Testing of Improved Thermal Scattering Library for Heavy Water in ENDF/B-VIII.β4

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1. Introduction

Thermal scattering data is important to accurately simulate the models of thermal neutron systems. At thermal energy range (below about 5eV), the scattering is affected by the chemical binding in the solid, liquid, or gas moderator material. These effects change the reaction cross section and energy-angular distribution of secondary neutrons. In order to correct these effects from binding atoms, the thermal scattering libraries have to be applied to the simulations of thermal systems.

Up to now, all of the thermal scattering libraries for light and heavy water, which have been used in various simulations, are produced from only two different models. First one is the General Atomic model (GA model) which was established in GASKET code by Koppel and Young in 1969. ENDF/B-VI.8 and JENDL 4.0 adopted this model for the thermal scattering library [1]. Second one is the IKE model which was established by Keinert and Mattes in 2004. ENDF/B-VII, JEFF 3.1 and RUSFOND-2010 are using this model for the thermal scattering library [2]. Recently, the thermal libraries of ENDF/B-VIII.β4 for light and heavy water have been improved based on the new model (CAB model) proposed by Damian from Centro Atomico Bariloche, Argentina [3,4,5]. For CAB model, the thermal scattering law for light and heavy water is computed by LEAPR module in NJOY code [7]. The new libraries for light and heavy water are based on the experimental data and the molecular dynamics simulation by GROMACS [6] to more accurately describe the frequency spectrum of waters than existing thermal libraries. Especially, heavy water library is taking into account two kinds of chemical bindings of D in D_2O and O in D_2O, which is the distinguishing characteristics in contrast with the existing thermal scattering libraries considering only one chemical binding of D in D_2O.

In this paper, we performed ICSBEP benchmark simulations [8] using MCNPX 2.7.0 code [9] to confirm the effects of the improved thermal scattering data for heavy water in ENDF/B-VIII.β4 by comparing the results of ENDF/B-VII.0 based ENDF70SAB library in ACE format for MCNP calculation [10].

2. Thermal scattering library

2.1 Thermal 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 ratio of the scattering nuclide mass to the neutron mass and $\cos\theta$ is the scattering angle in the laboratory system. The thermal scattering law contains dynamic and structural information about the target system and determines energy and angular distribution of secondary neutrons, which can be written

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

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

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

where

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

where $\rho(\beta)$ is the frequency spectrum which is a sum of discrete oscillator, solid-type spectrum and translational spectrum [7]. However, the thermal scattering law introduced above has limitations to describe dynamic and structural information of the materials accurately. Because the thermal scattering law neglects the coherent component of the scattering cross section and considers only the incoherent component, which is referred to as the incoherent approximation: $\sigma_{\mathrm{scatt}} = \sigma_{\mathrm{coh}} + \sigma_{\mathrm{inc}} \cong \sigma_{\mathrm{inc}}$. In addition, the Gaussian approximation is applied to Eq. (5), which means the intermediate scattering function is described by Gaussian form. The Gaussian approximation is known to have a limitation in the quasi-elastic range [11].

2.2 Improvement of Thermal Scattering Law for Heavy Water in ENDF/B-VIII.β4

As shown in Table I, the coherent scattering cross section of hydrogen is very small compared with incoherent scattering cross section. In other words, the fraction of $\sigma_{\mathrm{coh}}^{\mathrm{H}}$to $\sigma_{\mathrm{scatt}}^{\mathrm{H}}$ is just 2.14%. Hence, in case of the thermal scattering law for light water, the incoherent approximation would not affect much influence on calculation of the scattering cross section. On the other hand, the fraction of $\sigma_{\mathrm{coh}}^{\mathrm{D}}$ to $\sigma_{\mathrm{scatt}}^{\mathrm{D}}$ for the deuterium is about 73.2%, which means the effect from the coherent scattering cross section is not negligible and using the incoherent approximation can cause large uncertainties about calculating the scattering cross section of heavy water.

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	4.232

For the thermal scattering law data of ENDF/B-VII.0, to deal with the coherent scattering effect of heavy water the scattering function was computed by the Sköld approximation [12].

