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Toward a new polyethylene scattering law determined using inelastic neutron scattering

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ABSTRACT

Monte Carlo neutron transport codes such as MCNP rely on accurate data for nuclear physics cross-sections to produce accurate results. At low energy, this takes the form of scattering laws based on the dynamic structure factor, $S(Q,E)$. High density polyethylene (HDPE) is frequently employed as a neutron moderator at both high and low temperatures, however the only cross-sections available are for ambient temperatures ($\sim$300 K), and the evaluation has not been updated in quite some time. In this paper we describe inelastic neutron scattering measurements on HDPE at 5 and 294 K which are used to improve the scattering law for HDPE. We review some of the past HDPE scattering laws, describe the experimental methods, and compare computations using these models to the measured $S(Q,E)$. The total cross-section is compared to available data, and the treatment of the carbon secondary scatterer as a free gas is assessed. We also discuss the use of the measurement itself as a scattering law via the one phonon approximation. We show that a scattering law computed using a more detailed model for the Generalized Density of States (GDOS) compares more favorably to this experiment, suggesting that inelastic neutron scattering can play an important role in both the development and validation of new scattering laws for Monte Carlo work.

1. Introduction

Moderated neutron spectra are often calculated using Monte Carlo techniques, such as those contained within the MCNP code [1]. The computation of leakage spectra and emission time distributions from High Density Polyethylene (HDPE) is of particular interest to a variety of disciplines, ranging from thermal and cold neutron moderators [2–5], neutronic assemblies [6], shielding, multiplying assemblies [7], and moderated detector design [8,9].

The accuracy of a Monte Carlo calculation depends in large part on the fidelity of the underlying nuclear physics data employed by the code as well as the accuracy of geometry. At low energy, this requires sampling the double differential cross-section many times over the history of the neutron's random walk through the polyethylene (PE) layer. The double differential cross-section is [10,11]

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b}{4\pi}\frac{k_f}{k_i}S(Q,E) \tag{1}$$

where σ_b is the scattering cross-section summed over all the nuclei and k represents the initial and final momentum of the neutron. For polyethylene, σ_b is dominated by the large incoherent scattering cross-section for hydrogen (20.48 b). The $S(Q,E)$ function describes the structure and collective dynamics of the medium. When written using dimensionless variables [12], $S(Q,E)$ is referred to as the "scattering law" for the material. The momentum transfer, Q, and energy transfer, E, between the initial and final state of the neutron are defined as

$$\begin{aligned} Q &= |\mathbf{k_i} - \mathbf{k_f}| \\ E &= E_i - E_f \end{aligned} \tag{2}$$

where for the neutron mass m the free neutron dispersion is

$$E = \frac{\hbar^2 Q^2}{2m}. \tag{3}$$

MCNP computations involving ambient temperature HDPE ($\sim$300 K) make use of a model for $S(Q,E)$ applied to neutron scattering below 4 eV. This scattering law has been ported forward [13,14] from the original, detailed in the 1969 report ENDF-269 [15], with evaluations at 296 and 350 K [1]. Accurate computations for different HDPE temperatures, such as are needed for cold moderators, would require a new scattering law to be produced. The usual procedure is to use the NJOY code [16], which computes the cross-section in the incoherent approximation.

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Incoherent inelastic neutron scattering defines the scattering in terms of a generalized density of states (GDOS), which represents the number of harmonic phonon excitations per unit energy. It is therefore essential to employ an accurate representation of the GDOS to calculate the scattering law.

There are several different models for the HDPE scattering law presented in the literature [15,17,18]. These models are generally benchmarked against the rather limited set of measurements found in Ref. [15], which include the total cross-section measurement by Armstrong [19] and a suite of inelastic measurements of the double differential cross-section. The total cross-section measurement should probably be superseded by the more recent one by Granada et al. [20]. The inelastic measurements are on an absolute scale, but are of low energy resolution and cover a limited range of energy transfer about the $\sim$200 meV incident neutron energy. All comparisons have been made at ambient temperatures, which is not a sufficient test of a scattering law intended for use at low temperature, such as was done in Ref. [17]. The correction to the low energy portion of the scattering law suggested by Ref. [18] made comparisons to low temperature specific heat data as a test of the GDOS, a practice which also does not provide a sensitive check of the details of the model.

Inelastic neutron scattering (INS), on the other hand, can provide an efficient means with which to examine the HDPE scattering law over a broad range of incident neutron energies and sample temperatures [21]. The main region of interest for the computation of thermal and cold neutron moderator spectra conventionally extends from 0.1 meV to 4 eV, which is well within the reach of a modern neutron spectrometer. INS measurements often lack an absolute scale, which should not be detrimental because the overall normalization of the calculated $S(Q,E)$ is provided by the bound atom cross-section and by the sum rules for $S(Q,E)$ [10].

