New evaluation of thermal neutron scattering libraries for light and heavy water

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Abstract. In order to improve the design and safety of thermal nuclear reactors and for verification of criticality safety conditions on systems with significant amount of fissile materials and water, it is necessary to perform high-precision neutron transport calculations and estimate uncertainties of the results. These calculations are based on neutron interaction data distributed in evaluated nuclear data libraries. To improve the evaluations of thermal scattering sub-libraries, we developed a set of thermal neutron scattering cross sections (scattering kernels) for hydrogen bound in light water, and deuterium and oxygen bound in heavy water, in the ENDF-6 format from room temperature up to the critical temperatures of molecular liquids. The new evaluations were generated and processable with NJOY99 and also with NJOY-2012 with minor modifications (updates), and with the new version of NJOY-2016. The new TSL libraries are based on molecular dynamics simulations with GROMACS and recent experimental data, and result in an improvement of the calculation of single neutron scattering quantities. In this work, we discuss the importance of taking into account self-diffusion in liquids to accurately describe the neutron scattering at low neutron energies (quasi-elastic peak problem). To improve modeling of heavy water, it is important to take into account temperature-dependent static structure factors and apply Sköld approximation to the coherent inelastic components of the scattering matrix. The usage of the new set of scattering matrices and cross-sections improves the calculation of thermal critical systems moderated and/or reflected with light/heavy water obtained from the International Criticality Safety Benchmark Evaluation Project (ICSBEP) handbook. For example, the use of the new thermal scattering library for heavy water, combined with the ROSFOND-2010 evaluation of the cross sections for deuterium, results in an improvement of the C/E ratio in 48 out of 65 international benchmark cases calculated with the Monte Carlo code MCNP5, in comparison with the existing library based on the ENDF/B-VII.0 evaluation.

1 Introduction

The scattering of low energy neutrons with matter is described by the double differential scattering cross section. For scatterers without low energy resonances, the double differential cross section can be factorized as:

$$\sigma(E \to E', \mu) = \frac{\sigma_b}{2kT}\sqrt{\frac{E'}{E}}\mathcal{S}(\alpha,\beta) \tag{1}$$

where E and E' are the incident and secondary energies, μ is the cosine of the scattering angle in the laboratory system, σ_b is the bound atom scattering cross section, kT is the temperature in eV and $\mathcal{S}$ is the so called *thermal scattering law*. The scattering law encapsulates the condensed matter properties of the material, and (for isotropic materials) is dependant of two variables: the non-dimensional change in momentum α and the non-dimensional change in energy β:

$$\alpha = \frac{E' + E - 2\sqrt{E'E}\mu}{AkT} \quad \beta = \frac{E' - E}{kT} \tag{2}$$

where A is the ratio of the mass of the scatterer to the mass of the neutron.

In the evaluated nuclear data libraries the scattering law is tabulated over a grid of α and β values in the ENDF-6 format[1], and processing codes like NJOY[2] or GRUCON[3] are used to reconstruct the double differential scattering cross section using eqs. 1 and 2.

In this paper we briefly introduce the models used to evaluate the new libraries, and summarize the validation data collected since the libraries were first introduced in 2014.

2 Models

The new evaluations for light and heavy water are based on the CAB Model, which is an improvement over two older water thermal scattering evaluations: the General Atomics Model[4] and the IKE Model[5].

Compared with the General Atomics model used in ENDF/B-III to ENDF/B-VI, and the IKE model used in ENDF/B-VII and JEFF 3.2, the CAB Model introduces three main changes:

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- translational motion is modeled with the Egelstaff-Schofield diffusion model instead of free gas,
- the continuous spectrum is derived from molecular dynamics simulations[6] computed using an implementation of the TIP4P/2005-flexible water model[7] and the molecular dynamics code GROMACS[8],
- structure corrections are applied to heavy water using partial structure factors explicitly for deuterium and oxygen bound in heavy water.

