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Thermal Neutron Induced Capture and Fission Gamma-Ray Emission Measurements and Simulation Methods

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Abstract. In order to validate evaluated capture gamma-ray cascade data used in coupled neutron-photon transport simulations, measurements of thermal neutron induced capture were performed as a function of neutron time-of-flight (TOF). A large 16-segment NaI(Tl) gamma-ray multiplicity detector was used with a 14-bit digitizer to record event-by-event gamma-ray energy deposition. Histograms of the gamma-ray emission spectra in each of the detectors and coincidence spectra of all 16 detectors were generated for analysis. The spectra were produced by gating on specific TOF regions that correspond to either thermal neutrons (0.01 - 1 eV) or energies of specific resonances in the sample material. The experimental results were compared to simulations using a modified version of MCNP6.2 that can read capture gamma-ray cascades from a file, transport them through the detector geometry and output event-by-event energy deposition in each detector similarly to the processed experimental data. The Monte Carlo code, DICEBOX, was used to generate the capture gamma-ray cascades using ENSDF evaluated gamma-ray energies and intensities and RIPL-3 model parameters. The experiment was also compared to spectra produced by MCNP6.2 using ENDF/B-VIII.0 evaluated photon production data. It was observed that in some cases the ENDF/B-VIII.0 data can produce a reasonable single detector spectrum but coincidence modeling is not currently possible in standard MCNP6.2 calculations.

1 Introduction

Measurements are routinely performed at the Rensselaer Polytechnic Institute (RPI) Gaerttner Linear Accelerator (LINAC) Center to generate (and validate) nuclear data for different neutron reactions in multiple energy ranges including sub-thermal, thermal, resolved resonance, unresolved resonance, and fast energy regions. These measurements use neutrons generated by a 60 MeV pulsed electron LINAC coupled with various neutron and gamma-ray detector systems located at different flight path lengths using the time-of-flight (TOF) method.

The neutron capture process is an important compound nucleus absorption reaction occurring in many applications. When a neutron is captured in nucleus ^{A}Z, it creates a compound nucleus ^{A+1}Z that is excited to a state with energy equal to the sum of the neutron binding energy and kinetic energy of the incident neutron. The compound nucleus de-excites by emitting gamma-ray cascades with average multiplicities typically between 2 and 4. Capture gamma rays can be used to directly measure the capture cross section [1]. For some applications, including active neutron interrogation, the average energy of the emitted gamma rays and the energy deposition spectra provide information used to identify the specific materials interacting with neutrons. The experimental and simulation methods used at RPI enable the analysis of both types of gamma-ray measurements: single detector response and coincidence measurements, these methods are useful for a wide range of applications that measure gamma spectra and use this information.

Evaluated nuclear structure data stored in compiled libraries, such as the Evaluated Nuclear Structure Data File (ENSDF) [2], provide information about de-excitation schemes, including the energies and intensities of gamma rays emitted during de-excitation, that can be used with simulation tools. Methods were developed to test thermal neutron induced capture gamma-ray data generated from nuclear structure evaluations using RPI measurements [3]. To achieve this goal, a modified simulation technique was used based on MCNP6.2 [4] to more accurately model and transport neutron capture gamma-ray cascades. The results of the new simulation tool were compared with experimental neutron capture gamma-ray spectra measured at the RPI LINAC. Analysis methods are also under development for actinides, such as ^{235}U, where both fission and capture gamma-rays are emitted as a result of neutron absorption.

2 LINAC Experiments

Neutron capture gamma-ray measurements were performed using the RPI 16-segment NaI multiplicity detector located at a flight path distance of about 25.5 m (useful in the energy range from 0.001-3000 eV). The detected gamma-ray pulses are digitized, streamed to a computer, and saved for multidimensional post processing. The neu-

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tron capture gamma-ray spectra for different incident neutron energy ranges and observed multiplicities were derived for comparison with simulations. The neutron capture gamma-ray spectra for a single resonance in a particular material can be analyzed to determine spin dependence. In the case of U-235, resonance gamma-ray spectra was previously used to separate fission from capture gamma-ray contributions in order to simultaneously measure the capture and fission cross sections [5].

The data acquisition system includes a Struk 16 Channel 250 MHz 14-bit Digitizer (SIS3316-250-14) connected directly to each photo-multiplier tube. During the experiment, a sample was placed in the center of the detector and energy deposition from neutron capture gamma rays from the sample were digitized and saved to a file. The information included pulse waveform and neutron TOF which enables processing the data at different incident neutron energy windows.

