Generation and Validation of Thermal Neutron Scattering Cross Sections for Heavy Water by Using New Sköld Correction Factors

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1. Introduction

Recently, the thermal scattering libraries of ENDF/B-VIII.β4 for light and heavy water have been released with the new water model (CAB model) proposed by Damian [1] using NJOY2016 code [2]. The new thermal scattering library for heavy water has significant improvements in comparison with existing thermal scattering libraries. First, in order to take into account an effect from the coherent scattering of the oxygen atom in heavy water molecule, the thermal scattering library of ENDF/B-VIII.β4 for heavy water has introduced both D bound in D_2O and O bound in D_2O data, which is a notable difference from the existing thermal scattering library considering oxygen atoms as a free gas. Second, parameters and functions associated with the molecular vibrations and the coherent effect of heavy water have been calculated using the molecular dynamics simulation to more accurately describe the realistic motions of water molecule. Also, the molecular dynamics simulation by GROMACS v.4.5.5. code [4] is performed based on TIP4P/2005f water model for light and heavy water [3].

In this paper, we particularly focused on generating the thermal scattering cross section of heavy water that the intermolecular coherence is considered. The effect from the coherent scattering of heavy water molecules is considered by applying the Sköld approximation [5]. We also calculated the Sköld correction factor which is needed for applying the Sköld approximation by using GROMACS v.5.1.4. code. The thermal scattering cross sections based on newly calculated Sköld correction factor are generated by NJOY2016 code. Additionally, the Sköld correction factor is also calculated by EPSR (Empirical Potential Structure Refinement) code [6], which is the software for building atomic and molecular structural models of disordered materials such as liquids and glasses based on experimental diffraction data. Finally, the performances of generated thermal scattering cross sections are validated by performing ICSBEP benchmark simulation using MCNPX 2.7.0 code [7].

2. Calculation method

2.1 Theraml Scattering Law

At thermal energy range, the double differential inelastic scattering cross section for solid, liquid and gas moderator material can be described as a function of the thermal scattering law $S(\alpha,\beta)$:

$$\frac{\partial^2\sigma}{\partial\Omega\,\partial \mathrm{E}} = \frac{\sigma_b}{4\pi kT}\sqrt{\frac{E'}{E}}S(\alpha,\beta), \quad (1)$$

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. Also, the thermal scattering law depends on parameters of α and β. Here α is the momentum transfer parameter defined as

$$\alpha = \frac{E'+E-2\sqrt{EE'}\cos\theta}{AkT}, \quad (2)$$

and β is the energy transfer parameter defined as

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

where A is the mass ratio of the scattering nuclide to the neutron and $\cos\theta$ is the scattering angle in the laboratory system.

2.2 Sköld correction factor

As shown in Table I, the coherent scattering cross section of hydrogen is very small compared with incoherent scattering cross section. Hence, in case of the thermal scattering law for light water, the incoherent approximation (i.e. $\sigma_{scatt} = \sigma_{coh} + \sigma_{inc} \cong \sigma_{inc}$) would not affect much influence on calculation of the scattering cross section. On the other hand, the fraction of σ_{coh}^{D} to σ_{scatt}^{D} of the deuterium is about 73.2%, which means that the use of the incoherent approximation can cause large uncertainties on calculating the scattering cross section of heavy water. Also, as opposed to hydrogen having much larger scattering cross section than oxygen, deuterium has similar scattering cross section with the oxygen, which indicates that oxygen and deuterium have similar importance in scattering cross section of heavy water. Hence, the thermal library of ENDF/B-VIII.β4 for heavy water takes into account both the effect of D bound in D_2O and O bound in D_2O.

Table I: Coherent and Incoherent Scattering Cross Sections (Unit: barn)

	σ_{coh}	σ_{inc}	σ_{scatt}
1-H-1	1.7583	80.27	82.03
1-H-2	5.592	2.05	7.64
8-O-16	4.232	0.0	4.232

