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^{239}Pu(n,2n) and ^{241}Pu(n,2n) surrogate cross section measurements using NeutronSTARS

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Introduction

The goal of this project was to develop a new approach to measuring (n,2n) reactions for isotopes of interest. We set out to measure the ^{239}Pu(n,2n) and ^{241}Pu(n,2n) cross sections by directly detecting the 2n neutrons that are emitted. With the goal of improving the ^{239}Pu(n,2n) cross section and to measure the ^{241}Pu(n,2n) cross section for the first time. To that end, we have constructed a new neutron-charged-particle detector array called NeutronSTARS. It has been described extensively in Casperson et al. [1] and in Akindele et al. [2]. We have used this new neutron-charged-particle array to measure the ^{241}Pu and ^{239}Pu fission neutron multiplicity as a function of equivalent incident-neutron energy from 100 keV to 20 MeV. We have made a preliminary determination of the ^{239}Pu(n,2n) and ^{241}Pu(n,2n) cross sections from the surrogate ^{240}Pu(α,α'2n) and ^{242}Pu(α,α'2n) reactions respectively. The experimental approach, detector array, data analysis, and results to date are summarized in the following sections.

Experimental Approach

To measure the cross sections of interest we used the surrogate technique. By inelastically scattering alpha particles off the target we excited the desired nucleus over an equivalent incident-neutron energy range of 100 keV to 20 MeV. The desired reactions are ^{239}Pu(n,fxn), ^{239}Pu(n,2n), ^{241}Pu(n,fxn) and ^{241}Pu(n,fxn) which correspond to the surrogate reactions ^{240}Pu(α,α'fxn), ^{240}Pu(α,α'2n), ^{242}Pu(α,α'fxn) and ^{242}Pu(α,α'2n) respectively, see Figure 1.

We use surrogate reactions to overcome the difficulty or the impossibility (in some cases) of being able to make targets. For example, the desired reaction would be either the ^{241}Pu(n,2n) or ^{241}Pu(n,f) reaction. However, these are very difficult targets to make due to the short half-life (14.3 years) of the target nucleus. A 10-mg sample (typical mass needed for a direct measurement) would have an activity of 1.04 Curies, or 3.8×10^{10} decays per second. Instead, we can use a surrogate reaction to excite the same intermediate nucleus ^{242}Pu*. This way, we can measure the outgoing neutrons for the (n,2n) or the fission decay channel to measure the fission cross section as well as the average fission neutron multiplicity. The surrogate targets only need to have a mass of 100-200 micrograms and the ^{242}Pu isotope has a half-life of 375000 years. These two facts create a much lower activity target which is 0.8 microCuries, or 2.9×10^{4} decays per second. This target is a million times less radioactive and does not affect the performance of the detectors being used to make the measurement. Another reason we can use so little target material is because the alpha particle beam intensity is very high. A 1 nano-ampere particle beam has approximately 6×10^{9} particles per second and they are focused to a few millimeters creating a flux of approximately 6×10^{10} particles per second per cm^2.

The experiment consisted of a 14-day run conducted from the middle of April to the first week of May 2017 at the Texas A&M Cyclotron Institute using a 55-MeV alpha beam from the K150 Cyclotron. The alpha particles were used to inelastically excite the respective surrogate nuclei. The scattered alpha particles were detected with a silicon telescope array consisting of a 150 micron thick delta-E and 1000 micron thick E. Both detectors were made by Micron Semiconductor Inc. and were S2 model detectors. By knowing the energy of the beam, the angle of the scattered alpha particle and the energy of the scattered alpha particle we are able to reconstruct the excitation energy of the Plutonium nucleus. The silicon telescope enables us to perform particle identification as well as measure the energy and angle of the scattered particles, see Figure 2 and 3. Upstream of the target location there was another S2 silicon detector that was used to measure fission events. Fission fragments from a fission event escape the target with kinetic energies of approximately 100 MeV and were detected in the upstream silicon detector with a total efficiency of approximately 40%, see Figure 2. Surrounding the scattering chamber was the 2.2-ton neutron detector. The combined particle-neutron array was named NeutronSTARS, Neutron-Silicon Telescope Array for Reaction Studies. Extensive electronics developments and implementation was also undertaken for this project. We implemented new state of the art STRUCK 3316 digitizers to record the photomultiplier signals from the neutron detector array. The STRUCK digitizers also had to be adapted into the silicon data stream so we could perform online analysis to ensure the experiment was working correctly in real time. NeutronSTARS is described further in the next section.

