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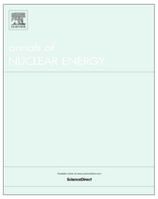

Thermal scattering law of $(C_2H_4)_n$: Integrating experimental data with DFT calculations

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ABSTRACT

Improvements in determination of the thermal scattering law of moderator materials (measuring, calculating and validating) are important for accurate prediction of neutron thermalization in nuclear systems. In this work a methodology for producing thermal scattering libraries from the experimental data for polyethylene $(C_2H_4)_n$ is discussed. Double differential scattering cross section (DDSCS) experiments were performed at the Spallation Neutron Source of Oak Ridge National Laboratory (SNS ORNL). New scattering kernel evaluations, based on phonon spectrum for $(C_2H_4)_n$, are created using the NJOY2016 code. Two different methods were used: direct and indirect geometry neutron scattering at ARCS and SEQUOIA, and VISION instruments, respectively, where the phonon spectrum was derived from the dynamical structure factor $S(Q,\omega)$ obtained from the measured DDSCS. In order to compare and validate the newly created library, the experimental setup was simulated using MCNP6.1. Compared with the current ENDF/B-VII.1, the resulting RPI $(C_2H_4)_n$ libraries improved both double differential scattering and total scattering cross sections. A set of criticality benchmarks containing $(C_2H_4)_n$ from HEU-MET-THERM resulted in an overall improved calculation of K_{eff}, although the libraries should be tested against benchmarks more sensitive to $(C_2H_4)_n$. The DFT + oClimax method is used and is shown to be most comprehensive method for analysis of moderator materials. The importance of DFT + oClimax method lies in the fact that it can be validated against all data measured at VISION, ARCS and SEQUOIA, and experimental total scattering cross section measurements.

1. Introduction

As the accuracy of simulations advances in many areas of nuclear science, code packages such as the Monte Carlo N-Particle code (MCNP), Goorley et al. (2016), are highly dependent on the accuracy of current Evaluated Nuclear Data Files (for example ENDF/B-VII.1), Chadwick et al. (2996). These evaluated libraries contain different nuclear reaction data, and most relevant for this work, these libraries contain thermal neutron scattering cross sections. Evaluations are widely used in neutron transport codes, and in this work we used MCNP 6.1. Due to the lack of the experimental data in the thermal region, it is difficult to validate the simulations against actual measured quantities. For the most current evaluations the only available data for validation and benchmarking are total cross section measurements.

For most moderators, the current ENDF/B-VII.1 libraries were created using a theoretical phonon spectrum (or density of states). The ENDF/B-VII.1 library for polyethylene was created by Koppel, Houston and Sprevak in 1969 and was converted to ENDF 6 format in 1989 at Los Alamos National Lab. In 2016 new polyethylene library, using molecular dynamics to calculate the phonon spectrum, created by North Carolina State University Nuclear Reactor Program has been added to ENDF as ENDF/B-VIII.b5 library. These libraries were created to correctly reproduce the energy dependent total neutron scattering cross section, with less attention to double differential scattering cross sections (DDSCS). Little to no DDSCS experimental data exist, and they were rarely used in the evaluations. In Kirouac et al. (1966) performed DDSCS measurements on polyethylene but due to the high incident energies, 830 meV and above, the data was not sufficient to be used for the validation or creation of new thermal scattering libraries. The goal of this work was to obtain new experimental DDSCS data, use it to derive an experimental phonon spectrum, and establish a streamlined

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methodology for the creation of new thermal scattering libraries for different moderator materials. The importance of this work lies in the fact that it is an review of thermal neutron scattering for polyethylene from experimental and simulation aspects. This work includes a method of combining both experiments and simulations to guide the creation of new thermal library. Additionally, using the methodology developed here, thermal scattering libraries for other nuclear materials (e.g., lucite, alpha-quartz, and ice-1 h) have been created (Ramić et al., 2018). The need for this work becomes self explanatory when one looks at the results from the Section 3.1.1 for $(C_2H_4)_n$ DDSCS and total cross section comparison. Polyethylene in ENDF/B-VII.1 evaluation is one of the better and more consistent evaluations.

2. Thermal neutron scattering theory

There are two parts to neutron scattering: incoherent and coherent. If we imagine a neutron beam incident on a system of multiple constituent particles, the incoherent part would represent the sum of the effects created by waves that do not interfere with each other, while the coherent part is represented by the sum of effects of waves that do interfere with each other, MacFarlane and Kahler (2010). Both parts include elastic and inelastic scattering components. For elastic scattering, there is no change in the neutron energy, while in the inelastic scattering, incident neutron energy can be up-scattered (increase in neutron energy), or down-scattered (loss of energy). The incoherent approximation for the coherently scattering polycrystalline material, where the inelastic neutron scattering (INS) data from randomly oriented crystallites is averaged, is valid when the volume of the reciprocal space covered by the INS experiment is much larger than the volume of the Brillouin zone of the crystal.

