Measuring and Simulating Neutron Capture γ-Ray Spectra using the RPI γ-Ray Multiplicity Detector

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Abstract. Accurate modeling of neutron induced capture γ-ray production is essential for many applications such as reactor and shielding calculations, understanding γ-ray heating in critical systems, and non-proliferation efforts. To determine the accuracy of nuclear data evaluations and simulation tools used to transport capture γ-cascades, the 16-segment γ-ray multiplicity NaI(Tl) detector at the Rensselaer Polytechnic Institute Gaerttner Linear Accelerator Center has been upgraded to measure neutron capture γ-ray spectra and multiplicity as a function of energy. Several samples including Fe, Mn, Co, Ta, and ^{235,238}U have been measured using the time-of-flight method for incident neutrons in the low-energy region from 0.01 – 100 eV. A new method has been developed to model the event-by-event capture γ-ray cascade energy deposition in the detector array using `DICEBOX` and a modified version of MCNP-6.2. The method has been validated using ^{22}Na and ^{60}Co coincidence sources and the well-studied thermal ^{56}Fe(n,γ) γ-ray intensities. Additional measured samples will be used for further validation and analysis. The new modeling capabilities coupled with measured γ-ray spectra can be used to test transport codes and nuclear data evaluations of neutron capture γ-rays used to simulate experimental results.

1 Introduction

Validating neutron capture γ-ray data is a top priority for many nuclear-related applications including radiation shielding, reactor simulations related to γ-ray heating and active neutron interrogation for non-proliferation [1]. To this end, the Rensselear Polytechnic Institute (RPI) γ-Ray Multiplicity Detector is used to measure neutron capture γ-ray cascades at the Gaerttner Linear Accelerator (LINAC) Center via the time-of-flight (TOF) method.

The goal of this work is to measure neutron capture γ-ray emission spectra and γ-ray cascade multiplicity as a function of neutron energy for important nuclear materials in the resolved resonance region (including ^{235,238}U and various structural materials) in order to validate the γ-ray data stored in nuclear data libraries. Measured γ-ray spectra are compared to simulated results using current simulation tools (MCNP-6.2 [2] and Cascading Gamma-Ray Multiplicity (CGM) [3]) and improved models (a modified version of MCNP-6.2 coupled to the Monte-Carlo γ-ray cascade code `DICEBOX` [4]) using the current nuclear data and forward modeling methods.

Previous work using the RPI γ-Ray Multiplicity Detector system includes measuring a natural Ta sample (including both ^{181}Ta and ^{180m}Ta isotopes) and a natural U sample (including both ^{235}U and ^{238}U isotopes) to validate the neutron capture yield measurement and begin performing γ-ray spectra analysis [5].

Recent measurements of low-energy ^{56}Fe(n,γ) spectra have been completed and the results were used to validate the system. The experimental methods, simulation techniques, and results are presented in the current paper.

2 Experimental Methods

The Gaerttner LINAC Center at RPI is used to measure and validate nuclear data [6]. Electrons are accelerated toward a water-cooled tantalum target to produce neutrons and the TOF method is used to determine the energy of the incident neutron interacting with samples in different detector systems at various flight paths.

For this work, the enhanced thermal target [7] was used to measure neutron capture γ-ray spectra in the low-energy region of about 0.01 - 100 eV. The LINAC operated at 25 pulses per second with a 500 ns pulse width. During the measurement, three ^{235}U fission chambers (referred to as monitors) were used to determine the neutron beam intensity during data collection. Monitor data were used to eliminate deviations and normalize the experimental data to account for fluctuations in the neutron beam intensity throughout the experiment.

The RPI γ-Ray Multiplicity Detector (shown in Figure 1) is used to measure neutron capture yield and γ-ray spectra as a function of neutron energy. The detector is a total-absorption type calorimeter composed of 16 segments of NaI(Tl) crystals in an array around the sample of interest located approximately 25 m from the neutron

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production target. The inside of the cylindrical detector is lined by a 1 cm thick ceramic $^{10}B_4C$ sleeve to prevent scattered neutrons from entering and being captured in the detector material. The system is approximately 75% efficient at detecting 2 MeV γ-rays and up to approximately 96% efficient when detecting γ-ray cascades [8]. The detector was energy calibrated using a ^{22}Na γ-ray source.

