A NEW METHOD FOR THE SIMULTANEOUS MEASUREMENT OF THE NEUTRON CAPTURE AND FISSION CROSS SECTIONS OF ^{235}U

D. G. Williams, R. Bahran, E. Blain,
B. McDermott, M. Rapp, Y. Danon*
Rensselaer Polytechnic Institute
Department of Mechanical, Aerospace
and Nuclear Engineering
110 8th St
Troy, NY 12180

D. Barry, G. Leinweber, R. Block, J. Hoole
Bechtel Marine Propulsion Corporation
Knolls Atomic Power Laboratory
P.O. Box 1072
Schenectady, New York 12301-1072

ABSTRACT

A new method for the simultaneous measurement of the neutron capture and fission cross sections of ^{235}U has been developed at the Gaerttner LINAC Laboratory located at Rensselaer Polytechnic Institute. This method uses a 16 segment γ-multiplicity NaI(Tl) detector. The prompt γ rays from a neutron interaction with the highly-enriched ^{235}U sample are detected and categorized based on the total energy deposited and the multiplicity (number of γ rays detected). Spectra of the events are recorded using the time-of-flight method and grouped based on γ energy and observed multiplicity. Above the resolved resonance region for ^{235}U (2.25 keV), the cross sections are presented as averaged values. The results of this research can be used to improve the accuracy of the capture-to-fission ratio for ^{235}U in the resolved resonance region and to provide additional capture cross section data to the limited amount that currently exists for ^{235}U in the unresolved resonance region.

I. INTRODUCTION

With the continued improvement of computing capabilities, the accuracy of nuclear data has become a primary factor impacting the accuracy of numerical Monte Carlo methods for nuclear systems. This limitation was recently reiterated by Forest Brown, one of the developers of MCNP at Los Alamos National Laboratory, during a colloquium presentation at Rensselaer Polytechnic Institute.[1] A specific example of the need for more accurate experimental data is ^{235}U in the unresolved resonance region, which begins at a neutron energy of 2.25 keV. According to Otuka *et al.*,[2] the most important physical constants in nuclear energy applications are the neutron cross sections of ^{235}U. Additionally the authors specify that the characteristics of fast reactor cores, analyzed through critical experiments using the Bystrye Fisicheskie Stendy (BFS) test core assemblies, are sensitive to the neutron capture cross section of ^{235}U in the keV region. In the unresolved resonance region for the neutron capture cross section for ^{235}U discrepancies of up to 10% exist between some of the evaluated libraries. While the JEFF-3.1.1 (Ref 3) and ENDF/B-VII.0 (Ref 4) libraries are nearly identical for ^{235}U in this region, they are generally lower than the JENDL-4.0 (Ref 5) library for energies between 2.5 and 5.5 keV and generally higher for energies between 5.5 and 10 keV. The data currently available varies in the keV region and is not accurate enough to resolve the discrepancies between the libraries. The neutron cross section is related to the yield of an experimental sample by Equation (1) where Y_x is the yield (number of events per incident neutron) for reaction x, typically capture, fission, or scattering.

$$Y_x(E) = (1 - e^{-n\sigma_t(E)})\frac{\sigma_x(E)}{\sigma_t(E)} \quad (1)$$

The remaining variables in Equation (1) are the number density in atoms per barn, n, the total microscopic cross section, σ_t, and the microscopic cross section of the particular reaction, σ_x.

As seen in Figure 1, the discrepancy between the different experiments would translate to a relative uncertainty of up to 30% on the ENDF/B-VII.0 values. One of the objectives for the development of this technique and subsequent measurement is to address the need for a more accurate measurement of ^{235}U cross sections in the unresolved resonance region. Due to the accuracy of previous measurements of the total and fission cross sections, the observed variations in the capture yield Y_γ for ^{235}U can be attributed to the capture and scattering cross sections.

