Sensitivity Analysis of H_2O Pulsed-Neutron Die-Away Experiments to the H-H_2O Thermal Scattering Law

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INTRODUCTION

Lawrence Livermore National Laboratory is conducting new Pulsed-Neutron Die-Away (PNDA) benchmark experiments to validate neutron thermal scattering laws (TSLs). TSLs are important data for modeling thermal fission reactors, criticality safety scenarios, and radiation protection and detection, *i.e.* any application with thermal neutrons. These simulations require high-quality nuclear data, with confidence in their quality established through validation.

PNDA experiments have several advantages over critical experiments when used to validate TSLs [1]. The target materials are very simple in terms of their geometry and composition, which reduces modeling uncertainties in the benchmark. The only reactions in the target material are absorption and scattering, which limits uncertainties from other nuclear data, especially those related to fission. Furthermore, the simple targets make the experiments less expensive and easily tunable to vary absorption and thermal scattering sensitivities. The integral parameters in well-conducted experiments have low uncertainties of 0.1% to 0.5%, which is useful for validation. Finally, they do not require fissile material and can be conducted in non-nuclear facilities at reduced cost and regulatory burden.

The validation process typically includes a quantification of uncertainties, both in the experiment and in the simulation. Simulation uncertainties may include those arising from the modeling (e.g., geometry and composition) or from nuclear data. The nuclear data uncertainties can be particularly important in neutronically-driven scenarios like those encountered in reactor physics and criticality safety. This work quantifies the uncertainty in the integral parameter of a simulation of a PNDA experiment caused by TSL uncertainties, demonstrating the experiment's tunable sensitivity. We focus on a light-water experiment and the H-H_2O TSL data. The conclusions can be applied to future validation studies of TSLs with PNDA experiments as well as to potential Bayesian model calibrations of the TSLs with said experiments.

Pulsed-Neutron Die-Away Experiments

The experiment uses a neutron generator to impinge a short (10^{-5} to 10^{-3} s), mono-energetic neutron pulse on a target sample. The target is the medium for which the TSL is being validated, *e.g.* H_2O. After the pulse, the neutron population moderates and reaches a thermal equilibrium in the target with a fundamental spatial mode and a characteristic decay-time eigenvalue. The eigenvalue can be extracted from the experimental measurements of the neutron flux and then used as an integral parameter in validation. Eq. 1 describes the neutron population at thermal equilibrium and in the fundamental spatial mode. Here, $\mathbf{r}$ is a spatial location, t is time, $R(t)$ is the background room return of neutrons, and α [s^{-1}] is the characteristic flux decay-time eigenvalue, or the integral parameter of the experiment.

$$\phi(\mathbf{r},t) = \phi_0(\mathbf{r})\exp(-\alpha t) + R(t) \qquad 1$$

The PNDA analysis requires calculating α, for both experimental and simulated data, by fitting Eq. 1 to a neutron counts vs. time curve. Fig. 1 shows a simulated pulsed die-away curve from MCNP6.2®. The neutron pulse ends at 0.66 ms, and the neutrons diffuse through the target material and travel to the detector. An exponential fit with non-linear least squares is performed on the neutron counts in the region where the fundamental mode is reached to determine α. The fit varies φ_0 and α to minimize the cost function, χ^2, between the MCNP6.2® tally and the exponential function.

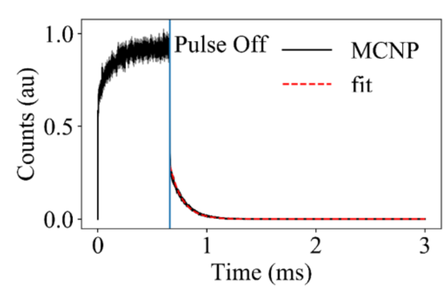

Fig. 1. Simulated die-away curve from a PNDA experiment.

The geometric dimensions of the moderating target determine the experiment's sensitivity to thermal scattering. When the target sample is large, α depends mostly on absorption, *i.e.* not on TSLs. When the target sample is small, the physics of thermal scattering highly influence α. Fundamentally, the sensitivity of α to thermal scattering increases as the target sample size decreases. From this understanding, we expect the α value of small targets to have the largest sensitivity to TSL uncertainties.

The PNDA experiment studied herein is based on the measurement of Ref. 2. It was a light water experiment with a cylindrical geometry. The cylinder's dimensions were manipulated to alter the physics of the system, i.e. to emphasize the sensitivity to absorption or scattering data. We took their geometric buckling values and used them to determine the dimensions of corresponding cylinders, as the actual dimensions were not reported. For each buckling, a fixed cylinder height of 18 cm was used, and the radius was varied to match the reported buckling. No experimental uncertainties were available, and we assumed a value 1% for all α values. While this is not a high-fidelity approach, it was adopted because this study focuses on the sensitivity of PNDA experiments to TSLs and less on a precise validation. In the future, new PNDA experiments will have high-fidelity models and precise catalogues of the experimental uncertainties for validation.

