

New Capabilities of the RPI γ -Multiplicity Detector to Measure γ -Production

Katelyn Cook^{1,*}, Ezekiel Blain¹, Amanda Lewis², Devin Barry², Michael Rapp², Adam Daskalakis², Peter Brain¹, Dominik Fritz¹, Adam Ney¹, Sukhjinder Singh¹, Benjamin Wang¹, and Yaron Danon¹

¹Gaerttner LINAC Center, Rensselaer Polytechnic Institute, Troy, NY 12180

²Naval Nuclear Laboratory, P.O. Box 1072, Schenectady, New York 12301

Abstract. Accurate modeling of γ -production in neutron capture reactions is critical for many applications including non-proliferation, safeguards and modeling nuclear reactors. To improve this work, the Rensselaer Polytechnic Institute (RPI) 16-segment γ -multiplicity NaI(Tl) detector at the Gaerttner Linear Accelerator (LINAC) Center has been upgraded by implementing a digital data acquisition system. The new digitized system records the γ -energy deposition distribution in each individual detector, and γ -multiplicity values as a function of neutron time-of-flight (TOF). With the new capabilities, high precision capture (and fission) yield measurements can be made, and the accuracy of simulation tools used to predict capture γ -cascades can be tested. To validate the updated system, an experiment was performed using a natural Ta sample to measure ^{181}Ta and ^{180m}Ta resonance capture yield by detecting prompt γ -rays emitted from neutron capture interactions as a function of both neutron energy and measured γ -multiplicity of each capture event. The results confirm earlier measurements and agree with theoretical yield in the low energy resonance region from 1 to 20 eV. A $^{238}\text{U}(n, \gamma)$ measurement was also performed to generate γ -spectra. For capture γ -cascades where the total γ -energy deposition is close to the neutron binding energy, γ -spectra were measured for individual resonance energies and observed γ -multiplicities. The results are comparable in shape to a recent measurement done using the Detector for Advanced Neutron Capture Experiments (DANCE) array at Los Alamos Neutron Science Center (LANSCE); however, differences need to be compared to Monte-Carlo n-particle simulations.

1 Introduction

Measuring γ -emission energy spectra in neutron capture reactions is important to help understand γ -heating in nuclear reactors. To increase the accuracy of capture γ -spectra measurements, updates have been made to the Rensselaer Polytechnic Institute (RPI) γ -multiplicity detector. The new system includes a SIS3316 16 channel 250 MHz 14-bit Flash Analog-to-Digital Converter (FADC) based data acquisition system. This enables digitization of the pulse wave for all events on each of the 16 detector segments resulting in detailed capture (and fission) γ -spectra to compare to simulations. With this information, event discrimination is flexible to both energy and time-of-flight (TOF). The experimental design also includes a new interface that controls the sample changer which allows the system to collect data more efficiently and automatically. The detector is used to measure capture and fission yields in the 0.01 eV - 3 keV energy range.

A major benefit of the new detector system is the ability to generate distributions of γ -energy, neutron energy, coincidence and γ -multiplicity. The γ -energy distribution can be measured in each individual detector or for each observed γ -multiplicity value. Using these distributions, information can be obtained about the capture γ -cascades which can help constrain models that are used for reaction calculations.

*e-mail: cookk4@rpi.edu

A natural tantalum sample (^{nat}Ta) was used to measure the ^{181}Ta and ^{180m}Ta resonance capture yield as a function of incident neutron energy and validate the new data acquisition system. The ^{nat}Ta data set was also used to compare experimental γ -spectra to simulations. In addition to the ^{nat}Ta measurement, capture γ -spectra of ^{238}U were measured to further understand important nuclear materials. The methods used to perform these measurements, compare to previous measurements and evaluate the accuracy of current simulations are described in this paper.

