

## Experimental validation of thermal scattering evaluations

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**Abstract.** In order to test the performance of new neutron thermal scattering law (TSL) evaluations it is desirable to have experimental data that is highly sensitive to the TSL and provides high fidelity information on the energy dependent performance of TSL evaluations. Three relevant experiments are discussed including: accurate thermal total cross section measurements, thermal neutron die-away experiments, and neutron leakage experiments. The experimental setups and results are reviewed and examples provided for some moderators including polyethylene, Plexiglas, and YH<sub>x</sub>. For the experiments performed thus far, there is generally good agreement between the measured total cross section and simulations using current TSL evaluations, however in certain energy ranges differences were observed. Similarly neutron die-away and leakage measurements for samples at room temperature are in good agreement with data computed from TSLs, however leakage measurements for polyethylene at 29K show discrepancies with TSL evaluations.

### 1 Introduction

In recent years there has been a resurgence in the field of thermal scattering law (TSL) evaluations and experiments. This comes after years that this field was dormant with little updates or new ENDF evaluations. Evaluated TSL is based on atomistic calculations that in most cases result in a phonon spectrum that can be processed using a code like NJOY [1] to create the scattering kernel for use in applications. In the evaluation assumptions are made on the material structure and crystallinity thus the experiments are also validating such assumptions by comparing them to real materials.

For neutron moderators such as water (H<sub>2</sub>O) and polyethylene (CH<sub>2</sub>) atomic mix of the nuclide cross sections will result in a very wrong scattering cross section in thermal energies. TSL evaluations take into account the change in relative velocity between the neutron and atom that originate from molecular vibration and rotation effects (primarily of H). A TSL evaluation is used to reproduce the thermal scattering cross sections and angular distributions below a few eV. In thermal reactor applications where the neutron flux is mostly isotropic, the highest sensitivity to the scattering kernel is from the total cross section and much less from the angular distributions. Figure 1 shows the thermal total cross section of CH<sub>2</sub> calculated using free gas Doppler broadening and using a scattering kernel in ENDF/B-8.0 [2], the large difference between the two is evident as the incident neutron energy decreases. Direct measurement of the scattering cross section is not easy but measurement of the total cross section can be ac-

complished with high accuracy (1-2%) by inferring it from neutron transmission measurements. Similarly the angular distribution of the scattered neutrons can be calculated from the TSL and thus the neutron slowing down rate and neutron leakage from a moderator are sensitive to this distribution.

TSL evaluations will affect results (multiplication factor) calculated for thermal criticality systems and criticality benchmarks, but these systems usually involve other materials that contribute to the uncertainty of the calculated multiplication factor, thus experiments that are sensitive only to the TSL are preferred for validation. These experiments must have the sensitivity to resolve differences between evaluations when they occur and cover the different physics of the TSL including temperature.

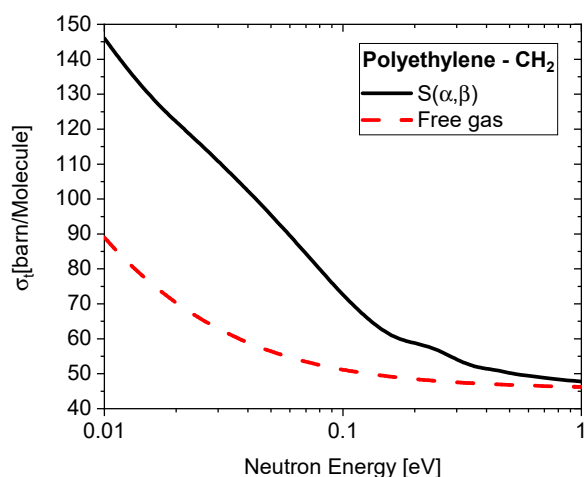
### 2 Experiments

Three types of experiments were developed and used, they include: high-accuracy total cross section measurements in the energy range from 0.0005 eV to 3 eV, neutron die-away measurements, and neutron leakage measurements. These experiments test different aspects of the TSL that are relevant to applications. These types of experiments are not new and were previously used, but in order to use them for validation of modern evaluations, experimental capabilities need to be re-developed and be available.