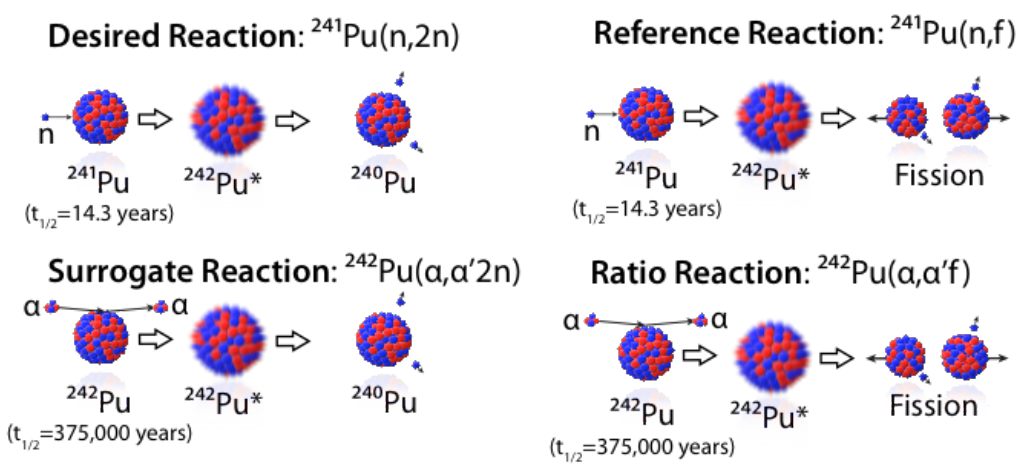

Figure 1. This figure illustrates a typical the surrogate reaction approach. For example, the desired reaction would be either the ^{241}Pu(n,2n) or ^{241}Pu(n,f) reaction (top two images). Instead, we use a surrogate reaction (bottom two images) to excite the same intermediate nucleus 242Pu*.

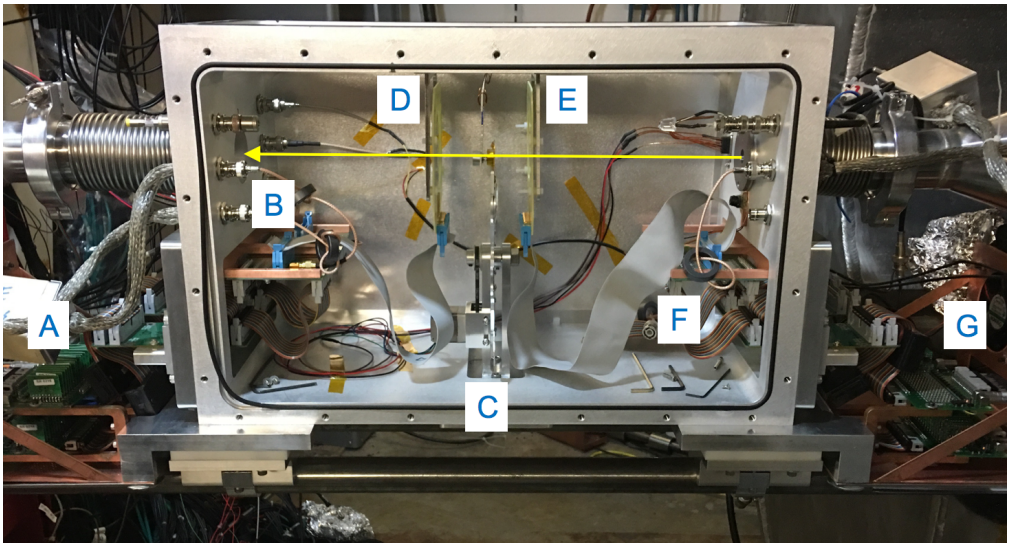

Figure 2. The NeutronSTARS nuclear-reaction chamber. The labels in the figure correspond to: A) Preamplifier board for the delta-E and E detectors; B) Breakout board, which separates the signal from the DC bias; C) Remotely controlled, stepper-motor-driven, 8-position target wheel; D) Downstream delta-E-E telescope; E) Upstream fission detector; F) Fission-detector breakout board; and G) Fission-detector preamplifier board. All of the apparatus in this figure were created for this project.

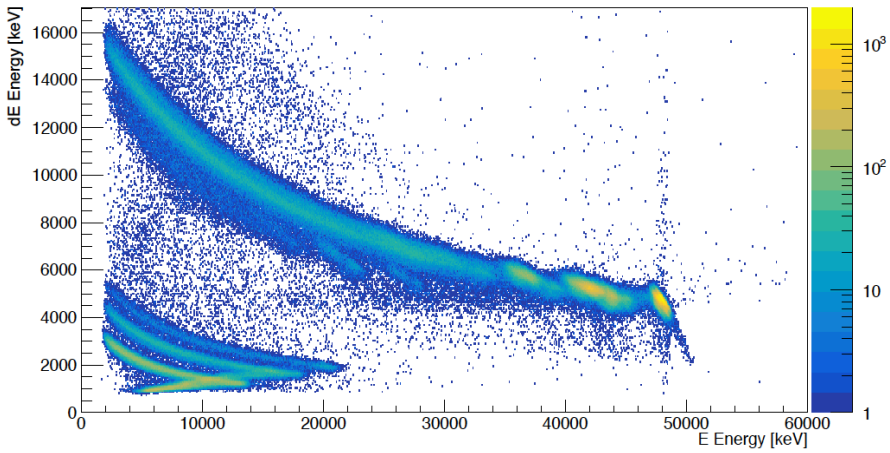

Figure 3. Particle-identification plot showing the energy lost in thin delta-E detector vs total energy for outgoing light ions in reactions following an incident alpha particle beam on Pu-242. The different A and Z charged particles appear as separate bands in the spectrum. There is clear separation between alpha particles (top band), He-3 (fainter strip below alpha band), and protons, deuterons and tritons (bottom three bands with protons on the bottom).

