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EXPERIMENTAL APPROACHES TO STUDYING THE FISSION PROCESS USING THE SURROGATE REACTION TECHNIQUE

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Over the past several years, Lawrence Livermore National Laboratory has been developing the surrogate technique to study the properties of fission and perform precision cross section measurements. The surrogate technique allows nuclear physicists to measure previously unattainable compound-nuclear cross sections of nuclei currently one to two nucleons away from a stable or long lived (~10,000 y) nucleus. The goal of the surrogate technique is to create the same excited compound nucleus as is produced in a specific desired direct neutron reaction, study the decay channels and extract the cross section of choice; (n,γ), (n,2n) or (n,f). This may be accomplished via an inelastic scattering reaction (e.g. (p,p'), (d,d'), (^{3}He,^{3}He')), a neutron transfer reaction (e.g. (d,p), (^{18}O,^{16}O)), or a pick-up reaction such as (^{3}He,^{4}He). To measure the decay channels (gamma-ray or fission) we have constructed the Silicon Telescope Array for Reactions Studies (STARS), the LIvermore BERkeley Array for Collaborative Experiments (LIBERACE) and the HYDRA array for precision fission studies. Preliminary results from a recent ^{238}Pu(n,f) measurement are presented.

KEYWORDS : ND2010, Nuclear Data, surrogate reaction, ^{238}Pu, fission, cross section

1 INTRODUCTION

Precise cross sections for neutron-induced reactions on minor actinides are important for the design of advanced hard neutron spectrum reactors and nuclear transmutation schemes [1,2]. Many of these actinides have relatively short half-lives (<100 years), making it difficult to produce and fabricate targets for neutron experiments. However, for many isotopes there exists a longer-lived actinide nearby; the neighbouring isotope can be used to form the same compound nucleus as the initial desired reaction. Decay from the compound state is assumed to be independent of the production reaction, allowing reactions with the neighbouring isotopes to be used as a surrogate for the reaction of interest.

Surrogate reactions often employ particle transfers or inelastic scattering reactions. The outgoing particle is easily identified and can be used to identify the energy of the compound system. The simultaneous measurement of the outgoing particle and an observable that characterizes the exit channel of interest (fission fragment or gamma ray) can then be used to deduce the cross section of the desired channel. For details of the formalism see [3-4]. Several variants of the surrogate approach exist: the absolute surrogate technique determines the cross section by combing the calculated probability for compound nucleus formation with the measured probability of decay [5]. Alternatively, one can examine two reactions relative to one another. The surrogate ratio method (SRM) [6]

allows an unknown cross section to be determined relative to a known one. Specifically, for a neutron-induced fission reaction, one has

$$\sigma_u(n,f;E) = \frac{N_{p-f,u}(surr,E)}{N_{p-f,k}(surr,E)} \times \sigma_k(n,f;E) \qquad (1)$$

Here σ_k and σ_u denote the known and unknown cross sections for neutron-induced reactions and $N_{p\text{-}f,k}$ ($N_{p\text{-}f,u}$) denotes the experimentally determined number of particle-fission coincidence events in the surrogate reaction that created the associated compound nucleus. Light ion reactions such as (d,p), (^{3}He,t), (^{3}He,^{3}He'), (^{3}He,α) and (α,α'), have been successfully used as surrogate reactions for (n,f) cross-sections in the actinide region [6-13].

This paper describes the detector systems employed for surrogate reactions by the STARS/LIBERACE collaboration, and presents preliminary results of the ^{238}Pu(n,f) cross section obtained from a surrogate ratio measurement. For the first time a single measurement covers the energy range from 500 keV to 20 MeV.

2 EXPERIMENTAL APPARATUS

To determine the decay probabilities for a particular exit channel, charged-particles have to be detected in coincidence with fission fragments, gamma rays or other observables characteristic of the exit channel. In the present setup, charged particles and fission products are detected by the STARS array and gamma rays are detected by the LIBERACE array.

2.1 Silicon Telescope Array for Reactions Studies (STARS)

The STARS array is comprised of a series of double-sided annular Si detectors for particle detection, placed downstream from the target, to measure light charged particles emitted in the reaction. Typical experiments employ two (ΔE-E) or three (ΔE-E1-E2) such detectors. Upstream from the target, an additional Si detector is used for fission tagging. Figure 1 shows a schematic for the STARS array.

