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Deducing the ^{237}U destruction cross-sections using the Surrogate Ratio Method (U)

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*We have deduced the destruction cross section of ^{237}U via the (n,γ) and (n,2n) reactions over an equivalent neutron energy range of 0 to 20 MeV using a new form of the Surrogate Ratio method [1-4] . The relative fission and neutron-evaporation decay probabilities of excited ^{238}U populated via the (α,α') inelastic scattering were measured using the **s**ilicon **t**elescope **a**rray for **r**eaction **s**tudies (STARS) coupled to the **Li**vermore **B**erkeley **ar**ray for **c**ollaborative **e**xperiments (LIBERACE). These relative probabilities were then combined with the ^{237}U(n,f) cross section deduced by Burke et al., [4] to deduce the (n,γ) and (n,2n) cross sections in a model independent fashion. These cross sections are then compared to the compound reaction cross section calculated using an optical model calculation tuned to reproduce scattering data in the transactinide region. Our results presented and the prospects for using this technique to deduce (n,x) cross sections on radioactive nuclei are discussed. (U)*

Introduction

In many situations the production of a specific radioisotope can be used to determine the characteristics of a high-flux environment. However the utility of such radiochemical data is dependent on the quality of the nuclear cross section data used to model the production and destruction of the specific radioisotope being studied. A prime example of this is the nucleus ^{237}U which can be produced either via ^{238}U(n,2n)^{237}U or by two step capture on ^{235}U (i.e., ^{235}U(n,γ)^{236}U(n,γ)^{237}U). Although the production cross sections are relatively well-known the destruction of ^{237}U via the (n,γ) and (n,2n) reactions remain unmeasured due to the short life of the nuclide ($t_{1/2}$=6.75 days) which renders direct measurements virtually impossible.

Recently, A significant amount of interest has been focused recently on deducing neutron-induced fission cross sections on short-lived radioactive nuclei using a combination of measurement and modeling referred to as the "Surrogate Ratio" method [1-5]. The approach used in these papers involves measuring the same exit channel probability for two similar compound nuclei (CN) formed in the same "surrogate" light-ion reaction. The ratio of these exit channel probabilities can then be used to determine an unknown cross section (which proceeds via one CN) relative to a known one (which proceeds through the other). This ratio can be referred to as an "external ratio" (ER) since it is a ratio between decay probabilities of *two different CN*. In this manuscript we present a complementary "internal ratio" (IR) method where a ratio of decay probabilities for two *different exit channels* in the *same CN* is used to deduce an unknown cross section relative to a known one. These ratio techniques, when used appropriately, remove and/or reduce many of the systematic uncertainties related to the direct Surrogate Method [6-10] which involves a measurement of absolute and not relative decay probabilities of a CN.

In the experiment discussed here the fission, and xn-emission decay probabilities of excited ^{238}U nuclei populated via inelastic α-particle scattering was measured using a highly-segmented Silicon particle detector array (STARS) coupled to an array of six segmented Germanium "clover" detectors (LIBERACE). STARS measured the scattered a-particle and fission fragment energies and angles while LIBERACE provided a tag on xn-emission by identifying discrete γ-ray transitions to different residual Uranium nuclei. The ratio of the measured fission-to-xnγ emission probabilities was then combined with the ^{237}U(n,f) cross sections determined by Burke et al., [1,2] to obtain the ^{237}U(n,γ) and ^{237}U(n,2n) cross sections for incident neutron energies between 0-20 MeV.

Theory

Surrogate reaction methods are based on the assumption that the decay of an excited compound nucleus (CN) into an exit channel ξ formed using two different entrance channel α and β can be related to each other using a combination of modeling and experimental observation via the relation:

$$\sigma_{\alpha\xi}(E_x) = \sum_{J,\pi} \sigma_\alpha^{CN}(E_x,J,\pi) G_\xi^{CN}(E_x,J,\pi) \quad (1)$$

Here $\sigma_{\alpha\xi}$ denotes the ``desired" surrogate reaction, E_x is the excitation energy of the CN, $G_\xi^{CN}(E_x,J,\pi)$ is the CN decay probability measured using entrance channel β and $\sigma_\alpha^{CN}(E_x, J,\pi)$ are the partial-wave components of the total compound reaction cross section for the "desired" entrance channel α.

