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How accurately can we predict synthesis cross-sections of superheavy elements?

David Boilley,^{1,2,*} Bartholomé Cauchois,^{1,2} Hongliang Lü,^{1,2} Anthony Marchix,³ Yasuhisa Abe,⁴ and Caiwan Shen⁵

¹*GANIL, CEA/DRF-CNRS/IN2P3, Caen, France*

²*Normandie Université, Unicaen, Caen, France*

³*Irfu-CEA, Université Paris-Saclay, Gif-sur-Yvette, France*

⁴*RCNP, Osaka University, Ibaraki-shi, Japan*

⁵*School of Science, Huzhou University, Huzhou, China*

Synthesis of superheavy elements beyond oganesson is facing new challenges as new target-projectile combinations are necessary. Guidance from models is thus expected for future experiments. However, hindered fusion models are not well established and predictions in the fission barriers span few MeVs. Consequently, predictions are not reliable. Strategies to constrain both fusion hindrance and fission barriers are necessary to improve the predictive power of the models. But, there is no hope to get an accuracy better than one order of magnitude in fusion-evaporation reactions leading to superheavy elements synthesis.

Keywords: superheavy elements; nuclear reactions; uncertainty analysis.

I. INTRODUCTION

After the recent successes which lead to fill-up the last line of Mendeleev's periodic table [1], the synthesis of superheavy elements is facing new challenges. The heaviest synthetic elements have all been created in collisions of two heavy nuclei. For a historical review, see Ref. [2]. However, one has to find new target-projectile combinations to extend further the periodic table. For cold fusion reactions, the expected cross-sections are too low with present facilities to expect to synthesise a new element in a reasonable timeframe. And, for hot fusion reactions, there is no available target in sufficient quantity anymore to be associated with the ⁴⁸Ca beam. Heavier projectiles must be used. Thus, one has to find new optimum target-projectile combinations. Guidance from models is expected to optimise future experiments, and various predictions have been continuously published in the scientific literature. Accurate predictions are necessary as a small change in the cross-section could mean months of beam time for an experiment.

However, a direct comparison shows that predictions disagree with each other [3, 4] even if the models can reproduce existing data. One of the reasons is the so-called fusion hindrance, i.e. the strong reduction in the fusion cross-section with respect to what is calculated by a simple extrapolation of fusion models with light nuclei. Its origin is well understood and it is now widely acknowledged that the dynamical trajectory for the fusing system must pass over a conditional saddle point in a multidimensional space in order to form a compound nucleus, in contrast to light systems for which the conditional saddle point lies outside the point of hard contact in heavy-ion reactions. Dissipation also plays a crucial role to understand the fusion hindrance. But, there is no consensus on the dynamical models, leading to large discrepancies in predictions.

The total fusion-evaporation cross-section is a combination of three steps described by three different models, namely the

capture cross-section that brings the two nuclei in contact, the formation probability to reach the compound shape and the survival probability accounting for neutron evaporation from this excited compound nucleus which competes with the predominant fission decay mode:

$$\sigma_{ER} = \sigma_{cap} \times P_{CN} \times P_{sur}. \quad (1)$$

Capture and survival phases can be described by extrapolated models used for the fusion of light nuclei. They are supposed to be the best known parts of the reaction. Thus, the physics used to estimate σ_{cap} and P_{sur} is well established; however, parameters entering the models are not well constrained leading to uncertainties in the predictions.

The formation probability that is very specific to heavy ions collisions, is responsible for the hindrance phenomenon. There is no consensus on the dynamical model nor on the parameters. The physics used to describe the formation phase faces several open questions, and, as pointed out in Refs. [3, 4], P_{CN} calculated by various models spans two or three orders of magnitude. See Fig. 1. Thus, there is no hope to produce reliable predictions without assessing the formation step.

References [3, 4] also show that once multiplied by σ_{cap} and P_{sur} , all models converge to experimental data. This means that the uncertainties in σ_{cap} and P_{sur} are large enough to compensate the discrepancies of the various formation models and can be adjusted to get experimental data [5, 6].

Thus, to improve the predictive power of the models, we also have to find ways to reduce uncertainties in σ_{cap} and P_{sur} . Capture cross-section can be directly measured and the models are well constrained. Discrepancies between two reasonable models are lower than an order of magnitude [5]. Regarding the survival probability, the decay of the compound nucleus formed in the collision is dominated by fission. This means that a small change in the fission width will not affect much the fission probability but will induce great variations of the survival probability. Therefore, it is not a surprise that the uncertainty analysis performed in Refs. [5, 6] has led to pin down the fission width as a key parameter that needs to be assessed in order to improve the predictive power of the models.

