Letter

Law of optimal incident energy for synthesizing superheavy elements in hot fusion reactions

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The superheavy elements (SHE) have the potential to transform and challenge our understanding of atomic and nuclear physics and chemistry. One of the biggest difficulties to synthesize elements beyond Oganesson is the accurate prediction of the optimal incident energies (OIE) for experiments. To this end, I present that an analytical formula could be universally applied in hot fusion reactions for calculating the OIE, based on the striking correlation found between the OIE and Coulomb barrier height of side collision B_{side} as well as Q value. The calculated OIE with my method are in remarkably good agreement with the available experimental data. Results from the time-dependent Hartree-Fock theory also quantitatively confirm the correlation between OIE and B_{side}. The predictions on the OIE for synthesizing $Z = 119$ and 120 elements are shown. This will contribute significantly to the future experiments for synthesizing the SHE as well as the nuclei on the "island of stability".

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The synthesis of the superheavy elements (SHE) is one outstanding research field, which provides essential information for several fundamental questions [1]. For example, where does the periodic table of elements end? Also, the field of SHE research puts nuclear and atomic theory to the test. In the past decades, the ^{48}Ca induced hot fusion reactions and cold fusion reactions with the ^{208}Pb and ^{209}Bi targets have been successfully used to synthesize the SHE with $Z = 102 - 118$ [2–4]. And the seventh period of the periodic table is now completely filled.

Currently, there are worldwide efforts to discover the SHEs beyond Oganesson. Due to the limitations of available targets with $Z > 98$, the projectiles heavier than ^{48}Ca have been proposed and investigated. The reactions $^{50}\text{Ti} + {}^{249}\text{Bk}$ [5], $^{64}\text{Ni} + {}^{238}\text{U}$ [6], $^{58}\text{Fe} + {}^{244}\text{Pu}$ [7], $^{54}\text{Cr} + {}^{248}\text{Cm}$ [8], and $^{50}\text{Ti} + {}^{249}\text{Cf}$ [5] have already been examined for the synthesis of SHE with Z = 119 and 120. Unfortunately, no correlated decay chains were observed.

Recently, the cross sections for the $^{242}\text{Pu}(^{48}\text{Ca}, 3\text{n})\,^{287}\text{Fl}$ and $^{243}\text{Am}(^{48}\text{Ca}, 3\text{n})\,^{288}\text{Mc}$ are measured in Dubna Gas Filled Recoil Separator (DGFRS-2) [9–11]. It is found that the maximum cross sections of the 3n-evaporation channel are much higher than the values measured in earlier experiments [12,13]. The main reason is that the projectile energies in the latest experiments are close to the optimal ones.

To synthesize the SHE, especially for the elements beyond Og with the possible cross sections lower than 1 pb, the optimal incident energy (OIE) is one essential factor. However, due to lack of the experimental data and the complex theoretical process [14,15], the predictions on OIE show great uncertainty and model dependence [16–27]. The discrepancy of predicted OIE could reach 10 MeV in different models, which could result in an order of magnitude difference in cross section. Efforts have been made experimentally to estimate the OIE [28–30]. It was pointed out that the OIE for the hot fusion reactions could be estimated by determining the Coulomb barrier distribution. However, the analytical and reliable method is still far from established. The synthesis of SHE can be theoretically divided into three steps. The evaporation residue cross section (ERCS) in the xn evaporation channel can be calculated as a sum over all partial waves J:

$$\sigma_{\text{ER}}^{xn}(E_{\text{c.m.}}) = \frac{\pi\hbar^2}{2\mu E_{\text{c.m.}}}\sum_J (2J+1)T(E_{\text{c.m.}}, J) \times P_{\text{CN}}(E_{\text{c.m.}}, J)W_{\text{sur}}^{xn}(E_{\text{c.m.}}, J). \tag{1}$$

