Capture cross sections for the synthesis of new heavy nuclei using radioactive beams

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We have measured the capture-fission excitation functions for the reaction of stable $^{39}$K and radioactive $^{46}$K with $^{181}$Ta using the ReA3 facility at the National Superconducting Cyclotron Laboratory. In addition the capture-fission excitation function for the $^{39}$K + $^{181}$Ta reaction was measured at Australian National University. The capture cross sections for the $^{46}$K + $^{181}$Ta reaction are larger than those for the $^{39}$K induced reactions in the near barrier region although the reduced excitation functions for the two reactions do not indicate any fundamental differences between the reactions. The results of the measurements are compared to modern phenomenological models and microscopic time-dependent Hartree-Fock calculations. The implications of these measurements for the synthesis of heavy nuclei at radioactive beam facilities are discussed.

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Formally, the cross section for producing a heavy evaporation residue $\sigma_{\text{EVR}}$ in a fusion reaction can be written as

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

where $E$ is the center of mass energy, $\mu$ is the reduced mass, $\ell$ is the orbital angular momentum, and $T$ is the probability of the colliding nuclei to overcome the potential barrier in the entrance channel and reach the contact point where the initial kinetic energy was dissipated. $P_{\text{CN}}$ is the probability that the projectile-target system will evolve from the contact point to the compound nucleus. $W_{\text{sur}}$ is the probability that the compound nucleus will decay to produce an evaporation residue rather than fissioning. To understand the synthesis of new heavy nuclei, one must understand each of the terms in this equation.

The capture cross section is, in the language of coupled channel calculations, the “barrier crossing” cross section. It is the sum of the quasi-fission, fast fission, fusion-fission, and fusion-evaporation residue cross sections. The barriers involved are the interaction barriers and not the fusion barriers. The subject of capture and fusion cross sections is the subject of a recent comprehensive review article [1]. There are several models for capture cross sections [2–6]. Each of these models was calibrated by fitting a set of fusion-capture data. In general, these models have been shown to predict the magnitudes of these capture cross sections within 50% and the values of the interaction barriers within 20% [7].

However, when the predictions of these models are compared with measured data for capture cross sections for reactions involving neutron-rich projectiles, such as $^{27}$Al + $^{197}$Au, $^{26}$Mg + $^{248}$Cm, $^{48}$Ca + $^{154}$Sm, $^{238}$U, $^{248}$Cm, and $^{64}$Ni + $^{238}$U, the agreement between prediction and data is much worse. For example, in Fig. 1, one notes that the agreement between models and data gets worse as the $Z$ of the completely fused system increases and the agreement is also worse at lower energies. While the capture cross section is not the least well known of the three factors affecting heavy element synthesis, it is vexing that this simple quantity is not better described. This work described in this paper addresses this issue.

A number of authors have tried to assess the possibility of using neutron-rich projectiles, especially those available at radioactive beam facilities, to synthesize new neutron-rich heavy nuclei [8–13]. (It should be noted that all the known isotopes of elements 100–118 are neutron deficient relative to $\beta$ stability.) The problem is that to make new superheavy ($Z > 118$) nuclei, the production cross sections are at the sub-picobarn level, and radioactive beam facilities do not have the requisite beam intensities of $>10^{13}$ pps.

Does that mean that radioactive beams have no role in the synthesis of neutron-rich heavy nuclei? Loveland [20,21] and Hong, Adamian, and Antonenko [22] have pointed out
that radioactive beams may be useful tools for producing new neutron-rich isotopes of elements 102–107 (that albeit are still neutron deficient relative to β stability) at rates $\geq 5$ atoms/day. (These reactions involve the use of light beams, such as O, Ne, Mg, etc., that can be produced at higher intensities.) In the ReA3 facility, radioactive beams are produced by projectile fragmentation and separated in flight before being thermalized in a gas catcher. After being thermalized, the $1^+$ ions of the stopped nuclei are extracted, bunched, and re-accelerated. For the $^{39,46}$K beams used in this work, the reaccelerated beam intensities are expected to be $2.1 \times 10^9$/s and $5.31 \times 10^8$/s for the Facility for Rare Isotope Beams (FRIB) project while the current ReA3 beam intensities are $2 \times 10^7$/s and $7.7 \times 10^4$/s, respectively.