* Corresponding author, boilley@ganil.fr

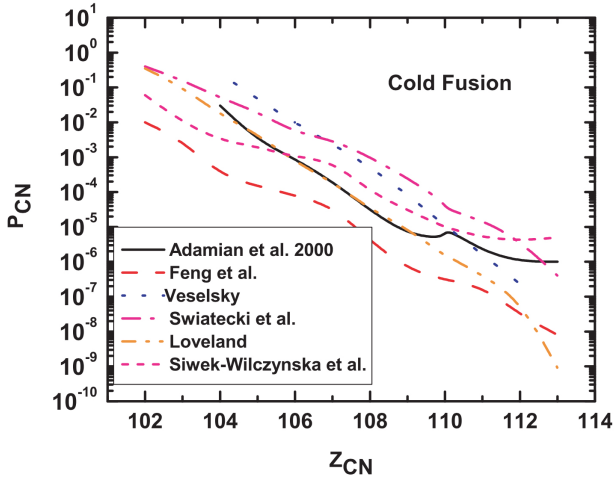


Fig. 1. Formation probability for cold fusion reactions as a function of the charge of the compound nucleus calculated by various models. Figure reproduced from Ref. [3]. See references therein for the models.

In this article, we shall focus on the uncertainty analysis of the survival probability and propose strategies to constrain the formation probability that accounts for the fusion hindrance.

II. UNCERTAINTIES IN THE SURVIVAL PROBABILITY

A. Various predictions

Compound nuclei formed during the collision decay through neutron evaporation or fission. Thus, the $1n$ survival probability is nothing else than a branching ratio

$$P_{1n} = \frac{\Gamma_n}{\Gamma_n + \Gamma_f}, \quad (2)$$

where Γ_n and Γ_f stand for the neutron evaporation width and the fission width respectively. The latter dominates. They can be calculated by a standard statistical model; see e.g. Ref. [7]. Uncertainty in the survival probability is easily deduced from the uncertainties in the widths, using the usual propagation formula:

$$\left[\frac{u(P_{1n})}{P_{1n}} \right]^2 = \left[\frac{\Gamma_f}{\Gamma_n + \Gamma_f} \right]^2 \left(\left[\frac{u(\Gamma_n)}{\Gamma_n} \right]^2 + \left[\frac{u(\Gamma_f)}{\Gamma_f} \right]^2 \right). \quad (3)$$

Here, we have assumed that Γ_n and Γ_f are independent from each other. When the ratio Γ_f/Γ_n is large, the relative uncertainty in the survival probability reaches its maximum value. This is very intuitive: as the fission decay mode dominates, evaporation events are very rare. Thus, a small change in the fission width will affect much the survival probability.

Uncertainty in the neutron evaporation width and in the fission width contributes equally. However, Γ_n mainly depends on the neutron separation energy, whereas the fission width mainly depends on the fission barrier, the damping energy of

the shell correction energy and the friction coefficient when taken into account. None of these parameters can be directly measured and are not well determined, inducing large uncertainties in the fission width and then, survival probability. As shown in Refs. [5, 6], the fission barrier is the dominating quantity among these three parameters.

Fission barriers can be calculated by various microscopic or macroscopic-microscopic models, while the other two parameters are generally adjusted to reproduce experimental data. Comparison between different calculations [8, 9] has shown that predictions in fission barriers, which is the most sensitive parameter, span few MeV. Consequently, calculated survival probabilities can differ by several orders of magnitude.

Even if one restricts the comparison to fission barriers that share the same shell correction energy, the predicted values span 1.5 MeV and can induce two orders of magnitude differences in the fusion-evaporation cross-sections [5, 6]. In one case, the fission barriers were estimated by the old method, i.e.

$$B_f = B_{LDM} - \Delta E_{shell}, \quad (4)$$

where B_{LDM} is the fission barrier calculated with a Liquid-Drop-Model and ΔE_{shell} is the shell correction energy at the ground state. The other case corresponds to fission barriers calculated directly by the same macroscopic-microscopic model.

Although the survival probability is the best understood part of the reaction leading to the synthesis of superheavy elements, the ambiguities in the fission barrier lead to discrepancies that span several orders of magnitude. This is more than for the formation probability that is supposed to be less understood.

Thus, reliable predictions require accurate fission barrier predictions. How well can we predict such a sensitive parameter?

