LAWRENCE LIVERMORE NATIONAL LABORATORY

LLNL-TR-739507

Surrogate 239Pu(n,fxn) and 241Pu(n,fxn) average fission-neutron-multiplicity measurements

J. T. Burke, B. S. Alan, O. A. Akindele, R. J. Casperson, R. O. Hughes, S. Fisher

October 4, 2017

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Surrogate $^{239}Pu(n,fxn)$ and $^{241}Pu(n,fxn)$ average fission-neutron-multiplicity measurements

J.T. Burke, B.S. Alan, O.A. Akindele, R.J. Casperson, R.O. Hughes and S. Fisher
September 2017

Introduction

We have constructed a new neutron-charged-particle detector array called NeutronSTARS. It has been described extensively in LLNL-TR-703909 [1] and 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. The experimental approach, detector array, data analysis, and results are summarized in the following sections.

Experimental Approach

This experiment used the surrogate technique to excite the desired nucleus over an equivalent incident-neutron energy range of 100 keV to 20 MeV. The desired reactions are $^{239}Pu(n,fxn)$ and $^{241}Pu(n,fxn)$, which correspond to the surrogate reactions $^{240}Pu(\alpha,\alpha'fxn)$ and $^{242}Pu(\alpha,\alpha'fxn)$, respectively; see Figure 1. The experiment was conducted Spring 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 recorded with a silicon telescope array. The neutron channel was detected by a 2.2-ton liquid-scintillator neutron detector surrounding the target.

NeutronSTARS Detector Array

A silicon telescope consisting of a thin (150-micron) delta-E and thick (1000-micron) E detector was assembled as described in Akindele et al [2]. A silicon fission detector was located approximately 2.0 cm upstream of the target location and the silicon telescope was located approximately 2.0 cm downstream of the target. The silicon telescope covered an angle range of approximately 35 to 65 degrees with respect to the beam. The ^{242}Pu and ^{240}Pu targets were mounted on a remotely controllable target wheel in the exact center of the vacuum chamber, which is also located inside the exact center of the NeutronSTARS neutron detector; see Figure 2.

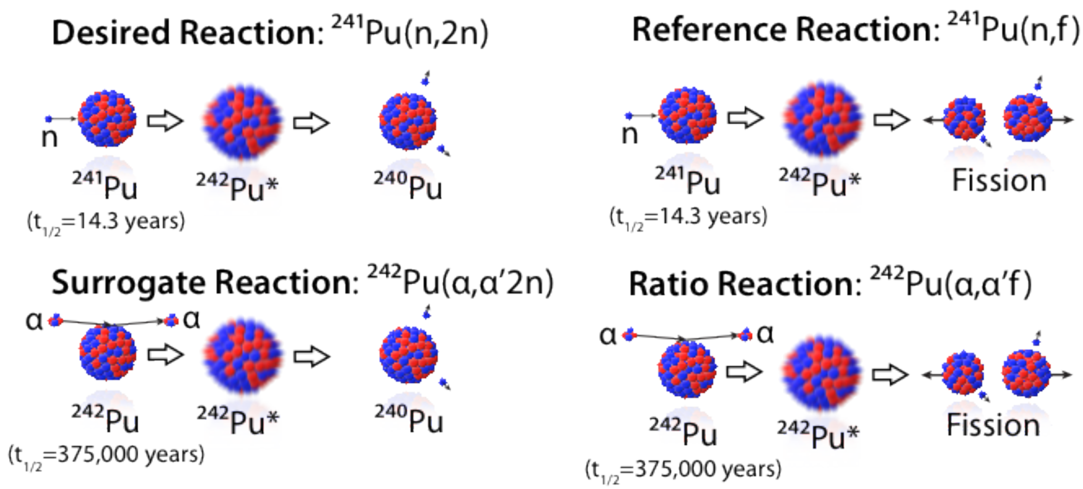

Figure 1. This figure illustrates a typical the surrogate reaction approach. For example, the desired reaction would be either the ^{241}Pu(n,2n) or ^{241}Pu(n,f) reaction (top two images). However, these are very difficult due to the short half-life (14.3 years) of the target nucleus. A 10-mg sample (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 use a surrogate reaction (bottom two images) to excite the same intermediate nucleus ^{242}Pu*. This way, we can measure the outgoing neutrons for (n,2n) or tag on fission to measure the fission cross section and also the average fission neutron multiplicity. The surrogate targets only need to have a mass of 100-200 micrograms and the specific activity is 0.8 microCuries, or 2.9×10^{4} decays per second. This target is a million times less radioactive.

