Survival-mediated capture and fusion cross sections for heavy-element synthesis

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Abstract

Formally, the cross section for producing a heavy evaporation residue, σ_{EVR}, in a complete fusion reaction is given as

$$\sigma_{\mathrm{EVR}}(E) = \frac{\pi h^2}{2\mu E}\sum_{\ell=0}^{\infty}(2\ell+1)T(E,\ell)P_{\mathrm{CN}}(E,\ell)W_{\mathrm{sur}}(E,\ell) \tag{1}$$

where E is the center of mass energy. P_{CN} is the fusion probability and W_{sur} is the probability that the compound nucleus will decay by emitting particles rather than fissioning. The first term represents the capture cross section. Notice that all terms depend on ℓ and the cross section depends on the product of these terms, which are not separable. When W_{sur} is zero or a small number due to low fission barriers at high angular momenta, the capture and fusion terms will be limited. For a series of $\sim$ 287 reactions leading to heavy evaporation residues with $Z_{CN} \leq 110$, we point out the implications of this fact for capture cross sections for heavy element formation reactions. From a comparison of calculated and measured evaporation residue cross sections we deduce values of the fusion probability, P_{CN}, for some of these reactions.

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I. INTRODUCTION

As shown in equation 1, the cross section for producing a heavy evaporation residue in a complete fusion reaction can be written as a non-separable product of three factors, that express the capture cross section, the fusion probability and the survival probability. One expects the survival probability to depend sensitively on the spin ℓ with low or zero fission barriers for higher ℓ values. By this we mean that many partial waves result in capture, but the higher partial waves result in non-surviving events. Thus, in the product of terms in equation 1, only those terms with a high survival rate, i.e., low spin, are relevant. We show, in Figure 1, the calculated spin dependence of the capture cross section for the reaction of ^{3}He + ^{233}U and the calculated spin dependence of the evaporation residue cross section for the 5n channel [1, 2]. The surviving spins are restricted due to angular momentum effects and thus the spin distributions in the capture cross sections are significantly different from those of the evaporation residues.

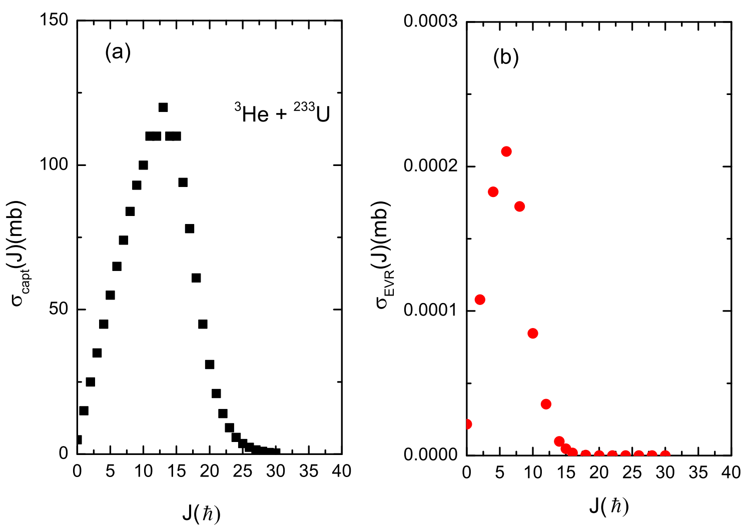

FIG. 1: Spin dependence of the (a) calculated capture cross section and (b) evaporation residue formation cross section for ^{3}He + ^{233}U. From [1, 2]

In this paper, we examine the impact of the restrictions on spin placed by the survival probabilities upon the effective capture and fusion cross sections for heavy element formation. By the term "effective " capture cross section, we mean the capture events resulting in non-zero survival probabilities. We do this in the context of a compilation of a large number of evaporation residue cross sections for heavy element formation. By using a simple formulation of the spin dependence of the capture cross sections and modern calculations of the survival probability of heavy evaporation residues, we attempt to deduce the impact of this spin restriction upon the synthesis of heavy nuclei. A preliminary account of this work has been presented elsewhere [2]. In [2], we introduced the concept of survival mediated cross sections and showed some examples of these effects. In this work, we show the full details of the calculations and draw some new conclusions, regarding P_{CN}, from the calculations.

II. METHODOLOGY

As explained in [2], the formalism for calculating the survival, against fission, of a highly excited nucleus is relatively well understood[3]. One starts with a single particle model [4] of the level density in which one allows the level density parameter to be a function of the excitation energy. Masses and shell corrections are taken from [5]. The deformation dependent collective enhancement of the level density is taken from [6]. The decay widths for decay by neutron, charged particle and γ-emission are calculated with standard formulas. Corrections for Kramers effects [7] are made to the fission widths. The fission barrier heights are calculated using liquid drop barriers and excitation energy dependent shell corrections.

The uncertainties in these calculations of W_{sur} are discussed in [2, 8, 9] Fission barrier heights are known to within 0.5 - 1.0 MeV [9]. A change of fission barrier height by 1 MeV in each neutron evaporation step can cause an order of magnitude uncertainty in the 4n-channel. To minimize sensitivity to this factor in this paper, we have restricted our attention to nuclei where $Z \leq 110$ where the barrier heights are better known. In the same vein, we have not treated reactions where the compound nucleus emits 6 or more neutrons.