In this paper, we report the first use of the ReA3 facility to study the capture-fission cross sections for reactions that are surrogates for possible use of radioactive beams in synthesizing new neutron-rich heavy nuclei. Specifically we report the results of measurements of the capture-fission cross sections for the reactions of $^{39,46}$K + $^{181}$Ta. These reactions were chosen to represent the best opportunities to study capture-fission cross sections at ReA3 given the beam intensities and energies that are currently available.

The experiment was performed using the Coincident Fission Fragment Detector (CFFD) [23] at the ReA3 facility. The CFFD consists of four large area parallel plate avalanche counters (PPACs) that are used to measure the time of flight and relative position of fission fragments from a binary event. Reconstruction of the velocity vectors of the coincident fragments allows one to calculate the masses and angular distributions of the fragments. The large solid angle of the PPACs is ideally suited for the low rate of fission events. A check of the measurements made at the ReA3 facility was made using beams of stable $^{39}$K from the 14UD Heavy-ion Accelerator Facility of the Australian National University (ANU).

The Coupled Cyclotron Facility (CCF) projectile fragmentation facility at the National Superconducting Cyclotron Laboratory (NSCL) was used in conjunction with the ReA3 reaccelerator to produce beams of $^{46}$K. The stable $^{39}$K beams at the NSCL were produced using only the ReA3 facility.

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FIG. 1. Sample predictions of capture cross sections for reactions synthesizing heavy elements. The labels FBD, Skyrme, Zagrebaev, QMD, and DNS indicate the predictions from [2], [3], [4], [5], [6], respectively. They refer to the Fusion by Diffusion, Skyrme, Quantum Molecular Dynamics, and DiNuclear Systems models. $V_B$ indicates the value of the Bass barrier energy [14]. The data for the $^{31}$Al + $^{197}$Au reaction is from [15], the data for the $^{48}$Ca + $^{154}$Sm reaction is from [16], the data for the $^{36}$Mg + $^{248}$Cm reaction is from [17], the data for the $^{48}$Ca + $^{238}$U and $^{48}$Ca + $^{248}$Cm reactions are from [18] while the data for the $^{64}$Ni + $^{238}$U reaction is from [19].
FIG. 2. The capture-fission excitation function for the $^{39}$K + $^{181}$Ta reaction. The labels MSU and ANU refer to the results of independent experiments conducted at ReA3 and ANU. The labels Sargsyan, Zagrebaev, and Wang refer to calculations of these cross sections using [3,4,13], respectively. $V_B$ denotes the position of the Bass barrier.

Cross-section measurements were made at seven energies between 180 and 210 MeV for the $^{39}$K reaction and five energies between 190 and 215 MeV for $^{46}$K reaction, spanning the respective Bass barriers. For $^{39}$K four energies were from a primary tune of the ReA3 accelerator and three additional energies were obtained by placing a 0.63 mg/cm$^2$ aluminum degrader foil upstream of the target. All $^{46}$K energies were from a primary tune of the ReA3 system. All beam energies at the NSCL were measured using attenuated beams striking a calibrated in-beam Si detector. A Ta target of thickness 0.938 mg/cm$^2$ was used for all measurements. This thickness was determined using alpha scattering measurements performed at Oregon State University.

The beam energies given herein are all “center of target” energies with the beam energy loss in the target being computed using SRIM [24]. For the stable beam experiments at ANU, the typical energy loss in passing through the target was 2.2 MeV, while in the ReA3 experiments, the typical beam energy loss in passing through the target was 6.8 MeV.

In Figs. 2 and 3 we show the measured capture-fission excitation functions for the $^{39}$K + $^{181}$Ta reaction (Fig. 2) and the $^{46}$K + $^{181}$Ta reaction (Fig. 3).

In Fig. 2, we show the results of independent measurements of the capture cross sections made at the ReA3 facility and at the Australian National University. These normalized measurements appear to agree within experimental uncertainties. We should also note that the capture-fission excitation function can be taken as the capture excitation function because straightforward calculations [4,25] for these reactions indicate that $\geq 99\%$ of the capture events undergo fission.