A processing code was written using the Julia language [6]. Coincidence events were determined based on a time window of 100 ns. For each coincidence event, the code records the time stamp and energies deposited in each of the detector segments. The result is a large table with all the events for each sample that is used for further analysis. The low level discriminator was set to 100 keV gamma-ray energy deposition in each detector to reduce background.

Before and during the experiments, the detectors need to be carefully aligned to have the same gain. Initial alignment was done by varying the voltage of each detector to align the spectrum of a Na-22 calibration source. A method to correct for detectors gain drifts as a function of time was developed to using the energy deposition spectrum in the beginning of the measurement and aligning subsequent measurements to it.

3 Simulation Methods

A modified version of MCNP6.2 and the Monte Carlo code DICEBOX [7], with nuclear structure information from ENSDF, were used to calculate the neutron capture gamma-ray cascade energy deposition and validate against the RPI LINAC experiments. Gamma-ray cascade data generated by DICEBOX was read by the modified MCNP6.2 code and transported through the RPI detector geometry. The output was a list of gamma-ray energy depositions in each of the 16 segments in the detector system for each neutron history that had a gamma-ray producing interaction, similar in format to the experimental results to allow for easy processing and comparison.

4 Results

Several measurements were done first to develop and validate the new experimental and simulation methods followed by additional measurements of other samples of interest.

4.1 Validation Measurements with Fe-56 Capture Gamma-Ray Spectra

A measurement of a 99.9% Fe-56 sample compared to simulation results is shown in Figure 1. The neutron energy was TOF gated between 0.01 to 0.1 eV and total gamma-ray energy deposition was gated between 2 and 12 MeV to produce the measured spectra. An alignment procedure was applied to the measured data to ensure all 16 detectors had the same gain throughout the experiment. DICEBOX calculated cascades using discrete levels and primary intensities from [8] were used in the simulation and gamma-ray emission was assumed to be isotropic. The experimental and simulated spectra are in very good agreement with respect to the individual detector spectrum and the coincidence spectrum; therefore, the simulation methodology was validated. As a reference, a simulation with ENDF/B-VIII.0 [9] (labeled as ACE) is also shown; it has the overall trend of the experimental spectra but does not agree with all the structure in the measurement. A comparison of the simulation with a coincidence spectrum can not reliably be done with the regular version of MCNP6.2 and ENDF/B-VIII.0 data alone, a cascade input file is needed.

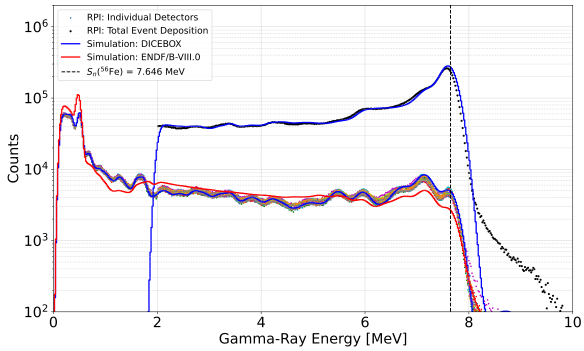

Figure 1. Comparison of measured and simulated capture gamma-ray spectra of an Fe-56 sample (0.052126 a/b). The MCNP6.2 and ENDF/B-VIII.0 simulation is not in as good agreement compared to the new simulation method using DICEBOX cascades. The neutron separation energy S_n for the compound nucleus is shown as a vertical line. (multi color points represent 16 individual detectors)

4.2 Mn-55 Capture Gamma-Ray Spectra

Similar measurements were done for a MnCu sample (80% Mn and 20% Cu) and the results are shown in Figure 2 compared to the ENDF/B-VIII.1β4 [10] evaluation. The new ENDF evaluation includes improvements to the Mn-55 capture gamma-ray spectra, but when cascades calculated using discrete levels and primary intensities from [11], a better agreement is obtained with the total energy spectra (coincidence).

4.3 U-nat Capture Gamma-Ray Spectra

A more challenging measurement was done for U-nat (and U-235). In the thermal neutron energy region of U-