T is the Coulomb barrier penetration probability. P_{CN} is the fusion probability to form a compound nucleus. W_{sur}^{xn} is the survival probability of the compound nucleus in the xn evaporation channel. The OIE, which corresponds to the maximal ERCS, is determined from both the fusion and deexcitation processes. In this Letter, considering that the compact configuration could enhance the fusion probability [31–34], and the Q value [$Q = M_{\text{P}} + M_{\text{T}} - M_{\text{C}}$; M_{P}, M_{T}, and M_{C} are masses of projectile (P), target (T), and compound nucleus (C), respectively] plays an important role for determining the most possible evaporation channel, which actually is the main criterion to define "cold" or "hot" fusion approaches, the systematics of the Coulomb barrier height in side-side collision of projectile and target (denoted with B_{side}) and Q value is investigated. The striking correlations between the OIE and B_{side} as well as Q value are noticed. Furthermore, a method that is valid for calculating the OIE in hot fusion reactions for synthesizing the SHE as well as the nuclei on the "island of stability" is presented.

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In this work, the B_{side} can be extracted from the effective nucleus-nucleus interaction potential V, which can be written as

$$V(Z_{1,2}, N_{1,2}, J, R, \theta_{1,2}) = V_{\text{N}}(Z_{1,2}, N_{1,2}, R, \theta_{1,2}) + V_{\text{C}}(Z_{1,2}, N_{1,2}, R, \theta_{1,2}) + \frac{(J\hbar)^2}{2\zeta_{\text{rel}}}. \quad (2)$$

Here, ζ_{rel} is the moment of inertia. θ_i is the angle between the symmetry axes of the ith nucleus and the collision axis. R is the centroid distance. The Coulomb potential is taken as the form in Ref. [35]:

$$V_C(Z_{1,2}, R, \theta_{1,2}) = \frac{Z_1 Z_2 e^2}{R} + \sqrt{\frac{9}{20\pi}}\left(\frac{Z_1 Z_2 e^2}{R^3}\right)\sum_{i=1,2} R_i^2 \beta_2^{(i)} P_2(\cos\theta_i) + \left(\frac{3}{7\pi}\right)\left(\frac{Z_1 Z_2 e^2}{R^3}\right)\sum_{i=1,2} R_i^2\left[\beta_2^{(i)} P_2(\cos\theta_i)\right]^2. \quad (3)$$

The double folding potential is employed to calculate the nuclear potential [33,36]:

$$V_N(R, \theta_1, \theta_2) = C_0\left\{\frac{F_{\text{in}} - F_{\text{ex}}}{\rho_0}\left[\int \rho_1^2(\mathbf{r}, \theta_1)\rho_2(\mathbf{r} - \mathbf{R}, \theta_2)d\mathbf{r} + \int \rho_1(\mathbf{r}, \theta_1)\rho_2^2(\mathbf{r} - \mathbf{R}, \theta_2)d\mathbf{r}\right] + F_{\text{ex}}\int \rho_1(\mathbf{r}, \theta_1)\rho_2(\mathbf{r} - \mathbf{R}, \theta_2)d\mathbf{r}\right\}, \quad (4)$$

where $F_{\text{in,ex}} = f_{\text{in,ex}} + f'_{\text{in,ex}}\frac{N_1 - Z_1}{A_1}\frac{N_2 - Z_2}{A_2}$. $C_0 = 300$ MeV fm^3, $f_{\text{in}} = 0.09$, $f_{\text{ex}} = -2.59$, $f'_{\text{in}} = 0.42$, $f'_{\text{ex}} = 0.54$. Z_1 (N_1), and Z_2 (N_2) are the proton (neutron) number of light projectile and heavy target, respectively. The nuclear density distribution functions ρ_1 and ρ_2 are chosen as two parameters of Woods-Saxon types:

$$\rho_1(\mathbf{r}, \theta_1) = \frac{\rho_0}{1 + \exp[(\mathbf{r} - \Re_1(\theta_1))/a_1]} \quad (5)$$

and

$$\rho_2(\mathbf{r} - \mathbf{R}, \theta_2) = \frac{\rho_0}{1 + \exp[(|\mathbf{r} - \mathbf{R}| - \Re_2(\theta_2))/a_2]}. \quad (6)$$