#### 2.1 Total cross section measurements

Over the years many cross sections and other nuclear data were measured at the Gaerttner LINAC Center at Rensse-

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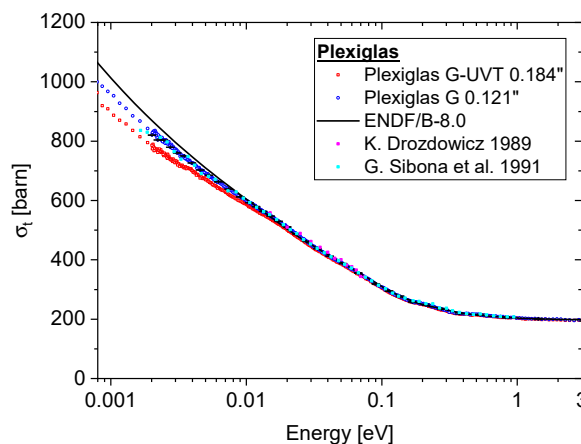
**Figure 1.** Comparison of the total cross section of CH<sub>2</sub> calculated using the scattering kernel in ENDF/B-8.0 vs. free gas Doppler broadened cross sections

laer Polytechnic Institute (RPI). The Center utilizes a linear electron accelerator (LINAC) that produces electron pulses that are 6 ns - 4 μs wide with an energy of about 55 MeV. These electrons are directed to a stack of water cooled tantalum plates to produce neutrons with an evaporation spectrum of average temperature of about 0.5 MeV and a spectrum tail up to the maximum electron energy. Different neutron production targets are used to moderate this spectrum and tailor it to the experimental needs (for example [3]). To cover the thermal region, a so-called enhanced thermal target (ETT) was previously developed [4]. This neutron production target provided good signal to background and energy resolution to enable resonance measurements for incident neutron energies from 0.002 - 20 eV [5].

In order to improve the measurements capabilities at RPI a cold moderator was recently designed and constructed as an add-on to the ETT and was named ETTC. The moderator is 2.54 cm thick piece of polyethylene operating at about 29K [6]. Below 0.01 eV the ETTC provides a flux enhancement of up to a factor of 8 over the ETT and thus enables measurement below the lowest Bragg edge of most materials of interest like Be, YH<sub>x</sub>, ZrH<sub>x</sub> and others. The ETTC has a usable energy range from 0.0005 - 3 eV where accurate neutron transmission measurements can be performed.

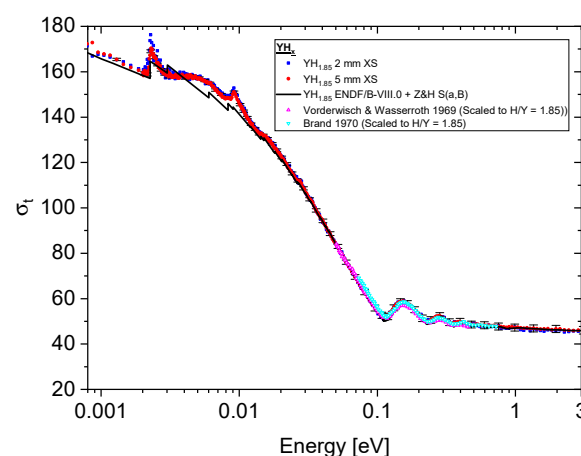
Recently both ETT and ETTC were used for measurements of room temperature total cross sections for several materials of interest to criticality safety applications including polyethylene, polystyrene, and Plexiglas. These materials are part of one or more criticality benchmarks. Other materials that are of interest to reactor applications such as YH<sub>x</sub> and Be were also measured. To illustrate the new capabilities, the ENDF/B-8.0 total cross section of Plexiglas is shown in figure 2 and compared with new RPI measurements and other experimental data found in EXFOR [7]. For Plexiglas two types of common variety (G and G-UVT) were measured and below about 0.02

eV they differ. The ENDF/B-8.0 TSL evaluation is closer to the measurement of Plexiglas G, but below 0.02 eV it is higher than the experiment. Such experiments indicate that the TSL can be reevaluated to achieve a better agreement.