In order to generate the scattering cross section with the coherent component of deuterium and oxygen, the thermal scattering cross section for heavy water used Sköld approximation:

$$S(\alpha,\beta) = S^{inc}(\alpha,\beta) + S^{coh}(\alpha,\beta) \quad (4)$$

$$S^{D}_{coh}(\alpha,\beta) = S^{D}_{inc}\left(\frac{\alpha}{\tilde{S}^{D}},\beta\right)\tilde{S}^{D}(Q) \quad (5)$$

$$S^{O}_{coh}(\alpha,\beta) = S^{O}_{inc}\left(\frac{\alpha}{\tilde{S}^{O}},\beta\right)\tilde{S}^{O}(Q) \quad (6)$$

where $\tilde{S}^{D}$ and $\tilde{S}^{O}$ are the Sköld correction factors [7] for deuterium and oxygen, respectively.

$$\tilde{S}^{D}(Q) = 1 + \frac{2}{3}[S_{DD}(Q) - 1] + \frac{1}{3}\frac{\sigma^{O}_{coh}}{\sigma^{D}_{coh}}[S_{DO}(Q) - 1] \quad (7)$$

$$\tilde{S}^{O}(Q) = 1 + \frac{1}{3}[S_{OO}(Q) - 1] + \frac{2}{3}\frac{\sigma^{D}_{coh}}{\sigma^{O}_{coh}}[S_{DO}(Q) - 1] \quad (8)$$

The $S_{DD}(Q)$, $S_{DO}(Q)$ and $S_{OO}(Q)$ are static structure factors regarding each atom in heavy water molecules.

2.2.1 GROMACS simulation

To calculate the static structure factor, we performed molecular dynamics simulation for heavy water molecules with TIP4P/2005f water model by using GROMACS v.5.1.4. The simulation system consists of 1084 heavy water molecules in the cubic box having a volume of 32.768 nm^3. The simulation was carried out using NVE ensemble (i.e. N: number of particles in the system, V: volume of the system and E: energy of the system are constant) and OPLS/AA (Optimized Potentials for Liquid Simulation/All Atom) force field at 293.6K for 10ps after taking procedures for the energy minimization and the equilibration.

The static structure factor is a mathematical description of how a material scatters incident radiation. Therefore, the static structure factor is a critical tool in the interpretation of scattering patterns obtained in X-ray, electron and neutron diffraction. And it can be calculated by Fourier transform of the radial distribution function as shown in equation (9).

$$S_{\alpha\beta}(Q) = 1 + 4\pi\rho\int_0^{\infty} dr\,(g(r) - 1)r^2\frac{\sin(Qr)}{Qr} \quad (9)$$

where Q is a wave number, ρ is the density of the simulation system, $g(r)$ is the radial distribution function, r is a distance from the center of the system, dr is an increment of the r. The radial distribution function gives the probability to find particles located at distance r and can be obtained from the result of the molecular dynamics simulation. As a result, Fig.1 shows Sköld correction factors calculated by equations (7) and (8). As shown in Fig.1, when comparing the Sköld correction factor from GROMACS simulation with ENDF/B-VIII.β4 data, the Sköld correction factors of deuterium show a high consistency. For the Sköld correction factor of oxygen, ENDF/B-VIII.β4 data shows higher peaks than GROMACS data.

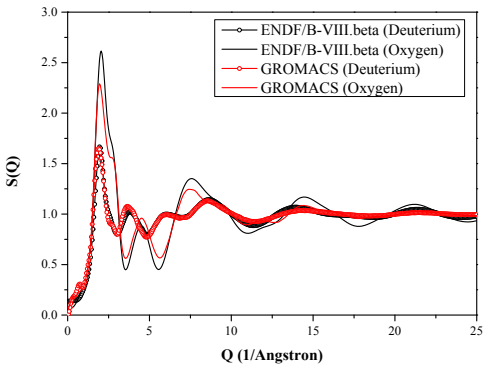


Fig.1. Comparison between Sköld correction factors obtained from ENDF/B-VIII.β4 data and calculated by GROMACS code (293.6K).

2.2.2 EPSR simulation

As described above, we also calculated Sköld correction factor using EPSR code. EPSR is a Monte Carlo code evolved from Reverse Monte Carlo (RMC) method, which also attempts to build a structural model of a glass or liquid, and which evolved to evaluate disordered materials structure on the basis of diffraction experiments.

To calculate the static structure factor, we used SPC/E (Extended Simple Point Charged) water model because EPSR code has a limitation to build TIP4P/2005f water model. The simulation system consists of 1000 heavy water molecules in the cubic box having a volume of 29.94nm^3. The simulation was carried out with the periodic boundary condition at 293.6K for 307 iterations.