In general the branching ratios, $G_\xi^{CN}(E_x,J,\pi)$ depend on the angular momentum and parity of the CN formed in the reaction. However, there are some circumstances under which the decay probabilities will depend solely on the excitation energy of the system [11,12]. This is referred to as the Weisskopf-Ewing limit. In the Weisskopf-Ewing limit the expression above simplifies to:

$$\sigma_{\alpha\xi}(E_x) = \sigma_\alpha^{CN}(E_x) G_\xi^{CN}(E_x) \quad (2)$$

Where $\sigma_\alpha^{CN}(E_x)$ is the reaction cross section describing the formation of the CN at energy E_x which can be derived from an optical model calculation, and $G_\xi^{CN}(E_x)$ denotes the Jπ-independent branching ratio for the exit channel ξ. ,$G_\xi^{CN}(E_x)$ is obtained from experiment by dividing the number of times that the CN decays via channel ξ by the efficiency for detecting the exit channel and the number of particles from the surrogate reaction entrance channel:

$$G_\xi^{CN}(E_{ex}) = \frac{N_\xi(E)}{\varepsilon_\xi N_{\beta\xi}} \quad (3)$$

Where $N_\xi(E)$ is the number of times that the exit channel ξ is observed, ε_ξ is the efficiency for detecting the exit channel ξ and $N_{\beta\xi}$ is the total number of observed surrogate reaction events. It can be very difficult to obtain this last quantity experimentally since the presence of any contaminants in the target will produce a background that is hard to quantify.

In the Weisskopf-Ewing limit, it is possible to obtain the *relative* probability for two different exit channels, ξ1 and ξ2 observed simultaneously in a single surrogate reaction by taking the ratio of the exit channel probabilities obtained using eqn. [3]. This ratio can then in turn be used to relate a known and an unknown neutron-induced reaction cross section by combining eqn. [3] with eqn. [1] in the Weisskopf-Ewing limit:

$$\sigma_{\alpha\xi 2}(E_\alpha) = \frac{G_{\xi 1}^{CN}(E_{ex})}{G_{\xi 2}^{CN}(E_{ex})} \times \sigma_{\alpha\xi 1}(E_\alpha) \quad (4)$$

where the ξ1 and ξ2 indices refer to the two different exit channels, E_x and S_n are the excitation and neutron separation energies of the CN, E_n is the equivalent "surrogate" neutron energy and $\sigma_{\alpha\xi 1}$ and $\sigma_{\alpha\xi 2}$ refer to the known and unknown (n,x) cross sections respectively. This method can be referred to as the "internal ratio" (IR) since both exit channels (ξ1 and ξ2) are for the same CN. This is in contrast to the "external ratio" (ER) method used by Plettner *et al.,* and Burke *et al.,* [2,3] where two identical exit channel probabilities are compared for two different, albeit similar, compound nuclei. Although both ratio methods assume that the Weisskopf-Ewing approximation is valid the ER approach may be less sensitive to violations of the Weisskopf-Ewing limit since there may be partial cancellation in the probabilities in the numerator and denominator of eqn. [4]. This will be discussed in greater depth in the conclusion section below.

Experimental Apparatus

The experiment was performed at the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory using STARS coupled to LIBERACE. Data were taken over a

period of 3.5 consecutive days using a 55 MeV beam of α-particles with an intensity between 2 to 5 pnA. Since the experimental set-up is nearly identical to that described by Burke et al., [1,2] we will present only a brief summary here.