As explained in [2], we began with the compilation of D$\ddot{u}$llmann of evaluated evaporation residue cross sections for reactions that produce heavy nuclei [10]. This compilation involves a large number of reactions producing nuclei from Z=80 to Z=122, although we have limited our calculations to cases where $Z_{CN} \leq 110$ and the number of emitted neutrons is 5 or less. We have only treated those cases where the experimental data is published in the open refereed literature. For each reaction (projectile, target and beam energy) we calculate the spin dependent capture cross sections using the Bass model [11, 12]. The Bass model is a semi-empirical model of fusion that uses an empirical nucleus-nucleus potential derived from an analysis of a wide range of experimental fusion cross sections. It should be applicable over a wide range of system masses and energies and offers a simple method of calculating a large number of cross sections. The predictive power of this model is expected for most systems to be comparable to experimental accuracy. At low energies, at or below the interaction barrier, the transmission probability is calculated using a parabolic potential [13] with a curvature of $\hbar\omega$ of 3 MeV. For some cases, this procedure fails and a full coupled channels calculation is employed to get the capture cross section and its spin dependence [3, 14, 15]. The input parameters for these calculations are taken from [3]. The survival probability is calculated using the formalism(s) of [3] for each value of the angular momentum, ℓ,. If we assume P_{CN} is 1, then, according to equation 1, we can estimate the evaporation residue formation cross section. If the calculated evaporation residue cross section is significantly greater then the measured cross section for this reaction, we have evidence that P_{CN} is less than 1.

To check the validity of this procedure, we calculated the EVR spin distribution for the reaction ^{176}Yb(^{48}Ca,4n)^{220}Th and compared it to the measured spin distribution of Henning et al. [16] (Figure 2) for the same reaction [2]. The measured and calculated spin distributions for this reaction are in general agreement. A similar agreement between calculated and measured spin distributions for the ^{208}Pb(^{48}Ca, 2n)^{254}No reaction was found in [6] using a calculation model similar to that employed in this work. Further measurements of the spin distributions for the survivors of fusion reactions leading to heavy nuclei would be useful in this regard.

A. Sensitivity of Calculations to the Choice of Calculation Models

We have chosen the Bass model [12] to calculate most of the capture cross sections in this work because of the simplicity of the model and its established [12] predictive power. What if we had used the computationally more complex coupled channels method to do these calculations? We show, in Figure 3, a comparison of the calculated spin distributions of the capture products using the Bass model and coupled channels calculations for a representative set of reactions. While there are differences in the calculated spin distributions using the two methods, there are no qualitative differences.

The issue of the uncertainties in the calculated spin distributions of the survivors of the

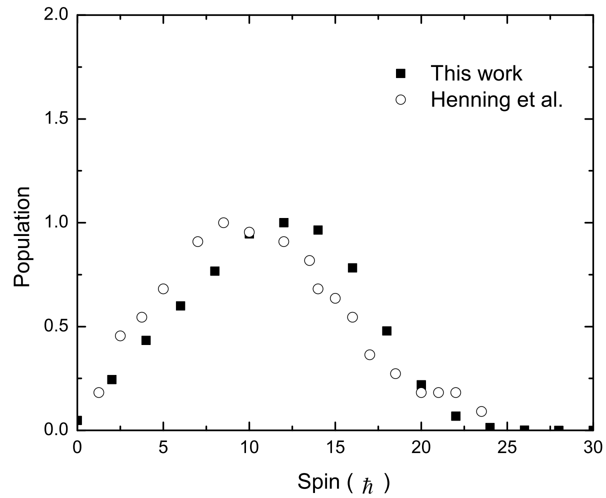

FIG. 2: Spin dependence of calculated and measured evaporation residue formation cross sections for the ^{176}Yb(^{48}Ca,4n)^{220}Th reaction. From [2].

fission-particle emission competition is more complex [9]. From the point of view of this work, we show, in Figure 2, the reasonable agreement between the calculated and measured spin distributions for a known case of a heavy element synthesis reaction. A similar results was found for the ^{48}Ca + ^{208}Pb reaction [6]. Loveland [9] has made a detailed examination of the strengths and weaknesses of models such as [3] and has placed limits on how well these models work.

III. RESULTS

For each reaction studied, we have tabulated the A and Z of the projectile, the target nucleus and the compound nucleus and the bombarding energy corresponding to the peak of the excitation function. We have also tabulated the calculated mean spin for capture process and mean spin of the calculated evaporation residues, and the measured evaporation residue cross section. These tables are found in the Supplemental Material[17].