II. EXPERIMENTAL CONDITIONS

This experiment was conducted using the time-of-flight (TOF) method with the electron linear accelerator (LINAC) at the Gaerttner LINAC Laboratory located at Rensselaer Polytechnic Institute as the source of neutrons. The

*Email: danony@rpi.edu

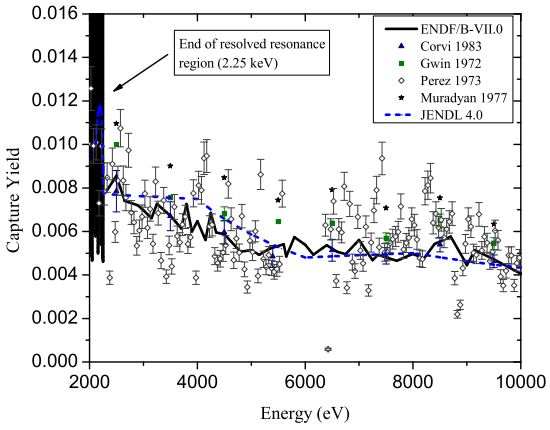

Figure 1: This graph demonstrates the spread of experimental values for the capture yield of ^{235}U and the discrepancies between the evaluated libraries of ENDF/B-VII.0 and JENDL 4.0. The experimental data was retrieved from EXFOR[6] and converted to capture yield for comparison with this experimental measurement.

60 MeV LINAC is a nine-section, radio frequency accelerator producing approximately 4x10^{13} neutrons/sec isotropically emitted from a water-cooled tantalum target.[7] Located at the 25 meter experimental station, a 16-segment NaI(Tl) γ multiplicity detector configured in a cylindrical geometry was used for the measurement.

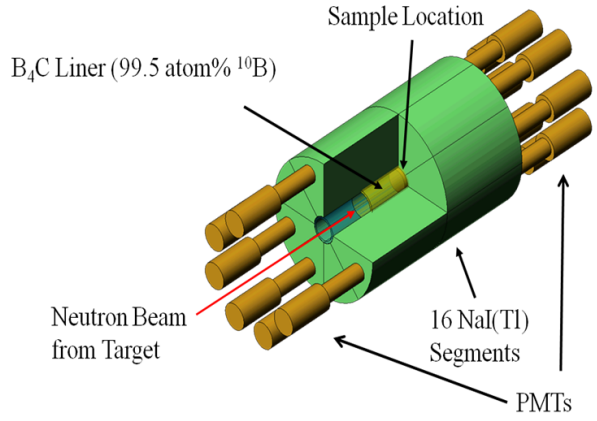

Figure 2: Cutaway image of the multiplicity detector showing the B_4C liner positioned between the neutron beam path and the 16 NaI(Tl) segments. The liner actually extends for the full length of the NaI(Tl) crystals. The sample location is in the center of the detector creating a 4π geometry.

A cylindrical hole centered on the axis of the detector allows the neutron beam to interact with the sample at the center of the detector. Between the central beam hole and the NaI(Tl) segments, there is a cylindrical B_4C liner 1.0 cm thick and enriched to 99.5% in ^{10}B designed to prevent scattered neutrons from interacting with the detector segments. The details of the multiplicity detector have been previously published.[8,9] Detected events are categorized based on the total γ energy deposited in the detector and the observed multiplicity. Each event in the data is recorded with the following tags: time-of-flight channel, multiplicity (1-15)*, and total γ energy group (up to four groups). A cutaway view of the multiplicity detector displaying the NaI(Tl) segments, boron liner, and sample location is shown in Figure 2.

Table 1: The conditions describing the experimental setup are summarized for the two categories of measurements: thermal and epithermal. Both the Enhanced Thermal Target and the Bare Bounce Target are based on the same tantalum plate design, but they moderate the neutron spectrum differently to achieve an increased flux in the desired energy range as indicated in the table. The conditions are chosen to enhance the quality of the data for the neutron energy region of interest in the designed experiment.

Experiment	Target	Pulse Width (μs)	Overlap Filter	Neutron Energy (eV)	Pulse Rate (pulses/s)
Thermal	Enhanced Thermal Target	0.5	None	$0.01 < E_n < 40$	25
Epithermal	Bare Bounce	0.015	Boron Carbide	$10 < E_n < 10000$	225

The experimental configuration was optimized for the desired energy range of the measurement, designated as thermal or epithermal. Elements considered in the design of the experiment include desired neutron flux, neutron overlap between LINAC pulses, and width of the resonances being measured. The configuration for each type of measurement is summarized in Table 1.

The sample consisted of multiple 0.5" diameter metallic uranium disks enriched to 93.33 $\pm$ 0.03 % in ^{235}U. The enrichment was determined by isotopic analysis conducted on three representative disks. The total mass of the sample ranged from 2.2 g for the thermal measurement to 20 g for the epithermal measurement. The motivation for increasing the mass of the sample for the epithermal measurement was to increase the interaction rate and thus the measured yield.

