Severe Inhibition of Fusion by Quasifission in Reactions Forming ²²⁰Th

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Evaporation residue cross sections have been measured following the fusion of ¹⁶O with ²⁰⁴Pb, forming the compound nucleus ²²⁰Th. These are compared with existing data for the same compound nucleus formed following fusion reactions with ⁴⁰Ar, ⁴⁸Ca, ⁸²Se, and ¹²⁴Sn projectiles. At energies where the reduced cross sections of *xn* evaporation residues would be expected to be the same for all reactions, those for the heavier projectiles are typically a factor of 10 smaller than those for ¹⁶O. This inhibition is attributed to strong competition of quasifission with fusion at the low angular momenta associated with evaporation residue formation.

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The creation of superheavy elements has been a longterm goal of nuclear physics research, following the prediction in 1966 [1] of the existence of an island of stability for nuclei far heavier than those found in nature. This is caused by the stabilizing influence of the spherical proton and neutron shells, which were expected to occur at Z =114 and N = 184, respectively. After having systematically [2] stepped up to element 112, recent experimental evidence [3,4] indicating the successful production (and long lifetimes) of elements 114 and 116 have stimulated increased experimental efforts worldwide in this area.

These elements are much more massive than the heaviest elements which can be used as target nuclei. In order to form them, relatively heavy projectiles must be fused with heavy target nuclei. The attractive short-range nuclear potential can overcome the large Coulomb repulsion even in such heavy systems, giving a potential barrier analogous to the fusion barrier in lighter systems. However, there is only a shallow potential pocket inside the barrier, and during the shape changes following capture, the systems often separate before forming a compact compound nucleus (CN). This process, called quasifission, is characterized by full damping of the initial relative kinetic energy, and significant (but not complete) mass equilibration between the two colliding nuclei [5]. Since passing over the potential barrier is not a sufficient condition for fusion, the barrier will be referred to as the capture barrier. The presence of quasifission inhibits heavy element formation, since those collisions leading to quasifission do not form a compact system, and thus cannot result in a heavy fusion product, referred to as an evaporation residue (ER).

The process of formation of heavy elements can be divided conceptually into three parts. The first step is that the capture barrier must be overcome. For light systems, this leads to formation of a compact CN. For collisions of heavy nuclei, quasifission competes with CN formation, with probability $P_{\rm QF}$. Thus the capture cross section $\sigma_{\rm cap}$ must be multiplied by the probability $P_{\rm CN} = (1 - P_{\rm OF})$ of reaching a compact CN, to give the fusion

cross section. The yield of ERs is then determined by the probability W_{sur} of surviving statistical fission decay from the compact configuration. Conceptually the ER cross section may therefore be written [6]

$$\sigma_{\rm ER} = \sigma_{\rm cap} P_{\rm CN} W_{\rm sur}; \tag{1}$$

here $P_{\rm CN}$ and $W_{\rm sur}$ are weighted averages over the low angular momenta leading to ER formation. Capture may well be described adequately in the coupled-channels framework, which has been successful in describing the fusion of lighter systems [7]. $W_{\rm sur}$ depends on both excitation energy E^* and angular momentum $\ell\hbar$, but should be independent of other entrance-channel conditions. $P_{\rm CN}$ also will depend on E^* and ℓ , but may depend sensitively on entrance-channel conditions such as mass asymmetry. It is important to determine $P_{\rm CN}$ experimentally, to improve our understanding (and thus models) of the competition between quasifission and fusion, allowing prediction of the most favorable reactions for producing superheavy elements.

Experimental studies of quasifission have usually concentrated on measuring mass distributions or angular distributions of the fission fragments themselves. It is established that quasifission gives wider mass distributions [5], and angular distributions more strongly peaked along the beam axis [8] than fission from compact shapes (fusion fission). Evidence of quasifission has been found for projectiles of 24 Mg and heavier in reactions with 208 Pb [8], and also in the mass-asymmetric reactions 16 O + 238 U [9] and 12 C + 232 Th [10], at beam energies below the average capture barrier energy.