**Figure 2.** The total cross section of Plexiglas comparing the RPI measurements with others and the cross section generated from the TSL in ENDF/B-8.0

Another example is a measurement of YH<sub>1.85</sub> plotted with other experimental data available in EXFOR and the ENDF/B-8.0 evaluation in figure 3. This comparison shows the ability to resolve all the Bragg edges at low energies and the wavy behaviors of the cross section between 0.1 and 1 eV. In general the evaluation is in good agreement with the experiment except for the Bragg edge structure between 0.002 to 0.02 eV. This illustrates the need to use a TSL library that is tailored to the actual material intended to be used in an application and not an ideal material often used for the evaluation. The reasons for disagreement could be related to a different crystalline structure in the measurements compared to the evaluation or inaccuracies in the physics embedded in the processing codes.



**Figure 3.** The total cross section of YH<sub>1.85</sub> comparing the RPI measurements with others and the cross section generated from the TSL in ENDF/B-8.0

## 2.2 Neutron die-away measurements

The use of neutron die-away measurements is an established method for measuring the slowing down time of a moderator [8]. It was used in the early days of reactor physics to measure neutron diffusion coefficients of different materials. In this method a pulsed neutron beam is entering a moderating material and the thermal neutron population inside the medium, or leaking from it, are measured as a function of time. To understand the physics it is useful to recall the fundamental mode solution of the time dependent diffusion equation for the neutron density  $n(\mathbf{r}, t)$  in a large medium:

$$n(\mathbf{r}, t) = A_0 \psi_0(\mathbf{r}) e^{-\alpha t} \quad (1)$$

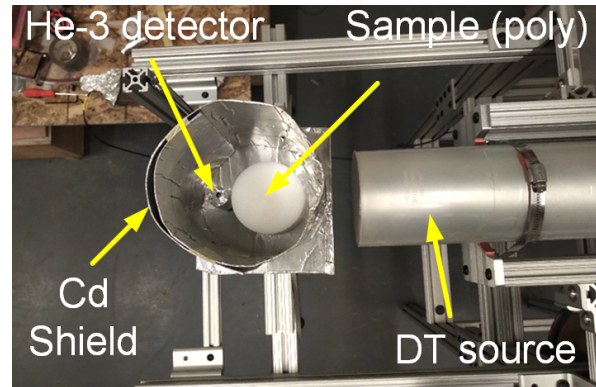
where  $\alpha = v(\Sigma_a + DB^2)$  and  $\psi_0(\mathbf{r})$  is the spatial distribution. In these equations  $v$  is the average thermal neutron velocity,  $\Sigma_a$  is the average thermal macroscopic absorption cross section,  $D$  is the effective diffusion coefficient, and  $B^2$  is the geometric buckling which is inversely proportional to the dimensions of the system. Thus a large moderator will have a small value of  $B^2$  and thus  $\alpha$  is small and dominated by  $\Sigma_a$  resulting in a slower flux decay. In a small (leaky) system additional terms are needed in order to correct the diffusion coefficient and usually a diffusion cooling coefficient  $C$  is added such that:  $\alpha = v(\Sigma_a + DB^2 + CB^4)$ . Measurements of the neutron die-away  $\alpha$  as a function of the buckling can provide information on the thermal diffusion coefficient and thermal absorption cross section of the moderator [9].

Now-a-days it is more advantageous to compare a neutron die-away measurement to detailed time dependent simulation using a code like MCNP [10] to check how a TSL library is performing in a time dependent calculation. The ability to use small samples of different geometries, and the relatively low cost of the experiments are advantages of this type of experiment.