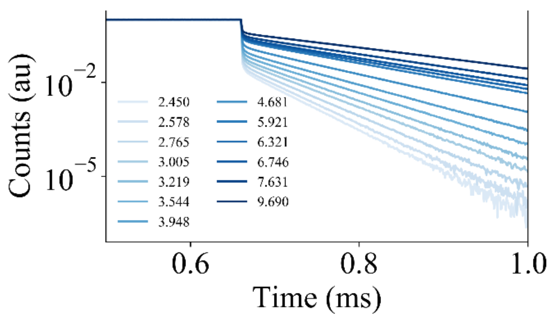

Fig. 2. Decay profiles for the historical H_2O experiments, differentiated by the radius (cm) of the target cylinder.

TSL Uncertainty Quantification

The H-H_2O TSL in the ENDF/B-VIII.0 library was made with the Centro Atómico Bariloche (CAB) model. The CAB model simulates molecular dynamics to calculate the continuous frequency spectrum, vibrational modes and partial structure factors. In this work, we use a parameterization methodology presented in Ref. 3 (and available at Ref. 4) to perform a Total Monte Carlo (TMC) uncertainty quantification of the α integral parameter in the PNDA experiment. This estimates the uncertainty of α to the TSL, which subsequently gives a measure of the sensitivity of the experiment to the desired TSL.

The double-differential thermal scattering cross section of a material can be calculated by means of Eq. 2. Here, E and E' are the incident and secondary neutron energies, σ_b is the bound scattering cross section, kT is the temperature in eV, α is a dimensionless momentum transfer term, β is the dimensionless energy transfer term, and $S(\alpha,\beta)$ is the symmetric form of the scattering law. $S(\alpha,\beta)$ is calculated with parameters derived from the CAB model and using a nuclear data processing code like LEAPR in NJOY.

$$\frac{\partial^2\sigma}{\partial\Omega\partial E} = \frac{\sigma_b}{4\pi kT}\sqrt{\frac{E'}{E}}\exp\left(-\frac{\beta}{2}\right)S(\alpha,\beta) \qquad 2$$

The UQ methodology involves sampling the model parameters of LEAPR. To sample the TSL, the model parameters of Eq. 2 and TABLE I are perturbed and used by NJOY to produce an ACE file for MCNP simulations. The nominal values of these parameters are those that are contained in the CAB model for H-H_2O. Except for the α and β grids, all samples were taken from a Gaussian distribution.

TABLE I. Parameters that are perturbed in the LEAPR file.

Parameter	Variable	Format
Free-atom cross section	σ_{free}	Scalar
Diffusion weight	w_d	Scalar
Oscillator energy 1	E_1	Scalar
Oscillator energy 2	E_2	Scalar
Oscillator weight 1	w_1	Scalar
Oscillator weight 2	w_2	Scalar
Continuous spectrum weight	w_β	Scalar
Continuous frequency spectrum	$\rho(\beta)$	Vector
Momentum grid	α	Vector
Energy grid	β	Vector
Structure correction functions	$S(Q)$	Vector

Fig. 3 shows 200 samples of the incoherent inelastic scattering cross section of this study and compares their mean to the ENDF/B-VIII.0 evaluation. It additionally shows the relative standard deviation of the cross section at different energies. Fig. 4 gives the resulting correlation matrix of the cross section from this sampling.

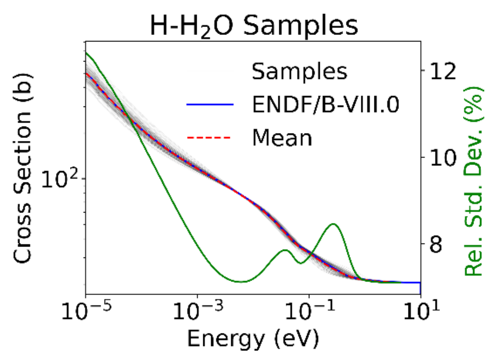

Fig. 3. The randomly sampled incoherent inelastic scattering cross section in this study. The sample mean is compared to the ENDF/B-VIII.0 evaluation.