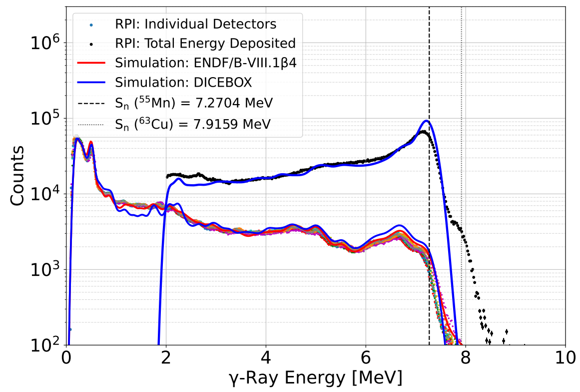

Figure 2. Comparison of measured and simulated capture gamma-ray spectra of a MnCu (0.0072185 a/b) sample. (multi color points represent 16 individual detectors)

nat, there is an appreciable contribution to the gamma-ray spectra from fission in U-235. A neutron TOF spectrum shown in Figure 3 allows us to use the strong 6.67 eV capture resonance in U-238 to isolate the U-238 capture gamma rays. This is because the low reaction rate makes the contamination from U-235 prompt fission gamma rays negligible. The agreement between the measured capture yield and ENDF/B-VIII.0 cross section simulated yield validates the use of a TOF window to restrict the capture reactions to a specific incident neutron energy range.

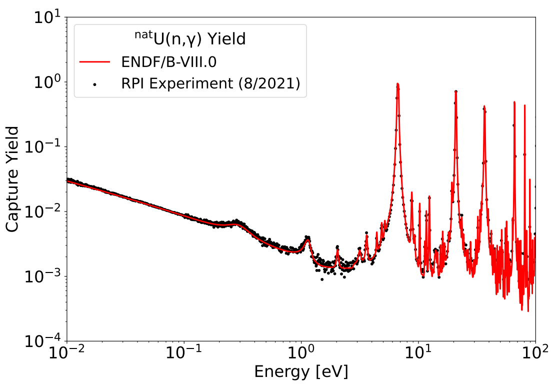

Figure 3. Comparison of measured and calculated capture yield for a U-nat sample (0.002436 a/b).

The measured capture gamma-ray cascade spectrum of U-238 in the 6.67 eV resonance was compared to the modified MCNP6.2 calculation using cascades from evaluated discrete levels and primary intensities from [12]. Figure 4 shows reasonable agreement; however in 2-3.5 MeV gamma-ray energy range, the simulation is lower than the experimental data which could be attributed to the lack of experimental capture gamma-ray data sets (shown in Figure 5). The gap in the Evaluated Gamma-Ray Activation File (EGAF) [13] data translates to a gap in the simulated response, which demonstrates how this method allows validation and assessment of the quality of capture cascade gamma-ray data. For this observation, it is assumed that the thermal capture gamma-ray emission spectrum and in the 6.67 eV resonance gamma-ray emission spectrum are similar. This is a good assumption for U-238 because it has target spin I=0 and spin J=0.5 for all strong s-wave resonances.

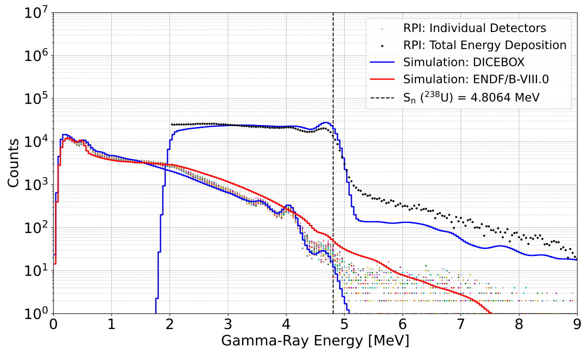

Figure 4. Comparison of measured and simulated capture gamma-ray spectra of a U-nat sample gated on the 6.67 eV U-238 capture resonance. (multi color points represent 16 individual detectors)

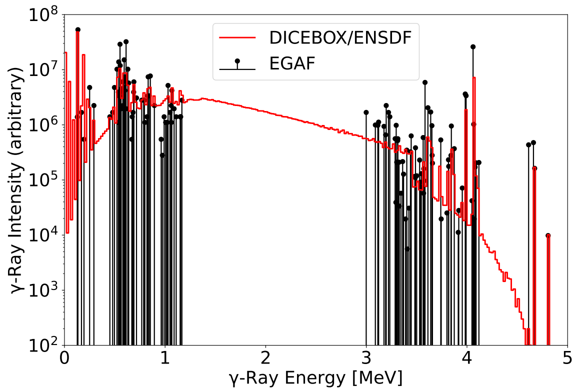

Figure 5. ENSDF data available for U-238 thermal neutron capture. Discrete levels and primary gamma-ray intensities were used in the DICEBOX cascade calculation (red) compared to EGAF lines which represent the available measured primary gamma-ray energies and intensities stored in the library.