The use of the Egelstaff-Schofield diffusion model leads to a better representation of the scattering for small exchanges of energy, but requires a refinement of the alpha and beta grids near zero. A slight discrepancy in the width of the quasielastic peak is still observable when this model is used and a possible solution was suggested[9], but it is not included in the distributed evaluation.

The processing code NJOY requires an increase of the size of the computing arrays and the precision of the calculations to process these libraries. A patch file with the necessary changes has been already submitted to the NJOY developers, and is included in the release of NJOY-2016.

More details of the models can be found in Refs. [10, 11]. The evaluations will be available in the new release of the evaluated nuclear data libraries ENDF/B-VIII.0 [12] and JEFF 3.3.

3 Experimental validation and benchmarking

The CAB Model was compared against double differential, quasi-elastic, angular differential, average scattering angle, and total cross section measurements. A selection of these calculations can be found in Ref. [10]. In addition to this, the libraries were compared with new heavy water angular differential measurements[13], and new total cross section measurements[14], and applied to the calculation of light water nuclear criticality benchmarks[11] from the ICSBEP handbook, and heavy water moderated nuclear criticality benchmarks[15] from the ICSBEP and IRPhE handbooks. For heavy water benchmarks, the use of the new library reduced $(C-E)/E$ in 39 of 65 benchmark cases compared to ENDF/B-VII data, and this number was increased to 48 when the heavy water libraries were combined with the ROSFOND-2010 evaluation for deuterium. More recently, the libraries were also applied to calculation of the isothermal reactor temperature for the EOLE[16] and IPEN/MB-01[17] critical facilities.

In all comparisons with experimental data, the new library performed equally or better than ENDF/B-VII. In some cases, the improvement found was very significant.

In particular, a very significant improvement was found in the temperature dependence of the total cross section for heavy water. One of the authors (D. Roubtsov) found an anomaly in the total cross section calculations when the ENDF/B-VII scattering kernel was used: the subthermal total neutron scattering cross section decreased when the temperature was increased (Fig. 1, top). This is an anomaly because the total cross section is expected to increase when temperature raises, caused by an increment in the upscattering probability. When the same calculations were repeated with the new model, no anomaly was found (Fig. 1, bottom).

To settle the dispute, the total cross section for heavy water at 20 and 50°C was measured in the Low Energy Neutron Source at Indiana University[14]. No anomaly was found in the experimental data, and the agreement with the new model was excellent (Fig. 2).

Comparison and analysis of the two models showed that the anomalous behavior of the total cross section in ENDF/B-VII is caused by a simplified description of the coherent component of the scattering cross section, which only includes deuterium-deuterium interference and is reduced too quickly when the temperature is increased from 293 to 350 K (Fig. 3, top). In the new model, deuterium-deuterium, deuterium-oxygen, and oxygen-oxygen interference terms are considered, and the correction factors used for the Sköld approximation are computed from molecular dynamics and validated with experimental structural data (Fig. 3, bottom). The structure factor corrections in the CAB Model change only slightly in this temperature range, and this is consistent with neutron diffraction measurements[18] which show that the structure of water changes only slightly over the whole liquid range.