Here, $\rho_0 = 0.16$ fm^{-3}. $\Re_i(\theta_i) = R_i[1 + \beta_i Y_{20}(\theta_i)]$ is the surface radius of the ith nucleus. The diffuseness parameter for light projectile (a_1) and heavy target (a_2) are 0.54 and 0.56 fm, respectively. The quadrupole deformation parameter of the ground nuclei in this work is from Ref. [37].

Recently, the supplementary experimental data of cross sections for synthesizing Cn, Fl, and Mc isotopes in the hot fusion reactions ^{48}Ca + ^{238}U, ^{48}Ca + ^{242}Pu, and ^{48}Ca + ^{243}Am are presented by Oganessian *et al.*. To evaluate the relation between OIE and B_{side}, the dinuclear system (DNS-sysu) model [33] is employed. The nucleus-nucleus interaction potential in the DNS-sysu model is calculated exactly same as Eqs. (2)–(6). I would like to emphasize that the Coulomb potential [Eq. (2)] is strict, and the nuclear potential with double-folding form [Eq. (3)] has been widely used and we

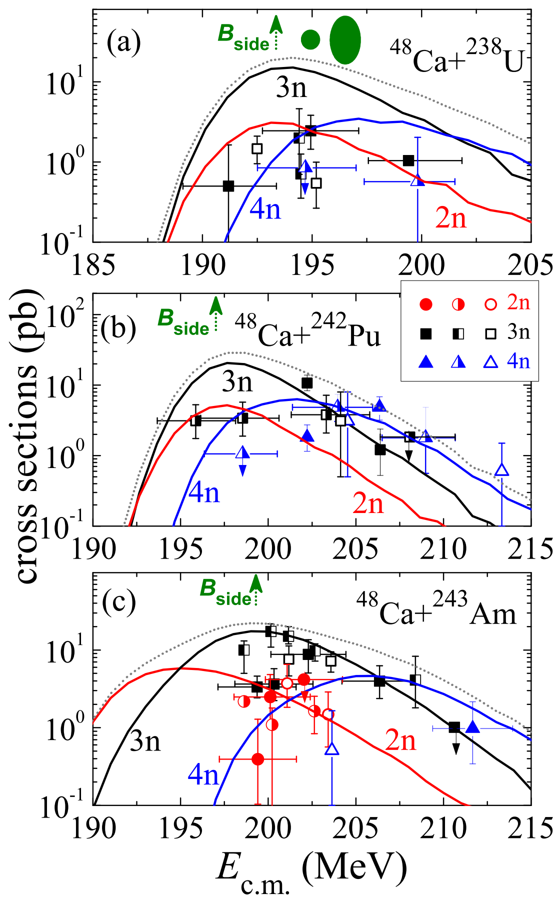

FIG. 1. The comparison of calculated ER cross sections with the experimental data for the reactions ^{48}Ca + ^{238}U [9,13,38,39], ^{48}Ca + ^{242}Pu [9,13,40], and ^{48}Ca + ^{243}Am [10–12,41]. The green arrows denote the values of B_{side}. The red, blue, and black solid lines denote the cross sections in the evaporation channels of 2n, 3n, and 4n, respectively. The gray dotted lines denote the total values. The circles, squares, and triangles denote the experimental data in the evaporation channels of 2n, 3n, and 4n, respectively. The open, half-closed, and closed symbols denote the data from different works.

