Pulsed-Neutron Die-Away Experiments for Plastics and Neutron Thermal Scattering Laws

Daniel Siefman1, Shauntay Coleman1, Jordan Northrop1,2, William Zywiec1, Jesse Holmes3, Michael Zerkle3, David Heinrichs1, and Catherine Percher1

1Nuclear Criticality Safety Division, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, CA 94550, USA
2School of Nuclear Science and Engineering, Oregon State University, 1701 SW Western Blvd. Corvallis, OR 97331, USA
3Naval Nuclear Laboratory, PO Box 79, West Mifflin, PA 15122, USA

Abstract. Pulsed-neutron die-away (PNDAs) can be useful benchmarks to validate neutron thermal scattering laws (TSLs). The experiment uses a neutron generator to impinge a short (~10^-4 s), mono-energetic neutron pulse on a target sample. After the pulse, the neutron population within the sample moderates and reaches thermal equilibrium with a fundamental spatial mode and characteristic decay-time eigenvalue. The eigenvalue can be extracted from the experimental measurements of the neutron flux and used as an integral parameter in validation. For certain materials and geometric configurations, the eigenvalue is heavily influenced by thermal neutron scattering of only the target material. For that reason, a PNDa experiment can have a higher sensitivity to TSLs than is commonly available in critical experiments. Herein, we present results for a series of new PNDa experiments conducted at Lawrence Livermore National Laboratory with plastic materials, e.g. high-density polyethylene and Lucite. We compare the experimental integral parameters to simulated results and report trends in the biases. We evaluate the bias with different Monte Carlo transport codes (MCNP6.2 and Mercury) and show no significant differences between the results of the two codes.

1 Introduction

Thermal neutron scattering laws (TSLs) are important nuclear data in simulations with neutron moderators, for example in thermal fission reactors, criticality safety scenarios, radiation protection, and radiation detection. TSLs account for changes in neutron scattering physics due to molecular binding effects in moderators. Without TSLs, the distribution of neutron energies and angles scattered by atoms in molecules are incorrectly calculated, which can potentially significantly bias simulations. These biases may contaminate analyses and limit their accuracy.

To understand the extent of any possible bias, there has been significant interest in developing benchmarks to validate TSLs. Validation can help assess the quality of nuclear data or determine if a costly re-evaluation is needed. Critical benchmarks are traditionally employed in validation, but they have problems when used for TSLs. Their bias is often dominated by fissile species making it very difficult to distinguish any bias made by the TSL. The presence of multiple materials and nuclear reaction channels in critical benchmarks can obfuscate the performance of any specific nuclear data type and complicate validation. Furthermore, the sensitivity of critical benchmarks to TSL data is limited [1]. Finally, they are expensive to conduct as they require rare materials and high-security/high-hazard facilities.

Pulsed-Neutron Die-Away (PNDa) experiments provide valuable performance data to validate TSLs and offer advantages over critical experiments [2, 3]. PNDa experiments have shown a greater sensitivity to TSL data than is commonly encountered in criticality experiments [4]. In comparison to criticality experiments, relatively few nuclear data (Fig. 1) affect the integral parameter of the PNDa experiment, helping to more easily discover sources of bias. Finally, the PNDa experiments are simpler in terms of geometry and materials, and at lower cost than criticality benchmarks.

Figure 1. Important cross section data affecting the HDPE and Lucite targets in PNDa experiments. From ENDF-B/VI.0, processed with NJOY [5].
For these reasons, Lawrence Livermore National Laboratory and the Naval Nuclear Laboratory are conducting new PNDA experiments [6–8] to validate TSLs. Ultimately, these experiments will be published as international benchmarks for TSL validation. This paper discusses the theory of the PNDA technique, the experimental setup (Fig. 2), the experimental results, and simulations of the experiments. It presents results for two plastic materials: High Density Polyethylene (HDPE) and Lucite, or poly(methyl methacrylate). The simulations use the Monte Carlo codes MCNP6.2 [9] and Mercury [10]. The paper discusses the simulation biases of the two codes. Before the benchmark is finalized, the bias conclusions should be considered as preliminary because many of the experiment’s characteristics (material density and composition, detector effects) have not yet been characterized.

