Characterization of a Liquid Ammonia Moderator

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Abstract.

Liquid ammonia is an attractive intermediate temperature moderating material, offering a high hydrogen density, low tendency toward radiation-initiated polymerization, and broad accessible temperature range. The LENS collaboration has characterized decoupled poisoned liquid ammonia moderators in an attempt to validate scattering kernels we are generating. Additionally, we have measured a borated water moderator in order to test a model for predicting detailed moderator performance at one level of intrinsic homogeneous neutron capture based on measurements at other levels of such capture. We describe both sets of measurements, as well as use them to illustrate some of the reproducibility challenges associated with moderator characterization measurements on compact neutron sources.

1. Introduction

Liquid ammonia (NH₃) is an attractive intermediate temperature moderating material, offering a high hydrogen density, low tendency toward radiation-initiated polymerization, and broad accessible temperature range relative to other intermediate temperature moderators. [1, 2, 3] Solid ammonia offers the same advantages at a broad range of cryogenic temperatures. Both forms offer many low-energy rotational modes providing effective neutron energy loss. A complication for the use of ammonia as either an intermediate temperature moderator or as a cold moderator is the significant neutron capture cross section for natural nitrogen; more than five times that of hydrogen on a per atom basis, nearly tripling the macroscopic capture cross section for the molecule. Ammonia enriched to high levels of ¹⁵N would avoid this additional capture, while retaining similar neutron scattering characteristics. Isotopically enriched ammonia would be practical for use at a production facility if merited, although it would likely require a recovery and re-use system.

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At the Spallation Neutron Source facility at Oak Ridge National Laboratory (SNS), we are considering the replacement of the ambient water moderator with one composed of liquid ammonia. Various previous studies concerning ammonia moderators have concerned solid ammonia at relatively low temperature. [1, 2] This is not our interest here: the ambient water moderator currently in use at SNS [4, 5] serves instruments that would benefit from an intermediate temperature spectrum, but not necessarily from a low temperature spectrum, and the power densities deposited in the moderator systems are not easily handled with a solid moderator material. In order to predict the practical performance of a liquid ammonia moderator, and thus evaluate its suitability for our use, we require two things: a validated scattering kernel with which to predict neutron thermalization, and a method for predicting the performance of a moderator of $^{15}$N-ammonia based on characterization and validation measurements performed with $^{14}$N-ammonia. Our group is in the process of measuring the inelastic and total neutron scattering from ammonia at various temperatures in order to generate scattering kernels. These efforts will be reported elsewhere. Here we describe integral measurements of prototypical liquid ammonia moderator performance intended to validate those scattering kernels. We have characterized ammonia moderators at temperatures of 220 K (liquid) and 185 K (just below the freezing point); while the solid ammonia measurements are not directly relevant for our desired application, they provide additional points of comparison for measurements reported in the literature (which were performed at 77 K for differently sized moderators), [1, 2] and were practical with relatively little additional effort. On a separate occasion, we performed comparable measurements of room temperature water and various concentrations of boric acid, with which we hope to test a model for extrapolating from measurements of natural ammonia to performance of $^{15}$N-ammonia with its lower capture rate. The details of those measurements, and the testing of that extrapolation model, will be reported elsewhere—however, the nature of the two experimental campaigns provided an opportunity to perform measurements on what should nominally have been the same moderator (distilled water in identical geometries). The comparison of these measurements provides insight into challenges for consistency and reproducibility in moderator characterization.

2. Measurements Performed

We have measured neutron wavelength spectra and wavelength-dependent emission time distributions (pulse shapes) from moderators composed of natural ammonia at 220 K and 185 K (liquid and solid, respectively). Measurements at these temperatures were repeated for moderators of size 100 mm by 100 mm by 25 mm, and size 100 mm by 100 mm by 15 mm. For each moderator vessel, the measurements were repeated with room temperature distilled water to provide a relative control. These control experiments were accomplished by removing the ammonia from the moderator vessel and replacing it with water, without removing or disturbing any physical components. Details of the LENS moderator test facility, our moderator performance characterization equipment, and our data analysis methods can be found elsewhere, [6, 7, 8, 9] and will not be repeated here. All measurements described were performed with cadmium decoupled moderators, using accelerator pulse widths of 13.5 $\mu$s.