The inelastically scattered a-particles were observed in a silicon telescope comprised of 140 μm ΔE and 1000 μm and thick E Micron "S2" type detectors. Each S2 detector has 48 rings on one side and 16 sectors on the other. For this experiment both detectors had pairs of adjacent rings and sectors bussed together to form twenty-four 1 mm wide rings and 8 sectors. The ^{238}U target was a self supporting metallic foil with a thickness of 6219±82 angstrom (1.1± 0.3 mg/cm^2 located 14.8±1 mm upstream from the front face of the ΔE detector. The ΔE and E detectors were spaced 3 mm apart. The beam spot on the target was approximately 3 mm in diameter. This geometry leads to an angular detection range in θ (the angle formed between the beam axis and the scattered alpha particle) from 38° to 60°. A "ray-trace" requirement on the front and back detectors excluded beam scattered from locations other than the target. A 4.44 mg/cm^2 aluminum foil placed between the target and the silicon telescope stopped forward flying fission fragments and also helped to mitigate the effect of delta electrons on the silicon detectors. Fission fragments were detected in a third 140 μm Micron S2 detector located ≈10 mm upstream of the target. The adjacent rings and sectors of this detector were also bussed together. The fission detector covered an angle range of 106° to 131° with respect to the beam axis.

The ΔE, E, and fission detectors were biased with 45 V, 105 V and 30 V respectively. The signals from the rings and sectors of the ΔE, E and fission detectors were fed through a total of 96 individual CHARGE8V Swan Research pre-amplifiers with sensitivities of 47 mV/MeV for the ΔE and E detector signals and 20 mV/MeV for the fission detector signal. These signals were fed into six 16-channel CAEN N568B shapers. The fast outputs of the CAEN N568B shapers were connected to LeCroy 1806 discriminators modified to be leading edge. The discriminator thresholds were set at 60 mV, which corresponds to an energy threshold of approximately 800 keV per channel. At least one hit in the ΔE and E detectors were required within an 80 ns coincidence interval to form the particle trigger. This master trigger rate ranged between 4 kHz to 6 kHz during the experiment. Once a valid trigger occurred, the delayed shaped slow output of the shaper channels were digitized using SILENA analog to digital converters (ADCs) with a 70 μs gate. Particle-fission timing was obtained using a Time-to-Amplitude converter module read out using an Ortec AD413 ADC.

The pre-amplifier signals from the EURISYS clover detectors, together with the Scionix BGO Compton-suppression shields were fed into 5 RIS corporation clover modules that provide high-resolution (14 bit) "leaf" energy and low-resolution (12 bit) side channel and timing information for the detector. A sixth clover was wired using "conventional" electronics comprised of NIM timing filter and shaping amplifiers coupled to CFDs and Ortec AD413 ADCs. The RIS clover modules, Silena ADC and Ortec AD413 ADCs were then read out using the FERA protocol into an Ortec CMC203 FERA driver/histogramming memory unit, which was in turn read out using a Sparrow

Corp. CAMAC-SCSI driver into the data acquisition computer. Online data monitoring was performed using one and two-dimensional arrays.

The ΔE and E silicon detectors were calibrated using a ^{226}Ra α-source. This calibration, while sufficient for the 140 μm ΔE detector which had a maximum a-particle energy deposition of ≈12 MeV, was insufficient for the 1000 μm thick E detector which had energy deposited in it in excess of 45 MeV. Therefore, elastically scattered α-particles were used to provide a higher energy calibration point. This point was obtained by subtracting the energy lost by the α-particles in the target, aluminum δ-shield, ΔE detector and Gold "sector-side" electrodes from the recoil-corrected elastically scattered α-particle energy.

The sectors of both detectors had a factor of approximately1.4 poorer energy resolution (δE/E) compared to the rings, so the energy from the rings was used for the analysis. However, the large scattering angle of the α-particles made it highly likely that the energy deposited in the ΔE detector would be collected over more than one ring. Therefore, in order to obtain the best energy resolution the energy was determined by adding together adjacent rings whenever two fired in a given event. The 1σ energy resolution of the combined detectors was taken as the sum of the squares of the individual uncertainties and ranged from 38-55 keV.

Analysis and Interpretation

Particle identification for inelastically scattered α's was accomplished by plotting front versus back detector energy. The total α-particle energy was then calculated from the sum of the detector energies plus the energy lost in the target (half-thickness assumed), the δ-shield and the Gold electrodes on the sector side of the S2 detectors. The bottom panel of figure 1 shows the α-particle ``singles" spectrum. The total energy of the ejected α-particle was then used to determine the excitation energy deposited in the ^{238}U by subtracting the α-particle energy and the calculated energy of the recoiling ^{238}U nucleus.