2 Experimental Setup

The RPI γ -multiplicity detector, shown in Figure 1, has 16 segments (20 L total volume) of NaI(Tl). The inside of the detector is lined with a 1 cm thick $^{10}\text{B}_4\text{C}$ ceramic sleeve enriched to 99.5 at.% in ^{10}B to absorb scattered neutrons from the sample, preventing the capture of neutrons in NaI. The detector system has about 75% efficiency to detect a 2 MeV γ -ray and up to approximately 96% efficiency for detecting γ -cascades [1]. Prior to the measurement, the detector was energy calibrated using a ^{22}Na source.

The measurement was conducted at the RPI Gaerttner Linear Accelerator (LINAC) Center using the TOF method. Neutrons were produced via a pulsed electron beam incident on a water-cooled tantalum target (referred to as the enhanced thermal target [2]). Neutrons traveled

down the collimated, 0.125 cm natural lead filtered and evacuated beam path to the sample located in the center of the NaI(Tl) γ -multiplicity detector 25.56-m from the target. The measurement was done for the low-energy region of about 0.01 - 100 eV. The LINAC operated at a repetition rate of 25 pulses per second and a pulse width of 500 ns. Three ^{235}U fission detectors (referred to as monitors) measure the neutron beam intensity during data collection to eliminate deviations and normalize the experimental data to account for neutron beam fluctuations throughout the experiment.

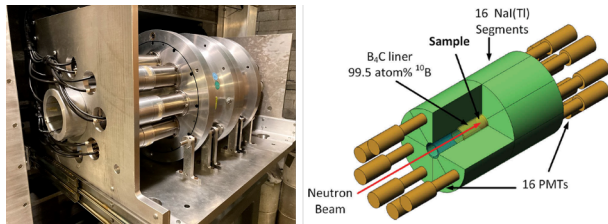


Figure 1. RPI γ -multiplicity detector at the 25-m flight station (left). Schematic diagram of γ -multiplicity detector to show individual detector segments, sample and boron liner (right).

The samples measured were 10 mil ^{nat}Ta and 20 mil ^{238}U . In addition to these samples, a blank aluminum sample-holder can was measured for background subtraction and a 100 mil $^{10}\text{B}_4\text{C}$ sample was used to calculate the neutron energy-dependent flux shape.

During the measurement, the γ -energy deposited in each detector segment was recorded for all events. The experimental data included information on neutron TOF, γ -energy, pulse height and γ -multiplicity. Experimental capture yields were calculated and compared to theoretical yields based on evaluated data. Neglecting multiple scattering in the sample, the capture yield at energy, E , is theoretically defined as

$$Y_\gamma(E) = \left[1 - e^{-N\sigma_t(E)}\right] \frac{\sigma_\gamma(E)}{\sigma_t(E)} \quad (1)$$

where N is the areal number density (atoms/barn) of the sample and σ_t and σ_γ are the total and capture microscopic cross sections, respectively. The experimental yield for TOF or energy bin i is defined as

$$Y_{\gamma,i}^{exp} = \frac{R_i - R_{b,i}}{C\phi_i} \quad (2)$$

where R and R_b are the sample and background count rate, respectively, ϕ is the smoothed, background-subtracted and corrected flux shape, and C is a normalization constant. The count rates are dead-time corrected, monitor-normalized, and grouped. Sample count-rates can be plotted as a function of incident neutron energy or TOF to analyze the data in real-time. In the processing code, for each event, if the sum of energies deposited in all 16 detectors is 1-20 MeV, the event is considered to be a capture event. Capture events are added to the TOF and/or γ -energy spectra for a defined range of incident neutron energies. The same technique is used to histogram γ -spectra in each detector segment (1-16) or for observed multiplicity events

(defined by the number of γ -rays emitted in a single capture event).

3 Results

3.1 ^{nat}Ta Capture Yield

A low-energy capture yield measurement of a 10 mil ^{nat}Ta sample is shown in Figure 2 compared to theoretical yields calculated using ENDF/B-VIII.0 [3], JEFF-3.3 [4] and NNL/ORNL [5] evaluations. The sample includes both ^{181}Ta and ^{180m}Ta ; however, only the JEFF-3.3 library has an evaluation for ^{180m}Ta , so it is used in each of the calculations. The theoretical yield calculations were completed using Equation 1 and evaluated Doppler-broadened cross section data from the respective data libraries (extracted from JANIS [6]). The RPI experimental capture yield was calculated using Equation 2.