NeutronSTARS – Neutron Silicon Telescope Array for Reaction Studies

Neutrons were detected by a 2.2-ton liquid-scintillator neutron detector surrounding the target chamber. A major portion of time and effort for this project was the complete refurbishment and re-invention of the neutron detector array at the Texas A&M Cyclotron Institute. In fact, the only part of the original array (dating back to the 1980's and 1990's) that was left untouched was the aluminum tanks that hold the liquid scintillator. It was deemed to be more expedient to simply re-use the existing aluminum tanks rather than fabricating new ones.

A liquid scintillator neutron detector works via 2 dominant nuclear reactions and their secondary effects. Neutrons enter the liquid scintillator by passing through the aluminum walls of the scattering chamber and then through the wall of the aluminum tank holding the liquid scintillator. Once in the liquid the neutron quickly thermalizes (within ~10 microseconds) and then begins to scatter around at low energies in the liquid, see Figure 4. Neutron capture on a proton forms a deuteron and emits a 2 MeV gamma-ray, ($n+p \rightarrow d+\gamma$). This represents about 10% of the neutron capture events in the liquid scintillator. The majority of the neutron captures occur on the natural Gadolinium isotopes and release between 6 to 8 MeV via multiple gamma rays ranging in energy from approximately 1-3 MeV, ($Gd+n \rightarrow (Gd+1n)+x\gamma$). The gamma-rays from both nuclear capture reactions then interact and scatter in the liquid scintillator producing scintillation photons. The scintillation light is emitted in the 400 nm wavelength range. The liquid scintillator must have a long mean free path for the scintillation photons as they scatter multiple times before striking the photomultiplier surface and finally being detected. To aid the photon scattering the inside surface of the aluminum tanks is coated with a titanium dioxide paint. The titanium dioxide provides a diffusive scattering surface for the scintillation photons.

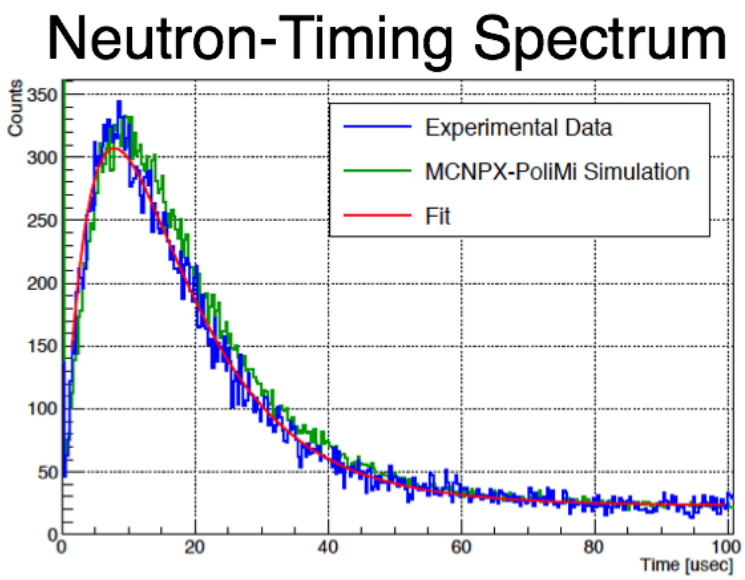

Figure 4. Courtesy of Oluwatomi (Tomi) Akindele who is currently a PhD student from UC Berkeley Nuclear Engineering Department working on this project for her PhD thesis. Tomi has performed and compared Monte Carlo modeling of the array using MCNPX-PoliMi and fit the data with empirical models from the literature to characterize the NeutronSTARS detector temporal response.

We performed a test run in December 2015 that was to serve as a benchmark measurement. Our goals were to measure the neutron multiplicity and the (n,2n) reaction using the surrogate reactions ^{236}U(α,α'fxn) and ^{236}U(α,α'2n) for the desired reactions of ^{235}U(n,fxn) and ^{235}U(n,2n) which are well known. The test run also served to ascertain the beam tune quality, beam induced backgrounds from the room, beam dump/faraday cup and any sources of scattering upstream that could produce neutrons.