B. Best estimate

Assuming that the model predicting the fission barrier is correct, there are still ambiguities in its parameters that induce uncertainties in the calculated physical quantities. There are few uncertainty analyses of the theoretical evaluation of the fission barrier. In Ref. [10], a very simple micro-macro model was used and leads to an uncertainty in the fission barrier of about 0.5 MeV. Another estimate based on a Bayesian analysis of a DFT model leads to a similar value for the uncertainty in the fission barrier of ^{240}Pu [11].

All these estimates rely on a fitting procedure on nuclear masses and give an uncertainty proportional to the RMS of the fit [12]. Whatever the model, this RMS has remained over few hundreds of keV this last decade. There is little hope to get an order of magnitude better. Thus, the uncertainty in the fission barrier for a given model will also remain at few hundreds of keV.

Constraint from experiments is possible although direct measurement of the fission barrier is not an easy task. The

157 heaviest nucleus which fission barrier has ever been measured
 158 is ^{254}No using the gamma multiplicities. Uncertainty at spin
 159 0 is estimated to be about 0.9 MeV [13–15]. This value is
 160 extrapolated from measurements at higher spins. The fission
 161 barrier can also be extracted by inverting a statistical decay
 162 model, namely the Kewpie2 code [7]. The result is model
 163 dependent and one has to be careful when using the value
 164 in another model. In particular, the extracted fission barrier
 165 depends on the value of the other parameters such as the fric-
 166 tion coefficient. An analysis based on Bayesian inference was
 167 performed to deduce the fission barrier from invented exper-
 168 imental data [16]. The uncertainty in the barrier depends on
 169 the experimental uncertainty and the number of data points.
 170 Values range from 34 keV for quite accurate data to 0.4 MeV;
 171 see Ref. [16] for details.

172 Consequently, it seems that there is an incompressible
 173 value of the uncertainty in the fission barrier that cannot be
 174 overcome. Although less important, other parameters also
 175 have large uncertainties. The friction coefficient is not better
 176 known than 20 years ago. The damping energy of the shell
 177 correction energy has not been carefully investigated since
 178 the Ignatyuk’s prescription [17] in 1975.

179 Because of these uncertainties in key parameters together
 180 with the amplification effect due to the fact that we want to
 181 estimate the cross-section of rare events, it is hardly conceiv-
 182 able to produce predictions with an accuracy lower than about
 183 one order of magnitude for cold fusion reactions. It is more
 184 for hot fusion reactions as we need fission barriers of several
 185 isotopes.

186 III. NECESSITY TO CONSTRAIN THE FUSION 187 HINDRANCE

188 For the less known part of the reaction, i.e. the formation
 189 step responsible for the hindrance of the fusion, there is no
 190 consensus on the model.

191 A. Common understanding of the phenomenon

192 Understanding and modelling fusion hindrance is a long
 193 standing problem. Back in the 1960s, the first predictions
 194 of fusion-evaporation cross-sections were far too optimistic
 195 [18]. Nix and Sierk showed for symmetric reactions that the
 196 dynamical trajectory for the fusing system must pass over a
 197 conditional saddle point in a multidimensional space in or-
 198 der to form a compound nucleus, in contrast to light systems
 199 for which the conditional saddle point lies outside the point
 200 of hard contact in heavy-ion reactions [19]. Later, Świątecki
 201 [20, 21] first introduced a dynamical model that broke with
 202 the idea that the capture process can be understood in terms
 203 of the static interaction of two spheres. The model empha-
 204 sises the role of rapid dynamical deformations away from the
 205 configuration of two spheres in contact. It also includes dis-
 206 sipation that leads to the requirement of even higher energies
 207 to fuse. Soon, experimental evidences confirmed the model
 208 and a lot of efforts were devoted to quantify the reduction in

209 the cross-section due to the fusion hindrance to the synthesis
 210 of superheavy elements. For a review, see Ref. [22].

211 Although dissipation plays a crucial role in understanding
 212 the fusion hindrance [23], associated fluctuations were not in-
 213 cluded so far. The Langevin equation that has been used ex-
 214 tensively to study the fluctuation-dissipation dynamics [24]
 215 has been adopted to study the dynamical diffusion over the
 216 conditional saddle that lies between the contact and the com-
 217 pound configurations [25–29]. This leads to more realistic
 218 dependence of the fusion cross section as a function of energy
 219 and a better estimate of the extremely low cross-sections.