![Example PNDA experimental setup for Lucite.](image)

**Figure 2.** Example PNDA experimental setup for Lucite.

### 2 Theory

The PNDA experiment is often described with one-speed, time-dependent diffusion theory in Eq. (1). This equation assumes thermal, monoenergetic neutrons at speed $v$, and gives the neutron flux $\phi$ at time $t$ and location $r$. The target moderator has a macroscopic absorption cross section $\Sigma_a$ [cm$^{-1}$] and a diffusion coefficient $D$ [cm]. $\Sigma_a$ and $D$ are treated as averaged across thermal energies assuming $1/v$ behavior of the cross sections.

$$\bar{\nu}D \nabla^2 \phi(r, t) - \Sigma_a \phi(r, t) = \frac{\partial \phi(r, t)}{\partial t}$$  \hspace{1cm} (1)

It is assumed that space and time are separable, leading to the general formulation of the initial neutron flux distribution. This is a triply infinite series that represents the expansion of the initial spatial distribution of neutrons in terms of orthogonal functions. The solution is then found with eigenfunctions and eigenvalues that consist of exponentially decaying terms, each with a decay constant.

If the neutron source is shut off and sufficient time is allowed to pass, the expansion can be simplified to only one term: the smallest spatial eigenvalue $B_0^2$. The diffusion equation at $t'$ is then given as Eq. (2). The eigenvalue $B_0^2$ is often called the “buckling” and governs the asymptotic transient behavior of the neutron population.

$$\phi(r, t') = A(r) \exp \left[-(\Sigma_a + \bar{\nu}D B_0^2) t' \right]$$ \hspace{1cm} (2)

The term $\bar{\nu}D$ can be expanded into a Taylor series of the buckling. This allows time-dependent behavior to be represented by Eq. (3), which is governed by a fundamental decay constant $\alpha_0$. The decay constant itself is given by Eq. (4), where $D_0$ [cm$^2$s$^{-1}$] is the asymptotic diffusion coefficient in an infinite medium of the target material and $C$ [cm$^4$s$^{-1}$] is the “cooling coefficient.” The cooling coefficient captures a geometric effect where more high-energy neutrons leak at the system boundaries, which causes the average neutron population to be at a temperature lower than thermodynamic equilibrium.

$$\phi(r, t') = A(r) \exp \left[-\alpha_0 t' \right]$$ \hspace{1cm} (3)

$$\alpha_0 = \bar{\nu}D B_0^2 D_0 - CB_0^4 + \cdots$$ \hspace{1cm} (4)

In this work, cylindrical targets were investigated. For cylinders, the spatial solution of the diffusion equation has a buckling in the form of Eq. (5). Here, $R$ is the radius of the cylinder, $H$ is the height, and $\delta$ is the extrapolation distance. $\delta$ can be calculated as 0.71$\lambda_r$, where $\lambda_r$ is the transport mean free path.

$$B_0^2 = \frac{\left(\frac{2.405}{R + \delta} \right)^2 + \left(\frac{\pi}{H + \delta} \right)^2}{\alpha_0}$$ \hspace{1cm} (5)

Eqs. (4) and (5) show how the experiment can be tuned to emphasize the physics of TSLs. The sensitivity to the absorption cross section or to the TSL is changed by altering a certain geometric dimension of the target, which causes changes in $B_0^2$. Large targets create small $B_0^2$, which causes $\alpha_0$ to depend mostly on absorption ($\Sigma_a$), i.e. not on TSLs. When the target sample is small, $B_0^2$ is large and $\alpha_0$ depends primarily on $D_0$, which itself is highly influenced by the physics of thermal scattering. Fundamentally, the sensitivity of $\alpha_0$ to thermal scattering increases with $B_0^2$, or increases as the target sample size decreases.

The PNDA analysis requires calculating $\alpha_0$, for either experimental or simulated data. The “wait time” method is the most commonly employed for this analysis. First, the neutron counts vs. time data is measured or simulated. Then an exponential is fit to these data to estimate the decay constant of the system after the pulse of neutrons. The fit must only be done after a sufficient amount of wait time so that only the neutron flux in the fundamental mode is used. Otherwise, the higher-order modes will not have sufficiently decayed away to provide a well estimated and reproducible decay constant. The sufficient amount of wait time depends on each sample’s size, as the magnitude of the buckling will determine the rate at which higher-order modes decay away. Larger samples with smaller bucklings require longer wait times. Section 4 provides further discussion of the method used in this work to calculate $\alpha_0$ from the experimental or simulated data.
It should be highlighted that one-speed diffusion theory does not need to be used to interpret the results of PNDA experiments. This theory is only needed to interpret the fitted eigenvalues to obtain diffusion parameters. The fundamental integral parameter, the decay constant of the neutron population, can be interpreted and used entirely without the diffusion theory and its associated assumptions.