The beam optics and room backgrounds proved to be acceptable and the rates were low enough as to not cause any issues. However, we notice within a month after the experiment that the energy resolution of the liquid scintillator was not correct. We determined that the energy resolution was essentially nonexistent despite results in the literature that lead us to believe otherwise. In order for the neutron detectors response to be precise enough for a surrogate reaction approach we had to completely overhaul the neutron detector. This became abundantly clear when we removed one of the existing photomultiplier tubes from each section of the neutron detector and looked inside to see the condition of the liquid scintillator, see Figure 5.

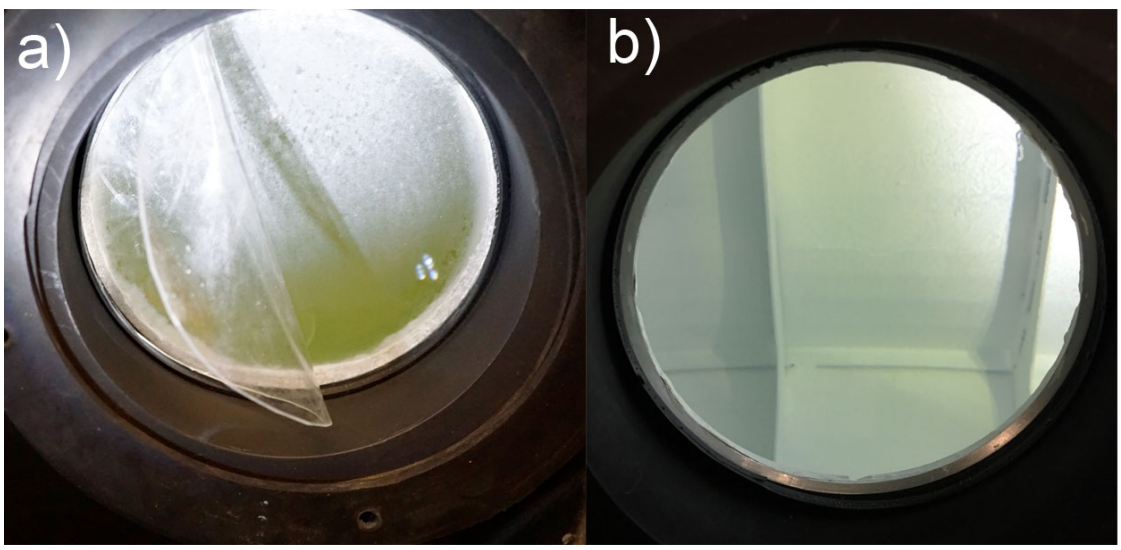

Figure 5. On the left in panel a) is the view of the site glass window that the photomultiplier tube is attached to. b) is a viewport now looking into the new EJ-335 liquid scintillator.

As you can see in Figure 5a, the old liquid scintillator had soured and was no longer a viable detector medium. The old liquid scintillator is nearly opaque and the photomultiplier window is covered in what we assume is precipitated Gadolinium. To achieve our goals, we drained and disposed of the old liquid scintillator by coordinating with the health and safety team at LLNL and Texas A&M University. Once the detector tanks were drained, they were cleaned with ethanol and wipes to remove any remaining contaminants from the tanks, see Figure 6. Each fish-eye lens that the photomultiplier tubes are attached to was cleaned. The tanks were sealed back up and then purged with dry nitrogen for 6-12 hours. After the tanks were purged they were filled with new liquid scintillator under dry nitrogen back fill gas until full. The EJ-335 liquid scintillator is composed of mostly pseudocumene and the smaller fraction of mineral oil (25-35%). The new liquid scintillator in the neutron detector is now Eljen Technologies EJ-

335 and contains 0.25% natural Gd by weight. In Figure 5b, the right panel shows the view the photomultiplier tube has into the new EJ-335 liquid. Then 4 gallons were removed from each tank to allow for the liquid scintillator thermal expansion. In total, twelve 55-gallon drums of EJ-335 liquid scintillator were put into the core sections of the neutron detector. All of this work was done solely by the LLNL team members see Figure 6 and 7.

Once the NeutronSTARS array was completed we began the task of instrumenting it with new 5-inch diameter photomultiplier tubes we purchased from ADIT. The photomultiplier tubes and bases were designed in conjunction with ADIT to provide high quantum efficiency (>20%) in the 400 nm wavelength range to best match them to the scintillation light wavelength of the EJ-335 liquid scintillator. The photomultiplier tube bases were made using a linear resistor network and used only passive circuit elements. The photomultiplier tubes were all tested and gain matched to their single photo-electron gains at LLNL in the Millennium-Falcon Laboratory before being shipped to the Texas A&M Cyclotron Institute for installation. The photomultiplier tubes were installed and mated to the glass viewports using circular 3-4 mm thick silicone sheets.

With all the apparatus in place and running, characterization with gamma ray sources and neutron sources were performed extensively to determine the response of the photomultiplier tubes prior to the actual beam time.

