Exploring an experimental route of synthesizing superheavy elements beyond $Z > 118$

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Role of the Coulomb interaction, mean fissility, mass asymmetry, and charge asymmetry parameters on the synthesis of heavy and superheavy elements has been examined with respect to the deformation parameters of the projectile and target nuclei explicitly in light of the experimental results. The observed facts are classified into four categories and are then used to study several unsuccessful as well as planned reactions to synthesize the new superheavy elements $Z = 119, 120$. Concrete inference is too difficult to draw from these results because of excessive deviations in evaporation residue cross-section data. It is found that the arbitrary choice of excitation energy for the experiments was the root cause of such large deviations. Such a complex issue can be resolved well by theoretical excitation function studies using the advanced statistical model or the dinuclear system model and choosing the excitation energy corresponding to the energy where the excitation function curve shows the maximum. We believe this method may help us to predict whether the estimated evaporation residue cross section can be measurable within the experimental limit of the existing facilities for the future reactions planned.

Elements with atomic number $Z \geq 104$ known as transactinides or superheavy elements (SHE) are produced using either cold [1] or hot fusion reactions [2][3]. These methods have enabled us to synthesize the SHEs up to the element oganesson $Z = 118$ [4]. The cold fusion reactions have been applied for SHEs from $Z = 104−113$ and the hot fusion reactions from $Z = 110−118$. In cold fusion reactions, $^{208}$Pb or $^{209}$Bi target nuclei are bombarded with the projectiles heavier than $^{48}$Ca and in hot fusion reactions the $^{48}$Ca projectiles are impacted on an actinide target. To produce the elements heavier than oganesson, the projectiles heavier than $^{48}$Ca are required as the target elements heavier than Californium ($Z = 98$) are not possible yet [5][6] and all such experimental attempts have been failed till date [7][8] irrespective of the fact that increasing stability was expected for superheavy elements $Z > 110$ [3][4]. Accordingly special effort is being invested to revamp the detection sensitivity close to $fb$. On the other side, we have taken up a project in pinning the possible reason of the said debacle.

A memory of entrance channel effects such as the Coulomb interaction parameter ($z$), compound nucleus fissility ($\chi_{CN}$), effective entrance channel fissility ($\chi_{eff}$), mean fissility ($\chi_m$), charge ($\alpha_Z$) and mass asymmetry ($\eta_A$) is often retained in the heavy-ion reactions [9][10], especially, producing compound nucleus (CN) of $A \approx 220$ [11]. A $\approx 230$ [12], actinide [13] and superheavy elements [14]. The entrance channel dynamics for the cold fusion reactions have been studied in detail about two decades ago [16]. While we have extended this study recently for the hot fusion reactions [17]. Furthermore, other entrance channel effects such as target deformation [18] and excitation energy [19] determine the quasifission characteristics [20], which are the important issues for a reaction to synthesizing a superheavy element [21]. Role of all these entrance channel effects has been examined explicitly by the experimental evaporation residue cross-sections of the heavy-ion reactions used for synthesizing the SHEs $Z = 104−118$ successfully. The same treatment has also been applied on the unsuccessful reactions which were employed for the SHEs $Z = 119−120$. Such works help us to establish certain systematic trends, which in turn allow us to plan for a most suitable reaction. Note that the beam energy required for this reaction is extremely important and it can be found from excitation function studies using either the advanced statistical model (ASM) [22][24] or the dinuclear system (DNS) model [16]. In this letter, we describe the full survey with a happy note that present ideas may lead to the synthesis of the SHEs beyond $Z = 118$.

As the projectile is made to incident on the target, a compound nucleus is formed at certain excitation energy, which cools down by evaporation of neutrons or any other light particles and the evaporation residue cross-section ($\sigma_{ER}$) is the measure of the formation of the superheavy nuclei. This $\sigma_{ER}$ is highly reduced due to re-separation of the nuclei by the quasi-fission in the same entrance channel and by a considerable probability of fission of the CN called fusion-fission [25]. The $\sigma_{ER}$ depends on specific entrance channel parameters $z$, $\chi_m$, $\eta_A$ and $\alpha_Z$ as mentioned above, which are defined in earlier papers (for example [16]) and has also been provided in Supplemental Material [26]. A slight change in the entrance channel parameters makes a large difference in the $\sigma_{ER}$ [27]. Such variations can be observed with the experimental $\sigma_{ER}$ versus any particular entrance channel parameter. We have studied the roles of the entrance channel in different classes of reactions that are made on the basis of deformation in the projectile and target nuclei.
They include the spherical-spherical, deformed-spherical, deformed-deformed and spherical-deformed. Fig. 1 displays the nature of experimental $\sigma_{ER}$ vs $z$ and $\chi_m$ in different deformation classes in semi-log plots. Similarly, trends of other entrance channel parameters on the $\sigma_{ER}$ have been shown in Fig. 2, Fig. 3 and Fig. 4.

