Generation of Thermal Neutron Scattering Library for Light Water Using Molecular Dynamics Simulation code, GROMACS

Haelee Hyun[a,b*], Do Heon Kim[a], Ser Gi Hong [b]
[a]*Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea*
[b]*Department of Nuclear Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea*
[*]*Corresponding author: hyunhl@kaeri.re.kr*

1. Introduction

Thermal neutron scattering cross-section is an important parameter used in the analysis and design of nuclear reactors because it determines the probability of neutron interaction with materials. In particular, light water is a crucial component since it has been widely used as a moderator and coolant in PWRs (Pressurized Water Reactor) and BWRs (Boiling Water Reactor). As a result, the thermal neutron scattering cross-section for light water has been extensively studied to precisely analyze the safety and efficiency of nuclear power plants.

For light water (H in H_2O) data in the ENDF/B-VIII.0 library [1,2], introducing molecular dynamics (MD) simulations led to significant performance improvements compared to the ENDF/B-VII.1 library [3]. However, while the JEFF-3.3 [4] is the newest version of the JEFF nuclear data library, MD simulations were not considered for generating light water data. As a result, there is no significant improvement in performance of light water data. Recently, a test version of the JEFF-4 (JEFF-4T2) library including newly evaluated H in H_2O data is being prepared for releasing the new version of the JEFF nuclear data library. It is expected that the light water data in the upcoming release of JEFF library will show significant improvements in performance, as it will be generated using both MD simulations and experimental data.

The JENDL-5 [5] library has generated new evaluation data for O in H_2O, which includes previously disregarded scattering effects of oxygen atoms in light water (Oxygen atoms contribute only about 5% of the total scattering cross-section of H_2O molecules.), which was generated by using GROMACS [6], an MD simulation code, to produce thermal scattering data for both H and O in H_2O.

In this paper, we generated a thermal neutron scattering library for H in H_2O using the MD simulation code GROMACS-2022.4 and nuclear data processing code NJOY2016 [7], and then compared it to existing light water libraries from ENDF/B-VII.1, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5.

2. Methods and Results

This section provides a simple introduction to the thermal scattering law (TSL) and outlines a procedure for generating thermal neutron scattering cross-sections for light water.

2.1 Thermal Scattering Law (TSL) for Light Water

The double differential scattering cross-section is given by Eq. (1).

$$\sigma(E, E', \mu) = \frac{\sigma_b}{2\pi kT}\sqrt{\frac{E'}{E}}e^{-\beta/2}S(\alpha, \beta), \qquad (1)$$

where E and E' are the incident and secondary neutron energies, σ_b is the characteristic bound cross-section, k is the Boltzmann constant and T is the temperature of the material in Kelvin. The thermal scattering law, $S(\alpha, \beta)$ depends on parameters of α and β. Here α is the momentum transfer parameter defined as

$$\alpha = \frac{E'+E-2\mu\sqrt{E'E}}{AkT}, \qquad (2)$$

and β is the energy transfer parameter defined as

$$\beta = \frac{E'-E}{kT}, \qquad (3)$$

where A is the mass ratio of the scattering nuclide to the neutron and $\cos\theta$ is the scattering angle in the laboratory system. The thermal scattering law, as defined by Eq. (4), provides the information of the energy and angular distribution of secondary neutrons.

$$s(\alpha, \beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\beta\hat{t}}e^{-\gamma(\hat{t})}\,d\hat{t}, \qquad (4)$$

where $\hat{t}$ is time measured in the unit of $\hbar/kT$ seconds. The intermediate scattering function $\gamma(\hat{t})$ is given by Eq. (5).

$$\gamma(\hat{t}) = \alpha\int_{-\infty}^{\infty} P(\beta)[1 - e^{-i\beta\hat{t}}]e^{-\beta/2}d\beta, \qquad (5)$$

where

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)}, \qquad (6)$$

where $\rho(\beta)$ is called as the frequency spectrum. In the light water system, the energy excitations resulting from the inter- or intramolecular bending and stretching motions of water molecules are described by the

frequency spectrum. This spectrum serves as an important input data in LEAPR module, which calculates TSL of target system in NJOY code.