The detailed measurements we have performed provide integral quantities we will use as a first level of comparison to calculations based on scattering kernels currently under development. Ammonia is a thermalizing moderator; robust and convenient integral metrics include the spectral temperature of the emitted neutron beams, the fundamental decay constant of the long-wavelength emission time distributions, and (somewhat less integral in nature) the wavelength-dependent metrics for emission time distribution width; mean emission time, full-width half-maximum (FWHM), and root-mean-square (RMS) emission time.

The spectral temperature $T_n$ of the neutron beam is estimated by fitting a linearized efficiency-weighted Maxwell-Boltzmann distribution in the range of 1–3 Å (beyond 3 Å the spectra depart
from the Maxwell-Boltzmann distribution). The fundamental decay time is estimated as the average of the decay times obtained by fitting the appropriate portion of each long-wavelength emission time distribution to an exponential decay. Mean emission times, RMS emission times, and FWHM were determined after subtraction of a linear background, and the RMS emission times $\sigma_t$ and FWHM $\delta t$ corrected for finite rectangular source pulse width $\Delta t_s \approx 13.5 \mu s$ and data acquisition channel width $\Delta t_c = 2 \mu s$ as

$$\sigma_t^2 = \sigma_m^2 - \frac{1}{12} \left( (\Delta t_s)^2 + (\Delta t_c)^2 \right)$$  \hspace{1cm} (1)

and

$$\delta_t^2 = \delta_m^2 - \frac{(1.96)^2}{12} \left( (\Delta t_s)^2 + (\Delta t_c)^2 \right).$$  \hspace{1cm} (2)

While these corrections are approximate, and rely on a square primary source pulse and a particular form for the emission time distribution, they are reasonable—perhaps more importantly, they are the same corrections used in earlier studies [2] and are therefore justified absent significantly better alternatives.

3. Results

Wavelength spectra measured for the 25 mm moderator are shown in Figure 1, and for the 15 mm moderator in Figure 2.

Emission time distributions at a single wavelength for a single moderator geometry are shown in Figure 3. Similar distributions were measured at other wavelengths, and for both moderator geometries, but are not shown here. The apparent fundamental decay time $\tau_1$ can be seen in Figure 3 between 50–100 $\mu s$. A second, longer-lived decay time $\tau_2$ can be seen beyond 150 $\mu s$, and is assumed to represent leakage from the reflector through an imperfect cadmium decoupler.

Spectra taken from the distilled water control moderators in both experimental campaigns (using the same moderator vessels, but with significant reassembly of the moderator-reflector system) are shown in Figures 4 and 5.

Numerical values for various metrics are shown in Table 1. The apparent total flight path length (to the emission time analyzer detector) $\bar{L}_T$ is determined by fitting the arrival time of the analyzed pulses. Since the analyzing crystal and the detector do not move between measurements, the variation in that fitted length can be assumed to reflect changes in the moderator position along the beamline upon cooling and reassembly. We make the following observations:
Figure 3. Emission time distributions measured at 1.35 Å for 25 mm thick moderators of water and ammonia.

Figure 4. Neutron wavelength spectra measured for a 25 mm thick water moderator on two separate occasions.

Figure 5. Neutron wavelength spectra measured for a 15 mm thick water moderator on two separate occasions.

Table 1. Measured parameters for ammonia and water moderators. All moderators were 100 mm by 100 mm transverse to the viewing direction.

| Material | Temperature (K) | Depth (mm) | $T_n$ (K) | $\tau_1$ (µs) | $\tau_2$ (µs) | FWHM (1.08 Å) | $\tilde{L}_T$ (m) |
|----------|-----------------|------------|-----------|---------------|---------------|---------------|-----------------|
| NH$_3$   | 220             | 25         | 280       | 18            | 68            | 14            | 9.2625          |
| NH$_3$   | 185             | 25         | 253       | 20            | 78            | 12            | 9.2625          |
| H$_2$O   | 302             | 25         | 357       | 21            | 108           | 23            | 9.2661          |
| NH$_3$   | 220             | 15         | 294       | 13            | 141           | 14            | 9.2755          |
| NH$_3$   | 185             | 15         | 270       | 14            | 139           | 9             | 9.2755          |
| H$_2$O   | 302             | 15         | 346       | 18            | 165           | 14            | 9.2786          |

- The intensity at the shortest wavelengths is slightly smaller for an ammonia moderator than for a water moderator of the same dimensions. This is as expected for the slab moderator configuration at LENS; the higher hydrogen density means that the viewed moderator surface is neutronically farther from the primary neutron source.
- The spectral temperature for the ammonia moderators is significantly higher than the physical temperature (20–50%), as might be expected for small moderators (also the case for water) and high capture cross section (specific to ammonia).
• The ratio between the spectral temperature and the physical temperature is larger for the solid ammonia than for the liquid ammonia, consistent with the loss of thermalizing efficiency associated with recoil.