never adjust the parameters. Therefore, the values of B_{side} calculated in this work have very small uncertainties. In Fig. 1, the experimental data are compared with the calculations in the DNS-sysu model. It can be seen that the calculated results are in good agreement with the experimental data. In particular, the calculated OIE, which is extracted from the total calculated cross sections (the peak positions of the gray dotted lines), are close to the incident energies with maximal cross sections in the measurements, especially for the reaction ^{48}Ca + ^{243}Am in which the feature of OIE with maximal cross section clearly emerges. The calculated Coulomb barrier height of the side collision is also shown. Note that the calculated OIE is close to the value of B_{side}. In hot fusion reactions with the deformed actinide nuclei, the barrier distributions are mainly related to the collision orientations. The phenomenon is associate with the behavior that the compact configuration could enhance the fusion probability, which was demonstrated in experimental measurement [31] as well as the theoretical models [32–34].

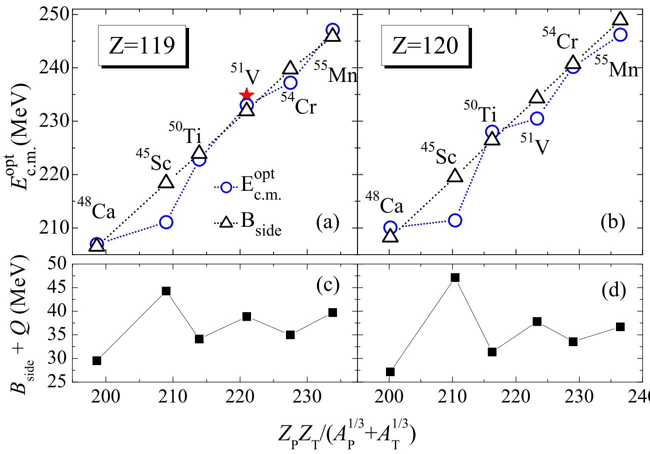

FIG. 2. The calculated OIEs for synthesizing the SHE $Z = 119$ (a) and 120 (b) based on the targets ^{244}Pu, ^{243}Am, ^{248}Cm, ^{249}Bk, ^{249}Cf, ^{254}Es, and ^{257}Fm. The values of B_{side} are also shown. The values of $B_{side} + Q$ for the reactions synthesizing SHE $Z = 119$ (c) and 120 (d). The red star denotes the OIE extracted from the experimental barrier distribution [30].

Furthermore, I extract the OIE from the calculated cross sections for synthesizing the SHE with $Z = 119$ and compare with the values of B_{side} in Fig. 2(a). Systematically, the values of B_{side} and OIE increase with increasing Coulomb parameters. The data extracted from the experimental barrier distribution is also shown for the reaction $^{51}V + ^{248}Cm$ [30]. The OIE calculated in the DNS-sysu model is 233 MeV, which is close to the value of 234.8 MeV presented in Ref. [30], as well as the value of 235 MeV calculated with an empirical method [26]. The corresponding B_{side} value is 231.9 MeV. One can see that OIE are close to the values of B_{side} for the most reactions. The linear relation between B_{side} and the Coulomb parameter is displayed. This is due to the fact that the diffuseness parameter is much smaller than the barrier position [42]. Besides, the quadrupole deformation parameters for the actinide targets are close within the range of $0.2 - 0.3$. One important feature is that the values of OIE fluctuate around B_{side}, except for the ^{45}Sc induced reaction in which the calculated value of OIE is 9 MeV lower than corresponding B_{side}. The excitation energy of the compound nucleus is determined from the sum of Q value and incident energy. The values of $Q + B_{side}$ are shown in Fig. 2(c). It can be seen that the value of $Q + B_{side}$ for the reaction $^{45}Sc + ^{254}Es$ is much larger than those in other reactions and is beyond the fluctuation. This is because of the small absolute Q value associating with the binding energy of ^{45}Sc. The high excitation energy reduces the survival probability of the compound nucleus. Consequently, the OIE for the reaction $^{45}Sc + ^{254}Es$ should be much lower than B_{side} value. The comparison of OIE and B_{side} in the reactions for synthesizing the SHE with $Z = 120$ are also shown in Fig. 2(b). Similarly, the OIE in the ^{45}Sc induced reaction is much lower than corresponding value of B_{side}. Also, as expected, the large value of $Q + B_{side}$ is noticed.

