Validation of S(α,β) thermal neutron scattering libraries using pulsed-neutron die-away experiments

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Abstract. Validation of the accuracy of S(α,β) thermal scattering law (TSL) evaluations for moderator materials is an important task for the development of high-performance nuclear engineering systems. Many recent thermal neutron scattering evaluations have had limited experimental validation. Like other nuclear data, validation of TSL libraries has historically been by integral criticality benchmarks. While sufficient for general study, these benchmarks often have limited sensitivity to the tested TSLs, and compounding uncertainties from other nuclear data can make validation ambiguous. In some cases, no criticality benchmarks exist that are sensitive to the TSLs of interest. With the development of high-performance next-generation thermal nuclear reactors, alternative validation of applicable TSLs is of high importance. By performing thermal neutron measurements via pulsed-neutron die-away (PNDA) experiments, along with parallel simulations, the integral performance of various TSL evaluations can be compared to measured experimental data. An experimental testbed using a D-T neutron generator, moderator sample, and thermal neutron detector was assembled at Rensselaer Polytechnic Institute. A Thermo Scientific D211 Deuterium-Tritium Neutron generator is used to generate 10 μs neutron pulses. Various targets of different sizes and geometries are used to moderate the neutrons. Multiple detector types and configurations were tested to optimize the experiment. The room-temperature polyethylene TSL evaluation is well vetted and is similar across different evaluations. This makes it an ideal evaluation to compare with the experimental results.

1 Introduction

Thermal scattering law (TSL) libraries are used to generate neutron cross sections in the thermal energy region. TSL libraries are used to reproduce the scattering cross sections in the thermal energy region that take into account the impact of molecular bonds between atoms in materials. TSL libraries are constantly being improved and updated for various materials. For example, ice has been shown to have different total thermal cross section than sub-cooled liquid water [1]. Each material has a unique TSL and without experimental data to provide a reference for the generation of these evaluations there are bound to be inconsistencies between the actual thermal cross sections and the theory generated TSL cross section.

At the Gaerttner Linear Accelerator Center at Rensselear Polytechnic Institute (RPI), a pulsed-neutron die-away assembly is undergoing design and construction for the utilization of experimentally determining the accuracy of TSL libraries. The most recent measurements involve utilizing a cold polyethylene moderator at 24K and comparing to MCNP 6.2 [2] simulations with ENDF/B-VIII.0 for nuclear neutron cross sections. For the TSL libraries, the ENDF/B-VIII.0 S(α,β) [3] is compared to the ENDF/B-VII.1 [4] evaluation for liquid water and polyethylene.

To test and validate the data collection and detector assembly, measurements were taken using distilled water in a glass beaker. Once the system was validated, measurements with room-temperature and low-temperature polyethylene were conducted.

2 PNDA Theory

Pulsed-neutron die-away (PNDA) measures the rate of neutron leakage from a target. This leakage time-dependent rate can be estimated by diffusion theory as an exponential function with decay constant as:

$$\lambda = (\Sigma_a + (D * B_g^2) - (C * B_g^4)) * v$$

Where; λ is the fundamental mode decay constant, Σ_a is the neutron macroscopic absorption cross-section, D is the diffusion coefficient which is defined as $D \approx \Sigma_a/(3 * \Sigma_t^2)$, B_g is geometric buckling, C is the neutron cooling coefficient, and v is the neutron velocity [5]. This equation is an extension of diffusion theory but can reasonably calculate the time-dependent rate of change of the flux leaking from a moderator. Utilizing a known geometric buckling, the absorption and scattering cross sections can be experimentally determined.

Since the sensitivity of the scattering cross section is correlated to the geometric buckling of the sample, the sample size can be adjusted to change the sensitivity of

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the experiment to the total neutron absorption or total neutron scattering cross section of the target material. Having smaller geometries (i.e., larger buckling) increases the sensitivity to the effects of the TSL in the measurement, and the effects of the neutron cooling coefficient. Utilizing larger geometries (i.e., smaller buckling) the impact of the scattering and neutron cooling coefficient becomes negligible, thus the total absorption cross section of a target can be measured.

3 Experimental Assembly

The PNDA experimental assembly at RPI is currently in initial construction and development. Various configurations have been tested and experimental validation measurements have been taken to test the capabilities of the equipment and viability of performing PNDA measurements at the Gaerttner Center. Previous PNDA experiments have been assessed and studied to provide inspiration and insight into designing a PNDA experimental system. The slab water geometry measurements of Lopez and Beyster [6] and the water sphere measurements of Nassar [7] were used as design references.

3.1 Equipment

The PNDA system utilizes several basic components: a neutron source that can undergo pulse operation, a measurement target, neutron detectors, room shielding, electronic equipment and structural components.