• The moderator vessel appears to shift approximately 3–4 mm upon cooling from 300 K to 200 K. This is not unreasonable. The viewed face of the moderator vessel appears to have moved some 12.4 mm upon removal and reinsertion. While we would expect a movement of 5 mm (due to the different size of the moderator vessel), the additional 7.4 mm motion provides an indication of the reproducibility of the moderator position.

• There is a long-lived tail on the emission time distributions for all moderators tested. It is significant in both length (i.e., the large decay time) and magnitude. It appears to vary quite a bit, possibly in response to the apparent shift in moderator position. We believe that it is due to in-leakage from the reflector through a geometrically imperfect cadmium decoupler. While its effects can be dealt with in assessing emission time distributions, we are concerned that it may significantly perturb apparent spectral temperatures and thermalization ratios.

• The reproducibility of the absolute intensity of the water moderator spectra from the two different campaigns is not as good as we would like. While the 25 mm data shown in Figure 4 match at short wavelengths as we might expect, the 15 mm data indicate a gain of 15% for the subsequent measurement. Even more troubling than this absolute change is the fact that the thermalization ratio changed for both geometries: a loss of 15% for the 25-mm geometry and 30% for the 15-mm geometry. Finally, the spectral temperature appears to have changed for the 15 mm case, being nearly 10 K lower in the second measurement. These discrepancies will be addressed further below.

Our ammonia moderator measurements, if taken at face value, provide the integral quantities, shown in Table 1, needed to validate scattering kernels used in moderator simulations. However, the consistency check performed on nominally identical water moderators are not satisfactory at this stage of data analysis. We have identified the following hypotheses for further consideration:

• The apparent shift in moderator position upon reassembly could result in a significant change in the target-to-moderator distance. We plan to perform a Monte Carlo sensitivity study. This alone would not explain changes in spectral temperature nor thermalization ratio.

• The apparent shift in moderator position upon reassembly could also result in different geometric imperfections in the cadmium decoupling. We will examine emission time distributions to see if consistent effects are seen there, and assess decoupler gaps in Monte Carlo studies.

• The second round of measurements (ammonia and water) took place a year after the first round (water and boric acid in water). The aluminum moderator vessels could have been contaminated with boric acid, causing unpredictable effects on subsequent moderator measurements. We are considering repeating the ammonia measurements with fresh moderator vessels built to the same specifications.

• The process we used to fill the moderator vessels with water or boric acid solution without removing them from the target-moderator-reflector assembly did not provide positive assurance that the material in question all made it to the desired location. We are considering various options for checking the moderator fill level remotely for future measurements.

• The absolute normalization of the spectra is done to delivered proton charge. Unfortunately, the proton beam diagnostics are not particularly well suited to the short-pulse low power operational mode we use for moderator characterization, and variations in beam position...
and energy could result in significant changes in primary neutron production. We have implemented a separate neutron detector system within the LENS reflector assembly in order to track primary neutron production independently from proton beam delivery.

- The absolute normalization of the spectra further depends on details of the data acquisition system, which was changed between these two measurements (the changes should affect only the proton normalization, not the neutron measurements). We are currently characterizing the data produced with both systems in order to assess differences in potential systematic errors between the two campaigns.

4. Summary
We have measured integral performance parameters of small decoupled ammonia moderators pertinent to scattering kernel validation. In the process of performing these measurements, we have seen inconsistent performance of water moderators used as control samples. The inconsistencies raise questions as to the reliability of these measurements, and thus offer insight into the qualities necessary for robust and reliable moderator characterization measurements:

- Consistent moderator positioning within the reflector assembly.
- Consistent and complete decoupling from the reflector assembly.
- Consistent and monitored moderator fill level.
- Consistent and monitored primary neutron source production.
- Robust moderator vessels immune to the effects of tested moderator materials, and/or characterization of the vessels themselves.
- A well-understood and self-consistent data acquisition system.

We expect that we will repeat this series of moderator characterization measurements, both to test our ideas for improving reliability and reproducibility and to verify that the data we have taken for ammonia are suitable for scattering kernel validation.

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