, again for the cases with the incident energy being 10 MeV above the barrier, the distribution of the radial momenta calculated at the contact radial point, which appears to be Gaussian-like with the mean value of zero. Its variance is consistent with that expected from the temperature calculated with the full energy dissipation. This, surprisingly, indicates that the radial degree of freedom of the relative motion is in thermal equilibrium with the internal nucleonic degrees of freedom at the contact point. These features are common over wide incident energies and also in the $^{48}\text{Ca} + ^{244}\text{Pu}$ system.

The results given above indicate that the SFM is very strong in the dissipations in the reaction towards the fusion. There is another model for the approaching phase, *i.e.*, the proximity potential model [18]. Analyses with the potential are now being made for comparisons.

It should be emphasized once more here that the passing over or the penetration of the Coulomb barrier is not the goal for fusion, but the beginning of the next process discussed in the previous section. In other words, the results of the present section provide initial conditions for Langevin dynamics towards the compound nucleus.

4 Remarks

The important question which channels are most favourable for the synthesis of SHE is not yet answered quantitatively. However, it is clear that so-called cold-fusion path has a merit in the survival probability and is inferred to have a demerit in the fusion probability, while the hot-fusion path does the opposite. A combination of the methods given in sects. 2 and 3 is expected to provide a reliable framework for quantitative predictions of the fusion probability which has been the origin of the most serious ambiguities in the predictions of the cross-sections. Calculations of residue cross-sections will be made soon with P_{fus} of eq. (3) and the statistical P_{surv} .

Of course, the frameworks used above are all in the classical mechanics. Therefore, the sub-barrier energy region is out of the scope of the above treatments. Possible quantum effects are to be investigated in both processes, especially because classically calculated probabilities have turned out to be so small that (dissipative) quantum tunneling could give rise to a comparable order of magnitude.

It would be meaningful to mention that the one-body friction model which is mostly used in discussions of the

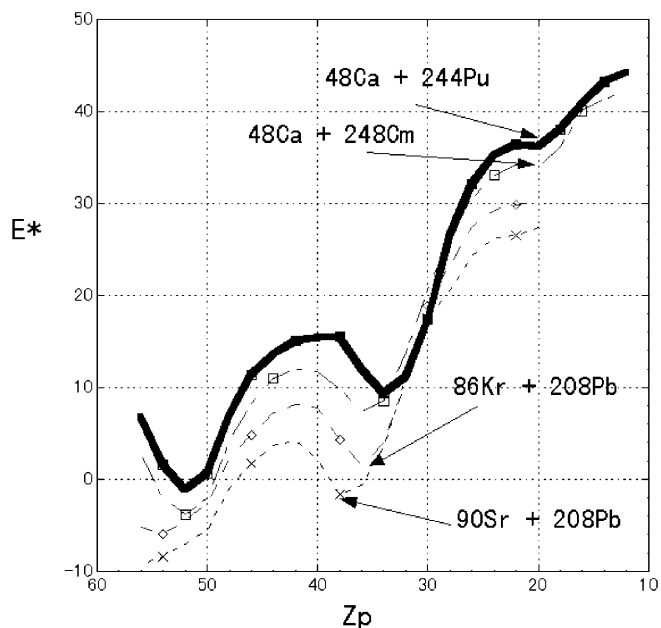


Fig. 4. Excitation energies of the compound nuclei with $Z = 118$ and $N = 176$, $Z = 120$ and $N = 178$, $Z = 114$ and $N = 178$, and $Z = 116$ and $N = 180$ are plotted against the possible projectile atomic number Z_p .

extra-push energy and in Langevin calculations is essentially temperature independent. There are many supporting evidences for such strong friction [19], but they are related to phenomena in rather high excitation, while reactions and compound nuclei formed in the synthesis of SHE are at very low energies, *i.e.* 10 MeV or a few tens of MeV. We, therefore, are not so sure that the one-body model is valid in formation and decay processes of SHE. If the friction is much weaker, extra-push energies should be much smaller and P_{form} much larger. Recent theoretical results by the linear response theory [20] would provide us with a reasonable T -dependence of the friction.

Lastly, another group of possible incident channels is to be pointed out. In the view of the cold-fusion path, excitation energies compound nuclei which are formed around the Coulomb barrier are crucial. Figure 4 shows examples of the compound nuclei with $Z = 118$ and $N = 176$, $Z = 120$ and $N = 178$, $Z = 114$ and $N = 176$, and $Z = 116$ and $N = 180$. The abscissa is the atomic number of projectiles and the ordinate is the excitation energy of the compound nuclei with the projectiles and their associate targets at the so-called Bass barrier [21]. We can readily see that there are two distinct minima which are related to ^{208}Pb and Xe and/or Gd. This feature is common over SHEs. The former is well known and has been investigated in the series of the experiments at GSI [22] and recently at Berkeley [16] while the latter is not yet well explored experimentally. Of course, there are small dips around $Z_p = 20$, which is related to ^{48}Ca projectile. These systems are rather in high excitation, but, as discussed above, are favourable in fusion probabilities and receive the benefit of relatively large P_{surv} due to small B_n .

The ridge line in fig. 1 and the lines in fig. 4 appear to be similar except for the effects of the shell correction energies of the projectiles and targets in the latter which give rise to the minima and the dips, but the two figures show the different physical quantities. Note that fig. 4 shows the usual Coulomb barriers, while the ridge line in fig. 1 shows the conditional saddles located far inside, and shows that there is a line of contact $((R_c - R_0)/R_0$ in fig. 1) of two nuclear droplets between them, though all the three lines become closer as mass asymmetry increases. This is the reason for the two-step treatment presently proposed with eq. (3).

The results given in sect. 2 have been obtained in collaborations with D. Boilley, B. Giraud and T. Wada, while those in sect. 3 are being worked out with G. Kosenko. To all of them the author would like to express his sincere thanks. Figure 1 is kindly made by C.W. Shen. This work is partially supported by the theory project of RIKEN Accelerator Research Facility (RARF).

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