3 Experiment

The LLNL experiment uses a D-T neutron generator, a shielding box to reduce room return, four He-3 neutron detectors, counting electronics, and the moderating targets. Fig. 3 provides a rendering of this experiment. The shielding box has 2.45-cm thick walls of 5%-borated HDPE (Shieldwerx™ 201HD) and 0.3175-cm thick cadmium sheets. The box is a neutron moderator and absorber that limits returning neutrons from the facility walls and equipment. The neutron generator was underneath the box and neutrons enter the box and the moderating target through a hole in the box. For the final benchmark, detailed impurity analyses will be taken of the borated HDPE, along with measurements of geometrical uncertainties of the box’s dimensions.

A D-T Thermo Scientific P 383 Neutron Generator creates the neutron pulses. When operating at 90 kV and 50 µA, a neutron output of about \(3 \times 10^8\) n/s is expected with an average neutron energy of 14.1 MeV. The experiment currently employs four LND, Inc. Model 25177 cylindrical He-3 detectors. The detectors are used in conjunction with electronics (combined high-voltage power supply, pre-amplifier, amplifier, and discriminator) from Precision Data Technology (model PDT20A-SHV-12V-C). The PDT operates at 12 V, providing 850 V to the detectors which have a pulse width of 50 ns. The neutron counts are registered with the Automated List Mode Module (ALMM) [11]. The ALMM provides time-tagged information with accuracy to 100 ns. The neutron detectors are connected to the ALMM’s channel input along with the trigger output of the generator itself.

Eleven HDPE targets and eight Lucite targets were measured. The dimensions and weights of each sample were recorded to reproduce them in simulations with MCNP6.2 and Mercury. The samples’ densities were reproduced from these volume and mass measurements. Ultimately, the target sizes are limited by physics and practical constraints. Physics limits the lower limit of the sample size. When the sample size is very small, the neutrons leak from the system more quickly than they can thermalize. This fact makes it very difficult to measure a statistically significant thermal neutron population within the temporal resolution of the detector and electronics, and with the existing background. Large targets are limited by practical concerns of fitting within the shielding box that reduces room return. Furthermore, there are diminishing returns on target size (i.e. sensitivity to absorption) as it becomes larger. Size increases create very small changes in buckling, and therefore \(\alpha_0\), because the buckling is inversely proportional to the target dimensions.

Each target was measured for thirty minutes. The generator was operated at 60 kV and 60 µA using a pulse width of 150 µs for the HDPE targets and 250 µs for the Lucite targets. The generator used varying frequencies and duty factors for each target. Smaller targets were operated at higher frequencies which allowed for more pulses to be generated in the same time window. The duty factor was manipulated to maintain a constant pulse width between measurements. The frequencies ranged from 400 Hz for the largest sample to 2000 Hz for the smallest sample. No dead-time corrections were applied to the counting results. While this is planned for future analysis, it has not yet been performed for two reasons. First, the number of counts registered per neutron pulse is low at 5-100 counts per 0.5 to 2 ms cycle. Only after hundreds of thousands of pulses is the die-away reconstructed. The second is that the data...
rejection needed to estimate the fundamental mode rejects the high-count-rate period when the generator is on. We believe that this limits dead time effects in the detector, but will we support this hypothesis in future work.

4 Results

The measurements were replicated with MCNP6.2 and Mercury simulations. The very simplified models had a mono-directional, 14.1-MeV-point source irradiate target cylinders that were inside the shielding box. This model does not include neutron scattering off the shielding box, detector effects, or potential spectral effects of the neutron source emitted by the generator. These modeling features will be included in the final benchmark evaluation. The model simulated $10^9$ neutrons in a single pulse and replicated the 150 or 250 $\mu$s pulse width of the experiment. It tallied the neutrons counts into bins of 1 $\mu$s, replicating the bin size of the post-processed experimental data.

The Mercury simulations used the same geometry and source term. They were performed with $10^9$ particles and employed path length fluence tallies over the target cylinder for direct comparison to MCNP, although this method does not replicate some of the spatial effects of the experimental detector. There may be more complex tallies that better reproduce the fluence seen directly by the detector in the experiment. Mercury simulations are run with ENDF/B-VIII.0 data processed with the FUDGE code [12], and stored in the Generalized Nuclear Database Structure (GNDS) [13]. These data are different from the ENDF/B-VIII.0 data used in MCNP simulations, which are processed with the LANL NJOY code and stored in ACE files [5]. The simulations assumed that the temperature of every sample was 300 K, although the experimental room was at an average of 291-292 K.