$$S(\alpha,\beta) = (1-f)S^{inc}(\alpha,\beta) + fS^{inc}(\alpha',\beta)S_{DD}(\kappa) \quad (7)$$

where

$$f = \frac{\sigma_{\mathrm{coh}}}{\sigma_{\mathrm{scatt}}} \text{ and } \alpha' = \frac{\alpha}{S_{DD}(\kappa)} \quad (8)$$

where $S_{DD}(\kappa)$ is a static structure factor which represents certain characteristic values of the spacing between D-D molecules to represent D-D interference in D_2O molecules. And the oxygen is approximated as a free gas. For the thermal data of ENDF/B-VIII.β4, the coherent component is also computed by the Sköld approximation. However, as opposed to hydrogen having much larger scattering cross section than oxygen, deuterium has similar scattering cross section with the oxygen, which indicates oxygen and deuterium have similar importance in heavy water. Hence, unlike light water, treating oxygen as a free gas could cause a large uncertainty for heavy water simulation due to the effect of oxygen interferences. Thus, the thermal library of ENDF/B-VIII.β4 takes into account both interferences of D-D and O-O of heavy water from the following equations,

$$S_{coh}^{D}(\alpha,\beta) = S_{inc}^{D}\left(\frac{\alpha}{\tilde{S}^{D}},\beta\right)\tilde{S}^{D}(\kappa) \quad (9)$$

$$S_{coh}^{O}(\alpha,\beta) = S_{inc}^{O}\left(\frac{\alpha}{\tilde{S}^{O}},\beta\right)\tilde{S}^{O}(\kappa) \quad (10)$$

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

$$\tilde{S}^{D}(\kappa) = 1 + \frac{2}{3}[S_{DD}(\kappa) - 1] + \frac{1}{3}\frac{\sigma_{coh}^{O}}{\sigma_{coh}^{D}}[S_{DO}(\kappa) - 1] \quad (11)$$

$$\tilde{S}^{O}(\kappa) = 1 + \frac{2}{3}[S_{OO}(\kappa) - 1] + \frac{1}{3}\frac{\sigma_{coh}^{D}}{\sigma_{coh}^{O}}[S_{DO}(\kappa) - 1] \quad (12)$$

The $S_{DD}(\kappa)$, $S_{DO}(\kappa)$ and $S_{OO}(\kappa)$ are static structure factors, which can be obtained from experimental data measured by Soper [13] or calculated by Fourier transform of radial distribution function of heavy water. Also, to overcome the limitation from the Gaussian approximation the thermal library of ENDF/B-VIII.β4 used a random jump diffusion model [11].

2.3 Thermal scattering libraries for heavy water

Figure 1 shows the comparison of total cross sections and thermal scattering cross sections of ENDF/B-VIII.β4 and ENDF/B-VII.0 with the experimental data measured by Kropff (EXFOR database #30283.002) [14]. The total cross section for heavy water is calculated by Eq. (13);

$$\sigma_{\mathrm{total}}^{\mathrm{D2O}} = \left(\sigma_{\mathrm{scatt}}^{\mathrm{D\ in\ D2O}} + \sigma_{\mathrm{abs}}^{\mathrm{D}}\right) * 2 + \left(\sigma_{\mathrm{scatt}}^{\mathrm{O\ in\ D2O}} + \sigma_{\mathrm{abs}}^{\mathrm{O}}\right) \quad (13)$$

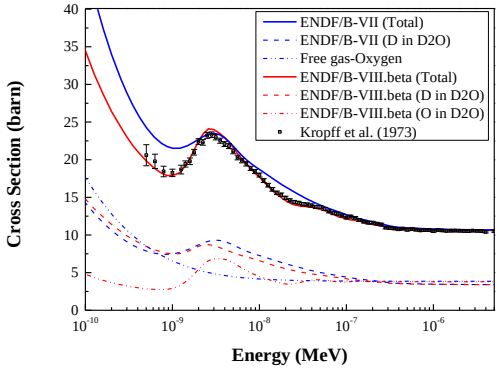


Fig.1. Total cross sections and scattering cross sections for heavy water at room temperature (293.6K).

Note that two dips are shown from measured total scattering cross section at 3meV and 10 to 30meV. The first dip is affected by D-D interference in D_2O molecule. Because both thermal scattering cross sections of ENDF/B-VIII.β4 and ENDF/B-VII.0 take into account D-D interference using Sköld approximation, first dip is observed from both total cross sections. However, the total cross section of ENDF/B-VIII.β4 shows better agreement with measured values, which corresponds to the effect of new Sköld correction factor. The second dip is due to the O-O interference. Compared with O in D_2O, free gas oxygen cross section has no fluctuations caused by interferences from D_2O molecule. Accordingly, discrepancies are observed between the total cross

section of ENDF/B-VII.0 and the measured total cross section at about 10 to 30meV.

3. Criticality benchmark problems

In order to estimate the effects of the thermal scattering libraries, 54 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 54 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 [15] was used for all nuclides except thermal scattering cross sections of D in D_2O and/or O in D_2O. The KNE71 is an ACE formatted library generated by Nuclear Data Center of the Korea Atomic Energy Research Institute, which have been disseminated through the KAERI/NDC website (http://atom.kaeri.re.kr/NDVG/).