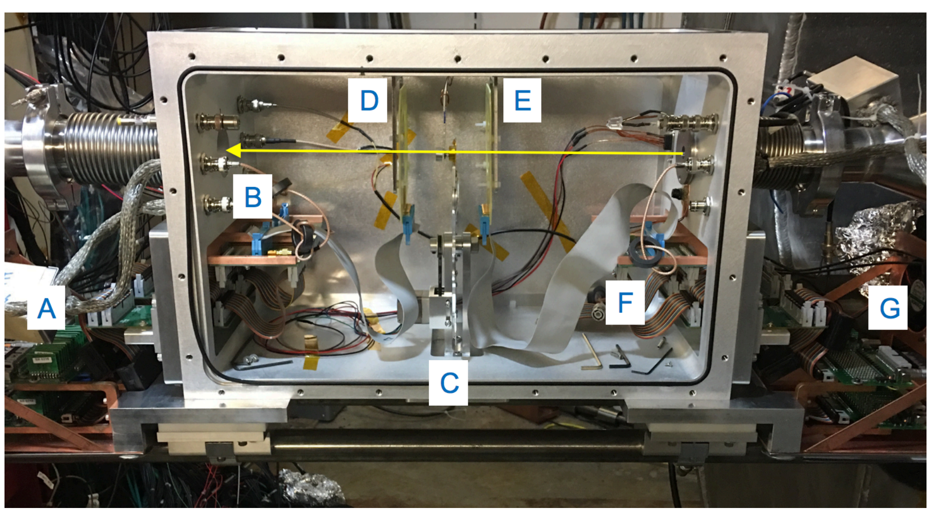

Figure 2. The NeutronSTARS nuclear-reaction chamber. The labels in the figure correspond to: A) Preamplifier board for the delta-E and E detectors; B) Breakout board, which separates the signal from the DC bias; C) Remotely controlled, stepper-motor-driven, 8-position target wheel; D) Downstream delta-E-E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

The neutron detector described in Akindele [2] consists of 2.2-tons of Eljen EJ-335 liquid scintillator (pseudocumene and mineral oil) doped with 0.25% natural Gd by weight. Any neutrons produced from excitation of the nuclei pass through the vacuum-chamber walls and enter the liquid-scintillator-filled tank surrounding the vacuum chamber. The neutrons are moderated and eventually thermalized by the hydrogen, carbon and oxygen in the liquid scintillator. They can capture on the Gd and then emit 6-8 MeV of energy in the form of a series of 1-2-MeV gamma rays. The gamma rays deposit energy in the liquid scintillator and cause it to scintillate, emitting photons primarily in the 400-nm wavelength. The liquid scintillator emits approximately 9500 photons per MeV of energy deposited. The scintillation photons are measured by photomultiplier tubes located on the outside of the liquid-scintillator-filled tank. The photomultiplier-tube signals are collected and timestamped for later correlation with the observed alphas events detected in the silicon telescope. The photomultiplier tubes were also calibrated and gain aligned prior to being installed on the NeutronSTARS detector array; see Figure 3.