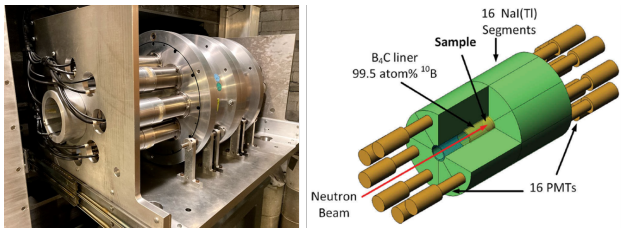

Figure 1. The RPI γ-Ray Multiplicity detector used to measure neutron capture yield and γ-ray emission spectra (left). Schematic diagram of the detector to show individual NaI(Tl) segments, sample, boron liner and PMTs (right).

A 16 Channel 250 MHz 14-bit Digitizer (SIS3316-250-14) replaced analog electronics to greatly improve the capabilities of the system by digitizing pulses generated during each coincidence event on all 16 detector segments. The energy deposited in each detected event can be determined using the digitized pulse data allowing for the generation and analysis of neutron capture γ-ray spectra as a function of energy.

The current work features measurement results from a 99.87% enriched ^{56}Fe sample to validate the experimental system. ^{56}Fe was chosen because the thermal neutron capture γ-ray data are available for the entire γ-ray energy spectrum up to the binding energy. The sample was 6 mm thick with a number density of 0.05159 ± 0.0003 atm/b [9]. A blank aluminum sample-holder can was measured for background subtraction and a 2.54 cm $^{10}B_4C$ sample was used to calculate the neutron energy-dependent flux shape. Additional samples include ^{55}Mn and ^{59}Co to further evaluate thermal neutron capture γ-ray data.

3 Updated Simulation Techniques (MCNP-6.2/DICEBOX)

MCNP-6.2/ACE and MCNP-6.2/CGM were the original simulation tools used by RPI for simulating neutron capture γ-ray production. MCNP-6.2/ACE extracts γ-ray data from the standard MCNP library that includes ENDF/B-VIII.0 [10] in ACE format. CGM is a code embedded into MCNP-6.2 that uses statistical models to produce correlated secondary γ-ray emissions. However, CGM does not always include the measured primary γ-ray intensities from thermal neutron capture which causes discrepancies between simulations and the experimental γ-ray spectra.

To more accurately simulate the experiment and perform event-by-event analysis, the original MCNP-6.2 modeling techniques were modified, as discussed in Ref. [5]. To summarize, the Monte-Carlo γ-ray cascade code DICEBOX is run prior to the MCNP-6.2 calculations to generate and tabulate γ-ray cascades. A modified version of MCNP-6.2 reads in the DICEBOX γ-ray cascade if a neutron reaction produces a γ-ray. Then the DICEBOX γ-ray cascade is transported isotropically through the detector geometry. The γ-ray energy deposited in each of the 16 detector segments are tallied throughout the simulation and the results can be analyzed using the same processing codes as the experimental data.

The γ-ray cascades generated with the DICEBOX code need to accurately represent the thermal neutron capture γ-ray data available in nuclear data libraries. To achieve this, DICEBOX uses many parameters to determine the de-excitation pathway of the compound nucleus. Discrete levels and model parameters, including total level density and photon strength functions, are extracted from the Reference Input Parameter Library (RIPL)-3 [11] database. Thermal neutron capture primary γ-ray intensities are manually added to the DICEBOX input file from the Evaluated Nuclear Structure Data File (ENSDF) [12] or other data sources to generate a more accurate calculation.

Figure 2 shows the thermal neutron capture γ-ray data for ^{56}Fe(n,γ), taken from the Evaluated γ-ray Activation File (EGAF) [13], compared to full γ-ray cascades modeled using DICEBOX and primary γ-ray intensities from the ^{57}Fe capture state decay scheme measured by Firestone, et al. [14]. The DICEBOX code predicts γ-ray intensities based on chosen models for each energy where there are no experimental γ-ray lines available in the nuclear data libraries. The ^{56}Fe(n,γ) measurement was chosen to validate this system because it has been previously measured across the whole γ-ray energy spectrum and does not require DICEBOX to calculate intensities based on statistical models. However, this is not the case for most isotopes, where statistical models are needed to fill the gaps in γ-ray data.