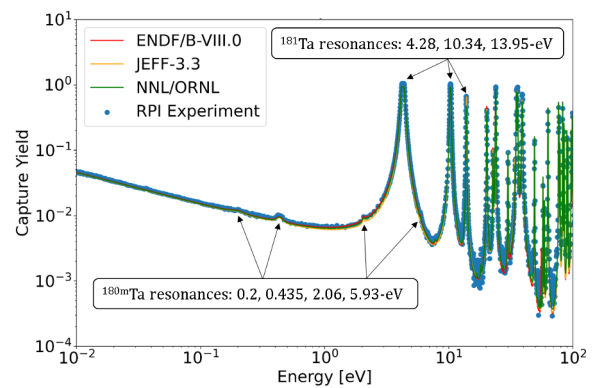


Figure 2. Low-energy capture yield results for a 10 mil ^{nat}Ta sample. Yield includes both ^{181}Ta and ^{180m}Ta isotopes. The measurement agrees with capture yields calculated from ENDF/B-VIII.0, JEFF-3.3 and NNL/ORNL evaluations.

The results confirm earlier measurements and agree with theoretical yield in the low energy resonance region from 1 to 20 eV, indicating the detector system is operating as expected with the new digitizer. Additionally, the data extends to 0.01 eV in the thermal region, where there is currently little experimental data.

3.2 ^{238}U γ -energy Spectrum

A low-energy measurement of a 20-mil ^{238}U sample was also completed to observe γ -spectra under specific resonances. Figure 3 shows two boxed resonances (36 and 66 eV) which correspond to incident neutron energies of interest. In these resonances, observed two-step cascades with total γ -energy deposition ± 0.5 MeV from the neutron binding energy (4.8 MeV for $^{238}\text{U}(n,\gamma)$) are histogrammed in Figure 4.

The results were compared to a recent measurement using the Detector for Advanced Neutron Capture Experiments (DANCE) array at LANSCE [7]. The data sets were normalized by the ratio of the area under the curves (total

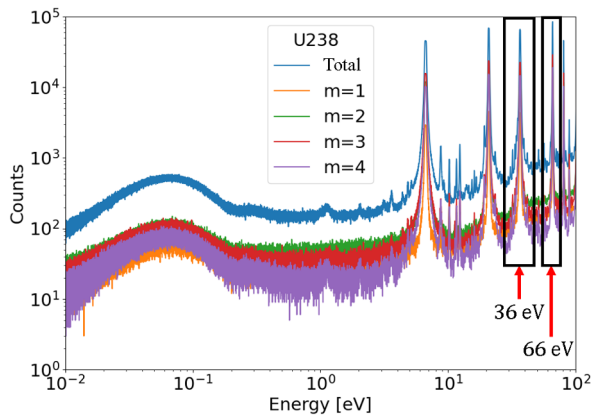


Figure 3. Low-energy capture yield results for a 20 mil ^{238}U sample used to generate γ -emission spectra. Several observed multiplicity spectra are shown and the total curve indicates the sum of all multiplicity events.

counts). Figure 4 shows general agreement between RPI and DANCE serving as a proof-of-concept; however, the differences need to be compared to simulation.