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

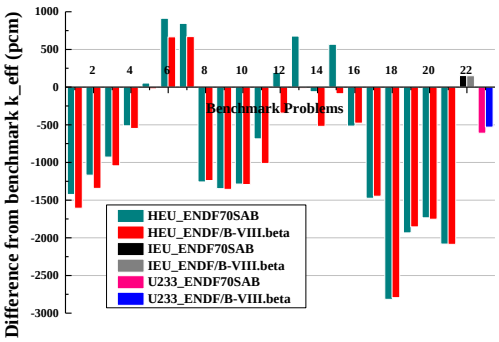
Category	Number of Benchmark Problems
HEU*	21
IEU*	1
LEU*	31
U233	1
WHOLE	54

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

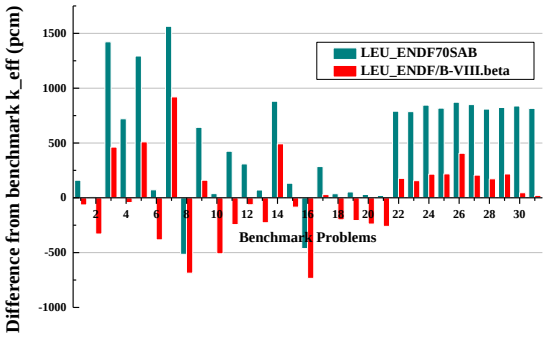
3.1 Results

In this paper, we compared the results of criticality calculations with different thermal scattering cross section libraries of ENDF/B-VIII.β4 and ENDF70SAB to confirm the effect of O in D_2O thermal data of ENDF/B-VIII.β4.

Fig.2. shows the comparison of the differences of the calculated k_{eff} from the benchmark k_{eff} for ENDF/B-VIII.β4 and ENDF70SAB. As shown in Fig.2. (a) for HEU, IEU and U233 cases, the ENDF/B-VIII.β4 shows similar results with the ENDF70SAB although the chemical bindings of D in D_2O and O in D_2O are applied. This might be caused by the spectrum hardening due to high enrichment of the fuel. For most of the thermal systems, however, fuel enrichment is maintained below 5%. Thus, the effect of the two chemical bindings is especially important for the LEU cases. Fig.2. (b) shows significant impact of O in D_2O thermal data in comparison to Fig.2. (a). Accordingly, the thermal data of ENDF/B-VIII.β4 for heavy water tends to produce better performances than ENDF70SAB.



(a) Categories of HEU, IEU and U233



(b) Category of LEU

Fig.2. Differences of calculated k_{eff} from benchmark k_{eff} with the thermal scattering libraries of ENDF/B-VIII.β4 and ENDF70SAB.

The root mean square (RMS) error is also introduced to represent the sample standard deviation of the differences between calculated and benchmark k_{eff}, which is defined as Eq. (14);

$$\mathrm{RMS}(\%) = \sqrt{\frac{\sum_{i=1}^{N}(k_{C,i}-k_{B,i})^2}{N}} \qquad (14)$$

where $k_{C,i}$ and $k_{B,i}$ are the calculated and benchmark k_{eff} for the i^{th} problem, respectively. N is the total number of problems in each case. The RMS error for heavy water problems are described in Fig.3. As discussed above, ENDF/B-VIII.β4 and ENDF70SAB library have similar RMS errors for HEU case and also the most remarkable improvement between two scattering libraries is shown in LEU category. This indicates that the addition of O in D_2O data from

ENDF/B-VIII.β4 could be more effective when simulating the LEU benchmarks with heavy water.

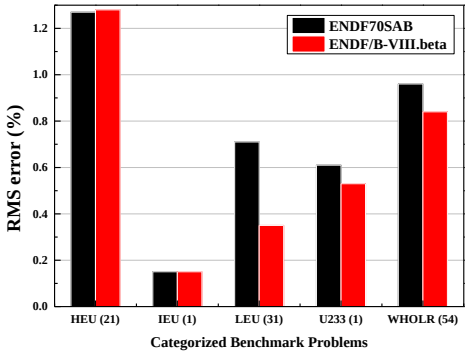


Fig.3. Comparison of RMS errors relative to benchmark K_{eff} values among different libraries.

4. Conclusion

In this study, 54 heavy water moderated/reflected benchmark calculations were carried out to confirm the effects of the improved thermal scattering data for heavy water in ENDF/B-VIII.β4. The ENDF/B-VIII.β4 library for heavy water consists of two chemical binding libraries of D in D_2O and O in D_2O, which is main difference from the former ENDF/B-VII.0 library with D binding in D_2O only. In order to confirm and validate the effects of each thermal library, differences of calculated k_{eff} from the benchmark k_{eff} are compared with each other and the RMS errors are also compared for each category. For HEU case, the results are similar in ENDF/B-VIII.β4 and ENDF/B-VII.0 scattering data. For LEU case, however, ENDF/B-VIII.β4 represents much better performance than ENDF70SAB. This implies the importance of the thermal scattering effect by oxygen interference in D_2O molecule.

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