III. DATA REDUCTION

A typical simultaneous measurement of the neutron capture and fission cross sections or their ratio, α, involves the identification of a capture versus a fission interaction on an event-by-event basis by the use of an in beam fission tag-

*While there are 16 NaI(Tl) segments, the limit of the observed multiplicity is 15 due to the configuration of the data acquisition system.

ging detector or detecting the prompt neutrons emitted from fission. In this experiment the events are categorized into three groups based on the total γ energy deposited in the detector. Within these γ energy groups, the observed multiplicity of each event is also recorded, allowing for a range of multiplicity to be chosen during the post experiment analysis. The three groups and their respective total γ energies are as follows: scattering group (350 - 620 keV), capture-fission group (2 - 8 MeV), and fission group ($>$ 8 MeV). The fission group is limited to fission events because the γ energy released from neutron capture in ^{235}U will not exceed 8 MeV since its neutron separation energy is only 6.53 MeV. The capture-fission group contains a mixture of capture and fission events. The scattering group contains events primarily resulting from scattered neutrons interacting with the B_4C liner and the subsequent detection of the 477 keV γ ray emitted from the ^{10}B(n,α)^{7}Li* reaction. This reaction occurs 94% of the time, while the reaction leaving the lithium nucleus in the ground state happens only 6% of the time.

The expression for calculating the experimental yield, Y^{exp}, of a particular reaction for a given energy is shown in Equation (2), where R_S and R_B are the dead-time-corrected, monitor-normalized count rates for the sample and background respectively and ϕ is the smoothed flux with the background subtracted. There are monitors, two fission chambers and one ^{6}Li glass detector, used to account for any neutron beam fluctuations during the experiment. The neutron flux shape is determined experimentally with a $^{10}B_4C$ sample and the multiplicity detector.

$$Y^{exp} = \frac{R_S - R_B}{\phi} \tag{2}$$

The fission yield is determined by using the data from the fission group and normalizing it to an accepted value from ENDF/B-VII.0 at a particular energy or resonance, where the microscopic cross section from the evaluated library is converted to yield by using Equation (1). The fission yield, Y_f, is given by Equation (3) where k_1 is a constant that includes the flux normalization and detection efficiency.

$$Y_f = k_1 Y_f^{exp} \tag{3}$$

The key to this measurement technique is determining the amount of fission to remove from the capture-fission group. This is accomplished by considering the data in the capture-fission group as a linear combination of the two interactions. Thus the capture yield, Y_γ, is expressed as shown in Equation (4) where k_2 and k_3 are the coefficients for the linear combination.

$$Y_\gamma = k_2 Y_{\gamma f}^{exp} - k_3 k_1 Y_f^{exp} \tag{4}$$

The coefficients, k_2 and k_3, are found by normalizing the capture-fission group data to accepted ENDF/B-VII.0 values at two particular energies or resonances (one of the energies can be the same as used for fission) resulting in a set of two equations with two unknowns. The energies chosen for normalization depend on the desired energy range of the experiment. In either case, thermal or epithermal, the cross section values at the chosen energy should be well-known and accepted as precise. This technique for the simultaneous measurement of the neutron capture and fission cross section has been tested by applying it to a thermal measurement of ^{235}U where the experimental data and the evaluations have lower uncertainty.

In the energy range of the thermal experiment, the B_4C liner is highly effective at preventing scattered neutrons from reaching the NaI(Tl) segments. As the incident neutron energy increases, the liner's attenuation of scattered neutrons decreases. With the decreasing effectiveness of the boron liner, the likelihood of a neutron penetrating the liner and subsequently being captured in the NaI(Tl) crystals increases. When a scattered neutron penetrates through the boron liner, is captured in the NaI(Tl), and the γ energy deposited in the detector exceeds 2 MeV, these events result in a scattering interaction being recorded in the capture-fission energy group. Such events have been labeled with the term "false capture" and are treated as a background to the capture spectra. This background can be represented as a third term in the expression for the capture yield as shown in Equation (5), where f_c is the false capture fraction and Y_s is the scattering yield.