Indirect evidence for quasifission has come from measurements of the cross sections of ERs. Yields lower than expected have been attributed to failure to form a compact shape, and have generally been interpreted [11] in the framework of the extra-push model [12]. Comparison of ER yields for Ar-induced reactions with those for more symmetric reactions, forming the same CN with Z between 80 and 90, indicated that extra kinetic energy is needed to force heavy nuclei together, as predicted. Where that energy was provided, through increased beam energy, the ER yields for the asymmetric reactions and the near-symmetric reactions were found to be essentially in agreement [13]. This result was taken to mean that even in the near-symmetric collisions, given the extra kinetic energy, quasifission does not compete with fusion at the low ℓ values leading to ER formation.

This conclusion appears to conflict with the evidence for quasifission seen in the fission properties observed in the much more mass-asymmetric collisions on Pb and U targets, and with more recent precise measurements for three reactions, each forming ²¹⁶Ra [14]. These show evidence both for inhibition of ER yields and for wider fission mass distributions in the reactions ¹⁹F + ¹⁹⁷Au and ³⁰Si + ¹⁸⁶W, compared with those for ¹²C + ²⁰⁴Pb, indicating that quasifission does compete with ER formation at low ℓ values.

To resolve this conflict, measurements of ER cross sections have been made for the reaction ${}^{16}\text{O} + {}^{204}\text{Pb}$, leading to the CN ${}^{220}\text{Th}$, at excitation energies between 30 and 50 MeV. ER cross sections have previously been measured for ${}^{220}\text{Th}$ formed in reactions with ${}^{40}\text{Ar}$ [15], ${}^{48}\text{Ca}$ [13], ${}^{82}\text{Se}$ [16], and ${}^{124}\text{Sn}$ [13] projectiles, at similar excitation energies. By comparing these with the new ${}^{16}\text{O}$ measurements, it will be shown that fusion with all the heavier projectiles is severely inhibited, and thus quasifission does compete with fusion at low ℓ values.

Beams of ¹⁶O with energies from 82.0 to 102.0 MeV, from the 14UD electrostatic accelerator at the Australian National University, bombarded a 204 PbS target \simeq $100 \ \mu g \ cm^{-2}$ in thickness, evaporated onto a $\sim 15 \ \mu g \ cm^{-2}$ C backing foil. The beam was pulsed, with 1 ns bursts separated by 640 ns. To determine the cross sections of ERs, an aluminum catcher foil 1.5 times thicker than the average ER range was placed immediately behind the target [14]. The stopped ERs undergo subsequent α decay from their ground states, with lifetimes from 110 ns to 2.7 min. The decay α particles were detected between beam bursts in an annular solid-state detector at backward angles. The energy spectra were fitted, constraining the relative intensities of primary and daughter decays, allowing determination of the individual xn, pxn, and αxn cross sections. The data were normalized to the yields of elastically scattered beam particles measured in monitor detectors at $\pm 22.5^{\circ}$. The relative solid angles of the detectors were determined by measuring far sub-barrier elastic scattering. Agreement of a measurement for the ${}^{16}O + {}^{208}Pb$ reaction with published data [17] confirmed the accuracy of the absolute cross sections.

Before comparing the ER yields from the reactions, it is desirable to determine whether the excitation energies for the different data sets are consistent. This was achieved by comparing the mean number N_{xn} of neutrons emitted in producing Th ERs. These values, offset in N_{xn} by 3, are plotted in the upper part of Fig. 1. Excellent 282701-2 agreement was found between the present data and the ⁴⁰Ar and ⁸²Se data, the line indicating the best fit. The ¹²⁴Sn data generally lie below this line, but a small energy offset of -2.5 MeV gave excellent agreement in the range 25 MeV $\leq E^* \leq 55$ MeV. The ⁴⁸Ca data agree well for $E^* \leq 40$ MeV, but N_{xn} at the two higher energies falls increasingly below the line. Shifts in E^* of -4.5 and -8 MeV were required to bring N_{xn} into agreement with systematics, which may be associated with beam and/or target degradation. The energy-corrected N_{xn} data are shown in the lower part of Fig. 1. The small scatter indicates that the excitation energy is defined to $\leq \pm 1$ MeV over all the reactions. Having established consistency in E^* , the ER cross sections can be compared.