The uncertainties in the cross sections measured at ANU and the general issue of the use of coincident fission fragments to deduce capture-fission excitation functions are discussed in [23,26]. The uncertainties in deduced quantities from radioactive beam experiments, such as this one, have been discussed in detail by [27]. The small number of measured points on capture excitation functions and the large uncertainties in the deduced cross sections can lead to significant uncertainties in deduced parameters of fusion barrier distributions. However, we are mindful of this difficulty and have not extracted interaction barriers from our data.

In Figs. 2 and 3, we compare our results with predictions of modern phenomenological models of the capture process [4,13,28]. The predictions of the coupled channel calculations of Zagrebaev overestimate the observed cross sections for both systems at above barrier energies. The empirical model of Wang and Schied [28] based upon a modified Woods-Saxon potential to describe the interaction agrees satisfactorily with the measurements of the interaction of stable $^{39}$K + $^{181}$Ta at above barrier energies, but overestimates the cross sections for the $^{46}$K + $^{181}$Ta reaction. The calculations of Sargsyan [13,29] appear to do the best overall job of representing the capture excitation functions for the $^{39}$,$^{46}$K + $^{181}$Ta reactions. This success is similar to that observed for the $^{48}$Ca + $^{208}$Pb reaction [13].

In Figs. 2 and 3, upper limit (UL) and lower limit (LL) estimates of the capture cross sections as calculated using time-dependent Hartree-Fock calculations (TDHF) are shown. (See [30] for details of similar calculations.) Especially for the $^{46}$K + $^{181}$Ta reaction, the TDHF predictions at above barrier energies do not agree with the measurements although they are compatible with the predictions of Sargsyan and Wang. Perhaps this indicates that these data can be used to challenge and improve the assumptions in time-dependent microscopic calculations. (It should be pointed out that heavy ions encounter semiclassical trajectories in TDHF, with fusion cross sections dropping to zero at the barrier. A comparison with data is therefore relevant at above barrier energies only.)
FIG. 4. (a) A simple comparison of the capture excitation functions for the $^{39,46}\text{K}+^{181}\text{Ta}$ reaction. (b) The reduced excitation functions for these reactions.

One might ask as to how the excitation functions for the two reactions compare, i.e., what is the effect of the neutron-rich $^{46}\text{K}$ relative to the stable $^{39}\text{K}$ projectile? The simplest comparison [Fig. 4(a)] indicates the reaction with the neutron-rich $^{46}\text{K}$ projectiles has a larger cross section for below barrier events. However, to compare these two reactions, we show [Fig. 4(b)] the traditional reduced excitation functions for the reactions. These reduced excitation functions are determined by plotting the cross sections vs $1/E_{\text{cm}}$ and extracting from that plot, the empirical capture barrier $V_B$ and the capture radius $R_B$. The reduced excitation functions do not show any significant difference between the reactions.

To understand the possible impact of these measurements on the production of neutron-rich heavy nuclei, we consider the reactions of the K isotopes with targets of $^{226}\text{Ra}$ and $^{227}\text{Ac}$ to form neutron-rich Bh and Hs nuclei. We use the formalism of Zagrebaev [4] to perform these calculations for the $^{46,47,48}\text{K}+^{226,227}\text{Ra}$ and $^{227}\text{Ac}$ reactions. (We have multiplied the calculated capture cross sections by 0.5 to reflect the results of our measurement.) We assume FRIB beam intensities will be $5.3 \times 10^8$/$\text{s}$, $3.5 \times 10^8$/$\text{s}$, and $3.5 \times 10^6$/$\text{s}$ for $^{46,47,48}\text{K}$, target thicknesses of 0.5 mg/$\text{cm}^2$, and values of $P_{\text{CN}}$ given by [12]. We find the production rates of $^{267,268,269,270}\text{Bh}$ are 0.07, 0.8, 0.8, 0.1 atoms/day while the production rates of $^{269,270,271}\text{Hs}$ are 0.1, 0.2, and 0.06 atoms/day. While such estimates are uncertain, they do indicate possible promise for the synthesis of neutron-rich heavy nuclei at FRIB. It may be that special efforts, targeted at the production of specific radioactive beams, will be able to increase the available beam intensities at FRIB [31].

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[29] These calculations were done using the formalism of [13] adapted to the $^{39,46}$K + $^{181}$Ta reactions by Professor Sargsyan.
[31] B. Sherrill (private communication).