Comparison of the Thermal Neutron Scattering Cross sections for Heavy Water obtained by GROMACS and LAMMPS Molecular Dynamics Simulations

Haelee Hyun[a,b*], Do Heon Kim[a] and 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

The most recently released thermal neutron scattering libraries (e.g. ENDF/B-VIII.0 and JEFF-3.3) have introduced computational chemistry simulations such as molecular dynamics (MD) and ab-initio molecular dynamics (AIMD). MD uses classical Newton's equation to simulate the designed system. In order to calculate the force and energy of the given system using MD, accurate classical potentials are needed. The potentials are usually calculated from empirical or semi-empirical relation such as Lennard-Jones. However, it is hard to obtain accurate classical potentials for some complex systems. In addition, since the classical potentials cannot completely describe effects from electronic polarization, AIMD can be run as an alternative.

Unlike MD, AIMD calculates the force and energy of the system based on the Schrödinger equation. In practice, AIMD has recently been used to generate the thermal neutron scattering libraries of some moderator materials such as graphite and beryllium [1]. However, AIMD is computationally much more expensive than MD.

In this paper, we focused on comparison of the simulation results of heavy water obtained by two MD software, GROMACS and LAMMPS [2,3], which are the most widely used strong MD simulation codes. ENDF/B-VIII.0 and JEFF-3.3 thermal neutron scattering libraries for heavy water have been generated based on GROMACS code simulation. In order to produce various thermal neutron scattering libraries in the future, we judged that experiences in both GROMACS and LAMMPS simulation would be important.

In this study, we have compared ENDF/B-VIII.0 scattering cross section of heavy water with those from GROMACS and LAMMPS simulation results.

2. Simulation Method and result

The scattering cross section of heavy water is generated by using NJOY2016 code in this paper. NJOY2016 requires the frequency spectrum and Sköld correction factor to produce the heavy water library. The physical meaning and calculation process of the two factors were described in the studies [4,5]. In this paper, we simply introduce the MD simulation procedures.

2.1 GROMACS simulation

GROMACS simulation is carried out with TIP4P/2005f water model [6]. This water model is also used for ENDF/B-VIII.0 heavy water data. To describe the motion of water molecules accurately, the strength of atomic bonds between oxygen-deuteron (OD) and angles between deuteron-oxygen-deuteron (DOD) has to be specified. Hence, the intramolecular potential which indicates the forces how strongly atoms hold each other is entered as an input parameter of GROMACS. The characteristic of TIP4P/2005f model is that the Morse potential is applied to describe forces of bonds, which Morse potential function is given by Eq. (1). For considering the angle bending, a harmonic potential function specified in Eq. (2) is applied.

$$V_{ODi} = D_r\{1 - \exp[-\beta(r - r_{eq})]\}^2 \quad (1)$$

$$V_{DOD}(\theta) = \frac{1}{2}K_\theta(\theta - \theta_{eq})^2 \quad (2)$$

where r_{eq} and θ_{eq} are the values of the bond length and angle at equilibrium and r_{ODi} is the instantaneous distance between the deuteron atom i and the oxygen atom. D_r and β are the parameters of the Morse potential that determine the bond strength and width [6].

We prepared the system that consists of 1082 molecules in a cubic box at 293.6 K for GROMACS simulation. The simulation has proceeded in the order of energy minimization, equilibrium, and production run. The equilibrium process was performed for 10 ps under NVT and NPT conditions, respectively. After the equilibrium process, the production run is performed for 10 ps under NVE condition.

Consequently, we have obtained the simulation results such as a velocity auto-correlation function and radial distribution function, which were converted into frequency spectrum and Sköld correction factor through an appropriate process [5] and used the results as an input data for NJOY to produce the scattering cross section for heavy water. Figure 1 shows the calculation results of frequency spectrum and Sköld correction factor.

2.2 LAMMPS simulation

For LAMMPS simulation, SPC/E (Extended Simple Point Charged) water model is used [7]. The reason why water models used in GROMACS and LAMMPS are different is that LAMMPS spent much more computational costs and gave some troubles (e.g. unexpected system expansion and increasing

temperature) when using TIP4P/2005f water model. SPC/E water model uses harmonic potential for describing both atomic bonds and angles. The harmonic bond potential is given in Eq. (3).

$$V_{ODi} = \frac{1}{2} K_r (r - r_{eq})^2 \quad (3)$$

where K_r is the parameter that determine the bond strength.

LAMMPS simulation system is prepared with 1500 molecules in a cubic box at 293.6 K. The simulation is processed in the order of the system equilibrium and production run. For equilibrium, NVT condition was applied twice, and NPT condition was applied once for 50 ps, respectively. After the equilibrium process, the production run is performed for 10 ps under NVE condition.