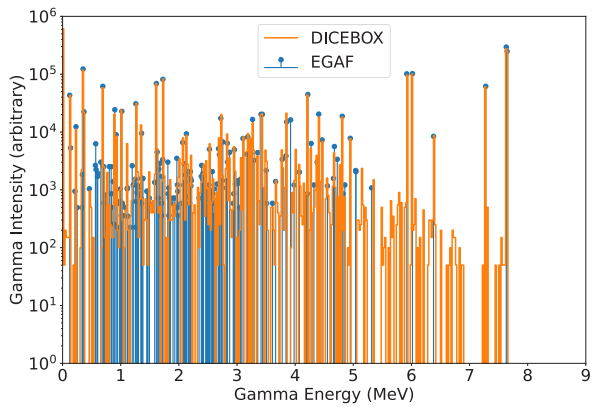

Figure 2. ^{56}Fe(n,γ) γ-ray lines (blue) from EGAF compared to a simulated DICEBOX γ-ray cascade spectrum (orange).

4 Results

Figure 3 compares the ^{56}Fe(n,γ) experimental γ-ray emission spectra to simulated results using the modified

MCNP-6.2/DICEBOX code. The experimental data is processed using a series of Julia [15] scripts. First, the processing code determines coincidence events based on a time window of 100 ns. The events recorded in each of the 16 individual detector segments during the coincidence window are considered to make up a neutron capture induced γ-ray cascade. The energy deposited in each of the 16 detector segments correspond to the energies of each γ-ray in the cascade. If the sum of the individual γ-ray energies in the cascade was above a 2 MeV threshold, the event was considered to be neutron capture. The γ-ray energies deposited in each detector segment and the total energy (sum of all segments) were added to the spectrum for a defined range of incident neutron energies.

The modified MCNP-6.2/DICEBOX simulation models the γ-ray production resulting from a neutron beam incident on the ^{56}Fe sample with neutron energies sampled uniformly from 0.01 - 100 eV. Detector geometry, sample material composition, and NaI γ-ray energy resolution are all used in the simulation as well. Since there are no statistical models added to the DICEBOX calculation of γ-ray cascades, the experiment is compared directly to the ^{56}Fe(n,γ) nuclear data from Firestone [14].

Compared to the original simulation tools, MCNP-6.2/ACE and MCNP-6.2/CGM, the modified MCNP-6.2/DICEBOX model represents the experimental data most accurately as shown in Figure 4 for individual detector segments and the sum of all 16 detector segments. The disagreement in the total energy (sum) curve is expected for the MCNP-6.2/ACE calculation because the simulation does not include coincidence events.

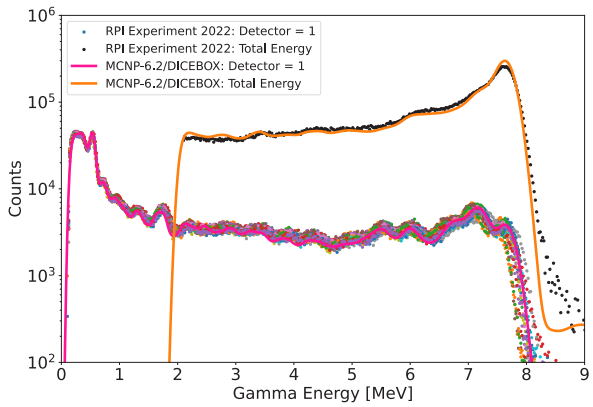

Figure 3. ^{56}Fe experimental neutron capture γ-spectra compared to MCNP-6.2/DICEBOX simulations. Individual detector γ-ray spectra are shown compared to simulated detector 1 results and the sum of all 16 detectors is compared to simulated total energy deposition. The high-energy γ-rays above the binding energy of ^{56}Fe (7.646 MeV) shown in both experimental and simulated results are a result of neutrons interacting in air.

5 Conclusions & Future Work

Experimental and simulation methods were developed and validated for determining the accuracy of thermal neutron capture γ-ray data using the RPI γ-Ray Multiplicity Detector. It has been demonstrated that results from RPI LINAC experiments can be compared to Monte Carlo simulations in order to test nuclear data and physics models. When neutron capture γ-ray cascade data are well-known, the γ-ray emission spectra can be accurately calculated using the modified MCNP-6.2/DICEBOX simulation method.

