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Improvement on the calculation of D_2O moderated critical systems with new thermal neutron scattering libraries

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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.

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*:

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

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

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

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

$$\beta = \frac{E'-E}{kT}. \tag{3}$$

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).

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

The modern thermal scattering laws are distributed in the evaluated nuclear data libraries, as part of the thermal scattering sublibrary (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 $S_{\mathrm{DD}}(q; T)$. 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, $\sigma_s(^2\mathrm{H}) \simeq \sigma_s(^{16}\mathrm{O})$, 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 $\phi(E) \propto E \exp(-E/kT_{\mathrm{eff}})$ at $E < 0.1$–1.0 eV with the neutron effective temperature $kT_{\mathrm{eff}} \approx kT \simeq 30$ 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

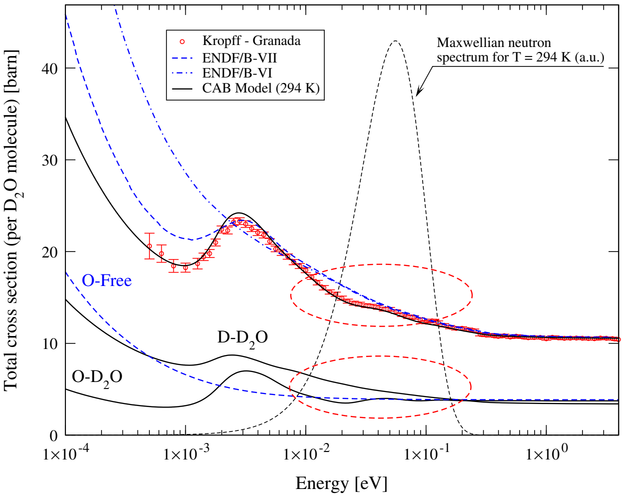

Fig. 1. Total cross sections for heavy water at 294 K (Kropff et al., 1974), compared with calculations using the CAB model, ENDF/B-VII (IKE model) and ENDF/B-VI (GA model) *vs.* the incident neutron energy. Ellipses mark the differences in the total cross sections at the energies which are very important for accurate modeling of the critical systems with the thermal neutron spectrum. The Maxwellian distribution for $T_{\mathrm{eff}} = 294$ 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).

discrepancy is that the dip found at $E \simeq 20$ meV in the total cross section is caused by interference effects in oxygen, and neither the GA model nor the IKE model include thermal scattering libraries for oxygen in heavy water. In our model, the inclusion of oxygen with an accurate description of its structure is reflected on the total cross section of O in D_2O, which differs considerably from the free-gas approximation used in the ENDF/B-VI and ENDF/B-VII thermal scattering laws.

3. Criticality benchmarks

The *International Criticality Safety Benchmark Evaluation Project* (ICSBEP) (Briggs, 2010) is a NEA-OECD project dedicated to compile, analyze and formally document critical experiments to be used as benchmarks for nuclear data and reactor calculation codes. The product of this effort are 558 reports or *evaluations* containing information on 4798 critical configurations, compiled in the *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, which is distributed annually as a DVD.

These evaluations include a description of all the important physical parameters (dimensions, compositions), and an analysis of the effect of their uncertainties in the multiplication factor of the system. For each system, a multiplication factor is given with its corresponding uncertainty, $k_{\mathrm{eff}}^{\mathrm{bench}} \pm \delta k_{\mathrm{bench}}$. Despite the system being critical ($k_{\mathrm{eff}}^{\mathrm{exp}} = 1.0$), the multiplication factor of the model $\left(k_{\mathrm{eff}}^{\mathrm{bench}}\right)$ could not be unity if simplifications were introduced in the preparation of the benchmark. The uncertainty associated to the multiplication factor includes not only the experimental error, but also the effect of the uncertainties in the parameters of the system (so that $\delta k_{\mathrm{bench}} \geqslant \delta k_{\mathrm{exp}}$).

To study the effect of the new CAB libraries on criticality calculations, we selected a series of heavy water moderated experiments from the ICSBEP Handbook (Table 1). As a baseline, all the systems were calculated using the KCODE mode of the Monte Carlo transport code MCNP5 1.60 (Brown, 2003) and the continuous-energy cross section data library based on the ENDF/B-VII.0 evaluation of nuclear data (Chadwick et al., 2006; Trellue et al., 2009).

The MCNP models of the heavy water ICSBEP benchmarks were run until a statistical uncertainty $\delta k_{\mathrm{eff}}^{\mathrm{calc}} \approx 20$ pcm was achieved, which is small compared with the benchmark uncertainties ($\delta k_{\mathrm{MCNP}}^{\mathrm{calc}} \ll \delta k_{\mathrm{bench}}$; 1 pcm is the change in k_{eff} by 1.0×10^{-5}).