![Figure 5. Geometry in an example Mercury simulation.](image)

Each die-away curve needs a specific time interval for its fit, or else the $\alpha_0$ integral parameter may be over or underestimated. It is possible to underestimate $\alpha_0$ by choosing 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 underestimate or have excessively noisy $\alpha_0$ values by including data that are too late in the curve where room return is significant. Moreover, we have observed that the standard deviation of $\alpha_0$ is particularly sensitive to the time interval of the fit, with incorrect intervals creating up to 5% overestimated uncertainties.

Our fitting approach is based on observed behavior: too early time bins create overestimated $\alpha_0$ values and too late time bins create noisy $\alpha_0$ values. Importantly, when using data in the fundamental mode, the $\alpha_0$ values are consistent (within fitting uncertainty) despite varying time intervals. Fig. 6 describes this behavior for an example MCNP simulation. Over a given time interval, e.g. 300 $\mu$s, the start point of the fit slid and the fit is performed for each new section of 300 $\mu$s. An iterative and expert-based analysis chose the 300 $\mu$s interval, and this interval was applied for every target. For a given die-away curve, the data region where plateaued $\alpha_0$ values occurred was chosen and the mean and standard deviation of the $\alpha_0$ values across the plateau is reported. The population standard deviation of the $\alpha_0$ values is propagated along with the fit uncertainty for the final reported uncertainties. This process is repeated for each target’s die-away curve, for both simulated and experimental data.

![Figure 6. Simulated die-away curve (black) and varying fitted $\alpha_0$ values. The solid red line is the mean across the plateau and the dotted red line is the ±1 standard deviation.](image)

4.1 High Density Polyethylene

Fig. 7 presents the measured die-away curves for the HDPE cylindrical targets. The legend gives each cylinder’s radius. The fundamental measurement is time-tagged neutron counts in the detectors. These counts are aligned with the time tags of the generator pulses to reconstruct a single die-away curve. This post-processing requires a defined time bin size, which here was set to 1 $\mu$s. Fig. 8 provides the measured decay constants from the LLNL experiment and by Sjöstrand et al. [14]. This comparison to previous measurements provides confidence in the experimental setup.

Fig. 7 shows behavior that is typical of PNDA experiments. The target size dictates the slopes of the die-away curves: larger samples have thermal neutron populations
Figure 7. Experimental and simulated (MCNP6.2) die-away curves for the eleven HDPE targets using ENDF/B-VIII.0 data. The legend gives the radius of the associated HDPE sample.

Table 1. HDPE target parameters, experimental decay constants, and simulated decay constants with MCNP6.2 and Mercury.

<table>
<thead>
<tr>
<th>Radius (cm)</th>
<th>Height (cm)</th>
<th>Buckling (cm$^{-2}$)</th>
<th>Decay Constant (ms$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Experiment</td>
<td>MCNP6.2</td>
<td>Mercury</td>
</tr>
<tr>
<td>2.62</td>
<td>5.16</td>
<td>0.9246</td>
<td>28.472 ± 0.152</td>
</tr>
<tr>
<td>3.89</td>
<td>7.70</td>
<td>0.4547</td>
<td>17.449 ± 0.043</td>
</tr>
<tr>
<td>5.32</td>
<td>10.87</td>
<td>0.2504</td>
<td>12.392 ± 0.010</td>
</tr>
<tr>
<td>6.55</td>
<td>13.18</td>
<td>0.1710</td>
<td>10.224 ± 0.007</td>
</tr>
<tr>
<td>7.78</td>
<td>15.56</td>
<td>0.1238</td>
<td>9.200 ± 0.017</td>
</tr>
<tr>
<td>9.05</td>
<td>18.26</td>
<td>0.0922</td>
<td>8.273 ± 0.010</td>
</tr>
<tr>
<td>10.44</td>
<td>21.03</td>
<td>0.0701</td>
<td>7.846 ± 0.011</td>
</tr>
<tr>
<td>11.71</td>
<td>22.94</td>
<td>0.0571</td>
<td>7.492 ± 0.006</td>
</tr>
<tr>
<td>12.86</td>
<td>25.48</td>
<td>0.0473</td>
<td>7.250 ± 0.008</td>
</tr>
<tr>
<td>14.61</td>
<td>28.02</td>
<td>0.0377</td>
<td>7.075 ± 0.006</td>
</tr>
</tbody>
</table>

that decay away slower than small samples. Larger samples also have slower decay of higher-order modes. In the fundamental mode, the die-away curve should exhibit pure exponential behavior. Right after the pulse, the large samples show more significant non-exponential behavior. The shielding box does not eliminate the background from room return, but has shown to significantly reduce it [7].