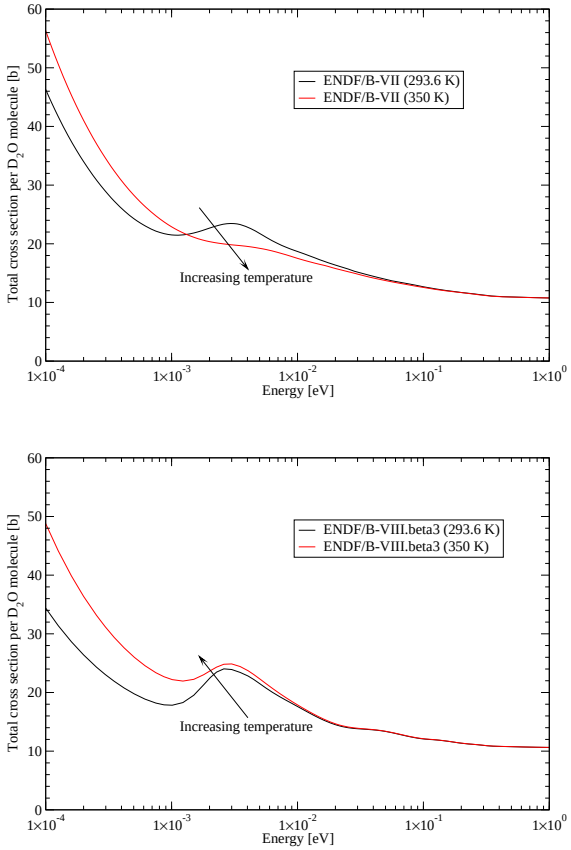

Figure 1. Total cross section for heavy water calculated with the ENDF/B-VII library (top), and the new model (bottom).

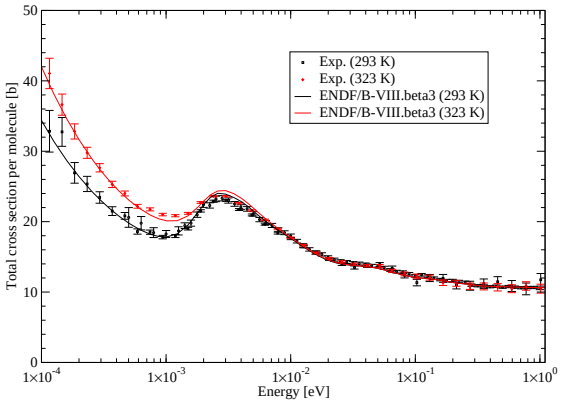

Figure 2. Total cross section for heavy water at 20 and 50 °C measured at the Low Energy Neutron Source, compared with calculations with the CAB Model.

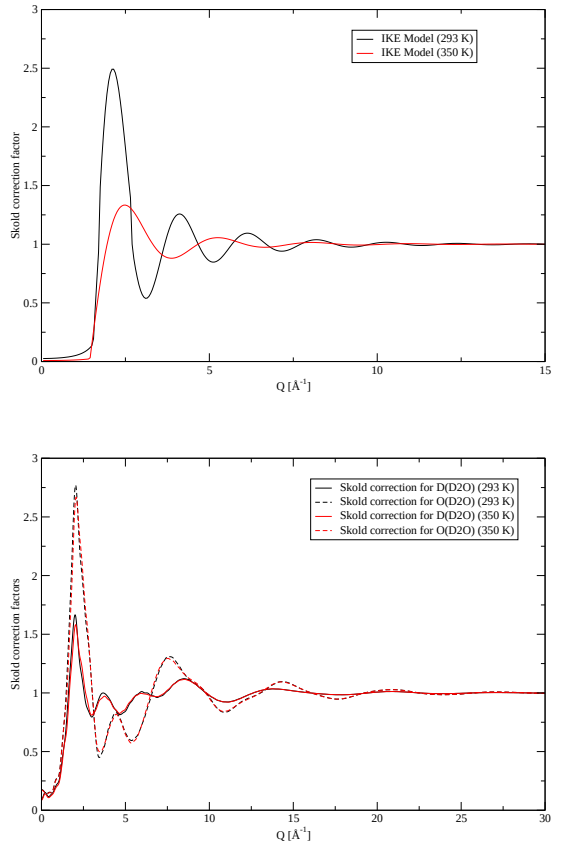

Figure 3. Sköld correction factors used in the IKE Model used in ENDF/B-VII (top) and the CAB Model (bottom).

4 Conclusions

The light and heavy water thermal scattering libraries produced from the CAB Model represent a state-of-the-art evaluation of the interaction of low energy neutrons with water. The libraries have been tested using experimental neutron interaction data and different types of reactor benchmarks, and the libraries perform equally or better than the evaluations available in previous versions of the evaluated nuclear data libraries.

The evaluations are accepted for release in ENDF/B-VIII.0 and JEFF 3.3, and are currently available for testing in the ENDF/A GForge server.

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