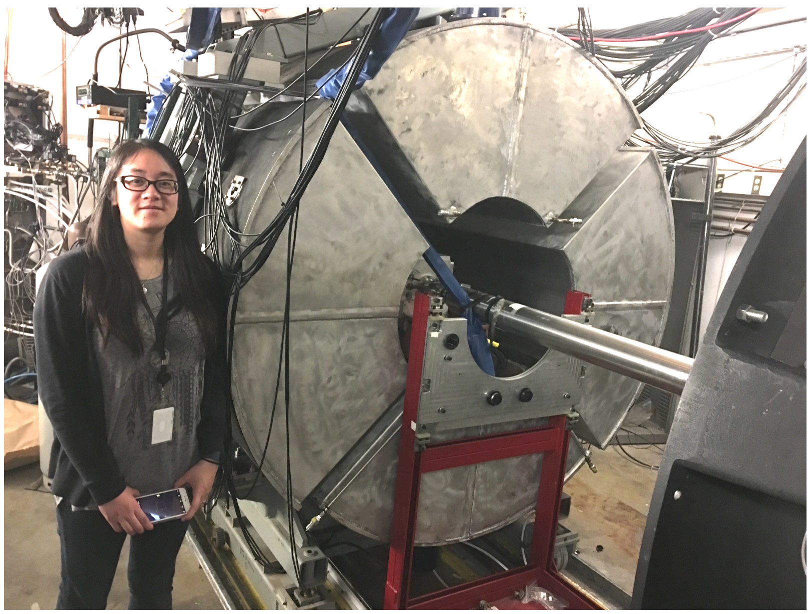

Figure 3. Dr. Barbara Alan standing beside the NeutronSTARS detector array. The central core with 2.2 tons of Eljen EJ-335 liquid scintillator is divided into 4 physical quadrants. Note the beamline passing through the center of the NeutronSTARS core. The beam passes from right to left through the array in this image.

Data Analysis

In the data analysis, the silicon-detector energy calibrations are determined by using a multiple-energy alpha-emitting Ra-226 alpha source. The photomultiplier tubes were calibrated prior to being installed as well as in situ with gamma ray sources (e.g., Co60, Cs137, Na22). The alpha particles are identified using the energy deposited in the delta-E and E detectors. Once the alpha particles have been identified, we can subdivide the data into energy bins that represent the excitation energy of the nucleus being studied; see Figure 4. For each excitation-energy bin we can then determine the average number of neutrons emitted from a fission event.

Neutrons are identified by their energy deposition in the liquid scintillator and their interaction time with respect to the inelastic alpha-particle events that excited the nucleus being studied. A prompt time region is defined from 2-60 microseconds after the initial alpha-trigger event. A background, or delayed, region is also defined in a temporal region from 400-450 microseconds or any suitable background time frame. The prompt-time and delayed-time neutron events are subtracted from each other, which removes room background and beam related backgrounds and yields a net number of neutrons; see Figure 5.

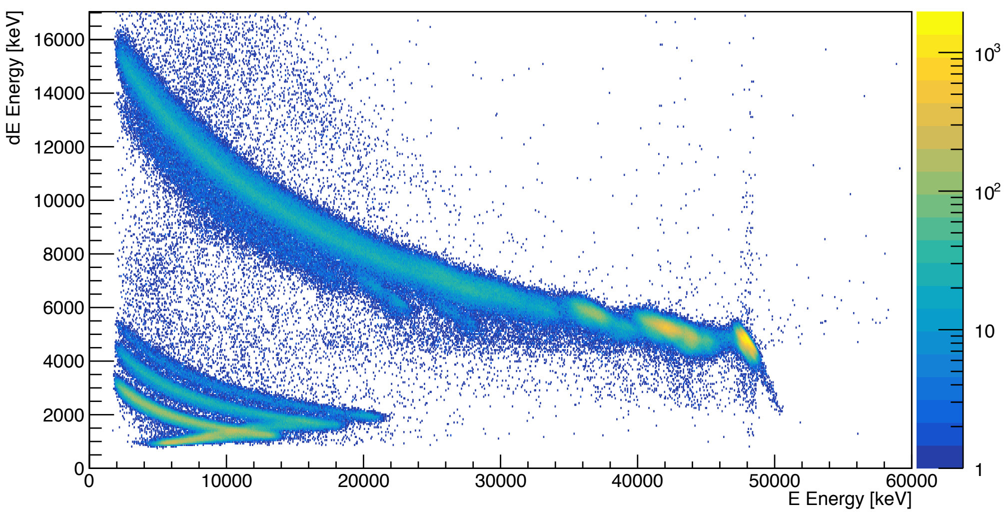

Figure 4. Particle-identification plot showing the energy lost in thin delta-E detector vs total energy for outgoing light ions in reactions following an incident alpha particle beam on Pu-242. The different A and Z charged particles appear as separate bands in the spectrum. There is clear separation 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).