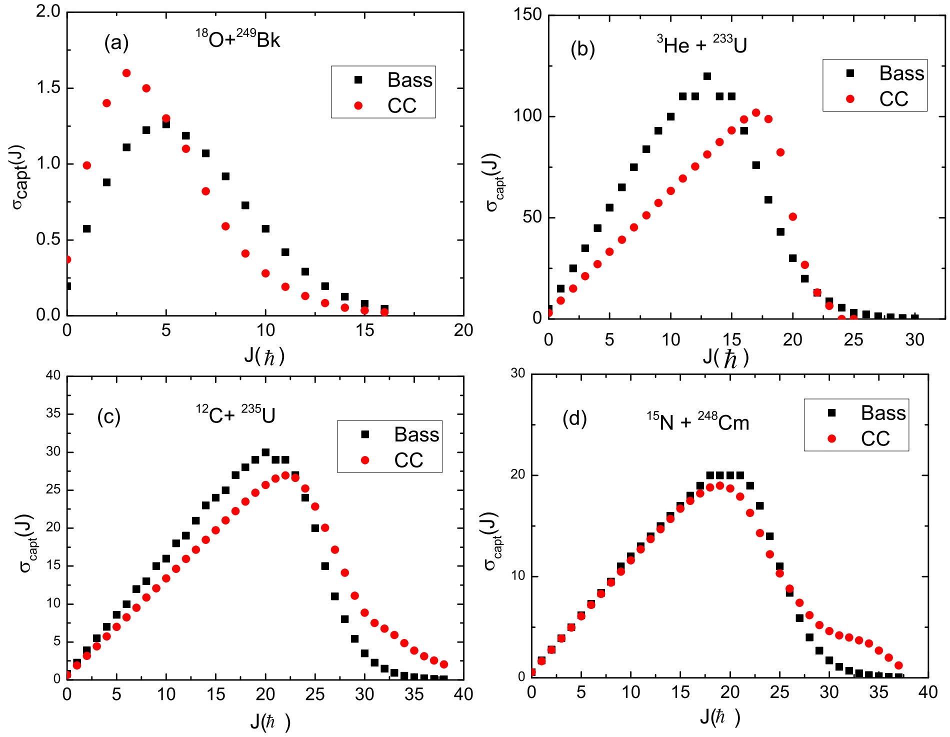

FIG. 3: A comparison of the calculated capture product spin distributions using the Bass model and coupled channels calculations (CC) for a representative set of reactions, (a) 91 MeV ^{18}O + ^{249}Bk, (b) 44.7 MeV ^{3}He + ^{233}U, (c) 71 MeV ^{12}C + ^{235}U and (d) 88 MeV ^{15}N + ^{248}Cm.

IV. DISCUSSION

As stated above, we have calculated the spin dependent capture cross sections and the spin dependent evaporation residue cross sections for 287 reactions. Statistical summaries of these calculations can be found in the Supplemental Materials. In this section, we will discuss these calculational results , sorting them by Z_1Z_2 product.

A. $\mathbf{Z_1Z_2 \leq 750}$

There are 68 cases where $Z_1Z_2 \leq 750$. Review of these data shows certain straightforward trends. As one goes from a 3n to a 4n to a 5n reaction, the excitation energy of the fissioning system increases, and the mean spin of the captured system increases significantly.

As a consequence, the surviving evaporation residues show a spin distribution that is an increasingly smaller subset of the products of the capture process. In Figures 4 and 5 we show typical plots of the spin dependent capture cross sections and the spin dependent evaporation residue cross sections for some cases of 3n, 4n and 5n reactions [18, 19] that demonstrate this effect. A further consequence of this behavior comes in evaluating the terms in equation 1. In evaluating these terms (capture, fusion and survival) one must use the spin restricted values of each term–which are quite different than the non spin-restricted values. Consider the 5n capture cross sections shown in Figure 4 and 5. Much of the capture distribution does not survive fission and must be excluded in using equation 1. Similarly calculations of P_{CN} should be spin restricted to be relevant for heavy element synthesis.

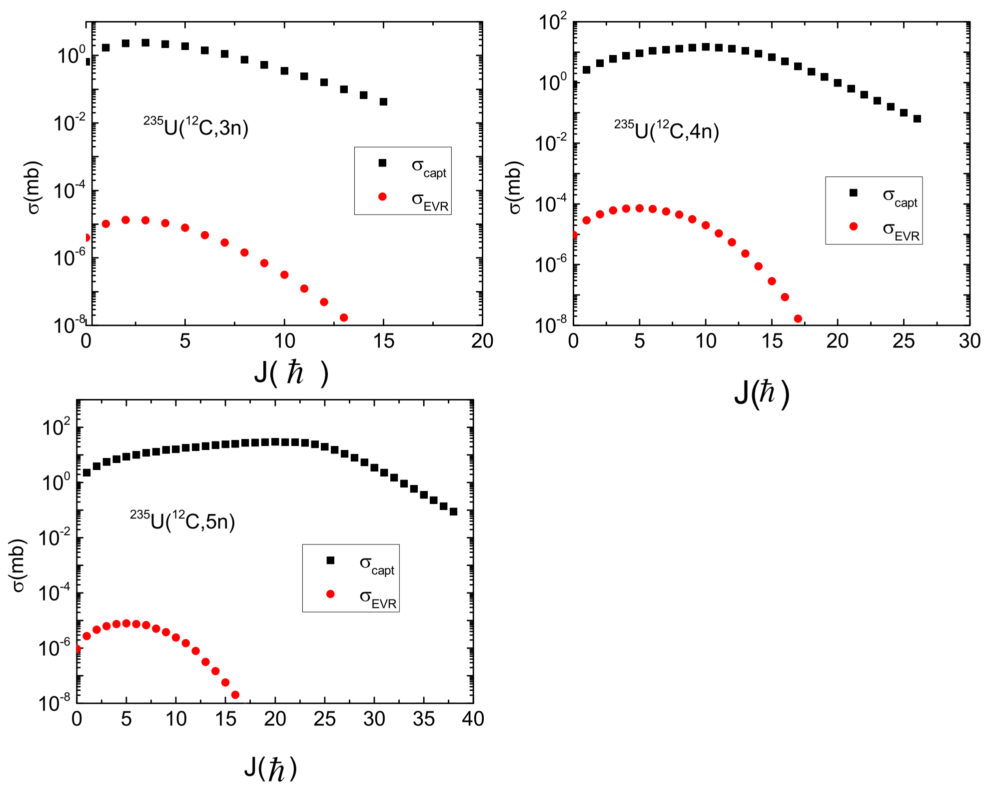

FIG. 4: Spin dependence of calculated capture and evaporation residue cross sections for the ^{235}U(^{12}C, 3-5n) reactions [18]

On average, we found that P_{CN} was 1 for this group of reactions, i.e., the calculated evaporation residue cross section agreed with the measured cross section within experimental