As shown in Fig.2, when comparing the Sköld correction factor from EPSR simulation with ENDF/B-VIII.β4 data, the Sköld correction factor of deuterium also shows a high consistency in common with the result of GROMACS simulation. For the Sköld correction factors of oxygen, the result from EPSR simulation shows the lower peaks than other results. And the Sköld correction factor calculated by EPSR has higher values at the initial values of Q than other data. It might be because the applied water model is different.

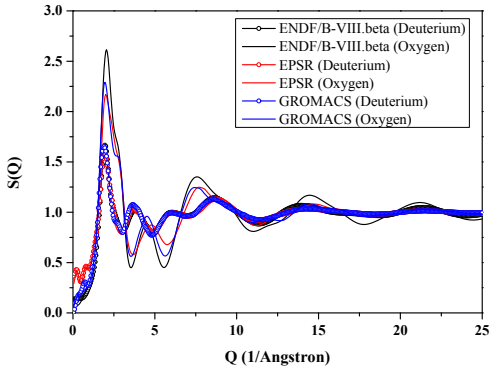


Fig.2. Comparison between Sköld correction factors obtained from ENDF/B-VIII.β4 data and calculated by EPSR and GROMACS code (293.6K).

2.3 Comparison of thermal scattering cross sections

Finally, we generated the thermal scattering cross sections using NJOY2016 code. The Sköld correction factors calculated by each molecular dynamics simulation code are used as an input of LEAPR module of NJOY2016 to generate the thermal scattering cross sections that the effect from coherent scattering is taken into account.

Fig.3 shows two dips on the scattering cross section at 3meV and 10 to 30 meV. The first dip is affected by the D-D interference and the second dip is caused by the O-O interference in D_2O molecule.

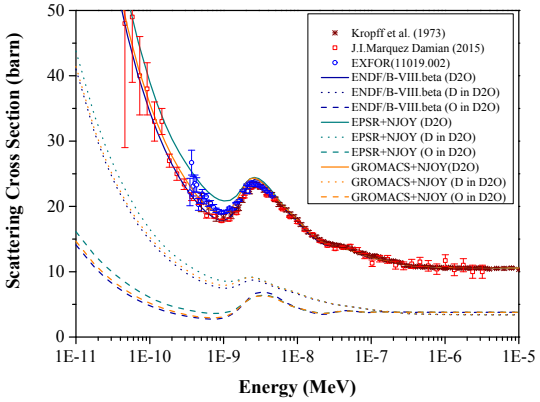


Fig.3. Scattering cross sections for heavy water and each atom at room temperature (293.6K).

Additionally, as shown in Fig.3, the thermal scattering cross section based on the Sköld correction factor calculated by GROMACS simulation shows a rather satisfactory accordance with the scattering cross section of ENDF/B-VIII.β4 for heavy water, which is assumed because Sköld correction factors are calculated by using the same water model and simulation code, GROMACS. The other way, as mentioned above, the Sköld correction factor calculated by EPSR simulation has shown relatively remarkable differences in comparison with ENDF/B-VIII.β4 data. Accordingly, the scattering cross section based on the Sköld correction factor calculated by EPSR simulation also shows prominent differences compared with the scattering cross section of ENDF/B-VIII.β4. The notable differences are shown under the energy range of 3meV. However, for the energy range over 3meV, all of generated scattering cross sections indicate almost complete accordance with the scattering cross section of ENDF/B-VIII.β4 for heavy water and the experimental data.

3. Criticality Benchmark Problems

In order to estimate the effects of the thermal scattering libraries, 59 heavy water moderated/reflected experiments are taken from the International Handbook of Evaluated Criticality Safety Benchmark Problems (ICSBEP Handbook) [8]. As shown in Table II, the 59 benchmark problems can be classified according to the principal fuel. All of benchmark calculations were done by using KCODE mode of the Monte Carlo Transport code MCNPX 2.7.0 along with the thermal scattering libraries. The ENDF/B-VII.1-based KNE71 library [9] was used for all nuclides except thermal scattering cross sections of D in D2O and/or O in D2O. All the MCNP benchmark simulations have been carried out at 293.6K. Also, the MCNP runs were terminated after a statistical uncertainty was reduced to below 20 pcm.