To determine $P_{\rm CN}$ from experiment, it is necessary to eliminate the dependence of ER cross sections on $\sigma_{\rm cap}$, which will be different for each reaction. The ER cross section can be written

$$\sigma_{\rm ER} = \pi \lambda^2 \sum_{l=0}^{\infty} (2\ell + 1) T_{\ell} W_{\rm sur}(E^*, \ell) P_{\rm CN}(E^*, \ell), \quad (2)$$

where T_{ℓ} is the probability of capture for ℓ , and λ is the reduced de Broglie wavelength. Division by $\pi \lambda^2$ gives the reduced cross section $\tilde{\sigma}_{\text{ER}}$. The ER yield is limited to low ℓ values by the decrease with ℓ of the CN fission survival probability $W_{\text{sur}}(E^*, \ell)$, and also, in principle, of



FIG. 1. Average number of neutrons emitted in producing Th ERs, as a function of excitation energy. The upper data set is offset by 3 neutrons, and shows the data at the E^* values quoted in Refs. [13,15,16]. For the lower data set, E^* has been shifted for the ⁴⁸Ca + ¹⁷²Yb and ¹²⁴Sn + ⁹⁶Zr reactions (see text), to bring them into agreement.

 $P_{\rm CN}(E^*, \ell)$. At beam energies sufficiently high above the capture barriers, T_ℓ is essentially unity for such ℓ values; i.e., the partial waves are fully populated. At these energies T_ℓ can thus be dropped, giving

$$\tilde{\sigma}_{\rm ER} = \sum_{l=0}^{\infty} (2\ell + 1) W_{\rm sur}(E^*, \ell) P_{\rm CN}(E^*, \ell).$$
(3)

If $P_{\rm CN}(E^*, \ell)$ were unity, at the same E^* all reactions should give the same $\tilde{\sigma}_{\rm ER}$, as long as $W_{\rm sur}(E^*, \ell)$ is independent of the entrance channel. The ratio of a measured $\tilde{\sigma}_{\rm ER}$ to that for a reaction with $P_{\rm CN}(E^*, \ell) = 1$ gives directly the factor $\overline{P}_{\rm CN}$, which is an average of $P_{\rm CN}(E^*, \ell)$ over the low angular momenta expected to lead to ER formation. Physically, the fusion inhibition factor $\overline{P}_{\rm CN}$ is the factor reducing the total ER cross section as a result of quasifission competition.

Figure 2 shows $\tilde{\sigma}_{\text{ER}}$ as a function of E^* for all but the ⁴⁸Ca reaction, where only *xn* ER cross sections are available [13]. The excitation functions rise with E^* , then saturate. The rise is associated with the ℓ values leading to ER formation becoming increasingly populated as the beam energy increases, and saturation is reached with full population. The values of $\tilde{\sigma}_{\text{ER}}$ for the ¹⁶O reaction are much higher than those for the heavier projectiles, and the saturation yields show a consistent trend to lower $\tilde{\sigma}_{\text{ER}}$ as the entrance channel becomes more mass symmetric.

Conclusions drawn from these results depend on the emission of neutrons, protons, and alpha particles being independent of the entrance channel, since they can strongly affect $W_{sur}(E^*, \ell)$. They may not be independent due to dynamical effects before and/or after capture. The former are associated with light projectiles, for which incomplete fusion of α -cluster projectiles can cause considerable increases in $\tilde{\sigma}_{\rm ER}$ [14]. The latter might be expected at high excitation energies, where the evaporation lifetimes can be similar to the time taken to attain a compact shape. Since the fusion time is expected to increase with increasing mass symmetry [18], emission of charged particles (favored when the system is highly deformed) could be enhanced for the more symmetric reactions. The importance of these phenomena can be investigated directly from the experimental data. The ratio of the summed xn cross sections to the total ER cross section should be independent of the entrance channel in the absence of such dynamical effects. These ratios are shown in Fig. 3, with each reaction showing increasing charged particle emission with E^* , as expected. For the ${}^{16}\text{O} + {}^{204}\text{Pb}$ reaction, the *xn*/ER ratio is substantially lower than for the other reactions, due to large yields of Ra nuclei. As in the ${}^{16}O + {}^{204}Pb$ reaction [14], this is attributed to incomplete fusion, resulting in the same products as αxn evaporation. For the other reactions, the data show no significant dependence of charged particle emission probability on mass asymmetry.