My aim is to find key quantities and underlying relations that determine the OIE in hot fusion reactions. Obviously, the OIE strongly depends on the Coulomb barrier which is related to the charge asymmetry of the combination, while the deviation from the B_{side} is due to the structures of the projectile and target. With this in mind, I show the systematics of Q values for the hot fusion reactions in Fig. 3(a). The nuclei mass tables Möller1995 [37] and WS2010 [43] are used and compared. It is shown that the Q value decreases almost linearly with the Coulomb parameter. To extract the systematic law, the reaction systems are selected for fitting that the discrepancies between OIE and B_{side} are smaller than 5 MeV. Indeed, the residual sum of squares in fitting Q values without the ^{45}Sc reduced reactions is about five times smaller than that including them. By the way of linear fitting, the relations of $Q(z) = -1.017z + 28.47$ MeV and $Q(z) = -1.047z + 32.49$ MeV are established based on the mass tables Möller1995 and WS2010, respectively. Here, $z = Z_PZ_T/(A_P^{1/3} + A_T^{1/3})$. Therefore, the OIE can be estimated by superposition of systematic value and fluctuation resulted from the Q value:

$$E^{opt}_{c.m.}(Z_P, A_P; Z_T, A_T) = B_{side}(z) + Q(z) - Q(Z_P, A_P; Z_T, A_T). \tag{7}$$

Here, $Q(Z_P, A_P; Z_T, A_T)$ is the fusion reaction Q value calculated with the masses of projectile (P), target (T), and compound nucleus based on the mass tables Möller1995 [37] and WS2010 [43].

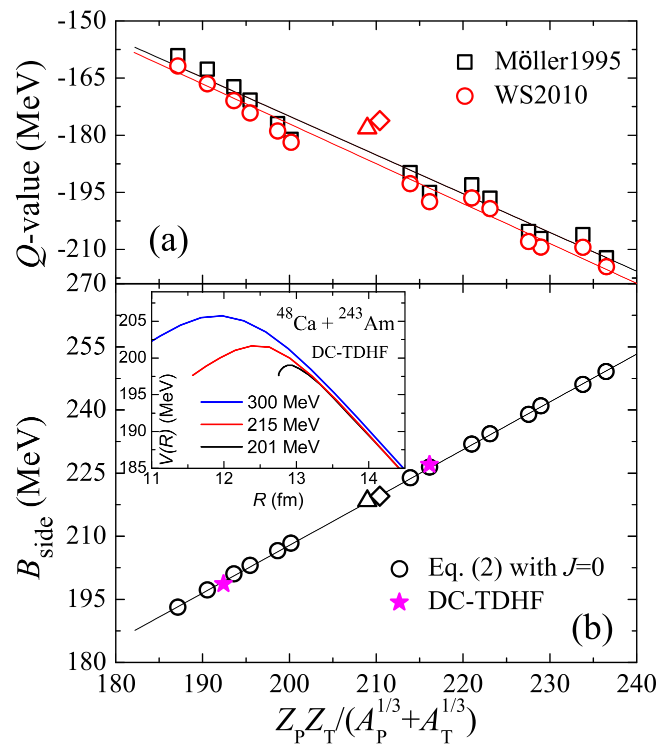

FIG. 3. Systematics of Q values (a) and B_{side} (b) in hot fusion reactions for synthesizing the SHE. The lines show the fitting of the data for the several hot fusion reactions with the discrepancy between OIE and B_{side} smaller than 5 MeV. The triangle and diamond in (a) denote Q values based on WS2010 for the reaction $^{45}Sc + ^{249}Cf$ and $^{45}Sc + ^{254}Es$, respectively. The stars in (b) denotes the calculated results for the reaction $^{48}Ca + ^{243}Am$ and $^{50}Ti + ^{249}Cf$ in the framework of the density constrained time-dependent Hartree-Fock (DC-TDHF) theory. The inset in (b) shows the energy dependence of dynamical interaction potential in side collisions calculated in DC-TDHF theory.