Events in the fission detector greater than 6 MeV were identified as fission events to remove light ions from direct reactions and charged-particle evaporation in a manner consistent with that described in [2]. A properly background subtracted gate on a master trigger-fission TAC was used to ensure that only prompt particle-fission events were included in the analysis. The α-particle in coincidence with the fission fragments are shown in the top panel of Figure 1.

The four separate leaves of each Ge detector were calibrated using a sealed ^{152}Eu source. An "add-back" procedure was used since the energy from a high-energy γ-ray is likely to be deposited in more than one of the 4 "leaves" of the EURISYS clover Ge detectors. Prompt (with respect to the master trigger) γ-rays from different leaves of a single ``clover" were added together to form a prompt ``add-back" energy. Similarly,

non-prompt γ-ray energies were added to form a non-prompt ``add-back" for background subtraction purposes.

The fission channel probability, $G_f^{CN}(E_x)$, can be obtained from the particle-fission coincident data using eqn. [3]. The fission detector efficiency can be expressed as the product of two parts. The first of these is simply the fraction of solid angle covered by the detector. The second part represents the increased efficiency resulting from the preferential emission of the fission fragments along at small angles with respect to the direction of the recoiling Uranium nucleus in a plane containing the fission fragment, the recoiling nucleus and the beam. This latter part was obtained fromm a comparison between the in-plane and out-of-plane fission rates in a matter identical to that used in references [1,2].

Determining the efficiency for detecting the evaporation channels through the observation of discrete γ-ray transitions requires a more detailed process since the γ-ray cascade in the residual nuclei are highly dependent on the structure of the low-lying states. The following procedure was used.

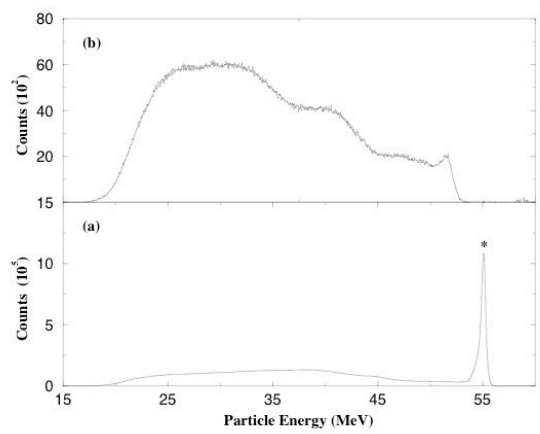

Figure 1: Total α-particle energy spectrum obtained gating on α-particle in the ΔE-E data (bottom) and in coincidence with fission fragments seen in the back detector (top). The elastic peak is marked with an asterick (*).

Gamma-ray spectra corresponding to several excitation energies bins less than the fission barrier and neutron separation energy were formed from the particle-γ coincident data. ^{238}U nuclei formed with these excitation energies can only decay via γ-ray decay. The most intense, non-coincident discrete γ-ray transitions corresponding to ^{238}U in these energy bins were corrected for internal conversion and relative efficiency (using data from a ^{152}Eu source) and then added together to form a "parallel path" sum of all non-coincident transitions de-exciting the nucleus following the procedure described in reference [13]. The transitions included in the "parallel path" sum were the 103.4 keV yrast $4^+ \rightarrow 2^+$, the 635.2 keV $1^- \rightarrow 2_1^+$, the 686.4 keV $3_1^- \rightarrow 2_1^+$, the 884.8 keV $1_2^- \rightarrow 2_1^+$, and the 1014.5 keV $2_2^+ \rightarrow 2_1^+$. Since this "parallel path" sum is for an energy region with unity probability for γ-ray emission it can be used to obtain an absolute "parallel path" xn evaporation residue detection efficiency, ε_{xn}:

$$\varepsilon_{xn} = \frac{\sum_{parallel} \gamma_i(E_{ex})}{N_\alpha(E_{ex})} \quad (6)$$