Figure 6. Initial loading of 6 barrels (330 gallons) of new EJ-335 liquid scintillator took place in December 2016. New photomultiplier tubes were also installed at that time. On the left: A 55-gallon drum of EJ-335 liquid scintillator is shown in foreground. Dr. Barbara Alan suited up is installing PMTs on NeutronSTARS. On the right Dr. Jason Burke cleaning out the top quadrant preparing for loading of new EJ-335 liquid scintillator.

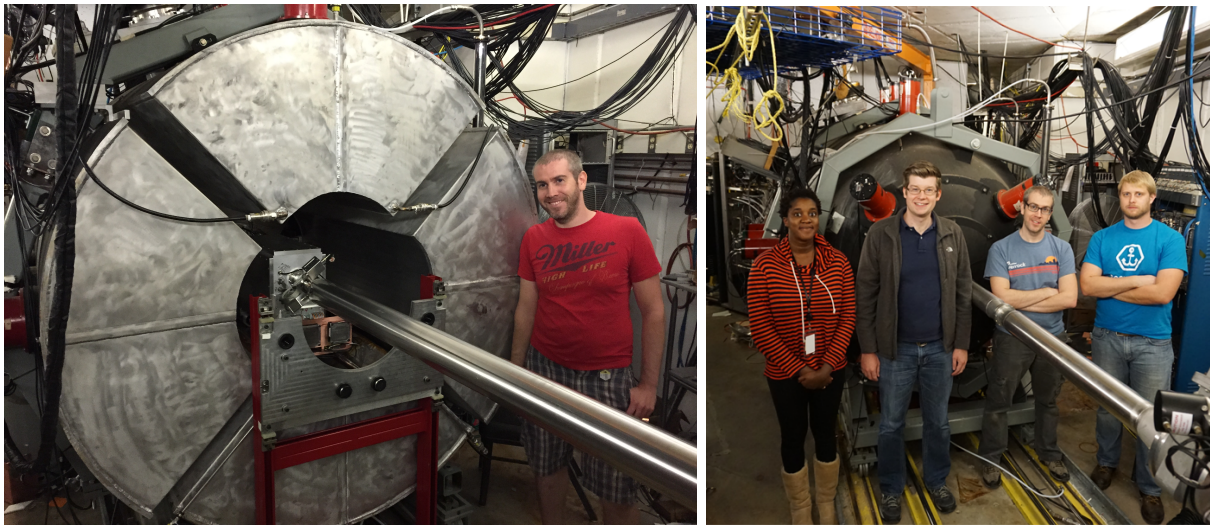

Figure 7. NeutronSTARS chamber installed on a K150 Cyclotron beam line at Texas A&M Cyclotron Institute. On the left, Richard Hughes (LLNL staff physicist) is standing in front of the NeutronSTARS core section. In the middle of the core is the scattering chamber with the silicond detectors. The beamline is running into the center of the array from left to right. On the right side we see Tomi Akindele (UC Berkeley PhD graduate student), Robert Casperson (LLNL staff member), Richard Hughes (LLNL staff physicist) and John Koglin (Lawrence Graduate Scholar/Penn State graduate student). Dr. John Koglin was recently hired into Design Physics Division at LLNL as a staff member.

Neutron detector efficiency

We characterized the response and performance of the NeutronSTARS array to neutrons using Cf252 and AmBe as neutron sources. With Cf252 we used the fact that 3% of the time Cf252 spontaneously fissions. By tagging on the fission events with the upstream fission detector we were able to measure the fission efficiency and the neutron detector efficiency. For every fission-event we looked at coincident neutron events. We determined the fission neutron multiplicity by adjusting the neutron detector efficiency in the fit parameters until we got agreement with the accepted neutron multiplicity for Cf252, see Figure 8. The fission detector efficiency is 40% for the experimental conditions and the single neutron detection efficiency was 50% +/- 1% with the calibrations and energy cuts used for this analysis.

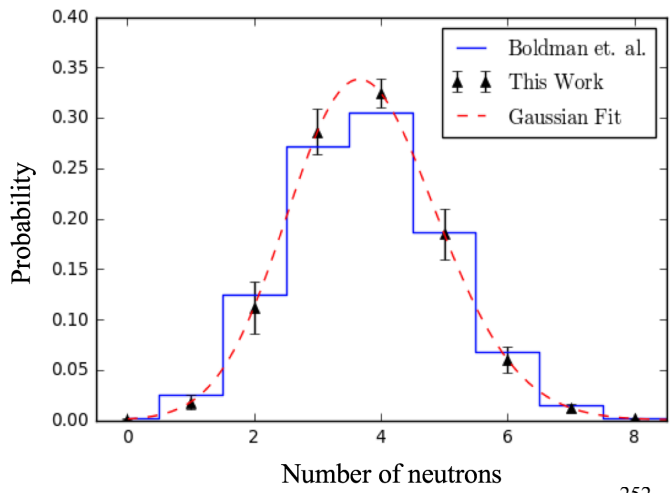

Figure 8. ^{252}Cf neutron multiplicity following fission obtained from a ^{252}Cf source located in the center of the NeutronSTARS detector. Plot courtesy of Oluwatomi Akindele for her PhD thesis project at LLNL.