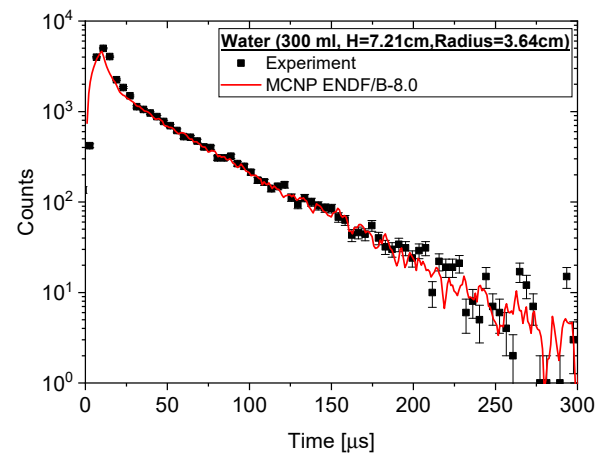
An example of the geometry used for such experiments is shown in figure 4, the geometry is very compact with flexibility on the sample size and geometry. The system was driven by a commercial pulsed DT source emitting  $10^8$  n/s with energy of about 14 MeV, operating at 100 Hz with a pulse width of 10  $\mu$ s.

An initial qualification of the system was performed using two measurements with a water cylinder - one with the sample, and one without it. The final result is the sample minus open spectrum. In figure 5 the experiment is compared with MCNP simulation of the geometry and He-3 detector using the ENDF/B-8.0 scattering kernel. The simulation was for the geometry with the sample only and did not include the surrounding room. The agreement between the slopes of the experimental and calculated die-away curves seems very good and can be further quantified by fitting the data.

Another experiment used a polyethylene cube sample and is shown in figure 6. In this case it seems that the simulation result has a slightly faster die-away (larger  $\alpha$ ) compared to the experiment. It is important to note that careful subtraction of the right background is needed. In the



**Figure 4.** A top view of the experimental setup used in this work showing the DT neutron source on the right, He-3 thermal neutron detector, polyethylene cylindrical sample, and a Cd thermal neutron return shield surrounding the sample and detector.

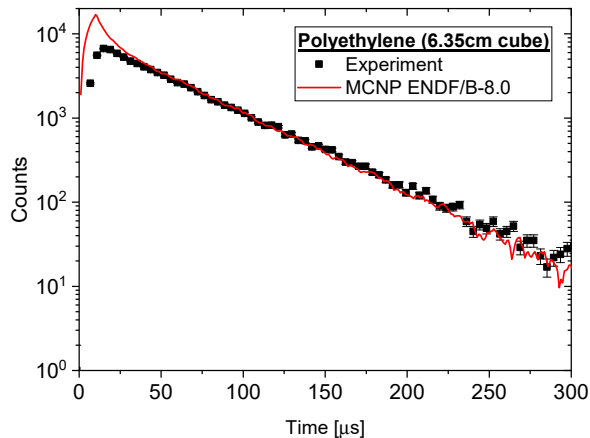


**Figure 5.** Measurement of thermal neutron die-away from a water cylinder, pulsed and measured from outside the cylinder and compared with MCNP simulation using the scattering kernel in ENDF/B-8.0.

region between 75-150  $\mu$ s where the background is negligible the agreement seems better. In this case (room temperature measurements) simulations with other available scattering kernels gives the same result and such experiment is insensitive to the very small difference between the evaluations.