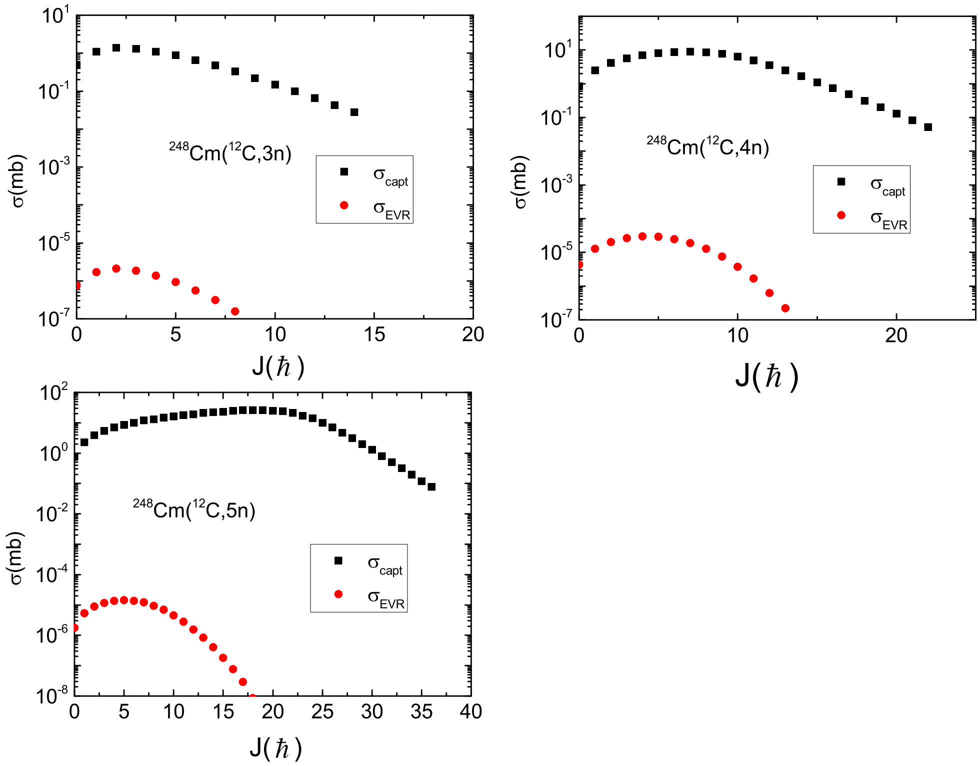

FIG. 5: Spin dependence of calculated capture and evaporation residue cross sections for the ^{248}Cm(^{12}C, 3-5n) reactions [19]

error. (This conclusion was based upon studying the 3n reactions, where these types of calculations should be most reliable.) A general sampling of some typical cases for Z_{CN} = 94-98, 101-103 and 104-105 is shown in Figure 6. [2]

The surviving evaporation residues have a most probable spin of $\sim 5\ \hbar$. The capture reactions have an average spins of 10-20 $\hbar$. The evaporation residue cross sections are significantly less than the capture cross sections. due to the fission of the completely fused system. Capture cross sections for these hot fusion reactions are typically 100-300 mb while the evaporation residue cross sections are $\sim$ nb to μb. Almost all of the reactions with Z_1Z_2 $\leq$ 750 are "hot" fusion reactions. For the few cases of "cold" fusion reactions, the mean spin of the capturing system is about $20\hbar$ while the surviving EVRs have J $\sim 6.7\hbar$

As discussed in[2], it is sometimes assumed that the relevant capture cross sections and P_{CN} factors can be evaluated for J=0. This assumption is not supported by the data shown