This paper will review the results of an inelastic scattering experiment on HDPE performed using the wide angular range chopper spectrometer (ARCS) [22,23] at the Spallation Neutron Source (SNS) [24]. Our goal is to compose a HDPE GDOS which is consistent with the primary experimental data for 5 and 294 K. The analysis approach will be to compare several theoretical GDOS models to the experimental data in the presence of instrumental resolution.

The models for the GDOS are chosen from a survey of the history of the current scattering laws and of the general literature. Throughout the remainder of the text, the models will be referred to using the acronyms identified by the bold letters

- **ENDF**: The current ENDF evaluation, with GDOS taken from the details presented in Ref. [15]. The ENDF HDPE file comments [25] indicate it is derived from a treatment of an isolated molecular chain [26] and employs a Debye model below 20 meV [27].
- **KM**: A composite model we have created using the GDOS presented by Lynch [28] for the high energy phonons, and the work of Kitagawa and Miazawa [29] at low energy. The KM work was generated in the same period as the ENDF law, but had moved beyond the isolated molecule treatment. Our model is derived by replacing the region below 71 meV in Ref. [28] by an equal area taken from the computation in Ref. [29] to describe the inter-chain modes. Data are extracted from digitized graphs.
- **ST/LH**: A composite model by Hill and Liu [17] using the GDOS correction suggested by Swaminathan and Tewari [18] for the low energy phonons, and the ENDF evaluation for the high energy phonons. ST/LH employs a different low energy density of states than is seen in KM. This is the most recent scattering law available in the literature. Data are extracted from digitized graphs.
- **Barrera**: A recent density functional theory (DFT) calculation for the inelastic scattering from HDPE [30]. This work represents an *ab initio* model considering the entire orthorhombic cell, without making a distinction between the intra- and inter- chain excitations, and without fitting parameters to measurements. The one phonon data presented at 20 K are extracted from the graph and inverted to obtain the density of states. Barrera does not show the relatively dispersionless C–H stretching modes, so we have modeled this feature as single Gaussian mode at 360 meV energy transfer with a FWHM of 10 meV and total area fraction of 0.31. This choice of area for the peak provides agreement with the measured value of the mean square displacement ($\overline{u^2}$).
- **ARCS**: The measured data is used in the one phonon approximation to the GDOS to examine whether the measurement itself can or should be used in lieu of a detailed calculation.

The paper is structured as follows. Section 2 reviews the basic theory behind the scattering law and gives an overview of the HDPE dynamics relevant to our measurements. Section 3 describes the samples and ARCS instrument, and provides a detailed discussion of the instrumental resolution. Section 4 describes the experimental results and analysis methods for elastic and inelastic scattering, and obtains the measured GDOS in the one-phonon approximation. Section 5 makes detailed comparisons between the $S(Q,E)$ calculated from each model, showing the KM and Barrera-based models provide the most accurate results. Results of the models are compared to the total cross-section at 294 K [31] before we conclude the analysis in Section 6.

2. Neutron scattering from HDPE

2.1. Incoherent neutron scattering

Neutron scattering from HDPE is dominated by the large incoherent scattering cross-section for an ensemble of H nuclei [10]. We will describe our inelastic $S(Q,E)$ observations by considering the incoherent scattering *only*, a practice referred to as the incoherent approximation. We expect this description to be appropriate for HDPE, however in practice the same treatment is also used to develop cross-sections for strong coherent scatterers such as graphite [32].

The general reasoning to apply this treatment in all cases, besides its relative simplicity, is that even if the material was a strong coherent scatterer, the multi-phonon terms would spread out the scattering in (Q,E) space to such a strong degree that the coherence would not be recognizable [10]. Also, it is surmised that multiple scattering events within the layer would blur out any fine details resulting from well defined dispersion relations.

Strongly coherent scatters now find wide use in moderated neutronic systems, such as cold moderators and ultra-cold neutron converters. Solid deuterium [73,33,74,34] and solid oxygen [35] are strong coherent scatterers used for the production of cold and ultra-cold neutrons (UCN, $\sim$100 neV). In these systems, the cryogenic solid can serve as both cold moderator and/or UCN converter.

The arguments in favor of the incoherent approximation become strained at low temperature and low energy for a number of reasons. For example, the sampling of just one or two scattering events from a thin layer is a possibility. The consideration of coherent scattering in thin layers is also important for simulations which include the moderator vessel's external structure. Recent