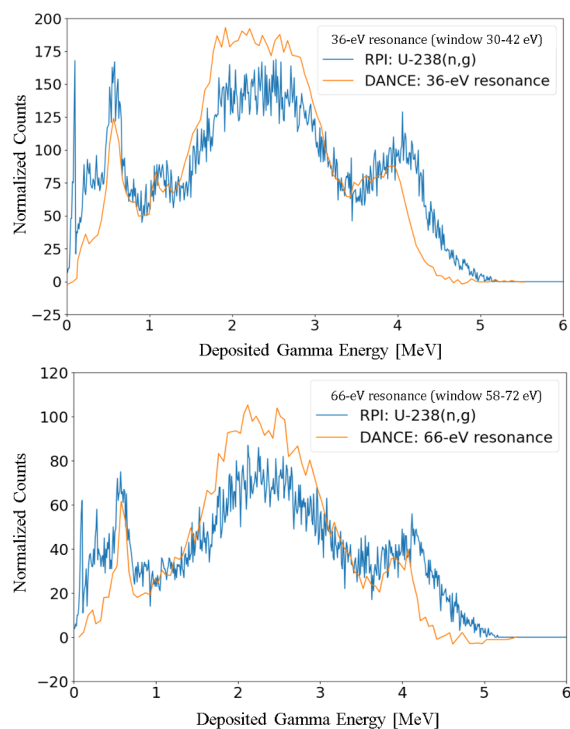


Figure 4. 36 and 66 eV resonance γ -spectra for observed two-step γ -cascades compared to a recent measurement using DANCE. The spectra are similar in shape; however the center of the DANCE spectra is higher than the RPI experiment where there are more counts in the high and low energy peaks.

4 Further Analysis

To accurately model event-by-event capture cascade spectra, a modification to the standard MCNP-6.2 [8] simula-

tion procedures are needed. First, capture γ -cascades are generated using an external code. In this work, DICEBOX [9] is used to write γ -cascades to a file. The cascade file is structured so the first column indicates the number of γ -rays in the cascade and the following columns correspond to the energies of each γ -ray. Next, a modification was made to MCNP-6.2 so that a γ -cascade is read in from the file when a neutron is captured. Each γ -ray in the cascade is then transported through the detector geometry and the energy deposition in each detector segment is tallied. Finally, the modified MCNP produces an event file which outputs the neutron history, cell (or detector segment) where the γ -ray was detected and the γ -energy deposited; enabling event-by-event analysis including coincidence.

A ^{22}Na γ -source was used to demonstrate that the modified MCNP returned the expected results. To produce an experimental γ -spectrum, a ^{22}Na source was measured in the sample position of the detector. The results were compared to two simulations: modified MCNP-6.2 using a cascade file generated to simulate the de-excitation after the decay of ^{22}Na and standard MCNP-6.2 using a photon source. Figure 5 shows the modified MCNP with the cascade file agrees with the measured spectrum because coincidence data is accounted for to observe the sum peak at about 1.78 MeV. Based on this, the modified MCNP simulation performed as intended and produced accurate results of the γ -spectrum.

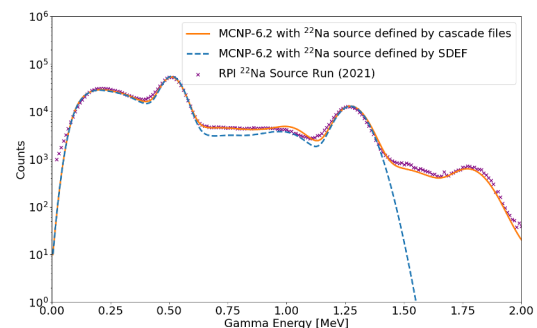


Figure 5. γ -energy spectrum for ^{22}Na source to test the modified MCNP. The standard MCNP with a photon source results in γ -lines from the de-excitation of ^{22}Na (511 keV and 1.27 MeV), but does not account for the sum peak at 1.78 MeV resulting from coincidence.

It is essential to evaluate the accuracy of capture γ -cascades generated from codes like DICEBOX. The DICEBOX calculated γ -cascade spectrum of the $^{181}\text{Ta}(n,\gamma)$ reaction was compared to measured γ -lines from the Evaluated γ -ray Activation File (EGAF) [10] in Figure 6. The DICEBOX calculation used primary γ -intensities extracted from the ^{181}Ta thermal capture data set in the Evaluated Nuclear Structure Data File (ENSDF) [11]. This is an area for further research, as there are missing γ -lines in the center of the ^{181}Ta spectrum in EGAF.