220 Very recently, we showed that the initial condition of the
 221 formation step slips due to the elimination of the fast vari-
 222 ables in a multidimensional description and this affects sig-
 223 nificantly the formation probability [30].

224 This point of view is challenged by so-called DNS model
 225 that was developed later. It is based on a very different con-
 226 cept, assuming a frozen configuration and exchange of nucle-
 227 ons [31–33]. Thus, it is contradictory with the models men-
 228 tioned above based on the evolution of collective degrees of
 229 freedom.

230 Although progress has been made in understanding the fu-
 231 sion hindrance, there is still no consensus on the dynamical
 232 treatment. Thus, it is almost impossible to perform an uncer-
 233 tainty analysis in such conditions. Moreover, many quanti-
 234 tative ambiguities remain. What is the barrier height of the
 235 conditional saddle that has to be overcome? It is clear that a
 236 small change in its value will have a great impact on the for-
 237 mation probability. What is the dissipation strength? What is
 238 the role of structure effects?

239 A comparison between models can give some insights on
 240 the amplitude of the discrepancies. Figure 1, reproduced from
 241 Ref. [3], shows that the formation probability for cold fusion
 242 reactions calculated by various models spans two or three or-
 243 ders of magnitude. Consequently, to improve the predictive
 244 power of the models describing the whole reaction leading to
 245 the synthesis of superheavy elements, one must constrain the
 246 formation probability responsible of the fusion hindrance.

247 B. Strategies to constrain the formation probability

248 Fusion-evaporation residue cross sections are of no help
 249 to assess the formation mechanism because of the survival
 250 probability that is also not well constrained as explained in
 251 the previous section. To get rid of the decay step, one should
 252 focus the step before in the reaction, i.e. the fusion cross-
 253 section.

254 The channel competing with the formation is the so-called
 255 quasifission process. It consists in reseparation into two frag-
 256 ments without reaching the compound state. On an experi-
 257 mental point of view, of course, understanding the competi-
 258 tion between quasifission and fusion is very important. How-
 259 ever, it is very difficult to distinguish the quasifission frag-
 260 ments from the fission ones as they have similar mass distri-
 261 butions. The two processes differ by their time scale and thus
 262 angular distribution [34]. Consequently, one lacks of reliable
 263 experimental fusion cross-sections.

One idea is to consider that reaction dynamics models must strive to reproduce experimental data on quasifission. However, constraining models on the dominating channel leads to unavoidable uncertainties. And, as for fission-evaporation competition, these uncertainties in quasifission modelling will have a large impact on the rare fusion events. Thus, reproducing quasifission data will definitively help to constrain models but might not allow to get a better agreement in the formation probability predictions.

Alternatively, hindrance could be constrained by comparing reactions leading to the same compound nucleus, one being hindered and the other not, in order to get rid of the ambiguities in the decay phase of the reaction. Such studies, currently under development, should naturally include uncertainty analysis.

IV. CONCLUSION

Although it is a difficult task, predictions of reaction cross-sections are useless without an uncertainty analysis [35]. Moreover, uncertainty analysis is a powerful tool to built up a hierarchy of the various sloppy points of a model. This leads to show that predictions of superheavy production cross-sections mainly suffer from the large discrepancies in the hindrance phenomenon modelling and fission barriers that can both affect the results by orders of magnitude.

There are two ways to estimate the accuracy of predictions. First is a comparison between various models. This leads to an estimate of the error in modelling. However, such a comparison can appear to be too pessimistic as some mod-

els might simply be wrong. For example, large fission barriers appearing in the comparison of Refs. [8, 9] would mean almost stable nuclei that would be easy to synthesise as the cross-section would benefit from the large survival probability. Second, assuming that the model is correct, we can perform an uncertainty analysis of the predictions in order to estimate the dispersion of the results due to the lack of constraints on many parameters. Such an analysis is too optimistic as it assumes that the model is correct. Therefore, it is not possible to apply it to the whole reaction as there are still too many open questions on the physics of the formation phase. However, for the sole survival step, there is no hope to get predictions more accurate than an order of magnitude. This is a very pessimistic finding in the case of superheavy elements that are produced in handful numbers.

If the absolute value of the predicted cross-sections cannot be accurately estimated, the ratio between two values will benefit from the strong correlation between them. This could be cross-sections at different energies or with different target-projectile combinations. Thus, the predicted trends would be correct. However, estimating the uncertainty in the ratio requires a careful evaluation of the covariances. This will be one of the priorities of our future research.

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