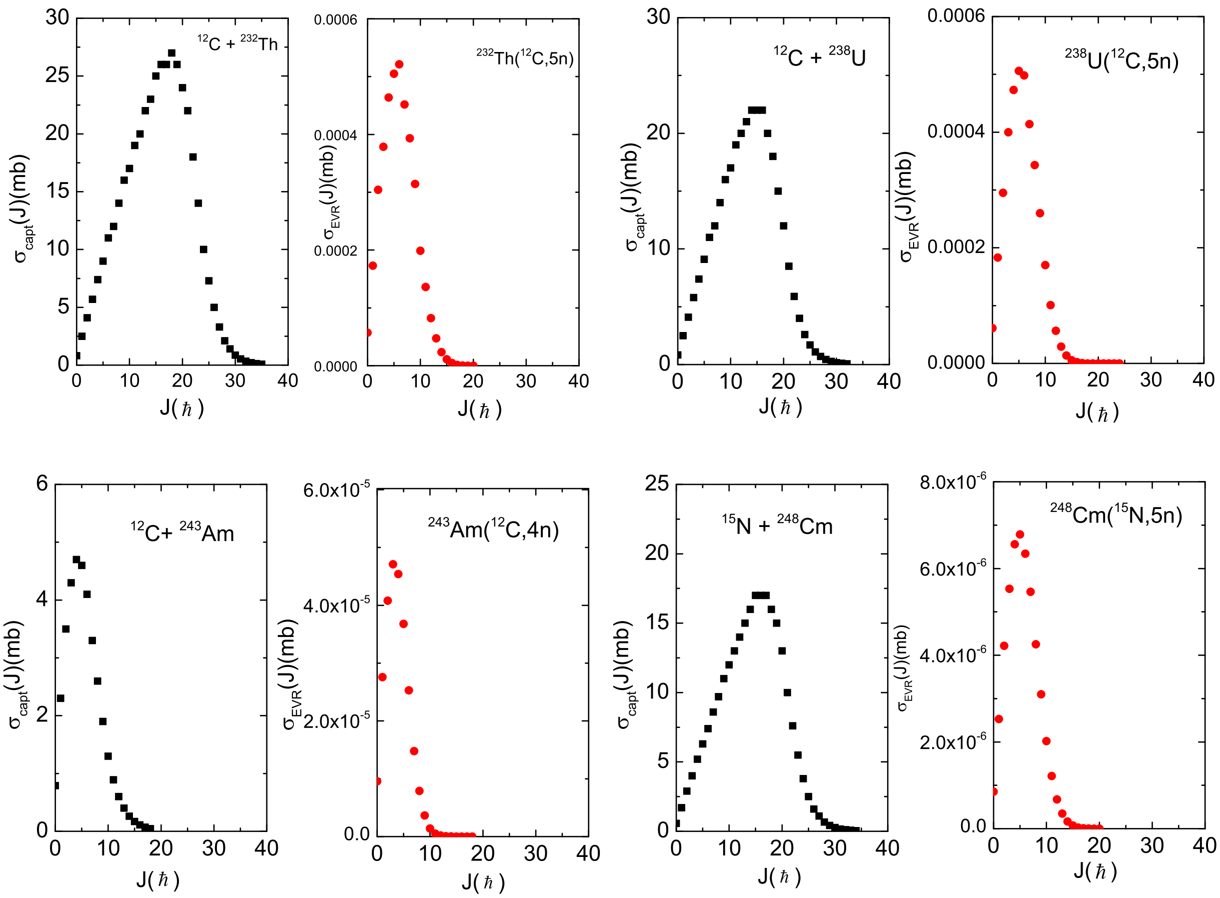

FIG. 6: Spin dependence of calculated capture cross sections and the EVR cross sections for the reactions of ^{12}C + ^{232}Th, ^{243}Am, ^{249}Cf and the reaction of ^{15}N with ^{248}Cm. [18, 20–22] where the lab frame beam energies were 70,73,70, and 86 MeV respectively. From [2]

in Fig.6.

B. $750 \leq Z_1Z_2 \leq 1000$

There are 37 cases in this category. Most of these reactions are sub-barrier reactions with small (nb) cross sections. Apart from the few cases of cold fusion reactions, one observes a smaller difference between the spin dependent capture and evaporation residue cross sections. The ratio of the mean spin of the EVR distribution relative to the capture distribution is 0.60 compared to 0.52 for the previous group. A sampling of these cases is shown in Figure 7 [2]. The mean spin of the surviving evaporation residues is less than that of the capture products. The evaporation residue cross sections are orders of magnitude less than the capture cross sections due to the effect of fission de-excitation. There are no cases of 3n reactions involving actinide nuclei or 1n reactions involving Pb or Bi target nuclei, so no conclusions about P_{CN} for this group are possible.

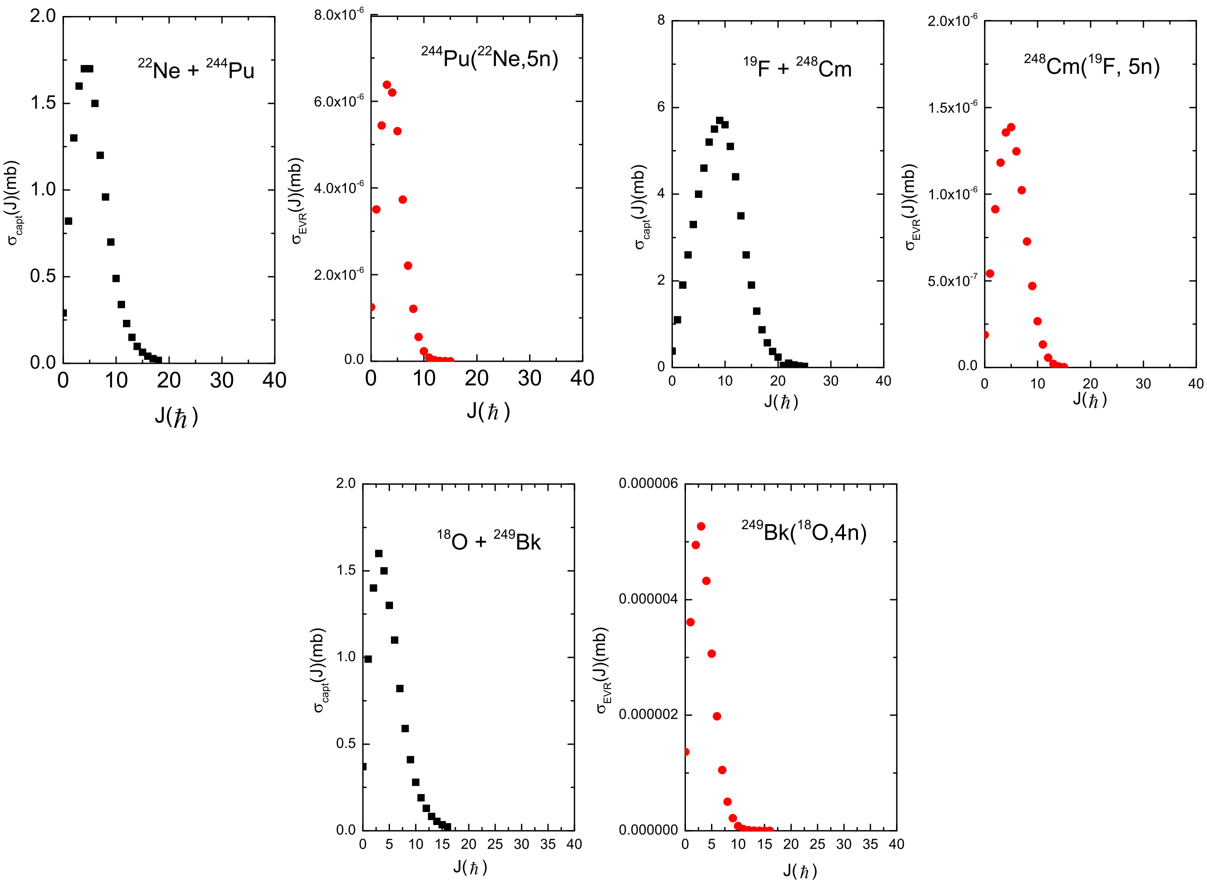

FIG. 7: Spin dependence of calculated capture cross sections and the EVR cross sections for the reaction of ^{22}Ne + ^{244}Pu, ^{19}F + ^{248}Cm and ^{18}O + ^{249}Bk. [23–25] where the laboratory frame beam energies were 114,106, and 93 MeV, respectively. From[2]

C. $1000 \leq Z_1Z_2 \leq 1500$

There are 69 cases in this category and they are almost exclusively hot fusion reactions. The clear case of a cold fusion reaction is the ^{207}Pb(^{40}Ar, 1n) reaction [26] where the calculated mean spin of the surviving evaporation residues is 21.6 $\hbar$ while the calculated mean spin of the capture products is 48 $\hbar$. A sampling of the calculated spin distributions for some randomly selected hot fusion cases is shown in Figure 8. One notices that the survivor distributions are a tiny fraction of the initial capture distributions and are, in fact, remarkably similar for all these reactions. While the calculated values of P_{CN} for this group are consistent with $P_{CN} = 1$, the dispersion of these values is unusually large and suggests that the estimation of P_{CN} from these calculations is not straightforward.