A strange characteristic has been noticed in the plots of Fig. 1-4. Though the ER cross-section data versus $z$ and $\chi_m$ can be presented on semi-log plot for spherical-spherical, deformed-spherical and deformed-deformed cases, but spherical-deformed case has to be differentiated with the range of compound nuclear atomic numbers, viz., preactinides ($z = 77 - 88$), actinides ($Z = 89 - 103$) and transactinides ($Z = 104 - 118$) as shown in Fig. 1-2. This strategy fails when we consider the parameter $\alpha_Z$ and $\eta_A$; there deformed-deformed case also has to be divided as the spherical-deformed case as shown in Fig. 3-4.

Having observed the nature of the reactions in Fig. 1-4, we may infer certain general facts as follows: (i) the fusion of two spherical nuclei yields the largest $\sigma_{ER}$, (ii) the second largest $\sigma_{ER}$ is obtained when both the projectile and target are deformed, (iii) fusion of either projectile or target nucleus is spherical or deformed yields the lowest $\sigma_{ER}$, (iv) the highest $\sigma_{ER}$ that can obtained is dictated by the properties of the entrance channel parameters such as Coulomb interaction parameter, mean fission parameter, charge asymmetry parameter, and mass asymmetry parameter. Furthermore, to get systematic trends of the curves seen in Fig. 1-4 every curve has been fitted with a suitable mathematical expression as given in Table I. These empirical equations can be useful while planning an experiment for synthesizing any new superheavy elemental isotopes. Before doing so we have first examined the reactions employed in the synthesis of SHEs $Z = 119 - 120$ and thus we have provided the expected $\sigma_{ER}$ in Table II. This exercise results in a gloomy picture because of the large uncertainties. The uncertainties are larger than the empirically predicted values. When the uncertainty with - sign is larger than the value itself, it implies that the net value is closed to zero and not at all negative. The similar scenario is observed in
FIG. 4. Variation of measured production cross-section as a function of mass asymmetry parameter for different projectile-target combinations [32][38][59] as mentioned in Fig[5]

TABLE I. The equations fitted with the data shown for In $\sigma_{ER}(x = z, \chi_m, \eta_A, \alpha_Z)$ as a function of the entrance channel parameter $x = z/\chi_m/\eta_A/\alpha_Z$ in Fig.1-4. Here, the first column stands for the type of reaction (TOR) with respect to the deformation parameter of the projectile and target nucleus. The last column gives the uncertainty in evaporation residue cross-section ($\sigma_{ER}$) as obtained from the least square fittings.

| TOR | In $\sigma_{ER}(x = z)$ | Range of atomic no. Z | Range of entrance channel parameter uncertainty |
|-----|------------------------|-----------------------|-----------------------------------------------|
| S-S | $-1.62 +4.55x-16.40x^{-2}$ | $75 \leq x \leq 105$ | $21 \leq x \leq 189$ $\pm 3.88 \sigma_{ER}$ |
| S-D | $-1.71 +6.81x-12.40x^{-2}$ | $75 \leq x \leq 105$ | $21 \leq x \leq 169$ $\pm 4.18 \sigma_{ER}$ |
| D-S | $-2.87 +11.38x-11.50x^{-2}$ | $90 \leq x \leq 105$ | $21 \leq x \leq 231$ $\pm 1.66 \sigma_{ER}$ |
| D-D | $-1.36 +13.38x-11.50x^{-2}$ | $90 \leq x \leq 105$ | $21 \leq x \leq 224$ $\pm 3.68 \sigma_{ER}$ |
| D-D | $-5.25 +12.78x-11.50x^{-2}$ | $825 \leq x \leq 105$ | $75 \leq x \leq 124$ $\pm 3.07 \sigma_{ER}$ |
| S-S | $-2.51 +29.47x+97.55x^{-2}$ | $75 \leq x \leq 105$ | $0.42 \leq x \leq 0.76$ $\pm 1.48 \sigma_{ER}$ |
| S-D | $-2.36 +28.98x-81.01x^{-2}$ | $75 \leq x \leq 105$ | $0.25 \leq x \leq 0.84$ $\pm 1.37 \sigma_{ER}$ |
| D-S | $-5.16 +14.54x-199.55x^{-2}$ | $75 \leq x \leq 105$ | $0.56 \leq x \leq 0.79$ $\pm 1.39 \sigma_{ER}$ |
| D-D | $-1.93 +92.99x-99.55x^{-2}$ | $75 \leq x \leq 105$ | $0.14 \leq x \leq 0.56$ $\pm 1.63 \sigma_{ER}$ |
| D-D | $-5.16 +92.99x-202.54x^{-2}$ | $75 \leq x \leq 105$ | $0.14 \leq x \leq 0.55$ $\pm 1.52 \sigma_{ER}$ |

Table III when we examine the reactions planned for future in different laboratories. To shed useful light on this issue we made use of theoretical tools as discussed below.

