Improvement on the calculation of D₂O moderated critical systems with new thermal neutron scattering libraries

J.I. Márquez Damián [a],[*],[1], J.R. Granada [a], D. Roubtsov [b]

[a] Neutron Physics Department and Instituto Balseiro, Centro Atómico Bariloche, Argentina
[b] Atomic Energy of Canada Ltd., Chalk River Laboratories, Chalk River, ON K0J 1J0, Canada

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ABSTRACT

To improve the evaluations in thermal sublibraries, we developed a set of thermal neutron scattering cross sections (scattering kernels) for the deuterium and oxygen bound in heavy water in the ENDF-6 format. These new libraries are based on molecular dynamics simulations and recent experimental data, and result in an improvement of the calculation of single neutron scattering quantities. In this work, we show how the use of this new set of cross sections also improves the calculation of thermal critical systems moderated and/or reflected with heavy water. The use of the new thermal scattering library for heavy water, combined with the ROSFOND-2010 evaluation of the deuterium cross sections, results in an improvement of the C/E ratio in 48 out of 65 benchmark cases calculated with the Monte Carlo code MCNP5, in comparison with the existing library based on the ENDF/B-VII evaluation.

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1. Introduction

Accurate nuclear criticality calculations are needed both for the design of nuclear reactors and for the verification of nuclear criticality safety conditions on systems that include significant amounts of fissile material. These calculations are based on neutron interaction data, which is distributed in evaluated nuclear data libraries. ENDF/B-VII.1 Chadwick et al. (2011), the current version of the ENDF evaluated nuclear data library, is an advance over previous versions of the library in the calculation of critical systems (Kahler et al., 2011). However, a number of heavy water moderated critical systems still cannot be calculated within the uncertainty of $\pm 1\sigma$ of the experimental value of k_{eff} (the effective multiplication factor). This is also true for other nuclear data libraries (Morillon et al., 2013; van der Marck, 2012).

The reasons for these discrepancies have been traced to the evaluations for deuterium (Kozier et al., 2011; Morillon et al., 2013) and oxygen (Kozier et al., 2013; Roubtsov et al., 2013; Taylor and Hollenbach, 2013) but, for thermal systems, they could also be related to the thermal scattering cross section of deuterium and oxygen bound in heavy water. In this paper, we present results of nuclear criticality calculations for heavy water moderated and/or reflected benchmark systems that are particularly sensitive to

modifications in the thermal scattering cross sections (scattering kernels). Using these international benchmarks, we study the effect of a new evaluation of the thermal scattering cross section for heavy water at room temperature on the value of k_{eff} of the thermal critical systems at zero power.

1.1. Thermal scattering

For thermal neutrons, the scattering cross sections are described in terms of a function $S(\alpha, \beta)$ known as the *thermal scattering law*:

$$\Sigma_s(E, \hat{\Omega} \rightarrow E', \hat{\Omega}') = \frac{\sigma_b N}{4\pi kT} \sqrt{\frac{E'}{E}} S(\alpha, \beta). \quad (1)$$

Here α is a dimensionless parameter related to the neutron momentum transfer:

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

and β is a dimensionless parameter related to the energy transfer:

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

In these equations, A is the nuclide-to-neutron mass ratio, μ is the scattering cosine, and other notations are standard (Williams, 1966; MacFarlane, 2010).

The scattering law is a property of the material, and depends on its dynamics and structure. (In condensed matter physics, the thermal scattering law $S(\alpha, \beta)$ is known as the *dynamic structure factor*, $S(\vec{q}, \omega)$ (Lovesey, 1984), and $\alpha \propto (\hbar\vec{q})^2, \beta \propto \hbar\omega$). In the Gaussian incoherent approximation implemented in the nuclear data processing code NJOY, module LEAPR (MacFarlane, 1994), the dynamics of liquids is represented by a generalized frequency spectrum $\rho(\omega)$, and the structure is introduced with the Sköld coherent correction (Sköld, 1967), based on partial (static) structure factors $S_{ij}(q)$. For molecular liquids, the vibrational spectrum can be subdivided roughly into three major parts: intra-molecular (broadened molecular vibrations), inter-molecular (hindered rotations and translations, also called librations) and low-energy self-diffusion (translational) parts. The contribution of different atoms in the molecule can be distinguished by introducing partial $\rho_i(\omega)$ terms, similar to the partial phonon density of states in crystalline solids.

For the neutron scattering by ^{1}H in hydrogenous materials, one can disregard the coherent corrections and use the incoherent approximation for the scattering kernel $S_H(\alpha, \beta)$. For the thermal neutron scattering by ^{2}H($\equiv$ D) in deuterated liquids, the incoherent approximation is, strictly speaking, not applicable ($\sigma_{coh}(^2H) \simeq \sigma_{incoh}(^2H)$), and one can calculate the coherent inelastic part of $S_D(\alpha, \beta)$ using the Sköld method and the known partial structure factors of the molecular liquid ($S_{DD}(q), S_{OD}(q)$, and $S_{OO}(q)$).

The modern thermal scattering laws are distributed in the evaluated nuclear data libraries, as part of the thermal scattering sub-library (see, for example, Refs. Chadwick et al. (2011); MacFarlane and Kahler (2010)), following the ENDF-6 format (Trkov et al., 2011) for representing the numerical data.