Table II: Number of Benchmark Problems

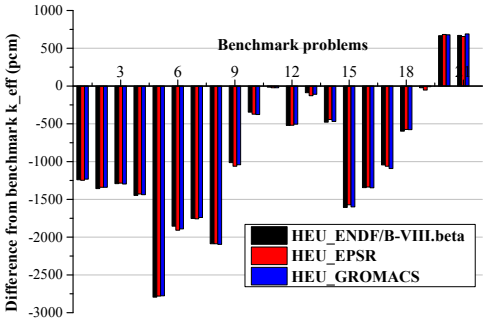
Category	Number of Benchmark Problems
HEU*	21
IEU*	1
LEU*	36
U233	1
WHOLE	59

*HEU: Highly Enriched Uranium
IEU: Intermediated Enriched Uranium
LEU: Low Enriched Uranium

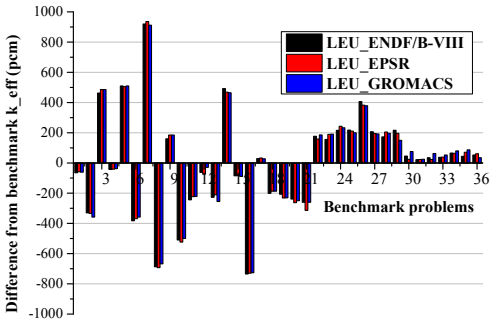
In this paper, we compared the results of criticality calculations regarding the thermal scattering cross sections to confirm the performance of generated libraries.

Fig.4 shows the comparison of the differences of the calculated k_{eff} from the benchmark k_{eff} with the thermal scattering libraries. The results of the benchmark problems show small differences about all of categories, which means the generated thermal scattering cross sections in this study has similar performance to the thermal scattering cross section of ENDF/B-VIII.β4. Especially, although the scattering cross section using the Sköld correction factor calculated by EPSR simulation has shown relatively large differences under the energy range of 3meV, the benchmark performance

was not unfavorable. This might be because the thermal neutron flux spectrum is biased at the energy range of 10~100 meV in the thermal system. In other word, in order to generate the scattering cross section of heavy water having better performance, precisely describing the second dip caused by O-O interference is more important than the first dip.



(a) Category of HEU



(b) Category of LEU

Fig.4. Differences of calculated k_{eff} from benchmark k_{eff} with the thermal scattering libraries of ENDF/B-VIII.β4 and generated thermal scattering libraries.

Also, the result of root mean square (RMS) errors for heavy water problems is described in Table III. The total RMS errors of ENDF/B-VIII.β4 and generated libraries indicate 0.807~0.808%, which also shows the generated libraries have similar performances with ENDF/B-VIII.β4 library in aggregate.

Table III: Comparison of RMS errors relative to benchmark k_{eff} values among different libraries (Unit: %)

Category	ENDF/B-VIII.β4	EPSR	GROMACS
HEU (21)	1.276	1.277	1.278
IEU (1)	0.151	0.158	0.126
LEU (36)	0.329	0.332	0.327
U233 (1)	0.531	0.532	0.532
TOTAL (39)	0.807	0.808	0.807

4. Conclusions

In this study, 59 heavy water moderated/reflected benchmark calculations were carried out to compare the performances of generated thermal scattering data for heavy water by using new Sköld correction factor calculated by EPSR and GROMACS simulations. The generated thermal scattering cross section using Sköld correction factor calculated by GROMACS shows the satisfactory accordance with the thermal scattering cross section of heavy water in ENDF/B-VIII.β4. On the other hand, the generated thermal scattering cross section using Sköld correction factor calculated by EPSR shows relatively large discrepancies under the energy range of 3 meV in comparison with the ENDF/B-VIII.β4 library. However, because the neutron flux spectrum in thermal system is biased at the energy range of 10~100 meV, that discrepancies did not seriously affect the benchmark performance of the thermal scattering cross section generated with EPSR simulation. We finally confirmed that the performances of both generated thermal scattering cross sections for heavy water represent similar performances with the thermal scattering data of ENDF/B-VIII.β4.

For further research, we would generate more improved thermal scattering cross section by using not only new Sköld correction factor but also new frequency spectrum of light and heavy water to more precisely consider the intermolecular and intramolecular interactions of water molecules.

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