Main goal of the present work is to find a way to synthesize the SHE, which is a system having a large number of nucleons having intrinsically a high density of excited states and thus a suitable statistical model can deal with its properties [97]. Excitation energy dependent evaporation residue cross-section with subsequent emission of $x$ neutrons can be well evaluated by the ASM as given by [22][24]

$$\sigma_{ERASM}(E_{cm}, \ell) = \frac{\pi}{k^2} \sum_{\ell=0}^{\infty} (2\ell + 1)T(E, \ell)P_{CN}(E_{cm}, \ell)P_{sur}^{\ell}(E^* + \ell)$$

Where $k$ is the wave number, average angular momentum $\ell = \langle \ell \rangle$, $P_{sur}^{\ell}$ is the survival probability of the compound nucleus to ground state by emitting neutrons or lighter particles, $P_{CN}$ is the compound nucleus formation probability, the excitation energy $E^* = E_{cm} + Q$, the reaction $Q$-value is calculated [88] and $T_{\ell}$ is the barrier penetration factor [22][24], which is a function of $E_{cm}$ and $\ell$. The value of $x$ depends on the $E^*$ used for the reaction. Note that $\ell$ takes a vital role and its value can be deduced as done in Capurro et al. [99]. While the $\sigma_{ER}$ by DNS model [10] is stated as

$$\sigma_{ERDNS}^{Z,A} = \sum_{\ell=0}^{\infty} (2\ell + 1)\sigma_{\ell}^{fus}(E_{cm}, \ell)P_{sur}^{\ell}(E^* + \ell)$$

Where $\sigma_{\ell}^{fus}$ is the partial capture cross-section which represents the transition of the colliding nuclei over the
TABLE III. Prediction on the possible outcome of the fusion reactions planned to synthesize the SHEs Z = 119-120. Type of reaction (TOR) is mentioned in 10th column in commensurate with Table I. Symbol 〈σER(z)〉(pb) is mentioned in the 11th column and β2 carry the same meaning as in Table II. Hence, when the uncertainty with - sign is larger than the value itself, it implies that the net value is close to zero and not at all negative. The energy E\text{ER}^\text{opt} indicates the optimal beam energy in MeV suggested for obtaining the maximum cross-section σ\text{ER}^{\text{opt}}(z) in pb using a particular model calculation.

| Name of lab | Reaction | z | χm | ηa | αz | Proj. | Targ. | Comp. | TOR | 〈σER(z)〉(pb) | E\text{ER}^\text{opt} (zn) |
|-------------|----------|---|-----|-----|-----|-------|-------|-------|-----|---------------|------------------|
| LBNL        | 26Kr + 294 Bi → 119 Pb \_ue [90] | 288.7 | 0.95 | 0.42 | 0.39 | 0 | 0 | 0.072 | S-S | 34.5±3.2 | 313 | 261(1n) |
| LBNL        | 26Kr + 294 Bi → 120 Pb \_ue [84] | 293.0 | 0.96 | 0.41 | 0.38 | 0 | 0 | 0.072 | S-S | 38.7±4.0 | 317 | 269(1n) |
| JINR        | 27Sc + 288 Pb → 119 Uue [90] | 249.0 | 0.84 | 0.69 | 0.65 | 0 | 0.24 | 0.08 | S-D | 1.6±3.1 | 217 | 18(3n) |
| RIKEN       | 27Sc + 288 Pb → 120 Uue [90] | 329.0 | 0.87 | 0.64 | 0.60 | 0.36 | 0.235 | 0 | D-D | 0.2±1.4 | 236 | 0.36nb(3n) |

Coulomb barrier and the formation of the initial DNS. Note that the deformation takes significant role in calculation of the fusion cross section. Ideally the dynamical deformation is required. However, static deformation is only used in the ASM and DNS models. Whereas the shell effect plays its role in the calculation of survival probability for both the models. Details of both the ASM and DNS models for excitation function study have been provided in Supplemental Material [26].