Because of the incomplete fusion for the 16 O-induced reaction, the yields of the *xn* products should give the





FIG. 2. Reduced ER cross sections as a function of excitation energy. The yields for the ¹⁶O-induced reaction are enhanced by incomplete fusion.

(corresponding to neutron evaporation only) to total ER cross sections. For the ¹⁶O-induced reaction a large yield of Ra nuclei formed following incomplete fusion suppresses the ratio. For the more symmetric reactions, no consistent trend appear as a function of mass asymmetry.



FIG. 4. Reduced xn ER cross sections (giving Th ERs). Above the saturation energy (35 MeV for ¹⁶O) the yields for the ¹⁶O-induced reaction are typically a factor of 10 higher than for the more symmetric reactions.

most reliable information on the dependence of the quasifission probability on the entrance-channel mass asymmetry [14]. The excitation energy dependence of $\tilde{\sigma}_{xn}$ for the five reactions forming 220 Th is shown in Fig. 4. Above the points of maximum yield (where the population of ℓ values leading to ERs saturates) the values of $\tilde{\sigma}_{xn}$ fall with E^* , with a similar slope for each reaction. The fall results from increasing competition from fission and charged particle evaporation, at an increasing number of stages in the evaporation cascade. The most significant feature shown by the data is that above the saturation energy, $\tilde{\sigma}_{xn}$ for the ¹⁶O + ²⁰⁴Pb reaction is typically 10 times larger than those for the more symmetric reactions. Measurements for reactions forming ²¹⁶Ra [14] suggest that the average fusion inhibition factor \overline{P}_{CN} may be less than unity even for an ¹⁶O projectile. However, taking $\overline{P}_{\rm CN} = 1$ for ¹⁶O + ²⁰⁴Pb, for the more symmetric reaction $\overline{P}_{\rm CN}$ is ~0.1. Furthermore, the data suggest a systematic trend to smaller \overline{P}_{CN} with increasing mass symmetry for the heavier projectiles (despite estimated systematic uncertainties of up to $\pm 30\%$ [13]), in agreement with the trends of $\tilde{\sigma}_{\rm ER}$ seen in Fig. 2.

These results show that in forming ²²⁰Th, fusion is inhibited increasingly severely for reactions more symmetric than ${}^{16}\text{O} + {}^{204}\text{Pb}$; significantly most of the inhibition already occurs for the ${}^{40}\text{Ar}$ -induced reaction. This is consistent with a picture of fusion in which the mass asymmetry dependence of the potential energy surface (PES) plays a very important role [6,9,14]. For massasymmetric collisions, the PES favors absorption of the light projectile by the heavy target nucleus, which leads to a compact CN with high probability. In contrast, for asymmetries smaller than the Businaro-Gallone ridge point, the PES favors the lighter nucleus taking mass from the heavier partner, leading to an elongated mass-symmetric configuration. The $\tilde{\sigma}_{xn}$ data show that the probability of forming a compact ²²⁰Th nucleus from this configuration can be smaller than 0.1, even at the low ℓ values associated with ER survival.

This conclusion emphasizes the importance of the entrance-channel mass asymmetry in determining the outcome of a reaction, even when forming a nucleus as light as ²²⁰Th. In reactions forming elements heavier than Th, inhibition of fusion due to quasifission is even more severe. The difference in inhibition due to mass asymmetry for reactions with beams close to Ca (hot fusion), and beams close to Ni (cold fusion), may not be as large as believed, because both are on the mass-symmetric side of the Businaro-Gallone ridge. We speculate that hot fusion is a viable mechanism for the production of superheavy elements [3,4], perhaps less due to the smaller entrancechannel mass asymmetry, and more due to enhancement of \overline{P}_{CN} associated with compact capture configurations resulting from collisions near the equator [9,19] of the heavy prolate target nuclei.

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