This value was found to be remarkably constant as a function of excitation energy, which is not surprising considering that the angular momentum imparted by inelastic scattering is far less sensitive to the energy of scattered particle than its angle of emission. It should also be noted that the ``parallel-path'' sum used emphasizes low-spin states since these transitions are usually the most intense in a rotational band. The average value obtained for ε_{xn} was 0.638±0.034%. The photo-peak efficiency for a six-clover Ge array in this geometry is expected to be approximately 0.84% at 1.33 MeV, indicating that the parallel path sum does capture the majority of decays of the compound nucleus even though the yrast $2^+ \rightarrow 0^+$ transition was not included in the parallel path sum. This xn-residue detector efficiency is applicable only for even-even residual Uranium isotopes since the low-lying states are significantly different for even-even and odd-mass nuclei due to the effect of the pairing interaction.

Using the deduced ε_f and ε_{xn} it is possible to relate the cross sections for ^{237}U(n,f) and ^{237}U(n,xn) for x=0,2 using eqn. [4] since these reactions result in the formation of an even-even residual nucleus.

If we combine this IR measurement with the results from [1,2] where the ^{237}U(n,f) cross section was deduced relative to ^{235}U(n,f) using the ER method we can obtain the ^{237}U(n,γ) and ^{237}U(n,2n) cross sections:

$$\sigma_{xn}(E_{ex}) = \frac{\sum_{xn} \gamma_i(E_{ex})}{\varepsilon_{xn} N_f} \times \sigma_f(E_{ex}) \quad (7)$$

where $\sum_{xn} \gamma_i(E_{ex})$ is the parallel path sum for ^{238}U (x=0) and ^{236}U (x=2), $\sigma_{xn}(E_{ex})$ and $\sigma_f(E_{ex})$ are the ^{237}U(n,xn) and (n,f) cross sections respectively.

α-particle energies greater than 47.8 MeV (corresponding to E_x < 7.2 MeV) are kinematically clear of inelastic scattering on light-ion contaminants in the target due to the recoil of the light target nuclei. This enables the determination of the ^{237}U(n,γ) cross section using not only the IR approach discussed above, but also by directly measuring the ``absolute probability'' of the decay of the excited ^{238}U CN and

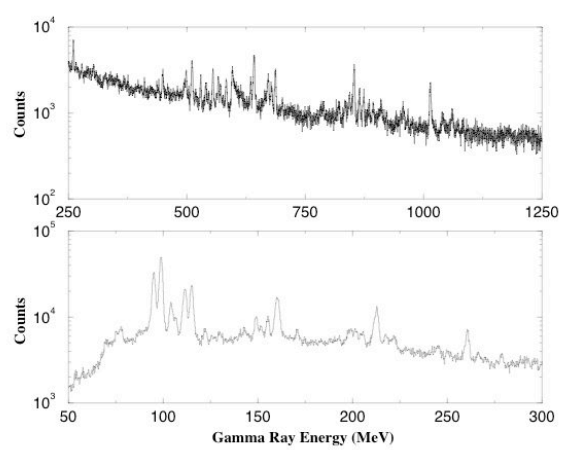

Figure 2: Total γ-ray spectrum in coincidence with α-particles. The transitions used to tag ^{236}U or ^{238}U residual nuclei are marked by open boxes and the major yrast transitions are marked with solid boxes. The low (E_γ < 250 keV) and high energy (250 < E_γ (keV) < 1250 keV) are shown separately in the bottom and top panels respectively.

applying the Weisskopf-Ewing approximation. The ^{237}U(n,γ) deduced using the both the IR and ``absolute probability'' methods and the ^{237}U(n,2n) cross section deduced using the IR alone are shown in the bottom and middle panels of Figure 3 and listed in Table 1.

The sum of the ^{237}U(n,2n) cross section derived in this work and the ^{237}U(n,f) cross section from Burke *et al.,* [1,2] should comprise the vast majority of the total compound reaction cross section for neutrons incident on ^{237}U for energies near the peak of the (n,2n)reaction ($E_n \approx 14$ MeV). The top panel of figure [3] shows that this is indeed the case when the the sum of the (n,f) and (n,2n) cross sections are compared to the compound reaction cross section deduced using the FLAP optical model [14].