For the neutron source, a D211 Thermofisher D-T source is used. The D-T source can generate 10^6 neutrons per second at an average energy of 14 MeV. This neutron source generates short pulses of 10 μs long at 100 pulses per second (100Hz).

Various measurement target samples have been utilized in the PNDA assembly. Validation of the measurements and analysis were done with a distilled water sample. Distilled water was chosen for its homogeneous composition (no crystalline structure), low impurity levels, and it is assumed to be a well-known moderator. Water was contained in glass or thin wall aluminum containers. Polyethylene samples have also been measured. Various sizes and geometries have been put though the PNDA system. For the low-temperature measurements, a slab geometry is utilized. 3 mm of cadmium surrounded the sample and detector to reduce room return and neutron background.

The detector used with this system was a LND25183. It is a He-3 gas proportional counter. The LND25183 was the preferred option due to its small size and insensitivity to gamma-rays.

System dead-time was determined utilizing a TOF clock. A Pu-Be source was placed near the He-3 detector with a polyethylene moderator. A measurement would then be taken with the detector system connected to the start and stop signal inputs on the TOF clock. Each cycle would start when it detected a count then stop when a second count is detected. By measuring the time between

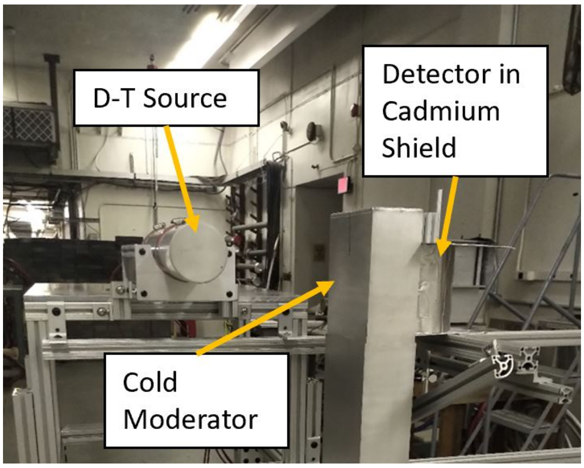

Figure 1. Cold Moderator System with Polyethylene Target, Installed in Modified PNDA Test Assembly

start count and the earliest second count the dead-time of the system can be determined. Using the electronics and detectors in this experiment a 3.5 μs dead-time was determined. A dead-time correction was applied to the experimental results utilizing the RPIXDR correction code [8])

3.2 Low-Temperature Moderator System

The low-temperature polyethylene measurements utilized the cryogenic cold moderator assembly. This system was utilized to perform TOF measurements on polyethylene and is thus optimized for those experiments [9]. Regardless of its original intended design function, it is found suitable for use in PNDA measurements. The cold moderator system consists of a moderator target inside a vacuum chamber. By utilizing a helium cryogenic cooler system, the target can be cooled down to 24K. A copper cold finger between the helium cryogenic system and the target acts as the thermal conductor. The polyethylene target is a 7" by 7" slab that is 1" thick with a 1/8" aluminum thermal conductor in the center. For more information on the cold moderator system refer to Reference [10].

4 Results

4.1 System Validation Measurements Using Water

Measurements of distilled water were initially performed to validate the accuracy of data measured by the PNDA system in comparison to MCNP 6.2 simulations. Distilled water was chosen as the target material for system validation for 2 primary reasons. The first is the TSL for water has been extensively validated at room temperature by Nasser and others. The second reason is distilled liquid water is a homogeneous material, it is easy to control for contaminants and there are no defects that could otherwise be present in a solid material such as polyethylene.

The experiment was conducted using a water sample housed in a 300ml borosilicate glass beaker. The sample and detector were both located inside the Cadmium can. For the validation the first measurement would be with the empty beaker as an open background measurement, then

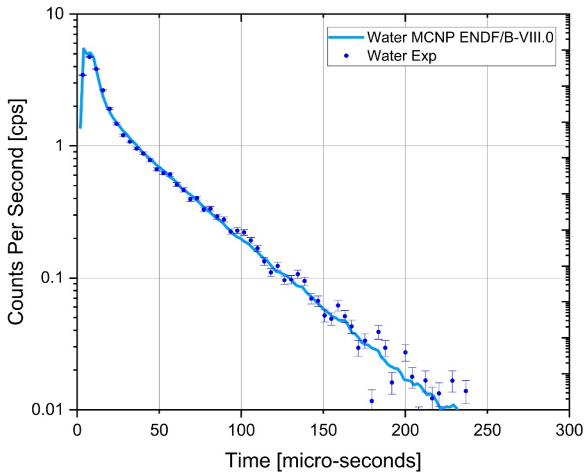

Figure 2. System Validation Measurements utilizing Water cylinder target, MCNP Water Cylinder Dimentions: 3.64 cm radius, 7.0 cm height