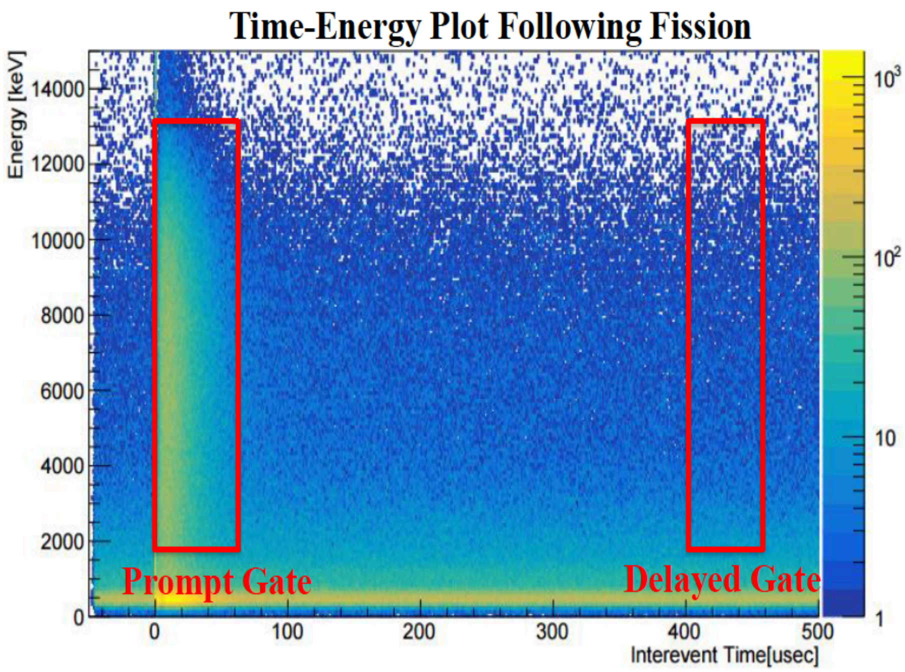

Figure 5. This plot shows the energy-versus-time response of the NeutronSTARS array to an incident neutron following fission. The prompt gate is triggered by observing a fission event in this case. The bright yellow energetic events in the prompt gate are the result of the neutrons capturing on Gd in the liquid and the extent in time is due to the moderation and capture time. The delayed gate is set well outside the prompt gate and used to subtract room background and beam related backgrounds.

Results: Average Fission-Neutron Multiplicity

In order to verify the analysis codes, analysis methodology, as well as the measured and simulated efficiency for NeutronSTARS, the spontaneous ^{252}Cf fission-source neutron multiplicity was measured with the array and compared to well established literature data. As can be seen in Figure 6 below, the NeutronSTARS detector array closely reproduces the expected neutron-multiplicity distribution and average neutron multiplicity, nubar. The single-neutron detection efficiency for NeutronSTARS with a 2 MeV energy cut on the liquid scintillator signal is approximately 54% +/- 1%.

Neutrons emitted from fission represent a background signal for the measurement of the 2n reaction channel. We therefore need to measure the neutrons emitted from fission events precisely in order to perform an appropriate subtraction. The surrogate experiment is designed so that we simultaneously measure the 2n and fxn exit channels within an experiment. As a result, we precisely measure the fission neutron multiplicity as a function of the compound-nucleus excitation energy over a range from the fission threshold to 20 MeV in a surrogate experiment. Figure 7 shows preliminary results of nubar for ^{241}Pu(n,fxn) from a recent surrogate measurement of ^{242}Pu (α,α'fxn). We have also performed the same analysis for ^{240}Pu(α,α'fxn) which is a surrogate reaction for ^{239}Pu(n,fxn), whose results are shown in Figure 8.