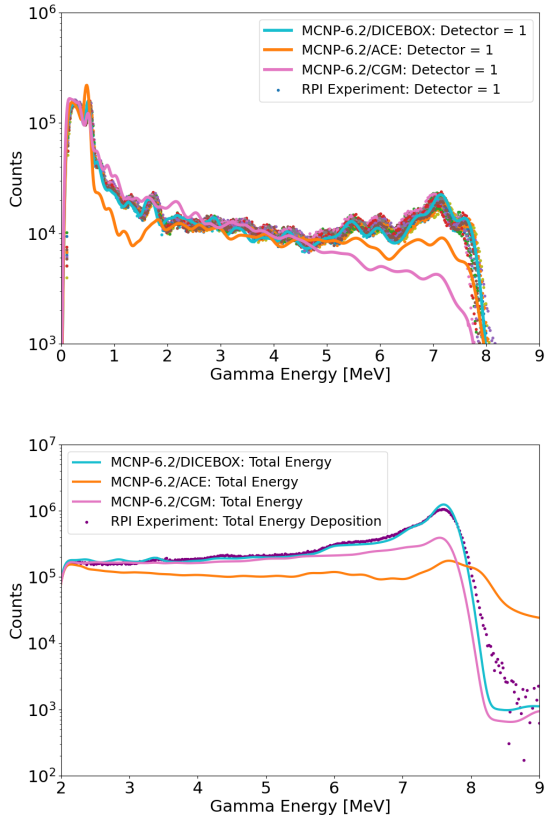

Figure 4. ^{56}Fe experimental neutron capture γ-spectra compared to MCNP-6.2 simulations. Individual detectors are shown compared to simulated detector 1 results (top) and the sum of all 16 detectors are compared to simulated total energy deposition (bottom).

The detector system was validated using experimental and simulated results of ^{56}Fe capture γ-ray production. The ^{56}Fe(n,γ) reaction was chosen because ^{56}Fe is an isotope with well-known neutron capture γ-ray data. The additional data collected using ^{55}Mn and ^{59}Co samples during the LINAC experiment will be used to further validate the system. Any new evaluations that include γ-ray data can be tested using the detector system and forward-modeling modified MCNP-6.2/DICEBOX method.

The RPI γ-Ray Multiplicity Detector system can now be used as a tool to test neutron capture γ-ray data and provide recommendations for isotopes with deficiencies in γ-ray data like ^{181}Ta and ^{235}U (see Figure 5). There are large gaps in the measured γ-ray lines from EGAF which results in the DICEBOX code relying on statistical models to predict the γ-ray intensities over large energy ranges. Additionally, there are efforts to explore the many differ-

ent models (level density and photon strength functions) that can be used in the DICEBOX code [16, 17] to further understand neutron capture γ-ray cascades.

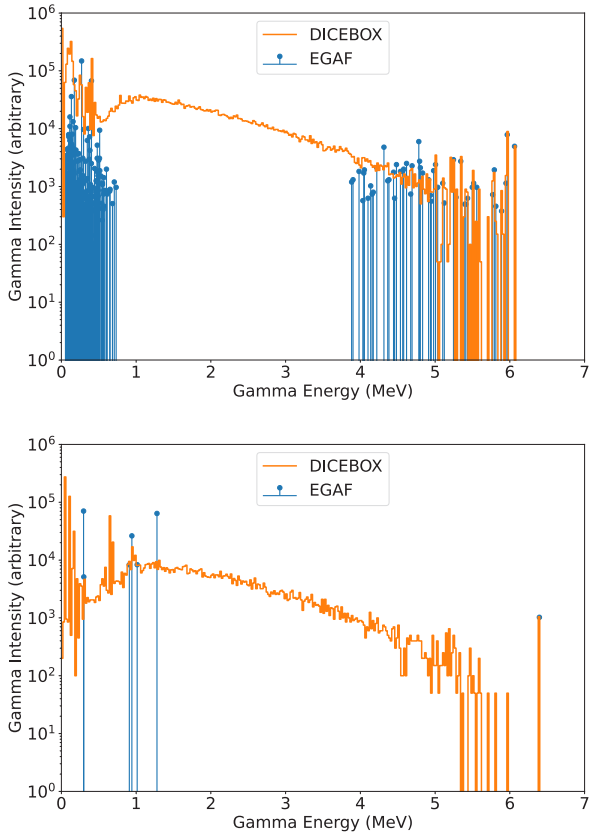

Figure 5. ^{181}Ta(n,γ) (top) and ^{235}U(n,γ) (bottom) γ-ray lines from EGAF compared to simulated DICEBOX γ-ray cascade spectra. It is clear there are gaps in the measured γ-ray data for these isotopes.

Acknowledgments

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