4.4 U-235 Capture Gamma-Ray Spectra

For neutron absorption reactions in U-235, there is an additional complication to the experimental data analysis because both capture and fission reactions produce gamma rays that can be detected and added to the spectra. One approach to separating the gamma rays from fission and capture is to select a resonance for which the capture cross section dominates and obtain the capture gamma-ray cascade spectrum for all events in this resonance. However, there will be a very small (but not negligible) background gamma-ray contribution from fission reactions. Reduction of this background is possible by selecting an additional resonance that is dominated by the fission cross section then setting a system of two equations and two unknowns to get the background corrected capture gamma-ray emission spectrum. This can work if the fission gamma spectrum is not varying between resonances which we verified by comparing 20 different resonances processed with a coincidence sum energy above the binding energy to isolate fission events. The results of the experimental analysis compared to simulated spectra are shown in Figure 6 for the 4.85 eV capture resonance (4.6-5.1 eV window) used in the analysis. The fission resonance used to subtract the prompt fission gamma-rays was located at 14.1 eV (13.4-14.3 eV window). The total gamma-ray energy deposition was required to be between 2 and 20 MeV. The simulation used DICEBOX cascades for U-235 thermal capture, the same incident neutron energy range of 4.6-5.1 eV, and the NONU card was enabled to eliminate fission reactions.

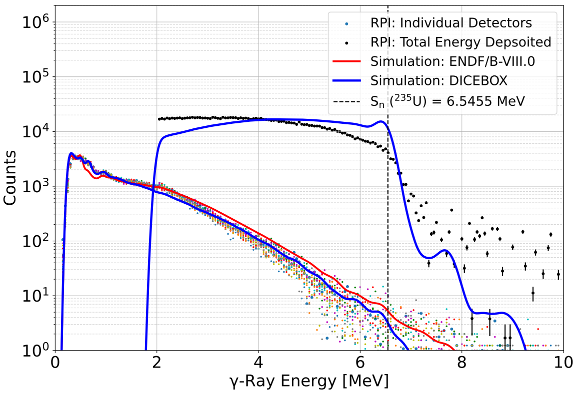

Figure 6. Comparison of measured and simulated capture gamma-ray spectra of a U-235 sample gated on the 4.85 eV U-235 capture resonance with the fission gamma rays subtracted. (multi color points represent 16 individual detectors)

The agreement between the experimental and simulated single detector spectra is surprisingly good considering the poor capture gamma-ray cascade data available (shown in Figure 7). There is a discrepancy in the coincidence spectrum and the binding energy peak is less visible in the experiment than it is in the simulation. It is possible that some components of the fission gamma-induced background spectra require additional correction or the detectors were not sufficiently aligned during the experiment.

5 Conclusions

The RPI multiplicity detector that was previously used for capture cross section measurements has been upgraded to provide data for testing neutron induced capture gamma-ray cascade spectra. This was accomplished by digitizing all 16 detectors to enable saving digitized energy deposition wave-forms to computer storage on an event-by-event basis. The new capability enables coincidence analysis as a function of neutron TOF. To compare the measurements to simulations, a modified version of MCNP6.2 was created to transport DICEBOX-generated capture gamma-ray cascades. In this version of MCNP6.2, for each neutron capture event, gamma-ray cascade energies are read from a file, transported through the detector geometry, and event-by-event energy deposition in the detector segments are saved to a file. The updated experimental and simulation methods allow for comparison of measured and simulated data including the capability to simulate experimental conditions such as discrimination level and energy resolution. Validation of the methods were done with an ^{56}Fe(n,γ) measurement which has well-known nuclear structure data for the compound nucleus de-excitation to its ground state. Results were also shown for neutron capture measurements of Mn-55 and U-235,238 (using resonances to separate capture and fission induced gamma rays emitted). This work represents the first steps in the creation of a validation method for capture gamma-ray cascades that is relevant to many applications.

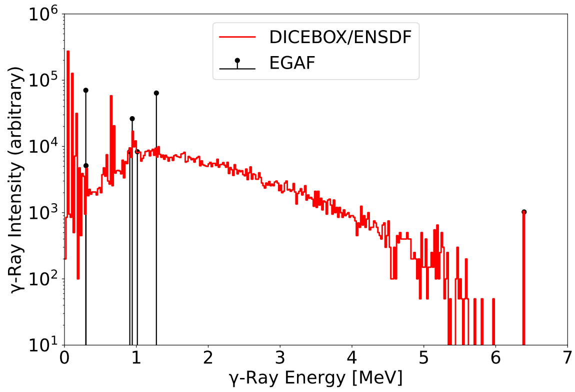

Figure 7. ENSDF data available for ^{235}U and used as part of the input for DICEBOX, it is plotted with EGAF which shows the available measured primary gamma-ray energies and intensities.

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