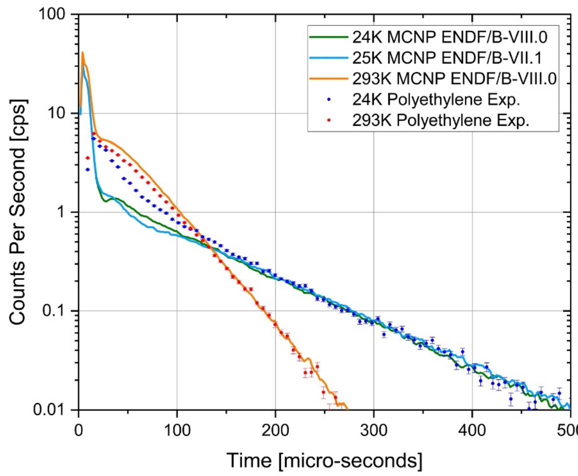

Figure 3. Cold Moderator System Measurements with Polyethylene Target vs MCNP Simulations

the beaker would be filled with water for the sample measurement.

Figure 2 shows the results of the distilled water validation measurements. The MCNP simulation results shown in Figure 2 only contain the sample and the active volume of the detector. The detector is modeled utilizing the active volume with an F4 tally and the reaction efficiency modifier: 103 (this modifier correlates to the (n,p) reaction in Helium-3). There is no room or Cadmium shielding in the MCNP simulation, only sample, detector and source. The source in the MCNP6.2 simulation is a directed spatially constant cone source that has an energy profile defined via a fast detector time of flight measurement.

The goal of the PNDA system validation measurements is to have experimental results that are equal to an "ideal" MCNP6.2 simulation of a PNDA experiment. With an "ideal" experiment having the target sample as the only source of thermalized neutrons in the system. Based on the water results it has been determined that the system can provide PNDA results that converge to an ideal simulation of a PNDA experiment, within reasonable error.

4.2 Low-Temperature Polyethylene Results

Initial measurements and simulations have shown few discernible discrepancies between the ENDF VIII.0 and ENDF VII.1 TSL libraries for room-temperature polyethylene. Thus a PNDA measurement with the unit at room temperature provides an excellent validation measurement that the system can provide results despite the modifications needed to utilize the cold-moderator assembly in the PNDA system. The Low-temperature TSLs were generated for ENDF/B-VIII.0 using NJOY2016 [11]. Discrepancies can be found computationally in low-temperature poly between ENDF/B-VII.1 and ENDF/B-VIII.0. Therefore, low-temperature PNDA measurements of polyethylene were taken to determine if the discrepancies can be experimentally discerned.

Using this system, special background correction methods were required. The cold moderator system is a single unit with the polyethylene contained within the system. Thus, a no-sample background measurement requires the entire assembly to be removed from the measurement. This includes the housing and copper cold finger contained in the low-temperature system. Background correction involved a standard sample minus no-sample measurement. Then a residual fit was applied to remove any room return neutrons that were measured due to the sample no longer covering the detector.

Figure 3 shows the results of the measurements taken with the cold moderator system. Good convergence between the experimental measurements and the MCNP6.2 simulations can be observed after 150 μs for both the room temperature and low temperature polyethylene measurements. In regards to validating TSLs it is the slope of the fully thermalized region that is important for validation. In this regard the data shows good correlation between the experiment and MCNP.

It is important to note that the MCNP simulation results shown in Figure 3 do incorporate the aluminum housing and copper cold finger. No TSLs are applied to the aluminum or copper in the simulation. The source term and detector modeling are the same as the water simulations and there is no room or shielding. The simulations only contain the source, detector and sample with cold finger.

The experimental assembly in its current form is capable of measuring the fundamental mode PNDA curve and could produce experimental data that can be utilized to discern the differences between TSL evaluations. The current measurements taken with water and polyethylene show the system is capable of producing results that are nearly identical to MCNP simulations of the target sample. If this experiment can be optimized and more controls of error and background are implemented, then it is feasible that this experimental setup can generate data that can discern the differences between two evaluations of a material at a specified temperature.

5 Conclusion

The PNDA system in development at RPI has shown promising results in its capabilities to collect neutron die-away data and be used for the validation of TSL libraries. Validation measurements with water in an ideal experimental assembly have shown excellent conformity between experiment and simulation. The low-temperature moderator system at RPI also provides a unique opportunity to look into the low-temperature range of TSL evaluations. However, more work is needed to optimize experiments utilizing the cold moderator for PNDA.

5.1 Acknowledgments

The first author has been supported by the Nuclear Regulatory Commission Fellowship Program under the grant NRC-HQ-60-17-G-0006. Any opinions, findings, and conclusions or recommendations expressed in this publication are those of the author(s) and do not necessary reflect the views of the US NRC. Special thanks to Dominik Fritz for the setup and operation of the cold moderator system.

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