Not only do we determine the average neutron multiplicity, but we also measure the neutron-multiplicity distribution as a function of energy from threshold to 20 MeV; see Figure 9. These distribution results are preliminary at this time, as we are making final checks on the analysis.

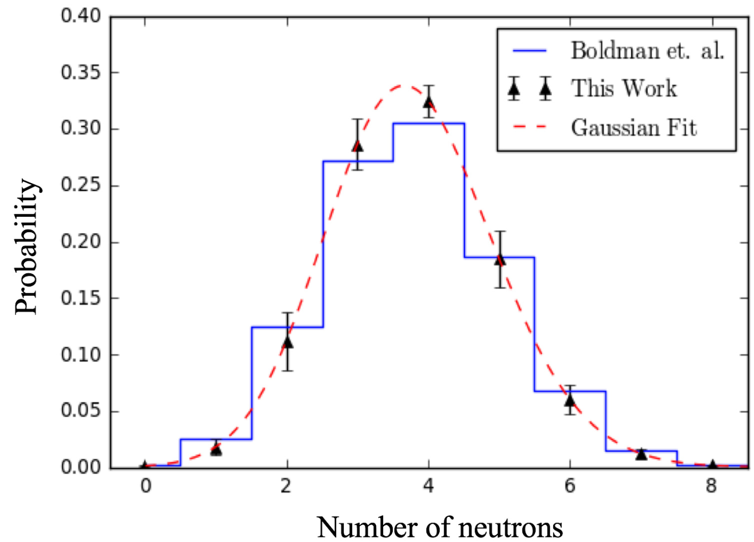

Figure 6. ^{252}Cf neutron multiplicity following fission obtained from a ^{252}Cf source located in the center of the NeutronSTARS detector.

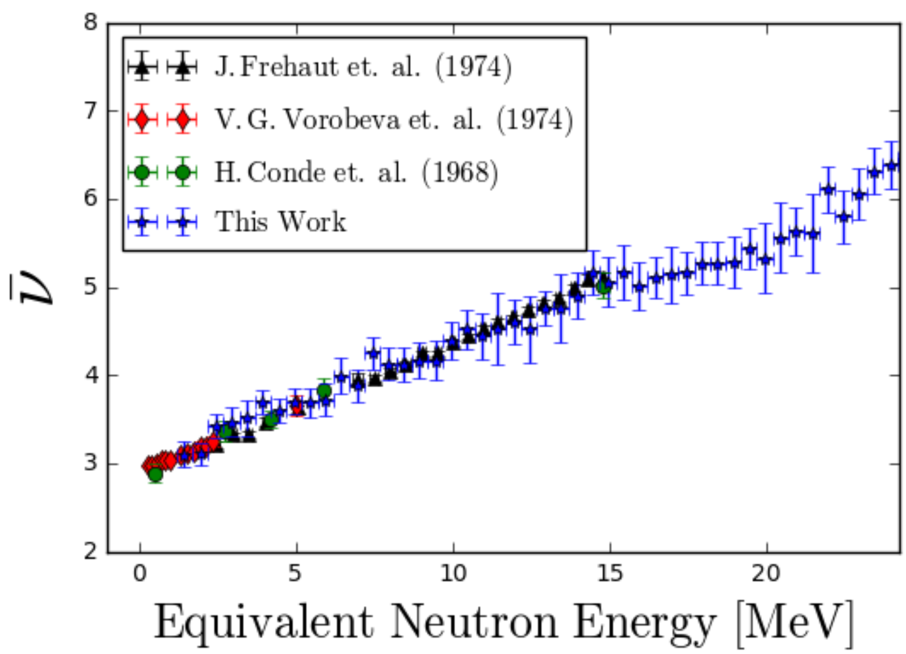

Figure 7. Energy-dependent ^{241}Pu neutron multiplicity following fission obtained from a surrogate reaction of ^{242}Pu(α,α'fxn) using NeutronSTARS. References for the data can be found in the Reference section Frehaut [3], Vorobeva [4], Conde [5].