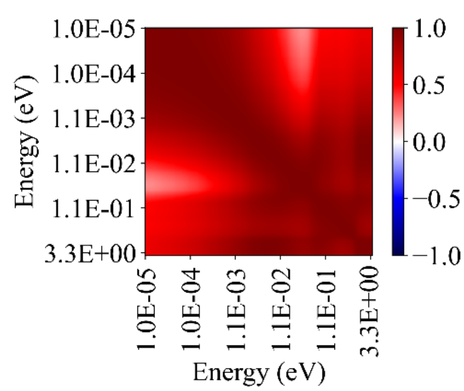

Fig. 4. Correlation matrix of the H-H_2O incoherent inelastic scattering cross section created by the random sampling.

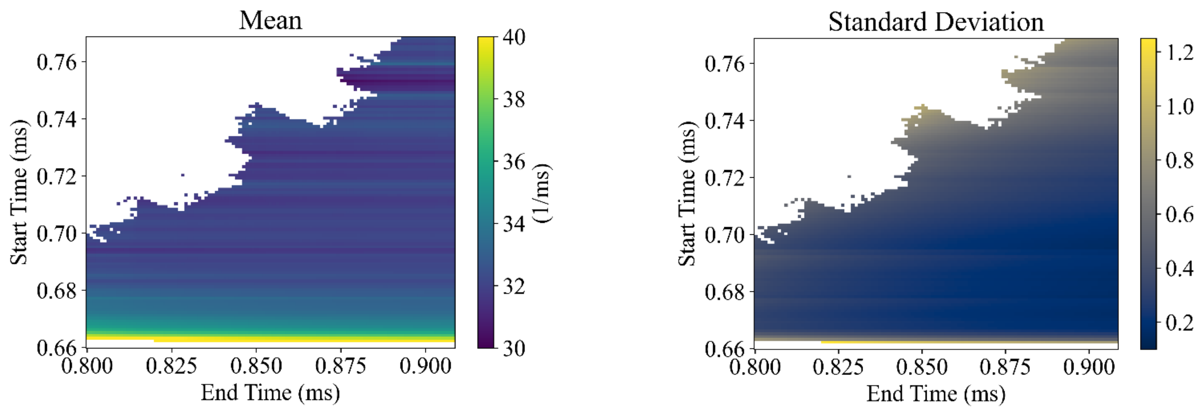

Fig. 5. Effect of data interval on the fit of α. The start time indicates the beginning of the data interval included in the fit, and the end time indicates the end of the time interval. At certain combinations of start and end times, the non-linear least-squares fitting algorithm did not converge and an α value could not be estimated, which is indicated by the white regions.

For every TSL sample, the models of the PNDA experiments were run. This creates a population set of α values from which the mean, standard deviation, and covariances can be estimated. We applied the Fast TMC approach to achieve quicker convergence [5]. This method applies a different random-number seed to each MCNP simulation to reduce the number of histories needed in each individual simulation. By doing so, the influence of the statistical uncertainty of each α (inherent to the Monte Carlo simulation) is reduced for a given number of random samples.

Each die-away curve needs a specific time interval for its fit, or else the α integral parameter may be over or under-estimated. It is possible to choose too early times, *i.e.* before either the flux is either fully thermalized or is in its fundamental spatial mode. It is also possible to include data that are too late in the curve where room return, *R(t)* in Eq. 1, is significant. These data are excessively noisy and can cause large uncertainty in the fitted α. Moreover, the standard deviation has shown to be particularly sensitive to the time interval of the fit, with incorrect intervals creating up to 5% over-estimated uncertainties in the α values.

In general, high quality fits create low uncertainties. If too early data are included, the curve does not follow as closely exponential behavior and thereby induces a larger fit uncertainty. If too late, *i.e.* noisy, data are included they create a large uncertainty in the fit. For this analysis, we fit the die-away curves over a range of intervals and record the α values and their statistical uncertainties. From these values, we choose the α that has the lowest uncertainty as the value for a specific sampling of the TSLs. Fig. 5 demonstrates this analysis, showing how the mean α can vary based on the fitting interval, and how the uncertainty of the fit also varies. The uncertainty of the fitted α is also important for estimating the standard deviation of α induced by the TSLs. Too large statistical uncertainties will create excessive noise in individual samples, masking the uncertainty from TSLs.

RESULTS

A total of 200 random samples of the H-H_2O TSLs were generated, and each was used to calculate the α value for the simulated PNDA experiments, following the previously outlined approach. TABLE II summarizes the mean calculated values and their standard deviations, and provides estimated experimental values from Ref. 2. The relative standard deviations of simulated α created by the TSLs, or σ_{xs}, shows a clear trend: the smaller targets have a larger TSL-based uncertainty than the larger targets. This confirms previously outlined theory that small targets are more sensitive to TSL data than large targets.