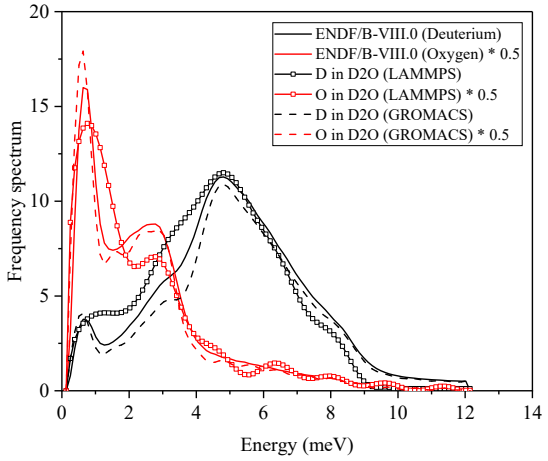
In the same manner as GROMACS simulation, the frequency spectrum and Sköld correction factor were obtained, and based on the results, we calculated the scattering cross section of heavy water.

2.3 Simulation results

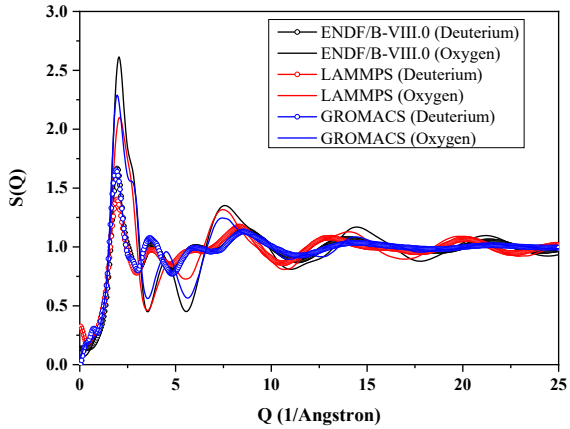
As shown in Fig.1, although all simulation codes use MD method, the results show the subtle difference. In case of the frequency spectrum (Fig.1. (a)), the peaks at around 0.6 meV and 2~3 meV stand for motions of intermolecular bending and stretching of oxygen and deuterium, respectively. In contrast with oxygen, deuterium has third peak at around 4~6 meV, which indicates the librational motions. The result from LAMMPS simulation represents the lowest peak of oxygen atom whereas the peak heights of deuteron are similar to all results. Overall, full shape of the spectrum calculated by GROMCAS simulation shows better accordance with ENDF/B-VIII.0 data. The reason is presumably because the same simulation code and water model were used.

In case of Sköld correction factor (Fig.1. (b)), all simulation results showed analogous shape. However, the result from LAMMPS simulation represents the lowest peak.

Combining the results, the scattering cross sections of heavy water were calculated and compared, which are shown in Fig.2. The scattering cross section shows two dips on the scattering cross section at 3 meV and 10 to 30 meV. The first dip is affected by the coherent effect due to deuteron atoms and the second dip is caused by the O-O interference in D_2O molecule. In case of LAMMPS based scattering cross section, it appears that the coherent effect was not properly reflected compared to the result from GROMACS simulation. The reason is most likely due to the low peaks of the Sköld correction factor because the Sköld correction factor is used as an input of NJOY2016 code to reflect the coherent effect.



(a)



(b)

Fig.1. Comparison of ENDF/B-VIII.0 data and GROMACS and LAMMPS simulation results (a) frequency spectrum (b) Sköld correction factor.

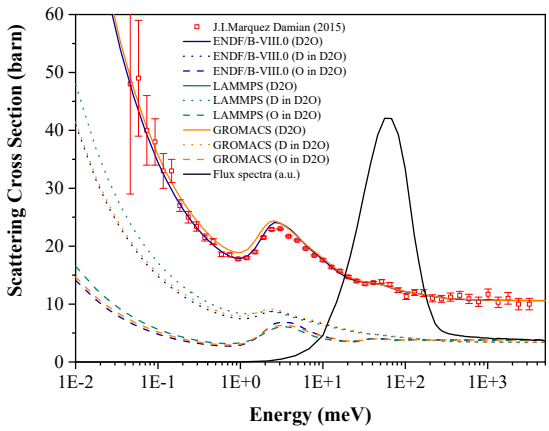


Fig.2. Comparison of the scattering cross sections for heavy water

Although there are some dissimilarities when comparing ENDF/B-VIII.0 and scattering cross sections generated by MD simulations, the differences in performance of those cross sections are not expected to be significantly large. Because MD based scattering cross sections have high similarity with ENDF/B-VIII.0 library above 10 meV range where the thermal neutron flux spectrum is biased. Estimating the performance of scattering cross sections for heavy water has been carried out in previous studies [4,5] and it shows that the first dip of the heavy water cross section has no significant impact on its performance.

3. Conclusions

We have generated the thermal neuron scattering library for heavy water by using major MD simulation codes, GROMACS and LAMMPS. Two water models, TIP4P/2005f and SPC/E, are applied to perform the GROMACS and LAMMPS simulation, respectively. The results from LAMMPS simulation showed a larger difference from ENDF/B-VIII.0 compared to the results from GROMCAS simulation. Nevertheless, it is confirmed that the coherent effects of deuteron atoms and oxygen-oxygen interference are reflected on the thermal neutron scattering cross section based on LAMMPS simulation.

For further research, we would generate more improved thermal scattering cross section by using AIMD simulation for heavy water. It is expected that this study would be a great foundation for generating various thermal neutron scattering libraries in the future.

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