D. $\mathbf{Z_1Z_2 \geq 1500}$

There are 113 cases of reactions in this category. The reactions range from 1n to 5n reactions with several examples of progressions like ^{208}Pb(^{48}Ca, 1-4n)([31]) to ^{209}Bi(^{48}Ca, 1-4n)[32, 33]. In these reactions, the mean spin in the capture cross section distributions increases from $\sim$ 10 $\hbar$ to $\sim$ 45 $\hbar$ while the mean spin of the surviving nuclei is $\sim$ 5 $\hbar$. Similar patterns are observed for symmetric reactions such as ^{100}Mo(^{100}Mo, 1-5n) [33] and ^{50}Ti-based reactions [34, 35] . Use of a ^{86}Kr projectile [85] in $^{121-123}$Sb (^{86}Kr, 3-5n) produces high spin capture products (J $\sim$ 60$\hbar$) but the surviving nuclei are of lower spin (6-7 $\hbar$). The oft studied ^{124}Sn + ^{96}Zr reaction [85] has a mean surviving spin of $\sim$ 6 $\hbar$ while the capture products show mean spins up to 73 $\hbar$. The average EVR spin of the 113 cases in this category is 5.8 $\pm$ 1.1 $\hbar$, despite large changes in the mean spin of the capture products. For the 1n out reactions, the mean evr spin is 6.0 $\pm$ 1.1 $\hbar$.

In this group, there are a number of cases of 1 n reactions, where the uncertainties in the deduced P_{CN} values should be minimal. Also, it is generally thought that systems with $Z_1Z_2 \geq 1600$ should show significant amounts of quasi-fission, making $P_{CN} \leq 1$ [37]. In Fig. 9, we show the deduced values of P_{CN} for 1n reactions in this group as a function of the traditional scaling variable Z_1Z_2. It should be noted that to the extent the techniques used in this paper to deduce P_{CN} are correct, this plot includes a substantial number of new "measurements" of P_{CN} not available from mass angle correlations, i.e., cases where $P_{CN} \leq 0.01$. The scatter in the data for a given Z_1Z_2 value reflects intrinsic uncertainty in our method of deducing P_{CN} and inadequacies in the use of a single variable like Z_1Z_2 to predict P_{CN}. (In Figure 10, we also show the use of an alternate scaling variable, x_{eff} [38] to sort out P_{CN} values, but with no substantial improvement in correlating P_{CN}. At any given value of x_{eff}, P_{CN} is undetermined within a factor of 10-100.)

These new data allow one to test some semi-empirical prescriptions of P_{CN}. For Pb and

Bi based reactions, Zagrebaev and Greiner [39] have proposed that

$$P_{CN}(E,\ell) = \frac{P^0_{CN}(Z_1Z_2)}{1+exp\left(\frac{E^*_B - E^*_{int}(\ell)}{\Delta}\right)} \tag{2}$$

where E^*_B is the excitation energy of the compound nucleus at the Bass barrier and E^*_{int} = $E+Q-E_{rot}(\ell)$, where Q is the fusion Q value and $E_{rot}(\ell)$ is the rotational energy and Δ is 4 MeV.

$$P^0_{CN} = \frac{1}{1+exp\left(\frac{Z_1Z_2-\zeta}{\tau}\right)} \tag{3}$$

where ζ=1760 and τ=45.

To compare our measured P_{CN} values with estimates of this model, we define a comparison metric [40], the theory evaluation factor, **tef**.

For each data point, we define

$$tef_i = log\left(\frac{\sigma_{theory}}{\sigma_{expt}}\right) \tag{4}$$

where σ_{theory} and σ_{expt} are the calculated and measured values of the P_{CN} factors. Then, the average theory evaluation factor is given by

$$\overline{tef} = \frac{1}{N_d}\sum_{i=1}^{N_d} tef_i \tag{5}$$

where N_d is the number of data points.

In Figure 11, we show the tef values for the comparison of the experimental and calculated values of P_{CN} [39] as a function of the scaling variable Z_1Z_2. The average tef value is -0.02, indicating the theoretical description of the P_{CN} factors for Pb and Bi based reactions is very good.

V. CONCLUSIONS

What have we learned from this study?

(a) The survival mediated capture cross sections for a series of 287 heavy element synthesis reactions have a mean associated spin $\sim 5\hbar$ even though the capture cross sections have mean spins ranging from 10 to 70 $\hbar$. (b) In estimating heavy element production cross sections, both the capture cross sections and the P_{CN} factors must be spin mediated, which in the case of the capture cross sections results in orders of magnitude lower cross sections. (c)

These concerns about the effect of spin mediation are more acute in hot fusion reactions compared to cold fusion reactions. (d) By comparing the measured and deduced values of the EVR cross sections for reactions where $Z_1Z_2 \geq 1500$, we have been able to deduce a set of new values of P_{CN} for 1n reactions for situations where ordinary measurements of P_{CN} are not possible. (e) The semi-empirical estimates of Zagrebaev and Greiner for P_{CN} in Pb and Bi based reactions appear to describe our P_{CN} data quite well.