Figure 7 shows the measured ^{181}Ta capture γ -spectrum at low incident neutron energies (0.01 - 0.04 eV) compared

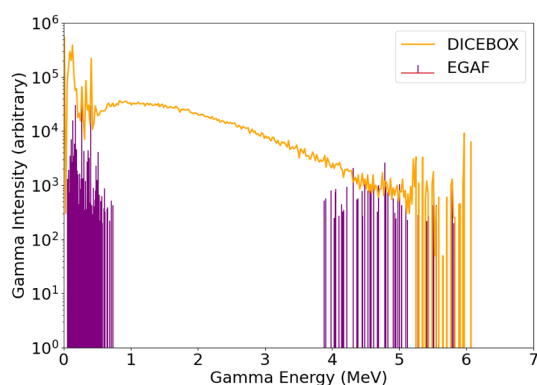


Figure 6. ^{181}Ta γ -cascade spectra reported in EGAF compared to simulated DICEBOX γ -cascade spectra. There are missing values in the center of the EGAF spectra. DICEBOX predicts the γ -energies and intensities where there are no experimental γ -lines in EGAF.

to four different MCNP simulations. If the total γ -energy deposited by a cascade was 1 - 20 MeV, it was considered a capture event and the γ -rays were tallied.

Each simulation had a 0.01 - 0.04 eV neutron beam incident on a 10 mil ^{181}Ta sample. MCNP-6.2/DICEBOX used the γ -cascades generated from DICEBOX and the modified MCNP. Two simulations used an MCNP feature called Cascading Gamma-Ray Multiplicity (CGM) which produced correlated secondary γ -emissions [12]. MCNP-6.2/CGM used the standard version of MCNP/CGM that has a fixed sample binding energy defined as 8.5 MeV. MCNP-6.2/CGM (w/ updated BE) used the modified MCNP/CGM with an updated binding energy for ^{181}Ta . MCNP-6.2/ACE used γ -cascade data from ACE files with the modified MCNP.

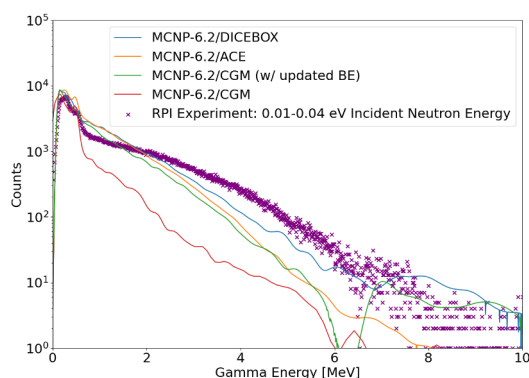


Figure 7. ^{181}Ta experimental capture γ -spectra compared to MCNP-6.2 simulations. These spectra are for one detector in the 16-segment array and only includes events where the total energy deposition in all 16 detectors is 1 - 20 MeV. The high-energy γ -rays above the binding energy of ^{181}Ta (6.0629 MeV) shown in both experiment and simulations are a result of neutrons interacting in air.

MCNP-6.2/DICEBOX appears to match the experimental data best; however, each of the simulated spectra are inconsistent with the measured γ -spectrum. Further research is currently underway to validate the experimental γ -spectra measurement using selected monotoxes which can inform the observed discrepancies in the ^{181}Ta γ -spectra.

5 Conclusion

A new data acquisition system was installed and validated by measuring resonance capture yield of ^{nat}Ta as a function of neutron energy. A low-energy capture measurement of ^{238}U was also completed to generate γ -spectra to further understand γ -heating in nuclear reactors. MCNP-6.2 was modified to compare simulated γ -spectra to experimental results, using $^{181}\text{Ta}(n, \gamma)$ as an initial test case. Future work will include confirming the method for comparing modeled and measured γ -spectra, identifying the differences between simulated and measured $^{181}\text{Ta}(n, \gamma)$ spectra, and further analyzing the $^{238}\text{U}(n, \gamma)$ data.

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