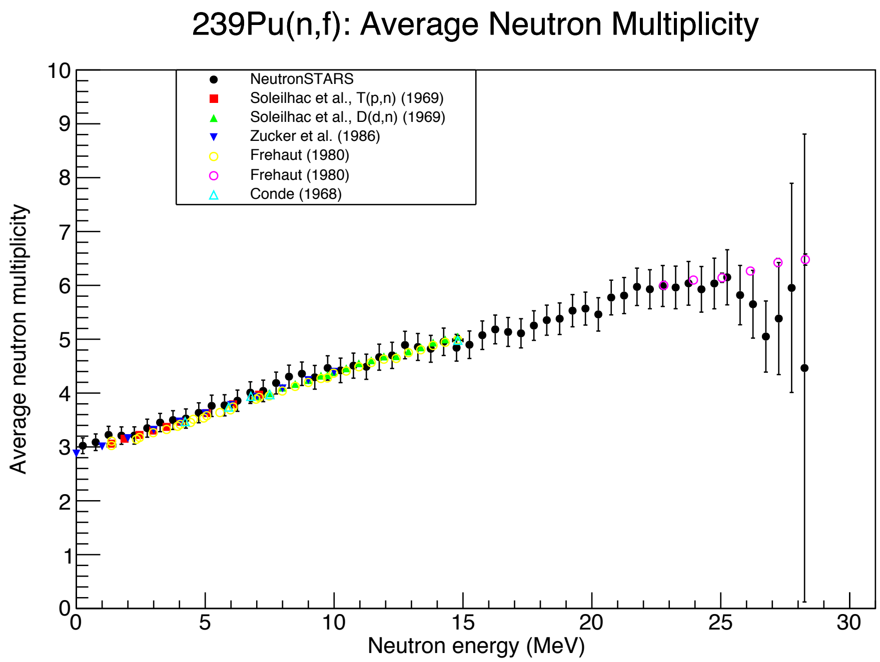

Figure 8. Energy-dependent ^{239}Pu neutron multiplicity following fission obtained from a surrogate reaction of ^{240}Pu(α,α'fxn) using NeutronSTARS. The data above 25 MeV should be disregarded as the Coulomb barrier is beginning to affect the results from the direct reaction. References for the data can be found in the Reference section Soleilhac[6], Zucker [7], Frehaut [8], Conde [5].

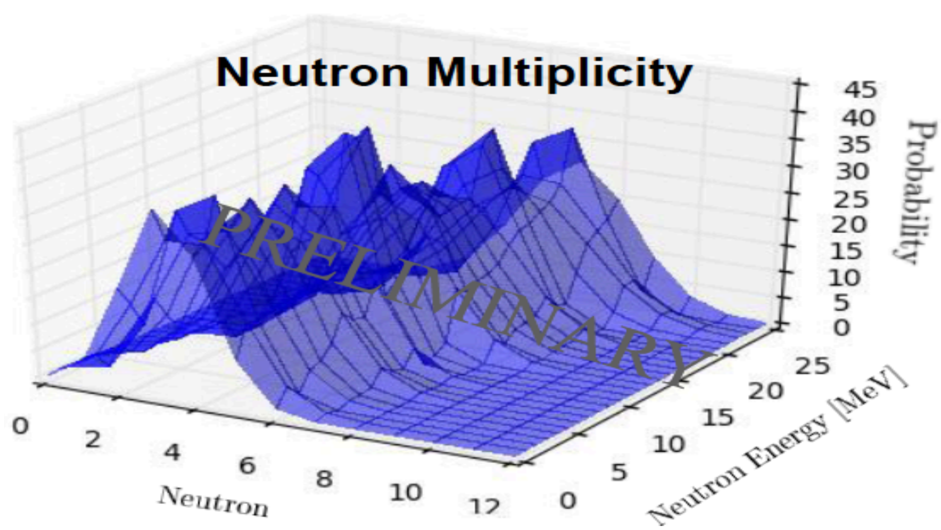

Figure 9. Shows the fission-neutron-multiplicity distribution from ^{240}Pu (α,α'fxn) surrogate measurement data.