Acknowledgments

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VI. APPENDIX

The Supplemental Materials are a vital part of this study, containing a heretofore unpublished compilation of experimental data and calculations for the ~ 287 reactions studied in this work. To be sure that this material is properly cited as part of this paper, we compile the 92 references cited only in the Supplemental Materials as part of the reference list of this paper. This list includes the following references. [41–63] [64–87] [88–110] [111–132].

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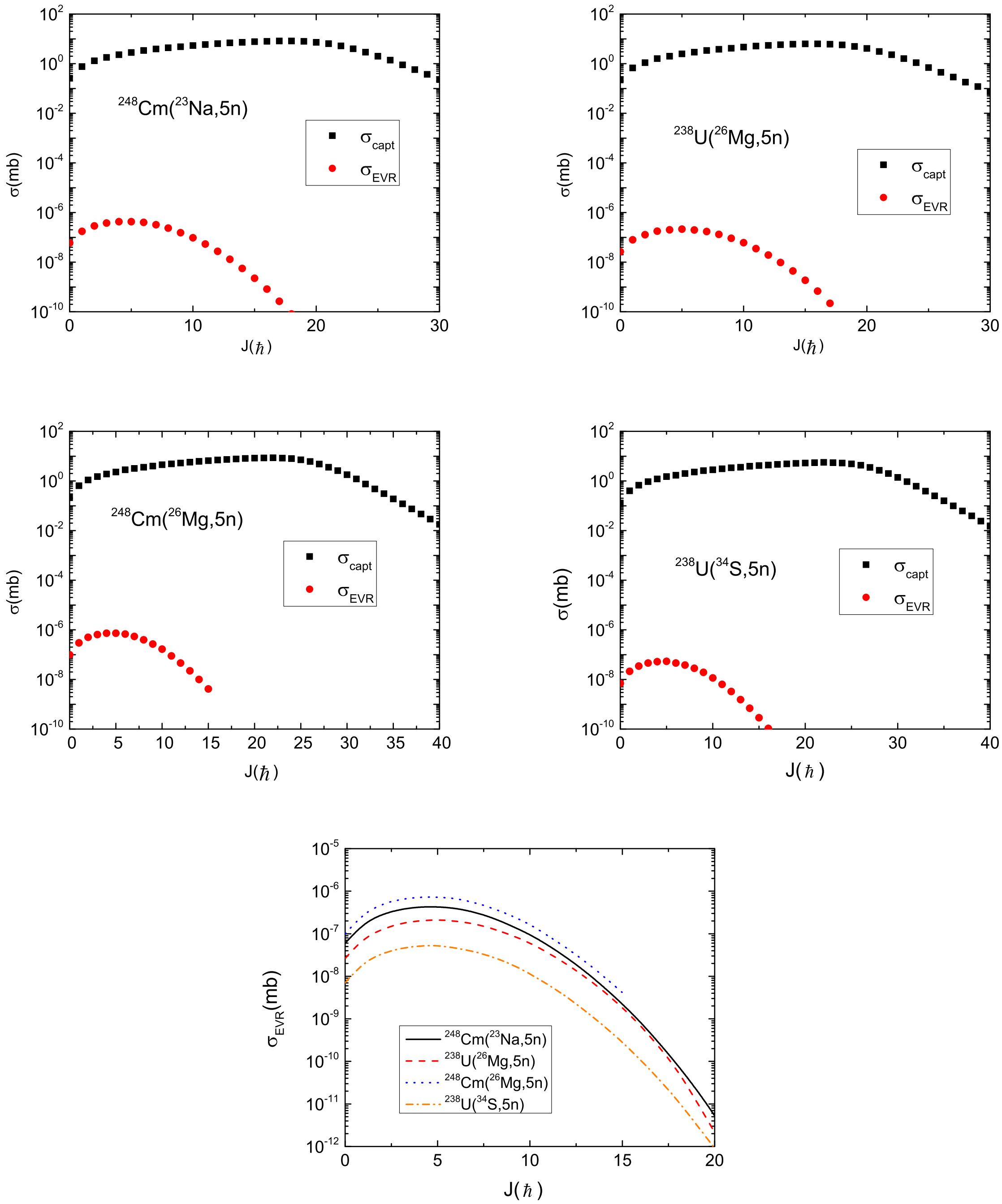

FIG. 8: Spin dependence of calculated capture cross sections and evaporation residue cross sections [27–30] where 1000 $\leq Z_1Z_2 \leq$1500 and the lab frame beam energies were 130,139,144 and 188 MeV respectively.