3.1. Results

The results of the criticality calculations are shown in Fig. 2, expressed as the ratio of calculation over expected experimental value (the benchmark multiplication factor):

$$C/E = k_{\mathrm{eff}}^{\mathrm{calc}} / k_{\mathrm{eff}}^{\mathrm{bench}}.$$

These results were computed using ENDF/B-VII.0 based libraries for all isotopes, with the exception of the thermal scattering libraries for deuterium and oxygen, which were replaced with the new $S(\alpha,\beta)$ model.

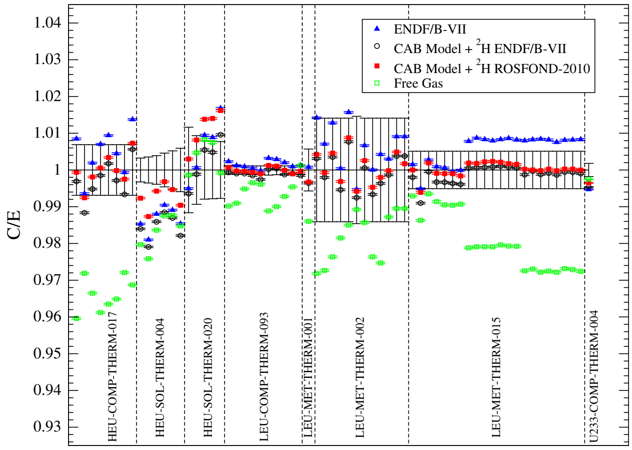

Fig. 2. Calculation/experiment ratio of the multiplication factor k_{eff} for 65 ICSBEP benchmarks modeling neutron criticality experiments in heavy water moderated/reflected systems.

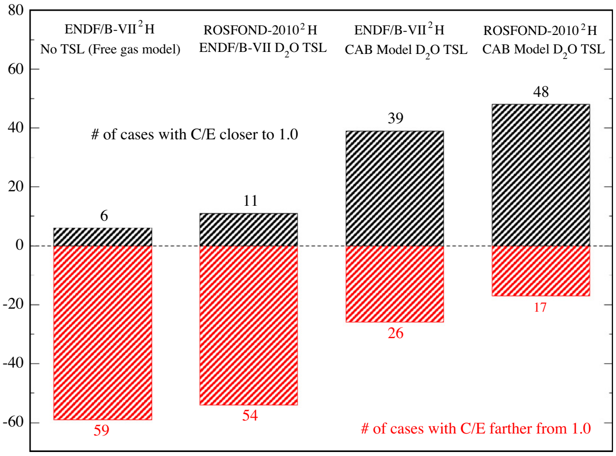

Fig. 3. Comparison of different combinations of ^{2}H library and thermal scattering libraries for heavy water, against results with ENDF/B-VII.0 cross sections. Values above zero represent the number of benchmark cases that show an improvement in the result, *i.e.* C/E being closer to unity than calculations performed with the ENDF/B-VII.0-based library.

The results obtained when the free gas approximation is used for heavy water are also shown in Fig. 2 to emphasize that the selected benchmarks are sensitive to the $S(\alpha,\beta)$ thermal scattering treatment for the neutrons interacting with deuterium and oxygen bounded in the heavy water.

The overall impact in the calculations with the CAB library is significant, with changes in the multiplication factor between

Table 1
Selection of ICSBEP criticality benchmarks used in the comparison.

Evaluation ID	# Of Cases	Title
HEU-COMP-THERM-017	8	RB reactor: lattices of 80%-enriched uranium elements in heavy water
HEU-SOL-THERM-004	6	Reflected uranyl-fluoride solutions in heavy water
HEU-SOL-THERM-020	5	Unreflected cylinders of uranyl-fluoride solutions in heavy water
LEU-COMP-THERM-093	10	Deuterium critical assembly with 1.2% enriched uranium varying coolant void fraction and lattice pitch
LEU-MET-THERM-001	1	RB reactor: natural-uranium rods in heavy water
LEU-MET-THERM-002	12	RB reactor: latices of 2%-enriched uranium elements in heavy water
LEU-MET-THERM-015	22	RB reactor: fuel assemblies substitution criticality experiments in lattices of 2%-enriched uranium in heavy water
U233-COMP-THERM-004	1	D_2O moderated lattice of $^{233}UO_2$–$^{232}ThO_2$

Table 2
Benchmark and computed multiplication factor for all considered benchmark systems. Benchmark and statistical uncertainty values in pcm for each multiplication factor are listed in parentheses. Calculated results in bold are within $\pm 1\sigma$ of the benchmark value; calculated results marked with ✓ are closer to the benchmark value than the reference ENDF/B-VII-based calculation, whereas calculated results marked with × are farther from the benchmark value than the reference calculation.