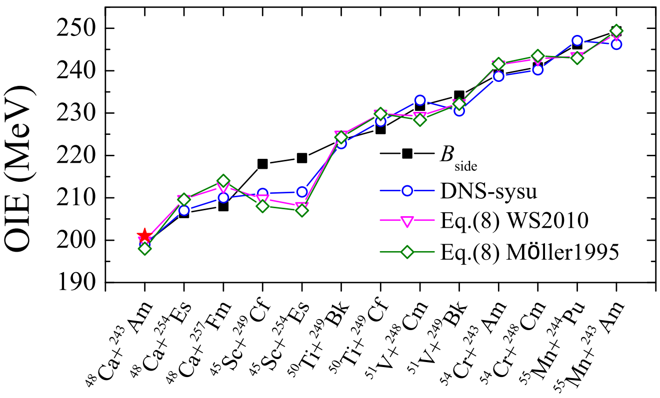

FIG. 4. The OIE calculated in the DNS-sysu model and Eq. (8) with different parameters based on the mass tables Möller1995 (M) and WS2010 (W) in the possible hot fusion reactions for synthesizing SHE with $Z = 119$ and 120. The calculated B_{side} (MeV) is also shown. The comparison of calculated results with experimental data [10] (denoted with star) in the reaction $^{48}Ca + ^{243}Am$ is also shown. The lines are used to guide the eye.

In Fig. 3(b), the fitting of B_{side} is also shown. We get $B_{side}(z) = 1.136z - 19.386$ MeV. I also show the fully microscopic approach DC-TDHF (Sky3D code with SLy5 [44]) result for the reaction $^{48}Ca + ^{243}Am$ that is the not system employed in the fitting. It is also the only reaction at present that the experimental OIE could be accurately extracted with error less than 1 MeV based on the abundant data. The TDHF approach describes the nuclear structure including the property of nuclear deformation and dynamics, naturally [45–47]. The inset shows the interaction potential of side collisions as a function of incident energy. From the perspective of the microscopic dynamics, the B_{side} value strongly depends on the incident energy. To get B_{side} related to OIE, it is reasonable to perform the DC-TDHF calculation at $E_{c.m.} = 201$ MeV which is the experimental OIE as shown in Fig. 1(c). The fitted value of B_{side} is 199.2 MeV, which remarkably agrees with the DC-TDHF calculation (198.7 MeV), indicating the high reliability of the interaction potential calculated in this work. On the other side, the value of B_{side} calculated in the DC-TDHF theory is close to the experimental OIE, in the microscopic way verifying the relevance between the OIE and B_{side} value. Meanwhile, it provides a method for the microscopic models to calculate the B_{side} for estimating the OIE that is the dynamical potential in side collisions with limit incident energy that two complex nuclei can just touch. As a example, for the reaction $^{50}Ti + ^{249}Cf$ with variation of the incident energy, 230 MeV is selected and the corresponding B_{side} value is 227 MeV, which is in good agreement with the fitted value (226.2 MeV).

Base on Eq. (7), one can write

$$E^{opt}_{c.m.}(Z_P, A_P; Z_T, A_T) = C_1 z + C_2 - Q(Z_P, A_P; Z_T, A_T). \quad (8)$$

Here, $C_1 = 0.119$ MeV and $C_2 = 9.084$ MeV for the case of Möller1995. $C_1 = 0.089$ MeV and $C_2 = 13.104$ MeV for the case of WS2010. The OIE can be simply estimated by this formula based on the values of z and Q.