The σ\text{ER}^\text{opt} can be calculated as a function of E\text{cm} using Eqn. (1) or (2) to find the optimum E\text{cm} in obtaining the maximum cross-section for zn channel (σ\text{ER}^{\text{opt}}(z)). We have first tested such possibility using the ASM through a successful reaction 62Ni + 208 Pb → 260 Ds for which the experimental σER at E\text{cm} = 239 MeV is known and thus marked in Fig. 8. At this energy, x = 1 is the major and x = 2 the minor channel yielding cross-section of 4.97 and 0.7 pb, respectively. Hence, total theoretical cross-section 5.67pb is in agreement with the measured cross-section 3.5±1.8pb [18]. Many such comparisons have been compiled in a recent article [100]. With this confidence level, we have calculated the σ\text{ER}^\text{opt} for the reaction 24Sc + 249 Cm → 224 Uue and the results shown in Fig. 8 (b) gives us a notion that the cross-section at the beam energy at which the experiment was carried out provides σ\text{ER}^\text{opt} = 6.4 fb only and thereby, beyond the reach of the experimental sensitivity at that point of time. On the other hand, if experiment be done at 208 MeV, it can lead to σ\text{ER}^\text{opt} = 3.97 pb, which is well within the present detection limit. This σ\text{ER}^\text{opt} versus E\text{cm}, study let us constitute the most vital input for an experiment, we call it the optimal energy criterion: the optimum beam energy for an allowed reaction with respect to the first four criteria must be determined by a suitable calculation to check whether the predicted largest σ\text{ER}^\text{opt} is within the detection limit of a certain setup. This fact can be represented well by the DNS model also as shown in Fig. 8 (c) and (d) and also in Table III and thus a model independent way. Significance of optimal energy selection for SHE synthesis has been experimentally demonstrated very recently by Tanaka et al. [101].

The above mentioned optimal energy criterion has been implemented on the planned reactions made by several labs [91, 102] for synthesizing SHEs Z = 119, 120 using the heavy-ion reactions and shown in Table [III]. Here, it is evident that all these reactions can be utilised...
to measure the $\sigma^{max}_{ER}$ at the optimal energy listed there. A remarkable point is that the first two reactions in Table III lead to a very large $\sigma^{max}_{ER}$ and thus should be attempted at the earliest possible. Furthermore, the optimal energy predicted by the ASM and DNS models differ by 1.9 MeV and the $\sigma^{max}_{ER}$ is the largest at the corresponding optimal energy, they agree within a factor of 8. Hence, we recommend conducting experiments at both the optimal energies to test the power of each calculation.

To summarize, a careful analysis of the experimental facts with the heavy ion reactions provides us certain criteria in the light of deformation parameters of the projectile and target systems (Fig. 1-4) that can be of great value to meet the challenges for synthesizing the new elements in the eighth period of the periodic table as mentioned above. The analysis based on these criteria does yield much less than 1 $\text{pb}$ evaporation residue cross-section for the unsuccessful experiments listed in Table [I] However, these cross-sections suffer from a large uncertainty. Such a scenario is noticed as well for the planned experiments shown on the 11th column of Table III. Root cause of such unusually large errors is nothing but the arbitrary choice of the beam energies used for the experiments. The optimal beam energy for obtaining the maximum evaporation residue cross-section can be chosen from the theoretical excitation function curves from the ASM as well as DNS model (Fig. 5). Furthermore, such curves can be used to find the expected cross-section at the specific beam energies used for the experiments. We saw the optimal energies and the evaporation residue cross-sections from these two models agree well within 9 MeV and a factor of 8, respectively.

Let us consider a very recent paper [103] that describes a despondent fact of not detecting any events from the $^{50}\text{Ti}+^{249} \text{Bk} \rightarrow^{295} \text{Uue} + 4n$ and $^{50}\text{Ti}+^{249} \text{Cf} \rightarrow^{296} \text{Ubn} + 3n$ reactions at cross-section sensitivity levels of 65 and 200 $\text{fb}$, respectively, at a beam energy of $E_{lab} = 281.5$ MeV. We provide theoretical cross-sections using ASM and DNS models for these reactions at this energy as well as at the optimal energies by each model in Table IV. Both the models predict a few tens of $\text{fb}$ cross-section at the beam energy used. Thus, the cause of debacle is well explained. Interestingly, the cross-sections shoot up to at least a few $\text{pb}$ at the optimal energies. Similar analysis of the proposed reactions in different labs shown in Table [III] gives us a pretty green signal. Especially, first two reactions in the Table producing $Z = 119$ can have a few hundred $\text{pb}$ cross-sections. Hence, we may see shortly that new members of the periodic table are being discovered with the presently available facility in the laboratories.

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See Supplemental Material for (a) Definitions of entrance channel parameters, (b) Advance Statistical Model and (c) Dinuclear System Model at http://link.aps.org/supplemental/............

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