Conclusion

We have successfully measured the ^{239}Pu(n,fxn) and ^{241}Pu(n,fxn) average neutron multiplicity over an equivalent incident-neutron energy from 100 keV to 20 MeV using the surrogate reactions ^{240}Pu(α,α'fxn) and ^{242}Pu(α,α'fxn) respectively. Our results agree with previous measurements and extend the energy range of the average fission neutron multiplicity data for ^{239}Pu and ^{241}Pu to 25 MeV. These results are also the first ever surrogate measurements using direct neutron detection to identify the outgoing channel. The fission neutron multiplicity data is also needed as it is a background for the (n,2n) cross section measurements we are performing.

These results will be written up for publication in a peer reviewed journal (e.g., Physical Review C). The results will also be made available to the EXFOR database and internally to NNSA Programs.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344, and supported by the by the U.S. DOE/NNSA Office of Defense Nuclear Nonproliferation Research and Development, Department of Energy National Nuclear Security Administration under Award Number DENA0000979, and the Department of Energy National Nuclear Security Administration through the Nuclear Science and Security Consortium under Award Number DE-NA-0003180.

References

1) “Surrogate Measurements of Actinide (n,2n) Cross Sections with NeutronSTARS”, R.J. Casperson, J.T. Burke, R.O. Hughes, O.A. Akindele, J.D. Koglin, B. Wang, A. Tamashiro, September 17, 2016, LLNL-TR-703909

2) “NeutronSTARS: A segmented neutron and charged particle detector for low-energy reaction studies”, O.A. Akindele, R.J. Casperson, B.S. Wang, J.T. Burke, R.O. Hughes, S.E. Fisher, A. Saastamoinen and E.B. Norman, Nuclear Inst. and Methods in Physics Research, A 872 (2017) p. 112–118 (LLNL-JRNL-736490)

3) Rept: Centre d`Etudes Nucleaires, Saclay Reports, No.4626 (1974) NSR: 1974FRZF “Measurement Of The Average Number,Nu-Bar, Of The Prompt Neutrons Emitted In The Pu-240 And Pu-241 Fission Induced By Neutrons Of Energy Between 1.5 And 15 Mev.” (In French). J.Frehaut, G.Mosinski, R.Bois, M.Soleilhac

4) “Energy Dependence of Average Number of Prompt Neutrons in Pu241 Fission”, N.P.Dyachenko, N.P.Kolosov, B.D.Kuzminov, A.I.Sergachev, V.M.Surin, Atomnaya Energiya, Vol.36, Issue.4, p.321 (1974)   NSR: 1974DY03

5) “Prompt nu-bar in neutron-induced fission of ^{239}Pu and ^{241}Pu”, H. Conde, J. Hansen and M. Holmberg, Research Institute of National Defence, Stockholm, Sweden, Journal of Nuclear Energy Vol. 22, pp. 53 to 60. Pergamon Press 1968

6) “Energy Dependence of it, for neutron induced fission of ^{235}U, ^{238}U AND ^{239}Pu from 1-3 to 15 MeV” M. Soleilhac, J. Frehaut and J. Gauriau, Journal of Nuclear Energy, Vol. 23, pp. 257 to 282. Pergmon Press 1969.

7) "Energy dependence of the neutron multiplicity Pv in fast neutron induced fission of ^{235,238}U and ^{239}Pu”, M. S. Zucker and N. E. Holden, Brookhaven National Laboratory, Upton, New York 11973, BNL-38491, DE86 015078 (1986).

8) “^{239}Pu nu-bar”, J. Frehaut Priv. Comm. to EXFOR 1980. Listing is in the nuclear data sheets.