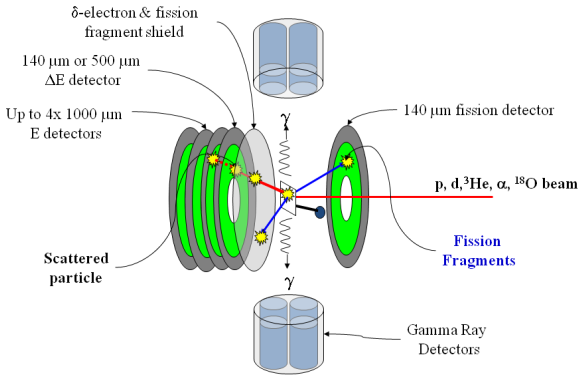

Fig. 1. STARS/LIBERACE detector array schematic.

2.2 Livermore BERkeley Array for Collaborative Experiments (LIBERACE)

The LIBERACE HPGe array is used to measure the gamma-rays emitted from the target following a direct reaction. Up to six Compton-suppressed clover detectors are placed around the target chamber, covering an angle range from approximately 30 degrees to 120 degrees with respect to the beam axis. The typical photo peak efficiency for the array is 2% at 200 keV and drops to approximately 1% at 1 MeV. For surrogate reactions, these detectors have been used to determine neutron capture (n,γ) cross sections [9,11], where particle-gamma coincidences are sought rather than particle-fission coincidences.

2.2 HYDRA

A new chamber, designed specifically for increased particle angular coverage, has been recently installed in the experimental hall as a complement to the STARS chamber. The HYDRA chamber consists of a 40 cm diameter chamber that is 22 cm deep. The bottom of the chamber has threaded holes with a 2.54 cm by 2.54 cm square hole pattern. The circumference has a threaded hole pattern with 10 degree angular increments between each hole. A 14 position in vacuum linear transfer arm enters the chamber from the bottom at the center. The target ladder can be rotated so the targets are perpendicular or at any angle desired with respect to the beam. The chamber can accommodate the standard Micron S1 and S2 detectors as well as arrays of individual Ortec transmission and single sided silicon detectors. An array of fission detectors based on solar cells is also being designed for installation. The chamber and hardware will allow for detailed fission studies, precision inelastic and elastic scattering cross section measurements over a wide angle range (10 to 170 degrees w.r.t beam).

^{238}Pu(n,f) via surrogate reaction

The ^{238}Pu(n,f) cross section was recently determined using the surrogate ratio method. The cross section was determined relative to the well-known ^{234}U(n,f) and ^{235}U(n,f) cross sections. The relevant compound nuclei ^{239}Pu, ^{235}U, ^{236}U were produced by inelastic α scattering on their respective ground states.

Thin targets of ^{235}U, ^{236}U and ^{239}Pu were synthesized by electroplating oxides of the actinide isotopes on 100 μg/cm^2 natural carbon foils. The target thicknesses were then determined by alpha counting. For the experiment described here, targets of 416(5), 322(2) and 140(1) μg/cm^2 were used for ^{235}U, ^{236}U and ^{239}Pu, respectively.

The targets were bombarded with a 55-MeV alpha beam from the 88 Inch Cyclotron at Lawrence Berkeley National Laboratory. The scattered alpha particles were detected by the STARS array, which was comprised of a 150 micron thick ΔE and a 1000 micron thick E detector. Fission fragments were detected in an additional detector 12 mm upstream of the target. The silicon telescope allowed a clear separation of the various scattered light ions (p, d, t, ^{3}He and ^{4}He) for particle identification. The energy and angle of the outgoing particles were recorded on an event-by-event basis throughout the entire five-day experiment. The excitation energy imparted to the nucleus can be reconstructed from the emission angle and energy of the light ion, the incident beam energy, and the energy losses in the target, shields, and detector system materials. The excitation energy is used to determine an equivalent neutron energy. The number of particle-fission coincidence events as a function of neutron energy was then used to determine the unknown cross section described in Eq. 1.

The ^{238}Pu(n,f) cross section was extracted by using two different reference surrogate reactions. Preliminary results are described in the following sections.

2.3 ^{238}Pu(n,f) derived from ^{239}Pu/^{236}U ratio

The ratio of the number of particle-fission events for ^{239}Pu(α,α'f) and ^{236}U(α,α'f) was used to obtain the ^{238}Pu(n,f) cross section relative to the known ^{235}U(n,f) cross section:

$$\sigma_{^{238}Pu}(n,f;E) = \frac{N_{p-f}(^{239}Pu(\alpha,\alpha'),E)}{N_{p-f}(^{236}U(\alpha,\alpha'),E)} \times \sigma_{^{235}U}(n,f;E) \qquad (2)$$

Using the ENDF/B-VII evaluation for the ^{235}U(n,f), we obtain the ^{239}Pu(n,f) result shown in Figure 2.