The DDSCS represents the number of neutrons scattering into a solid angle, subtended by a detector onto the sample, $d\Omega$, at energy transfer, $d\hbar\omega$. For mono-atomic sample, as seen in Ramirez-Cuesta (2004), in incoherent approximation DDSCS can be represented as:

$$\frac{d^2\sigma}{d\Omega d\hbar\omega} = \frac{\sigma_b}{4\pi}\frac{\mathbf{k}'}{\mathbf{k}}S(Q,\omega), \tag{1}$$

where $\mathbf{k}'$ and $\mathbf{k}$ are wavevectors of final and initial neutron states respectively, $\hbar Q$ is the momentum transfer, σ_b represents bound scattering cross section, $S(Q,\omega)$ is the scattering law.

In NJOY2016, the code used for creating of ENDF thermal scattering libraries, DDSCS in incoherent approximation, is represented as MacFarlane and Kahler (2010):

$$\frac{d^2\sigma}{d\mu dE} = \frac{\sigma_b}{2k_BT}\sqrt{\frac{E'}{E}}S(\alpha,\beta), \tag{2}$$

where E and E' are the incident and scattered neutron energies respectively, μ is the cosine of the scattering angle, k_BT is the equilibrium temperature in eV (25 meV at 293.6 K), and $S(\alpha,\beta)$ is the scattering law. The relationship between $S(Q,\omega)$ and $S(\alpha,\beta)$ is given as:

$$S(Q,\omega) = \frac{\hbar S[\alpha(Q),\beta(\omega)]}{k_BT}, \quad \alpha = \frac{\hbar^2Q^2}{2Am_nk_BT}, \quad \beta = \frac{\hbar\omega}{k_BT}, \tag{3}$$

where m_n is the mass of neutron, A is the ratio of the mass of the scattering atom to the neutron mass. The relationship between wavevector $\mathbf{k}$ and energy of neutron is:

$$E = \frac{\hbar^2k^2}{2m_n}, \tag{4}$$

α and β are related respectively to the momentum transfer and energy transfer:

$$\alpha = \frac{E' + E - 2\mu\sqrt{E'E}}{Ak_BT}, \beta = \frac{E' - E}{k_BT}. \tag{5}$$

In the incoherent and Gaussian approximation, MacFarlane and Kahler (2010), the $S(\alpha,\beta)$ is:

$$S(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\beta\hat{t}}e^{-\gamma(\hat{t})}d\hat{t}, \tag{6}$$

where:

$$e^{\gamma(\hat{t})} = \alpha\int_{-\infty}^{\infty} P(\beta)[1 - e^{i\beta\hat{t}}]e^{-\beta/2}d\beta, \tag{7}$$

with:

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)}, \tag{8}$$

where $\rho(\beta)$ is the density of vibrational states (GDOS), and $\hat{t}$ is the time measured in units of $\hbar/k_BT$ seconds. Generalized density of states (GDOS) is DOS weighted by squared atomic eigenvectors of vibrational modes.

Since $(C_2H_4)_n$ is a hydrogenous material, the incoherent elastic component can be represented as:

$$\left(\frac{d^2\sigma}{d\mu dE}\right)_{incoh,el} = \frac{\sigma_b}{2}e^{-2WE(1-\mu)}, \tag{9}$$

where W is the Debye-Waller coefficient, and is equal to:

$$W = \frac{\lambda}{Ak_BT}, \tag{10}$$

where λ depends on GDOS:

$$\lambda = \int_{-\infty}^{\infty} P(\beta)e^{-\beta/2}d\beta. \tag{11}$$

The connection between $P(\beta)$ and $\rho(\beta)$ can be observed in Eq. 8. The total scattering cross section for $(C_2H_4)_n$ is equal to the sum of incoherent elastic and inelastic parts.

If we expand the time-dependent part of the exponential $e^{-\gamma(t)}$ from Eq. 6 we obtain:

$$e^{\gamma(\hat{t})} = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{1}{n!}\left[\alpha\int_{-\infty}^{\infty} P(\beta)e^{i\beta\hat{t}}e^{-\beta/2}d\beta\right]^n, \tag{12}$$

thus, we can rewrite Eq. 6 in terms of phonon expansion:

$$S(\alpha,\beta) = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{1}{n!}\alpha^n\frac{1}{2\pi}\times\int_{-\infty}^{\infty} e^{i\beta\hat{t}}\left[\int_{-\infty}^{\infty} P(\beta')e^{i\beta'\hat{t}}e^{-\beta'/2}d\beta'\right]^n d\hat{t}, \tag{13}$$

At n = 0 there is no exchange of energy between neutron and the scattering molecule, hence the scattering is elastic. According to Ramirez-Cuesta (2004), n = 0 quantum event (mode) correspond to Rayleigh line in Raman scattering, and it corresponds to elastic line in the inelastic neutron scattering.

3. Measurements and analysis

The experimental data have been measured at the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL). Although the focus of this report is solely on $(C_2H_4)_n$, three different instruments have been used to measure different moderator materials and subsequent reports on remaining moderator materials will be released. The instruments used for data collection are Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) Granroth et al. (0120), a wide Angular-Range Chopper Spectrometer (ARCS) Abernathy et al. (2012), and VISION Seeger et al. (2009). ARCS and SEQUOIA are similar in geometry and produce