Neutron Multiplicity

With the data from the two-week long run on ^{242}Pu and ^{240}Pu targets by knowing the neutron detector efficiency, we could then use the detected alpha particles to gate on different excitation energies and measure the neutron multiplicity we observed at that energy. The results were corrected for the neutron detector efficiency for measuring 1n, 2n, 3n, etc... events. In this way we were able to determine the fission neutron multiplicity of ^{239}Pu(n,fxn) and ^{241}Pu(n,fxn) see Figures 9 and 10. We found excellent agreement in both cases to previous results. We also were able to measure the neutron distribution, not just the average number of neutrons emitted, see Figure 11.

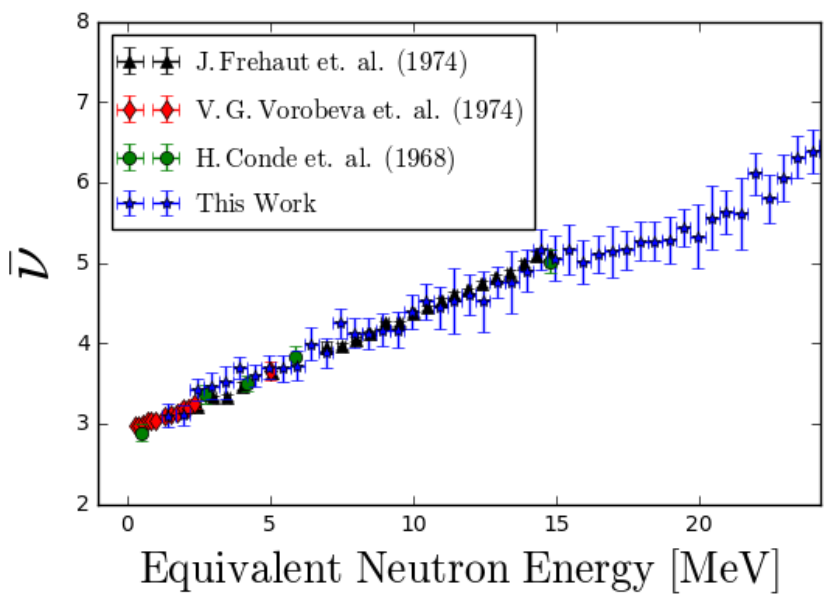

Figure 9. Energy dependent ^{241}Pu neutron multiplicity following fission obtained from a surrogate reaction of ^{242}Pu(α,α'fxn) using NeutronSTARS, see references 2-8. The plot is courtesy of Oluwatomi Akindele for her PhD thesis project at LLNL.

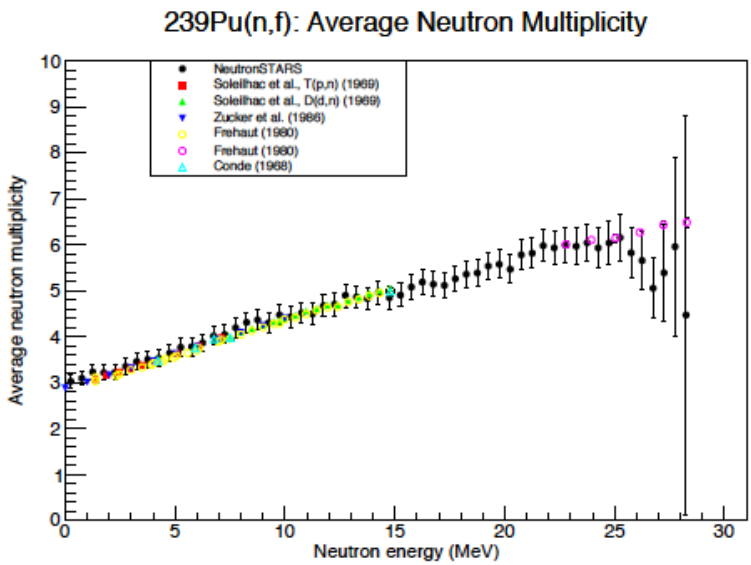

Figure 10. Energy dependent ^{239}Pu neutron multiplicity following fission obtained from a surrogate reaction of ^{240}Pu(α,α'fxn) using NeutronSTARS, see references 2-8. This plot was produced by Dr. Barbara Alan.