$$Y_\gamma = k_2 Y_{\gamma f}^{exp} - k_3 k_1 Y_f^{exp} - (f_c) Y_s \tag{5}$$

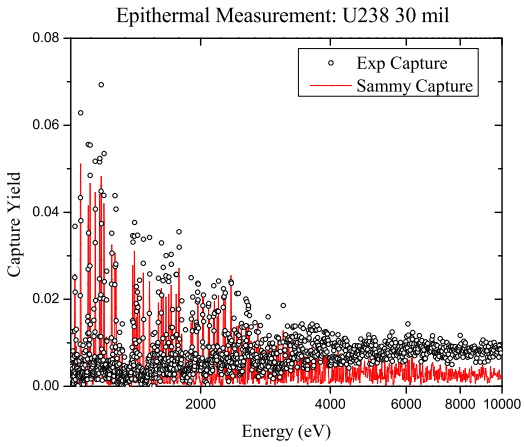

Figure 3: This graph shows the comparison of the capture yield between the epithermal measurement and the SAMMY calculation for a 30 mil (0.030 in) thick sample of ^{238}U. The increase in the capture yield of the measurement is due to the contribution from false capture. The impact of false capture in the ^{238}U measurement is large since this material has a high scattering cross section.

The impact of the false capture fraction on a measure-

ment of a material with a large scattering cross section is demonstrated with the capture spectra recorded for a ^{238}U sample as shown in Figure 3. For this nucleus the fission cross section is almost zero so that only capture and scattering reactions take place. As the neutron energy increases above 600 eV the measured yield separates from the yield calculated with the shape fitting code SAMMY[10] using ENDF/B-VII.0 parameters. The SAMMY code is used because it accounts for multiple scattering in the sample and includes the experimental resolution function.

The false capture fraction has been simulated with MCNP-Polimi[11] in order to track the γ ray production and energy deposition on a particle by particle basis. A graph of the simulated false capture fraction versus the incident neutron energy is shown in Figure 4 using the ENDF/B-VII.0 and JEFF-3.1 libraries.

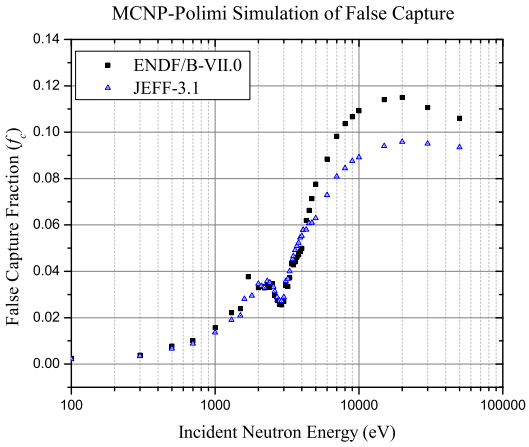

Figure 4: MCNP-Polimi was used to simulate the false capture fraction, f_c, of scattered neutrons from a natural lead sample 27 cm thick. The capture of the neutron by the NaI(Tl) crystal at energies above 3 keV is sensitive to the capture cross section of iodine as seen by the discrepancy between the ENDF/B-VII.0 and JEFF-3.1 simulations. The structure appearing near 3 keV is due to a large scattering resonance of iodine at about 2.8 keV.

The simulated false capture fraction based on the ENDF/B-VII.0 library was used to correct the ^{238}U experimental data and is presented in Figure 5. A point-by-point false capture correction is a challenge in the resolved resonance region which extends up to 20 keV for ^{238}U.

IV. RESULTS

The first step used to test the newly developed measurement technique was to apply it to the ^{235}U data obtained from a thermal measurement. This region provided two advantages: the cross sections are well known and the boron liner is very effective so the contribution from false capture

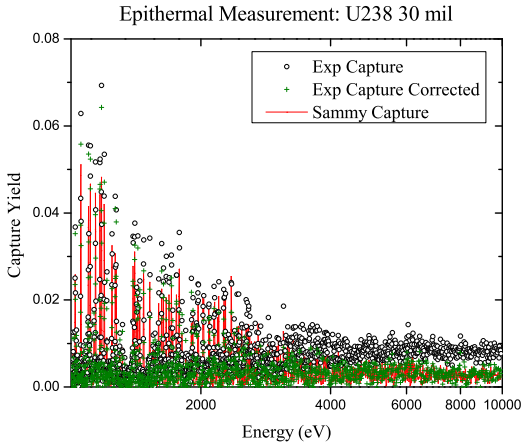

Figure 5: This graph shows the corrected experimental capture yield in comparison with the non-corrected experimental capture yield and the SAMMY calculation. The corrected capture yield has a much improved agreement with the SAMMY calculation. The resonance structure in the resolved resonance region and the large scattering cross section for ^{238}U present challenges to applying a point wise correction on a fine energy grid.

is negligible. The fission group was normalized at the accurate thermal energy (0.0253 eV) to determine the constant, k_1. Based on this normalization, the measured fission data and the SAMMY calculation agree very well from the thermal point up to 40 eV as shown in Figures 6 and 7.