In this approach target nuclei with ground state spins of 1/2$^+$ and 0$^+$ for ^{239}Pu and ^{236}U are compared respectively. It should be noted that the original desired reaction nuclei have considerably different ground state spins of 0$^+$ and 7/2$^+$ for ^{238}Pu and ^{235}U respectively. Spin states populated by neutron capture are expected to be considerably different from those populated by the surrogate reactions considered here. This should lead to a large discrepancy in the fission cross section at low energies (<2MeV) if the states participating in the fission near the barrier are populated in a significantly different manner. It is surprising that good agreement with the direct neutron capture fission cross sections is observed at low energies.

At higher energies, the surrogate reaction results agree well with previous measurements up to ~17 MeV.

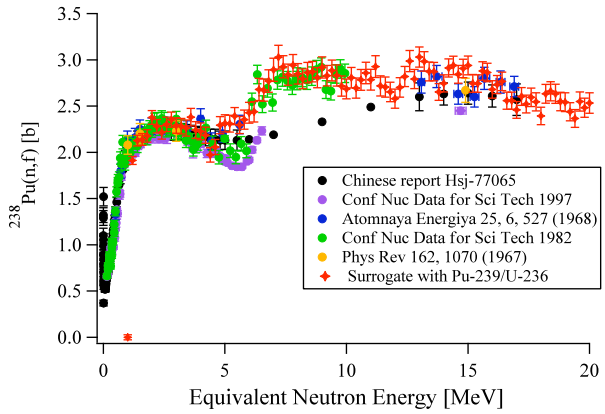

Fig. 2. Preliminary ^{238}Pu(n,f) cross section results obtained from the ^{235}U(n,f) ratio measurement are shown in red and are compared with direct neutron induced measurements.

2.4 ^{238}Pu(n,f) derived from ^{239}Pu/^{235}U ratio

The ^{238}Pu(n,f) cross section has also been determined using the surrogate ratio of ^{239}Pu(α,α'f) to ^{235}U(α,α'f). The ratio is described in equation (3) and is the same as equation (2) except with the ^{235}U (α,α'f) coincidence rate in the denominator and ^{234}U(n,f) (from JEFF 3.1) is the reference cross section.

$$\sigma_{^{238}Pu}(n,f;E) = \frac{N_{p-f}(^{239}Pu(\alpha,\alpha'),E)}{N_{p-f}(^{235}U(\alpha,\alpha'),E)} \times \sigma_{^{234}U}(n,f;E) \qquad (3)$$

The resulting ^{238}Pu(n,f) cross-sections, shown in Figure 3, agree with previous measurements within the uncertainties over the energy range measured. There is, however, a slight increase in the cross section across the energy range; this increase is most dramatic below 5 MeV. The higher cross-sections reported here may be attributed to spin effects due to the ground state spin differences between ^{235}U(7/2$^+$) and ^{239}Pu(1/2$^+$). This discrepancy will be investigated further by pursuing direct reaction calculations.

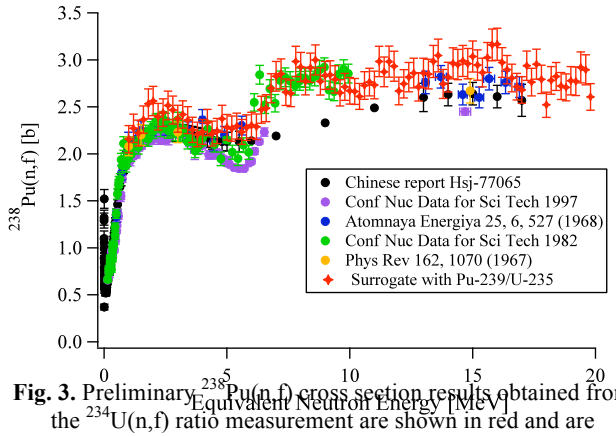

Fig. 3. Preliminary ^{238}Pu(n,f) cross section results obtained from the ^{234}U(n,f) ratio measurement are shown in red and are compared with direct neutron induced measurements.

CONCLUSIONS

Preliminary results for the ^{238}Pu(n,f) cross section have been deduced over an energy range from approximately 500 keV to 20 MeV in a single measurement for the first time. The results agree well with the separate measurements over most of the energy range. The region from 5 to 12 MeV clearly disfavours the results from Hsj-77065 [14]. More work is needed to fully interpret this surrogate experiment. In particular the low energy region (<2 MeV) needs to be examined for the effects of anisotropic fission fragment distributions.

Future surrogate reactions will focus on additional minor actinide cross sections. The new HYDRA chamber will allow us to begin performing precision large angle scattering

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