2. Thermal scattering libraries for heavy water

2.1. Existing libraries

The evaluated nuclear data libraries include scattering law files for heavy water produced from two essentially different models: one initially published by Koppel and Young at General Atomics (Koppel and Houston, 1978) (GA model), and another proposed by Keinert and Mattes at Institut für Kernenergetik und Energiesysteme, Stuttgart (Keinert et al., 1984; Mattes and Keinert, 2005) (IKE model).

The IKE model includes several improvements over the GA model. While both models are based on frequency spectra originally measured by Haywood in the 1960s (Haywood, 1967), the IKE model incorporates newer measurements which include temperature dependence, $\rho(\omega; T)$. The IKE model also includes a correction for the coherent component of the scattering in deuterium, whereas the incoherent approximation is used in the GA model. However, the IKE coherent correction is not complete, because it only includes the D–D partial structure factor $S_{DD}(q; T)$. In both models, the translational self-diffusion of the liquid is approximated as a molecular free gas.

Oxygen is treated as free atomic gas at a given temperature T in both models. For the light water (H_2O) this is a well-justified approximation because the scattering cross section of oxygen is small compared to the cross section of ^{1}H and the scattering in hydrogen is predominant. For heavy water, $\sigma_s(^2H) \simeq \sigma_s(^{16}O)$, and both D and O components have similar importance. Moreover even-even nuclei (^{16}O and ^{18}O) constitute 99.96% of natural oxygen, causing coherent scattering (which is not included in the free gas approximation) to be predominant.

2.2. New libraries

The new libraries, called the *CAB models for water* (Marquez Damian et al., 2014), are based on experimental data and molecular dynamics (MD) simulations. To improve over the existing libraries, MD simulations (Marquez Damian et al., 2013) using GROMACS (Van Der Spoel et al., 2005) were performed to calculate the frequency spectrum $\rho_i(\omega; T)$ for deuterium and oxygen in liquid heavy water at a room temperature ($T \simeq 300$ K) and normal pressure (1 atm). It is known (Marti et al., 1996) that the modern MD simulation packages are capable of predicting accurately the vibrational frequency spectra using flexible models of water (González and Abascal, 2011), provided the simulations run long enough to cover the translational self-diffusion time scale $\simeq 10$ ps with a time step small enough ($dt \simeq 0.1$ fs) to resolve the intra-molecular vibrations.

Coherent corrections were done using experimental data. Partial structure factors $S_{ij}(q; T)$ measured by Soper (Soper and Benmore (2008)) were used to calculate the Sköld correction factors for deuterium and oxygen scattering kernels.

The CAB libraries were validated by comparison with many single-neutron scattering quantities obtained experimentally over years of studying heavy water with neutrons, and we found an improvement over the scattering law files available in the modern evaluated nuclear data libraries. Details on the models and their validation can be found in Ref. Marquez Damian et al. (2014).

To analyze the effects of these libraries on critical systems, we compared the measured values of the total neutron cross section for heavy water (Kropff et al., 1974) with calculations with our model, ENDF/B-VI (GA model) and ENDF/B-VII (IKE model) (Fig. 1). Although the improvement found at very low ($E < 1$ meV) energies is important, the neutron flux expected in the thermal critical systems is low at these energies (*e.g.*, in heavy water, we expect that the neutron flux is $\phi(E) \propto E \exp(-E/kT_{eff})$ at $E < 0.1 - 1.0$ eV with the neutron effective temperature $kT_{eff} \approx kT \simeq 30$ meV at room temperature conditions). Thus the improvement for cold neutrons would have little impact on the thermal systems.

On the other hand, the $\simeq 9\%$ discrepancies found at thermal energies ($E \simeq 10-30$ meV) between calculations using the existing evaluated data libraries and experimental data have more importance from the perspective of the thermal critical systems: the neutron flux is higher at this energy range. The reason for this
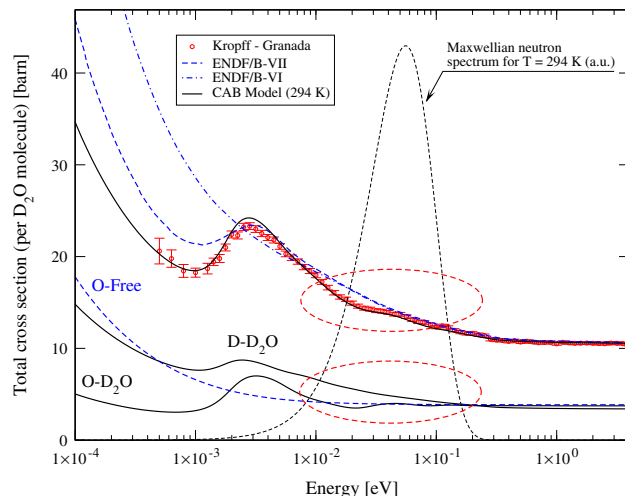

Fig. 1. Total cross sections for heavy water at 294 K (Kropff et al., 1974), compared with calculations using the CAB model, ENDF/B-VII (IKE model) and ENDF/B-VI (GA model) *vs.* the incident neutron energy. Ellipses mark the differences in the total cross sections at the energies which are very important for accurate modeling of the critical systems with the thermal neutron spectrum. The Maxwellian distribution for $T_{eff} = 294$ K, which would be expected for fully thermalized neutrons, is shown for reference. The differences in σ_{tot} can be traced to the total cross sections of O in D_2O (compare the curves at the bottom).