Table 1. ^{237}U(n,γ) and (n,2n) cross sections determined in this work. The equivalent neutron energy has a relative uncertainty of ±100 keV at every energy bin.

Energy (MeV)	Reaction	Cross Section (mb)
0.18(8)	(n,γ)	0.997(243)
0.30(8)	(n,γ)	0.791(183)
0.46(8)	(n,γ)	0.473(219)
0.74(8)	(n,γ)	0.286(132)
1.1(1)	(n,γ)	0.124(062)
6.1(4)	(n,2n)	0.06(0.19)
6.9(4)	(n,2n)	0.25(0.12)
7.7(4)	(n,2n)	0.76(0.19)
8.5(4)	(n,2n)	0.74(0.21)
9.3(4)	(n,2n)	0.87(0.18)
10.1(4)	(n,2n)	0.83(0.18)
10.9(4)	(n,2n)	0.94(0.19)
11.7(4)	(n,2n)	1.16(0.21)
12.5(4)	(n,2n)	1.08(0.23)
13.3(4)	(n,2n)	1.19(0.24)
14.6(4)	(n,2n)	1.36(0.24)
14.9(4)	(n,2n)	1.18(0.24)
15.7(4)	(n,2n)	1.04(0.23)
16.5(4)	(n,2n)	0.89(0.22)
17.3(4)	(n,2n)	0.90(0.20)
18.1(4)	(n,2n)	0.63(0.25)
18.9(4)	(n,2n)	0.50(0.14)
19.7(4)	(n,2n)	0.18(0.12)

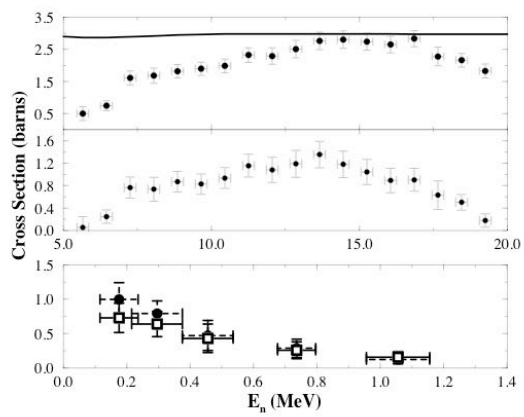

FIG. 3: ^{237}U(n,γ) cross section deduced using the Internal Ratio (IR) and absolute probability approaches (top), the ^{237}U(n,2n) cross section deduced using the IR method (middle) from this work and the sum of the ^{237}U(n,2n) from this work and the ^{237}U(n,f) from Burke et al., [1] (bottom).

Conclusions

The ^{237}U(n,γ) and (n,2n) cross sections have been determined using an "internal ratio" (IR) to the fission channel probability. The IR method may be applied to determine a host of (n,xn) cross sections through the observation of discrete γ-ray transitions in various residual nuclei. The IR method and the ER method presented in [3] both assume that the exit channel probabilities are independent of spin and parity (the Weisskopf-Ewing limit). However, recent theoretical calculations [15] seem to indicate that there is considerable cancellation of any residual spin dependence between the probabilities in the numerator and denominator of the ER. These ratio methods share another advantage over ``absolute probability" surrogate measurements in that the contribution from pre-equilibrium particle emission is ``folded" into the cross section when it is multiplied by the known ``reference" cross section.

The IR method offers less likelihood of cancellation since the exit channels in the numerator and denominator are likely to have different dependence on the spin and parity of the CN. The similarity between the higher energy (n,γ) cross section values obtained using the IR and absolute probability approach may reflect this limited cancellation. On the other hand, the IR method does have the advantage over the ER method that the number of surrogate reaction events in the numerator and denominator are identical.

A comprehensive theoretical and experimental ``benchmarking" of both the ER and IR methods is necessary to quantitatively establish the limits of validity of these techniques. This effort is underway and we will report the results in the near future.

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