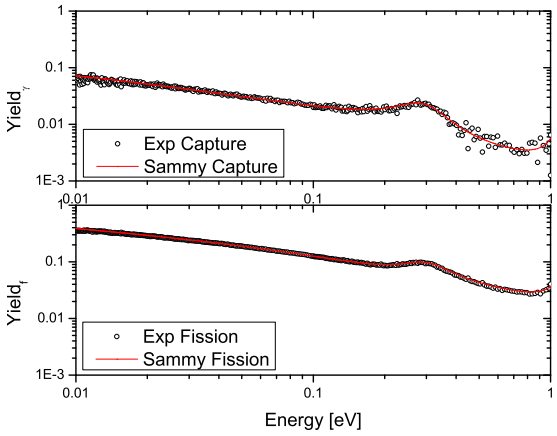

Figure 6: The graph depicts the excellent agreement between experiment and calculation for ^{235}U in the thermal region. The thermal point was used for the normalization of the measured values.

After the fission group data were normalized, the thermal point and the 11.7 eV resonance were chosen as the normalization points in order to determine the constants, k_2 and k_3. A comparison of the measured yield to the calculated yield determined from SAMMY for both capture and fission show very good agreement as demonstrated in Figures 6 and 7.

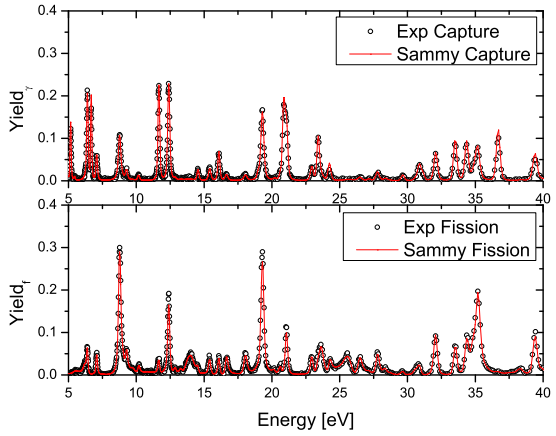

Figure 7: The graph shows excellent agreement between experiment and calculation for ^{235}U in the neutron energy range from 5 to 40 eV. The 11.7 eV resonance was used as the second normalization point for the measured capture yield.

After verification in the thermal region, the new technique was applied to an epithermal measurement of ^{235}U. For the analysis, the fission group with a multiplicity range of 3-11 was used along with the capture-fission group with a multiplicity range of 2-8. The scattering group was not used in this analysis. The normalization points for the epithermal measurement were 11.7 eV and 19.3 eV. These resonances were chosen because they are accurately known based on many experiments and agreement among the evaluated libraries. Using these low energy calibration points allows this measurement to accurately determine the capture yield at higher energies within the resonance region and the unresolved resonance region. The variations in the evaluated libraries and previous measurements indicate that the values at higher energies are not accurately known. Although the determination of the false capture fraction requires further refinement, a measurement of the neutron capture yield in the unresolved resonance region was conducted using the false capture correction found from the MCNP-Polimi simulation with a conservative estimate of 20% uncertainty on the correction. The results from the current measurement are consistent with previous experiments as well as the ENDF/B-VII.0 evaluation and the JEFF-3.1 evaluation as shown in Figure 8. In order to remove the dependence of the false capture correction on the simulations and the evaluated databases, the determination of the false capture fraction is also being investigated experimentally with carbon and lead samples. These materials provide an opportunity to measure the false capture contribution since their dominant neutron cross section, scattering, is orders of magnitude above capture and their cross section is effectively flat[†] in the energy range of the epithermal measurements. The planned outcome of investigating the false capture fraction through simulation and experiment is an energy-dependent correction factor with its associated uncertainty. As the false capture correction and the uncertainty analysis are refined, the uncertainty in the measurement is expected to approach 5% along with possible adjustments to the values of the yield. The current results will be finalized after the refinement is completed.