Figure 4 shows the comparison of calculated results from the DNS-sysu model and formula (8) with B_{side} values in the hot fusion reactions for synthesizing the SHE with $Z =$ 119 and 120. One can see that the values of OIE from the DNS-sysu model are close to those based on the formula (8). Clearly, the B_{side} values strongly overestimate the OIE for the ^{45}Sc induced reactions. The formula (8) is also analyzed by quantitatively comparing the calculated OIE with experimental data for the reaction $^{48}Ca + ^{243}Am$ in Table I. The parameters fitted based on nuclei mass tables Möller1995 and WS2010 are evaluated. It can be seen that the formula reproduces the experimental data well. Furthermore, the parameters C_1 and C_2 are optimized by using the WS2010 mass table. This is consistent with the fact that the root-mean-square deviation in the mass table WS2010 is down to 0.441 MeV, which means more accurate estimations of nuclear masses than that in Möller1995. As mentioned above, the experimental OIE for the reaction $^{48}Ca + ^{243}Am$ could be accurately estimated. Here, I would like to emphasize that the OIE calculated with the Eq. (8) for the reaction $^{48}Ca + ^{243}Am$ is in better agreement with the experimental data than B_{side} value from the DC-TDHF calculation, supporting the importance of the Q value considered in Eq. (8). In addition, more than 10 MeV difference between B_{side} and OIE can be seen for the ^{45}Sc induced reactions.

TABLE I. The values of OIE in the c.m. frame from DNS-sysu, experimental data (Exp.), and Eq. (8) in this work with different parameters based on the mass tables Möller1995 (M) and WS2010 (W). The calculated values of B_{side} (MeV) are also shown.

		OIE (MeV)		
Reactions	B_{side}	DNS-sysu	Exp.	This work Eq. (8)W (M)
$^{48}Ca + ^{243}Am$	199.2	199.0	201 [10]	199.9 (198)
$^{45}Sc + ^{249}Cf$	218	211.0		209.8 (208.0)
$^{45}Sc + ^{254}Es$	219.4	211.4		208.0 (207.0)
$^{48}Ca + ^{254}Es$	206.4	207.0		209.7 (209.6)
$^{50}Ti + ^{249}Bk$	223.7	222.8		224.9 (224.3)
$^{51}V + ^{248}Cm$	231.9	233.0		229.3 (228.4)
$^{54}Cr + ^{243}Am$	239.1	238.7		241.4 (241.6)
$^{55}Mn + ^{244}Pu$	246.2	247.1		243.4 (243)
$^{48}Ca + ^{257}Fm$	208	210.0		212.7 (214)
$^{50}Ti + ^{249}Cf$	226.2	228.0		229.8 (229.8)
$^{51}V + ^{249}Bk$	234.1	230.5		232.3 (232.2)
$^{54}Cr + ^{248}Cm$	240.8	240.2		242.8 (243.5)
$^{55}Mn + ^{243}Am$	249.3	246.2		248.7 (249.4)

In summary, the OIE matters whether the SHE beyond Og can be successfully synthesized. However, due to the complex processes of theoretical models, the great uncertainty and model dependence were shown. In this Letter, the systematics of B_{side} and Q value in hot fusion reactions is investigated. The striking correlation between the OIE and B_{side} as well as the Q value is found. Based on that, an analytical formula is presented to calculate the OIE, which could be universally applied in hot fusion reactions. The calculated results with my method are in remarkably good agreement with the available experimental data. The important fact is that the method is weak dependence of theoretical models. Furthermore, by using the TDHF theory, the reaction ^{48}Ca + ^{243}Am is investigated for estimating OIE. A method for the microscopic theory to calculate B_{side} determining the OIE is proposed, where B_{side} equals the dynamical potential in side collisions with limit incident energy that two complex nuclei can just touch. Results from the TDHF theory quantitatively confirm the relation between the OIE and B_{side}. Finally, I predict the OIE for synthesizing the $Z = 119$ and 120 elements in the possible combinations.

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