## 2.3 Neutron leakage measurements

A neutron leakage experiment is similar to a die-away experiment. In a leakage experiment a moderator is placed near a pulsed neutron source, and the detector is placed at a long distance, about 15m for the experiment described here. This measurement is thus similar to a time-of-flight (TOF) experiment where the detection time is related to the neutron energy. For the experiments reported here, the RPI LINAC was used with the ETTC target mentioned in section 2.1 and a polyethylene moderator was used as the sample. This setup allowed us to test the concept and measure the leakage for a room temperature and cold polyethy-

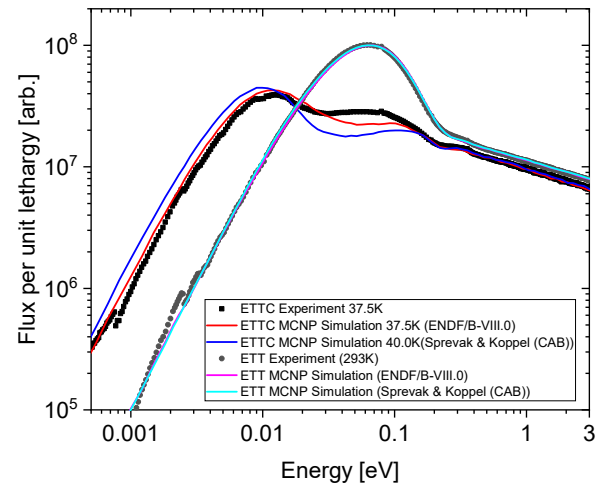


**Figure 6.** Measurement of thermal neutron Die-Away from a polyethylene cube, pulsed and measured from outside the cube and compared with MCNP simulation using the scattering kernel in ENDF/B-8.0.

lene moderator. The moderator size was 17.78 cm x 17.78 cm and 2.54 cm thick and measurements were done at two temperatures; 293K and 29K. The experimental and simulation results with two different TSL evaluations are shown in figure 7. The simulation in this case included the evaporation neutron spectrum produced by the tantalum target and included the full geometry of the ETT target and the added polyethylene moderator. The room temperature measurement also serves as validation of the simulation and shows excellent agreement with the experiment regardless of the TSL evaluation used (ENDF/B-8.0 or CAB [11]). The visible Bragg edge near 0.0025 eV and 0.0008 eV are from lead (or its oxide) that was in the beam between the moderator and detector. At low temperatures the two evaluations do not agree between them and show a departure from the experiment. This example shows the high sensitivity of this type of setup to the TSL evaluation. Because of the good agreement between the simulation and experiment at room temperature, the likelihood of a geometry mismatch between the them is low. There could be several other reasons to the disagreement between the evaluations and experiment which could relate to the models used and approximations that might not be valid at such low temperatures or approximations in the processing code NJOY [1] used to prepare the data for MCNP.

### 3 Conclusions

Experimental data is needed for validation of TSL evaluations, this data should be sensitive to the features of TSL such as cross section, angular distribution, and temperature. Three types of experiments; total cross sections, die-away, and leakage were discussed as possible experiments that can help validate TSL evaluations. The scattering cross section resulting from using the TSL is an important quantity for thermal systems and criticality applications thus accurate and energy detailed measurements are needed. This was accomplished by measuring the to-



**Figure 7.** Measurement of neutron leakage from the ETTC moderator at 29K with size of 17.78 x 17.78 x 2.54 cm<sup>3</sup> (7 x 7 x 1 inch<sup>3</sup>)

tal cross section from 0.0008 - 3 eV using the neutron time-of-flight methods. These measurements had accuracy of about 1.5% dominated by systematic uncertainties. Knowledge of the exact composition and crystalline structure of the sample is very important for correct interpretation of the measured cross sections. It is also important to note that accurate knowledge of the capture cross section of elements in the sample is also needed, in many cases the ENDF or other evaluations libraries can be used, especially for well know elements such as H and C (where capture is very small). The die-away and leakage method provide data that can be compared to time-dependent simulation that help test the performance of a TSL library by looking at leakage of thermal neutrons at different time scales.

All mentioned methods provide a comprehensive data set that validates the TSL and also provide information about discrepancies and informs the evaluation process on where improvements are needed.

### 4 Acknowledgment

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