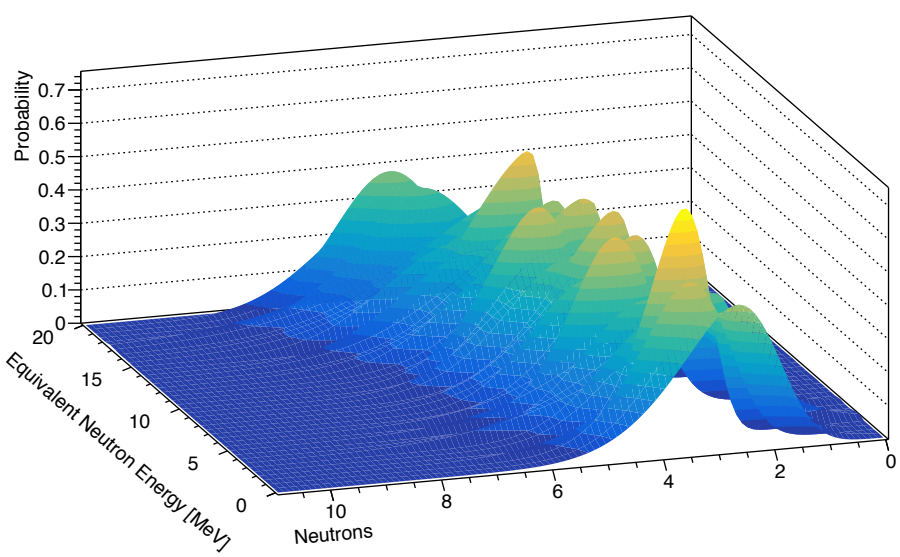

Figure 11. Shows the fission neutron multiplicity distribution from ^{241}Pu (α,α'fxn) surrogate measurement data. The fission neutron multiplicity distribution goes from 0 to 20 MeV equivalent neutron energy. Notice that the distribution is quite dynamic as the incident neutron energy increases. The plot is courtesy of Oluwatomi Akindele for her PhD thesis project at LLNL.

^{239}Pu(n,2n) and ^{241}Pu(n,2n) cross sections

Similarly, we used the alpha events to determine the excitation energy of the nucleus and the measured coincident fission events and neutron events to determine ^{239}Pu(n,2n) and ^{241}Pu(n,2n) cross sections. The cross section is determined as a ratio of the number of 2n events to the number of fission events as can be seen here

$$\sigma(239\mathrm{Pu(n,2n)}) = \frac{\mathrm{N(240Pu(\alpha,\alpha'2n))}}{\mathrm{N(240Pu(\alpha,\alpha'f))}} \times \frac{\varepsilon_{fission}}{\varepsilon_{2neutron}} \times \sigma(239\mathrm{Pu(n,f)}),$$

where $\sigma(^{239}\mathrm{Pu(n,2n)})$ is the (n,2) cross section for ^{239}Pu, $\mathrm{N}(^{240}\mathrm{Pu}(\alpha,\alpha'2n))$ is the net number of 2n events, $\mathrm{N}(^{240}\mathrm{Pu}(\alpha,\alpha'f))$ is the net number of fission events, $\varepsilon_{\mathrm{fission}}$ is the fission detection efficiency, $\varepsilon_{\mathrm{2neutron}}$ is the 2 neutron detection efficiency and $\sigma(^{239}\mathrm{Pu(n,f)})$ is the known (n,f) cross section for ^{239}Pu. The same equation applies to ^{241}Pu(n,2n). This equation is applied to each energy bin that is measured and the results are shown in Figures 12 and 13 below.

For the ^{239}Pu(n,2n) result we agree with previous measurements however there is still scatter in the result we have to date around previous measurements, see Figure 12. Figure 13 shows the first ever result for the 241Pu(n,2n) cross section. The results are valid in the range of the (n,2n) cross section between approximately 6 to 12 MeV. Above 12 MeV the 3n channel turns on and is biasing the result of the (n,2n) measurement.

Further refinement of the background subtraction and the neutron detection threshold should improve these results. We expect to be able to improve the detection efficiency by performing a secondary photomultiplier tube gain calibration which will provide even better energy resolution in the neutron detector. This will allow us to lower our neutron energy detection cut and increase our statistics by as much as a factor of 2. Doing so will also reduce the background events proportionally. The net effect will be a reduction in the scatter of the cross section and a reduction in the uncertainty of the cross section in both cases.