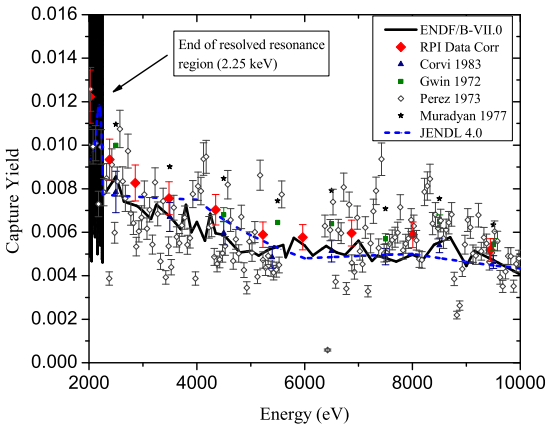

Figure 8: Within the estimated uncertainty, the results for the current ^{235}U measurement of the capture yield are consistent with both evaluated libraries and previously measured values. Based on the spread of the existing data, an accurate measurement is needed to address the discrepancies. The data presented was retrieved from EXFOR.[6]

V. CONCLUSIONS

The new technique for the simultaneous measurement of the neutron capture and fission cross sections that has been developed at Rensselaer Polytechnic Institute has shown promising results. The results from applying the method to ^{235}U in the thermal region compare very well to calculations for the capture and fission yield. The results of the epithermal measurement for capture are of comparable accuracy to previously measured data for ^{235}U in the unresolved resonance region. Through improvements in the determination of the false capture correction, which are being obtained with measurements of carbon and lead, along with a refined

[†]Except for two resonances near 3 keV.

quantification of the uncertainty in the measurement, this technique has the potential for much better accuracy of the ^{235}U capture measurement.

ACKNOWLEDGMENTS

The authors sincerely thank the technical staff at the RPI LINAC (Peter Brand, Matt Gray, Martin Strock and Azzedine Kerdoun) for their efforts in operating and maintaining the LINAC and assistance with setting up experiments.

REFERENCES

[1] F. Brown, "Computers and Monte Carlo - past, present, future." Colloquium Presentation at Rensselaer Polytechnic Institute. February 2011.

[2] N. Otuka, T. Nakagawa, and K. Shibata, "Uranium-235 neutron capture cross section at keV energies," *J. Nucl. Sci. Technol.*, vol. 44, p. 815, 2007.

[3] A. Santamarina, D. Bernard, P. Blaise, *et al.*, "The JEFF-3.1.1 nuclear data library: JEFF report 22," Tech. Rep. NEA No 6807, Nuclear Energy Agency, 2009.

[4] M. B. Chadwick, P. Oblozinsky, M. Herman, *et al.*, "ENDF/B-VII.0: next generation evaluated nuclear data library for nuclear science and technology," *Nucl. Data Sheets*, vol. 107, p. 2931, 2006.

[5] K. Shibata, O. Iwamoto, T. Nakagawa, *et al.*, "JENDL-4.0: A new library for innovative nuclear energy systems," in *Proceedings of the International Conference on Nuclear Data for Science and Technology*, (Jeju Island, Korea), April 26-30 2010.

[6] EXFOR, "Experimental nuclear reaction database." Complied by the Nuclear Reaction Data Centres Network (NRDC), retrieved from http://www.oecd-nea.org/janis/.

[7] M. E. Overberg, B. E. Moretti, R. E. Slovacek, and R. C. Block, "Photoneutron target development for the RPI linear accelerator," *Nucl. Instrum. Methods Phys. Res. A*, vol. 438, p. 253, 1999.

[8] R. E. Slovacek, R. C. Block, Y. Danon, C. Werner, G.-U. Youk, J. A. Burke, N. J. Drindak, F. Feiner, J. A. Helm, and K. Seemann, "Neutron cross-section measurements at the Rensselaer LINAC," in *Proceedings Topical Meeting. Advances in Reactor Physics*, vol. II, (Knoxville, Tennessee), p. 193, American Nuclear Society, April 11-15 1994.

[9] R. C. Block, P. J. Marano, N. J. Drindak, F. Feiner, K. W. Seemann, and R. E. Slovacek, "A multiplicity detector for accurate low-energy neutron capture measurements," in *Proc. Int. Conf. Nuclear Data for Science and Technology*, (Mito, Japan), p. 383, May 30-June 3 1988.

[10] N. M. Larson, "Updated users guide for SAMMY: Multilevel r-matrix fits to neutron data using Bayes equations," Tech. Rep. ORNL/TM-9179/R8, Oak Ridge National Laboratory, 2008.

[11] Polytechnic of Milan, *MCNP-POLIMI v1.0, Monte Carlo N-Particle Transport Code System to Simulate Time-Analysis Quantities*, 2002.