2.2 GROMACS Simulation

The ENDF/B-VIII.0, JEFF-4T2 and JENDL-5 libraries have implemented MD simulations using GROMACS code to generate the frequency spectrum of light water. In order to parameterize the target system consists of water molecules, ENDF/B-VIII.0, JEFF-4T2 and JENDL-5 libraries have used a flexible water model, TIP4P/2005f [8]. This model is well known for accurately describing actual water molecules. Hence, we also accepted TIP4P/2005f to produce the thermal neutron scattering data for light water.

For this study, we employed GROMACS-2022.4 software to create the system containing 1995 water molecules in a cubic box at a temperature of 293.6 K and a density of 0.999 g/cm^3. The simulation has proceeded in the order of energy minimization, equilibrium, and production run. The equilibrium process was performed for 100 ps under NVT and NPT conditions, respectively. After the equilibrium process, we performed a production run for 50 ps under NPT condition to generate data for analysis.

Using trajectories from the production run, we computed a velocity auto-correlation function (VACF) and converted it into the frequency spectrum using Fourier transform method. We then utilized this data as input of the LEAPR module in NJOY2016 code to generate the thermal neutron scattering library for H in H_2O data.

2.3 Simulation Results

Fig. 1 shows the differences between the frequency spectra utilized for generating H in H_2O in the ENDF/B-VII.1 and ENDF/B-VIII.0 libraries, and the one created by GROMACS for this study. Unfortunately, the spectra utilized in JEFF-3.3, JEFF-4T2 and JENDL-5 cannot be plotted since the data is unreleased.

In the ENDF/B-VIII.0 and GROMACS spectra, three peaks are detected in the energy range of about 0.005, 0.035, and 0.065-0.075 eV, which arise from the intermolecular interactions of water molecules. Each peak corresponds to a distinct motion of the water molecules, hydrogen bond bending, hydrogen bond stretching, and libration, respectively. However, despite using the same water model and simulation code, discrepancies are observed among the frequency spectra due to variations in simulation conditions such as simulation time, applied ensemble, and equilibration conditions. Moreover, the ENDF/B-VIII.0 spectrum is a combination of both experimental data and simulation results, which further contributes to the observed differences.

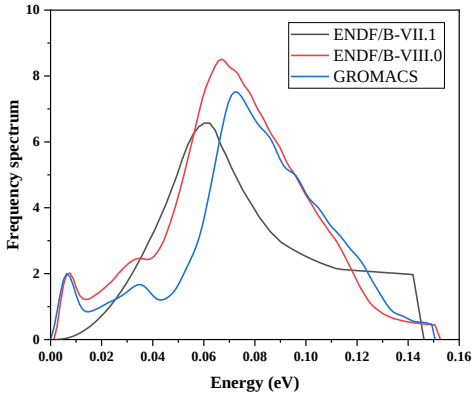

Fig. 1. Comparison of frequency spectra

The peaks related to hydrogen bond bending and stretching motions are absent in the ENDF/B-VII.1 spectrum. Although the JEFF-3.3 spectrum is not included in the Fig. 1, the shape of the ENDF/B-VII.1 spectrum is similar to that of the JEFF-3.3 spectrum. It is known that both spectra use IKE model developed by M. Mattes and J. Keinert [9], which is based on experimental data measured in 1960s.