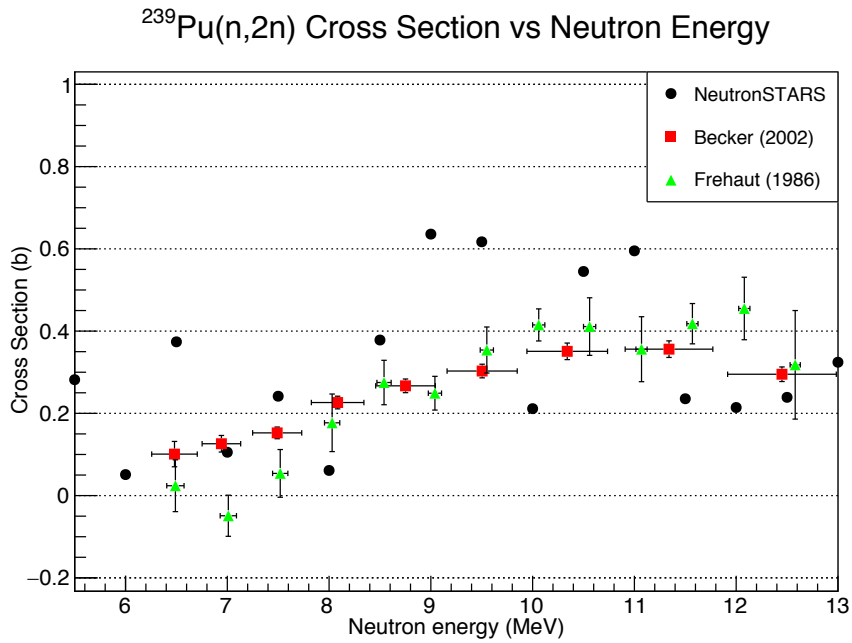

Figure 12. Shows the ^{239}Pu(n,2n) cross section determined from the ratio of the number of alpha events inducing 2n events divided by the number of alpha fission events multiplied by the known ^{239}Pu(n,f) cross section; $\sigma(^{239}\mathrm{Pu(n,2n)})=(N(^{240}\mathrm{Pu}(\alpha,\alpha`2n))//N(^{240}\mathrm{Pu}(\alpha,\alpha`f)))\cdot\sigma(^{239}\mathrm{Pu(n,f)})$ The efficiency of the fission detector and the neutron detector are also included in this equation. This result is from the analysis performed by Dr. Barbara Alan.

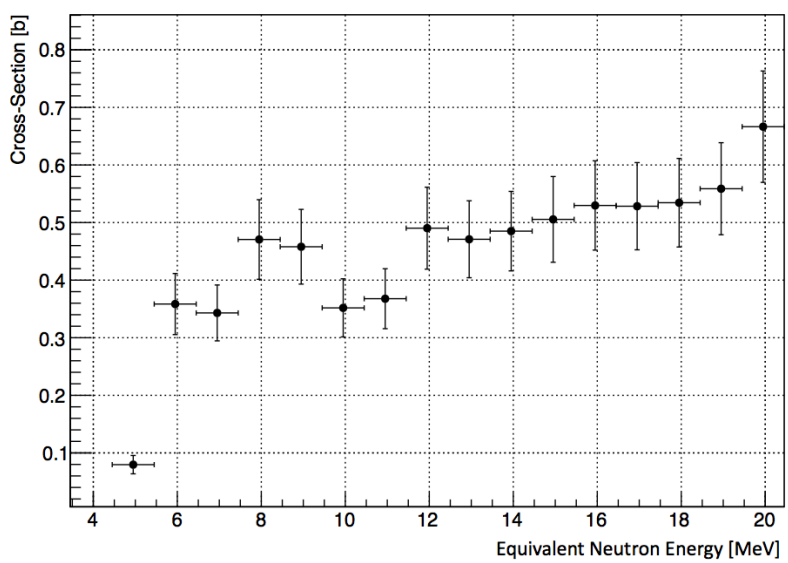

Figure 13. Shows the ^{241}Pu(n,2n) cross section determined from the ratio of the number of alpha events inducing 2n events divided by the number of alpha fission events multiplied by the known ^{241}Pu(n,f) cross section; $\sigma(^{241}\mathrm{Pu(n,2n)})=(N(^{242}\mathrm{Pu}(\alpha,\alpha`2n))//N(^{242}\mathrm{Pu}(\alpha,\alpha`f)))\cdot\sigma(^{241}\mathrm{Pu(n,f)})$ The efficiency of the fission detector and the neutron detector are also included in this equation. Plot from analysis performed by Tomi Akindele as part of her PhD thesis at LLNL. The increasing cross section above ~14 MeV is an indication that some number of 3n events are being misidentified as 2n events once the 3n threshold is crossed.

Conclusion

We have designed, fabricated, characterized and performed experiments with a new charged particle and neutron array called NeutronSTARS. We have developed and performed the first ever direct neutron detection surrogate reactions in the world. We have determined the fission neutron multiplicity of both ^{239}Pu and ^{241}Pu over an energy range from 100 keV to 20 MeV equivalent neutron energy for the first time in a single measurement. We have made a determinination of the ^{239}Pu(n,2n) and ^{241}Pu(n,2n) cross sections using the surrogate approach. There still remains improvements to be made in the analysis of the data we have in hand which should further reduce the uncertainties and provide an even more robust result. For instance, there is still likely some number of single neutrons contributing to a background underneath the 2n region. This is due to an imperfect background subtraction which we are working to improve.

The most important conclusion is that we have proven that this approach has merit and can be applied to a whole host of nuclear cross sections of interest over a wide range of isotopes. The results to date will be written up for peer reviewed publication in the coming months.

Auspices
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