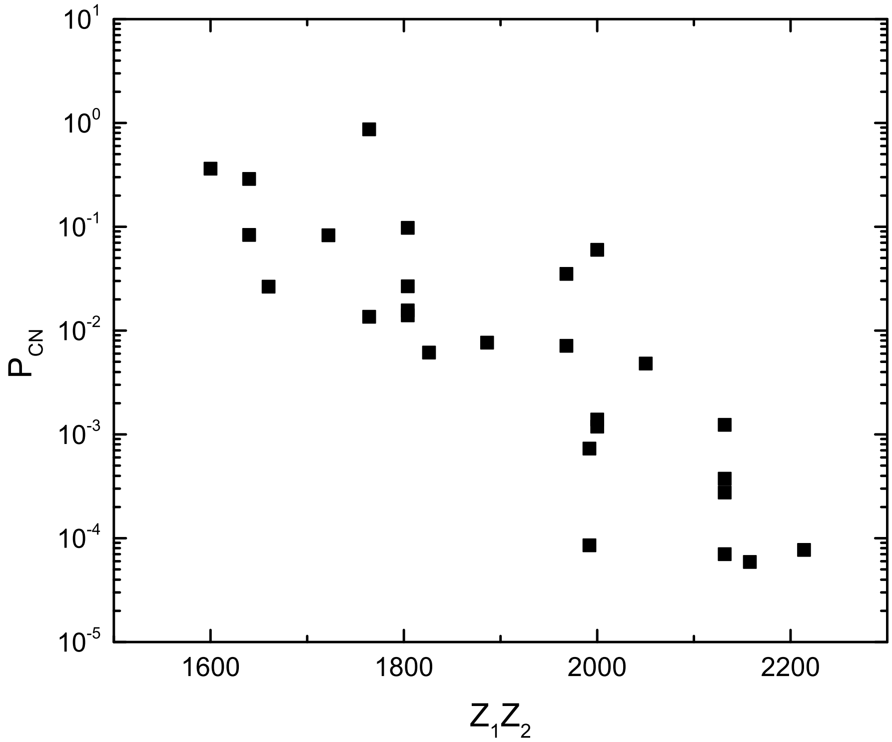

FIG. 9: Deduced P_{CN} values for 1 n reactions with $Z_1Z_2 \geq 1500$.

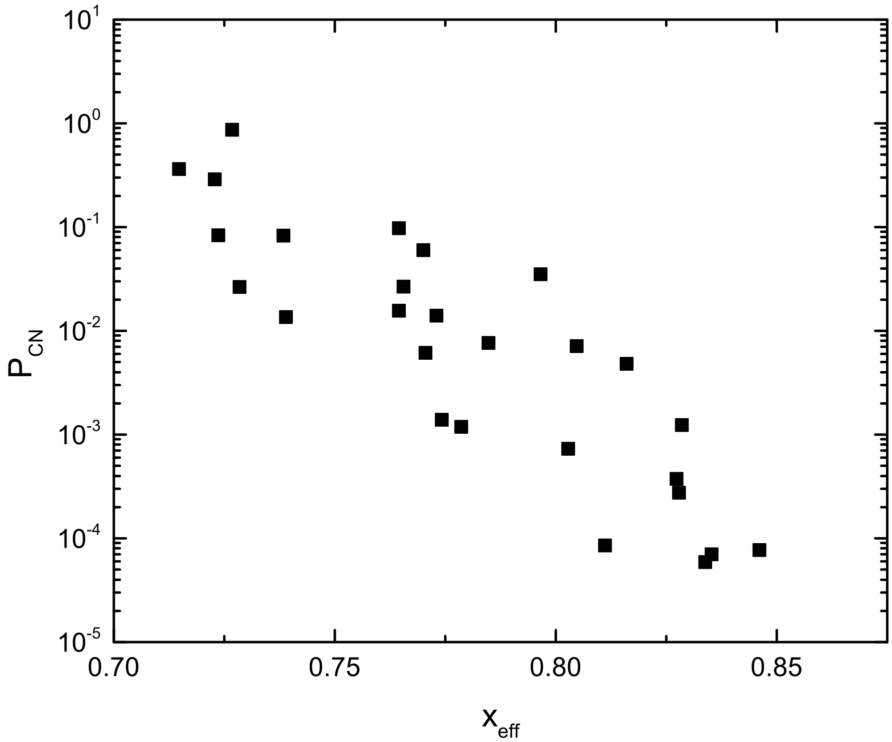

FIG. 10: Deduced P_{CN} values for 1 n reactions with $Z_1 Z_2 \geq 1500$ as a function of x_{eff}.

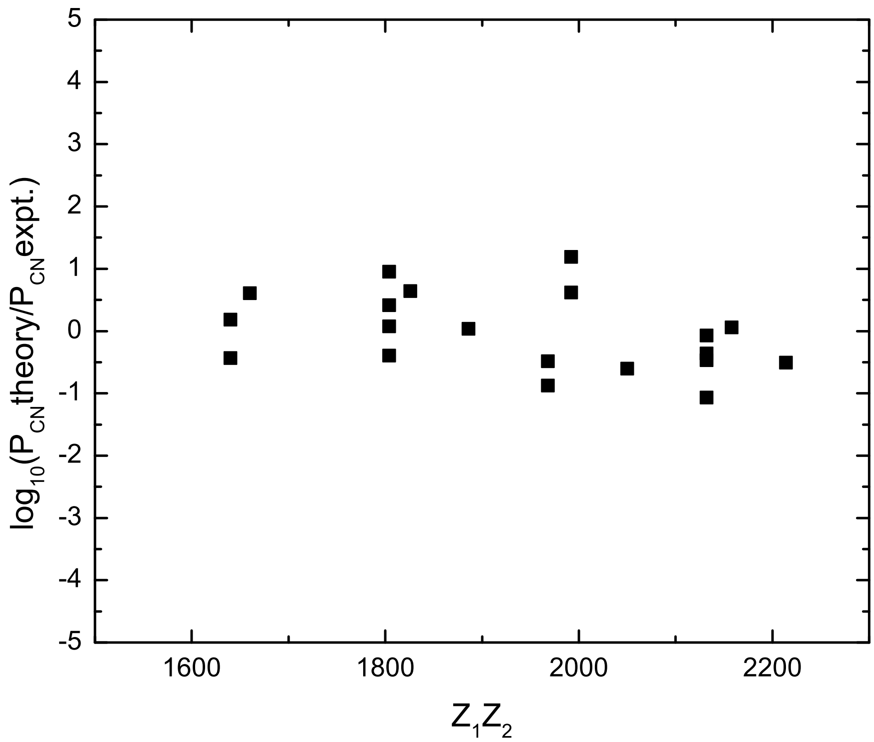

FIG. 11: Comparison of the calculated [39] and measured values of P_{CN} for 1 n reactions with $Z_1Z_2 \geq 1500$ as a function of Z_1Z_2.