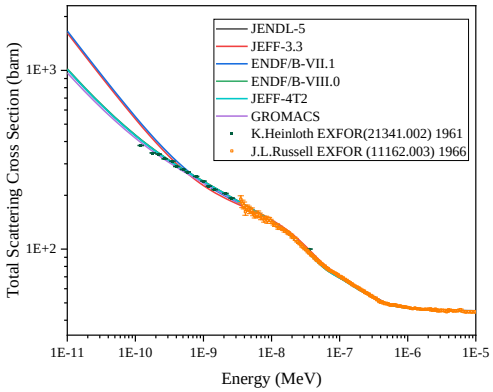

Fig. 2. Comparison of total scattering cross-sections for light water

The scattering cross-sections for light water are depicted in Fig. 2. As previously mentioned, the calculated scattering cross-sections from ENDF/B-VII.1 and JEFF-3.3 are comparable owing to the similar frequency spectra of these libraries. However, when compared to the remaining libraries, significant discrepancies were observed in the low-energy range of 1.0E-11 – 5.0E-10 MeV. On the other hand, the scattering cross-sections calculated from the ENDF/B-VIII.0, JEFF-4T2, JENDL-5 and GROMACS simulation in this study exhibit a similarity, and they are also in good agreement with the EXFOR data. These results suggest that the MD simulation methods can provide reliable estimates of scattering cross-sections.

3. Conclusions

In this study, we have compared the MD-based (ENDF/B-VIII.0, JEFF-4T2, JENDL-5) and experiment-based (ENDF/B-VII.1, JEFF-3.3) scattering cross-sections for light water. The results showed that there are significant discrepancies between the MD-based cross-sections and experiment-based cross-sections in the low energy range (< 5.0E-10 MeV). When comparing scattering cross-sections of light water obtained through experiments (EXFOR data) with the evaluated cross-sections, it was observed that the MD-based cross-sections showed higher agreement than experiment-based cross-sections. Also, we have calculated the frequency spectrum and generated the cross-section of light water using GROMACS and NJOY code. Although the second and third peaks of frequency spectrum were found to be lower than corresponding values in the ENDF/B-VIII.0 data, the cross-section calculated using GROMACS in this work showed good accordance with the other MD-based cross-sections.

It is concluded that the MD simulation methods can provide reliable estimates when generating the thermal neutron scattering cross-sections. In addition, the performance of our GROMACS-based cross-section will be validated through criticality benchmark calculations using the International Handbook of Evaluated Criticality Safety Benchmark Problems (ICSBEP).

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REFERENCES

[1] D. A. Brown, et al., ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data, Nuclear Data Sheets, Vol.148, p.1-142, 2018.
[2] J. I. Marquez Damian, J. R. Granada, D. C. Malaspina, CAB models for water: a new evaluation of the thermal neutron scattering laws for light and heavy water in ENDF-6 format, Ann. Nucl. Energy, Vol.65, 280, 2014.
[3] M. B. Chadwick, et al., ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data, Nuclear Data Sheets, Vol.112, p.2887-2996, 2011.
[4] A. J. M. Plompen, et al., The joint evaluated fission and fusion nuclear data library, JEFF-3.3, The European Physical Journal A, Vol.56, 181, 2020.
[5] O. Iwamoto, et al., Japanese evaluated nuclear data library version 5: JENDL-5, Journal of Nuclear Science and Technology, Vol.60, 1, 2023.
[6] D. Van Der Spoel, E. Lindahl, B. Hess, G. Groenhof, A. E. Mark, H. J. C. Berendsen, GROMACS: Fast, Flexible, and free, Journal of Computational Chemistry, 26(16), 1701-1718, 2005.
[7] D. W. Muir, et al., The NJOY Nuclear Data Processing System, Version 2016, LA-UR-17-20093, 2016.
[8] M. A. Gonzalez, J. L. F. Abascal, A flexible model for water based on TIP4P/2005, The Journal of Chemical Physics, 135, 224516, 2011.
[9] M.Mattes, J. Keinert, Thermal Neutron Scattering Data for the Moderator Materials H_2O, D_2O and ZrH_x in ENDF-6 Format and as ACE Library for MCNP(X) Codes, INDC/NDS report INDF(NDS)-0407, 2005.