This result is corroborated by Fig. 6, which provides the correlations between the α values of each target in the experiment; the smaller sized targets are much more tightly correlated. For example, the 2.45 cm and 2.58 cm radius cylinders have a correlation coefficient of 0.96. Meanwhile, the 2.45 cm and 9.69 cm cylinder have a correlation coefficient of 0.41. As the TSL data are the solely considered source of uncertainty, this demonstrates that the small targets have a larger sensitivity to thermal scattering effects. The larger targets still have a sensitivity to TSLs, as demonstrated by the non-zero correlation coefficient, but are less sensitive.

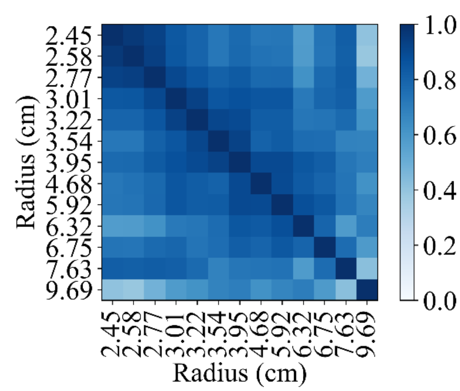

Fig. 6. Correlation coefficients between the different sized H_2O target cylinders.

TABLE II. Data for the validation of the experiment that include the mean calculated α, or C, and the experimental value, E. The uncertainty of C due to TSLs, σ_{XS}, is calculated with the observed uncertainty of the sample population, σ_{obs}, and the average statistical uncertainty of each α, σ^2_{stat}, by means of $\sigma_{XS} = (\sigma^2_{obs} - \sigma^2_{stat})^{0.5}$. The Z-score is a normalized bias measure.

Radius (cm)	Mean C (ms^{-1})	E (ms^{-1})	(C-E)/E (%)	σ_{obs} (%)	σ_{stat} (%)	σ_{XS} (%)	Z Score
2.45	32.78	34.5	-5.08	3.18	0.43	3.15	1.06
2.58	30.65	33.2	-7.59	2.75	0.39	2.72	1.65
2.77	27.84	30.2	-7.90	2.69	0.38	2.66	1.65
3.01	25.05	26.4	-5.26	2.74	0.34	2.71	1.01
3.22	22.95	23.4	-2.09	2.61	0.33	2.59	0.38
3.54	20.39	20.9	-2.24	2.37	0.33	2.35	0.40
3.95	17.99	18.3	-1.58	2.44	0.31	2.42	0.26
4.68	15.08	14.4	4.46	2.08	0.31	2.06	0.68
5.92	12.03	11.4	5.86	1.76	0.32	1.73	0.83
6.32	11.29	10.9	3.14	1.90	0.34	1.87	0.43
6.75	10.70	10.3	3.40	1.88	0.36	1.85	0.45
7.63	9.72	9.4	3.50	1.60	0.36	1.56	0.47
9.69	7.59	8.3	-8.62	1.40	0.48	1.32	1.18

In comparison to criticality experiments, these PNDA experiments have larger uncertainties induced by the assumed TSL uncertainty, indicating a larger sensitivity to TSLs themselves. Ref. 3 reports a 0.046% uncertainty in k_{eff} caused by TSLs for the OPAL reactor. Testing reported in Ref. 4 gives uncertainties of 0.12% to 0.25% in the k_{eff} of critical benchmarks. The high uncertainty of the PNDA experiments to the TSL data vs. critical experiments further highlights their utility in validating this kind of nuclear data.

The relative biases, (C-E)/E, show an underprediction of the simulations for small samples, or those more sensitive to TSLs. The Z-score for the α values was calculated to evaluate the importance of the biases, assuming an experimental uncertainty of 1%. The Z-score is the absolute difference between C and E normalized by their combined uncertainties. It indicates how many standard deviations away from zero a bias is. TABLE II shows that all target samples agree within two standard deviations. This validation is qualitative given the many assumptions and approximations taken to replicate the historical experiment. It also ignores calculated and experimental correlations that would be considered by evaluating the χ^2 of the data set. These deficiencies highlight the need for the new benchmark-quality experiments that Lawrence Livermore National Laboratory is conducting.

CONCLUSIONS

We have presented an uncertainty quantification of an H_2O, pulsed neutron die-away experiment's integral parameter to H-H_2O TSL data. The integral parameter of smaller-sized H_2O targets showed uncertainties up to 3.15%, which exceeds those reported for criticality experiments by an order of magnitude. The smaller targets also had the largest uncertainties, confirming a theoretical understanding: smaller targets are the most sensitive to thermal scattering.

Future work will include incorporating other sources of nuclear data uncertainty, especially absorption cross sections in H and O that are important for large target geometries. The experiments could also be incorporated into a Bayesian Monte Carlo data assimilation scheme [6]. Finally, this whole analysis scheme may be used with new experiments that will be conducted at Lawrence Livermore National Laboratory.

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