Fig. 9 gives the biases of simulations with both MCNP and Mercury for each HDPE target. The general trend is that the bias increases as the sample size decreases (as buckling increases). Mercury and MCNP show very similar trends in their biases. The average absolute difference in their bias was 0.3% and the maximum difference was 1.1% for the largest target.

The general conclusions on the bias are also not very reliable. The unreliability is caused by the lack of characterization of the HDPE samples. For instance, Fig. 4 shows clear color differences (yellow tint) between the HDPE targets, indicating likely material differences. These differences need to be modeled in the material compositions and densities of the targets, and were not modeled in this work. These properties will be fully characterized for the final benchmark, which will help to improve the confidence in bias analyses.
### 4.2 Lucite

Table 2 gives the target characteristics and fitted experimental and calculated decay constants for the Lucite experiments. The simulations used a Lucite density that was derived from the mass of the sample and the measured radius/height, assuming the sample was a perfect cylinder. In future experiments, more targets will be added to give more data points at intermediate to high bucklings. These additional measurements will help to better understand and attribute the bias of the simulations.

Fig. 10 gives the fitted experimental decay constants and compares them to values in the literature from Drozdowicz et al. [15], Drozdowicz & Woznicka [16], and Krynicka et al. [17]. The figure shows strong agreement between the low buckling/large volume targets, but increasing discrepancies as the buckling increases/target volume decreases. At large bucklings, the LLNL experiments agree better with Drozdowicz & Woznicka. Ignoring possible errors in the experimental setup, a major source of potential discrepancies may come from different target material characteristics. These differences include the composition and density of the target, which are likely to be more important when the target size is smaller.

Fig. 11 provides the biases of the MCNP and Mercury simulations for the Lucite targets. The biases of each code show similar trends with buckling changes: bias decreasing until a buckling of ~0.1 cm$^{-2}$ and then increasing biases as the target size decreases. There is an offset between the MCNP and Mercury simulations which has not yet been explained. Without a total uncertainty quantification of the model – due to uncertainties in the mass, composition, and geometry of the target – it cannot yet be said if these differences are statistically significant. The reported uncertainties only include those arising from the exponential fit, and not from the systems’ modeling.

### 5 Conclusions

This document presented new PNDA experiments and simulations that will serve to validate the TSL data of HDPE and Lucite. The experiments showed good agreement with other works in the literature that investigated...
the same materials. Simulations with MCNP and
Mercury showed trends in the bias that correlated with the in-
tegral parameter’s sensitivity to TSLs. As the target size
becomes smaller and the sensitivity to TSLs increases, the
bias increased as well. The biases between MCNP and
Mercury agreed well, showing no significant differences
between the two codes.

Future work will include adding more targets of dif-
ferent sizes to expand the experimental data set. To create
a high-quality benchmark will also require characteriza-
tion measurements of the experimental setup. These de-
tails will be added to the models of the experiment to help
eliminate any sources of bias that may originate from mod-
eling. These modeling errors may be incorrectly attributed
to the nuclear data. With accurate characterization, an un-
certainty quantification of the experimental models can be
done. This uncertainty quantification will help to better
analyze the source of bias. The future characterization
measurements include geometry, composition, and den-
sity of the targets and shielding box. Other simulation im-
provements will also include high-fidelity modeling of the
neutron generator and He-3 detectors.

References

(2022)
146, 13004 (2017)
247, 09016 (2021)
Nuclear Science and Engineering pp. 1–11 (2022)
[6] D. Siefman et al., Lawrence Livermore National
Laboratory LLNL-TR-820718 (2021)
Livermore National Laboratory LLNL-TR-833263
(2022)
[8] D. Siefman, W. Zwyiec, C. Percher, D. Heinrichs,
Transactions of American Nuclear Society (2022)
(2016)
[10] P. Brantley, M. McKinley, Mercury web site
(2022), https://wci.llnl.gov/simulation/computer-
codes/mercury/
UR-17-29472 (2017)
Livermore National Laboratory LLNL-TR-828141
(2021)
[13] G. Gert et al., Lawrence Livermore National Labora-
tory LLNL-CONF-828782 (2022)
Atomenergi AE-12 (1960)
[15] K. Drozdzowicz et al., Nuclear Instruments and Meth-
ods 178, 513 (1980)
[17] E. Krynicka et al., Nuclear Instruments and Meth-
ods in Physics Research Section B: Beam Interac-
tions with Materials and Atoms 251, 19 (2006)