Benchmark case ID	Benchmark	ENDF/B-VII	CAB model +ENDF/B VII ^{2}H	CAB model + ROSFOND-2010 ^{2}H
HEU-COMP-THERM-017-001	1.00000(690)	1.00849(23)	**0.99690(24)**✓	**0.99939(24)**✓
HEU-COMP-THERM-017-002	1.00000(690)	**0.99357(23)**	0.98835(23)×	0.99243(23)×
HEU-COMP-THERM-017-003	1.00000(690)	**1.00192(22)**	**0.99481(22)**×	**0.99810(22)**✓
HEU-COMP-THERM-017-004	1.00000(690)	1.00701(22)	**0.99852(21)**✓	**1.00054(21)**✓
HEU-COMP-THERM-017-005	1.00000(690)	1.00942(20)	**1.00174(21)**✓	**1.00339(21)**✓
HEU-COMP-THERM-017-006	1.00000(690)	**1.00447(22)**	**0.99713(22)**✓	**0.99982(22)**✓
HEU-COMP-THERM-017-007	1.00000(690)	**0.99935(23)**	**0.99335(23)**×	**0.99750(22)**×
HEU-COMP-THERM-017-008	1.00000(690)	1.01374(20)	**1.00562(20)**✓	1.00728(20)✓
HEU-SOL-THERM-004-001	1.00000(325)	0.98533(20)	0.98400(20)×	0.99233(20)✓
HEU-SOL-THERM-004-002	1.00000(355)	0.98101(20)	0.97907(20)×	0.98734(20)✓
HEU-SOL-THERM-004-003	1.00000(390)	0.98802(21)	0.98591(21)×	0.99425(20)✓
HEU-SOL-THERM-004-004	1.00000(460)	0.99044(22)	0.98846(21)×	**0.99681(21)**✓
HEU-SOL-THERM-004-005	1.00000(520)	0.98910(22)	0.98695(22)×	0.99467(22)✓
HEU-SOL-THERM-004-006	1.00000(590)	0.98537(23)	0.98212(22)×	0.99040(22)✓
HEU-SOL-THERM-020-001	0.99660(1160)	**0.99158(23)**	**0.99019(24)**×	**0.99957(24)**✓
HEU-SOL-THERM-020-002	0.99560(930)	**0.99624(25)**	**0.99446(26)**×	**1.00373(25)**×
HEU-SOL-THERM-020-003	0.99570(790)	1.00511(26)	**1.00113(26)**✓	1.00944(26)×
HEU-SOL-THERM-020-004	0.99550(780)	1.00433(26)	**1.00029(26)**✓	1.00946(24)×
HEU-SOL-THERM-020-005	0.99590(770)	1.01262(25)	1.00545(25)✓	1.01201(25)✓
LEU-COMP-THERM-093-001	1.00049(120)	1.00281(19)	**0.99983(20)**✓	**1.00121(20)**✓
LEU-COMP-THERM-093-002	1.00064(120)	1.00184(21)	**0.99968(21)**✓	**1.00037(20)**✓
LEU-COMP-THERM-093-003	0.99794(110)	**0.99886(20)**	**0.99697(21)**×	**0.99752(21)**✓
LEU-COMP-THERM-093-004	0.99981(110)	**1.00030(22)**	0.99831(22)×	**0.99918(22)**×
LEU-COMP-THERM-093-005	0.99856(110)	**0.99835(21)**	0.99601(22)×	**0.99756(22)**×
LEU-COMP-THERM-093-006	1.00085(140)	1.00407(20)	**1.00106(20)**✓	**1.00208(19)**✓
LEU-COMP-THERM-093-007	1.00035(130)	1.00325(21)	**1.00076(20)**✓	**1.00139(19)**✓
LEU-COMP-THERM-093-008	0.99946(110)	1.00149(21)	0.99822(21)✓	**0.99937(22)**✓
LEU-COMP-THERM-093-009	0.99992(110)	**1.00084(22)**	**0.99896(23)**×	**0.99889(22)**×
LEU-COMP-THERM-093-010	0.99824(110)	0.99947(22)	0.99675(23)×	**0.99787(22)**✓
LEU-MET-THERM-001-001	0.99900(570)	**0.99978(24)**	**0.99543(24)**×	**0.99573(24)**×
LEU-MET-THERM-002-001	1.00000(1410)	1.01421(22)	**1.00310(23)**✓	**1.00422(23)**✓
LEU-MET-THERM-002-002	1.00000(1410)	**1.00707(24)**	**0.99798(23)**✓	**0.99926(23)**✓
LEU-MET-THERM-002-003	1.00000(1410)	**1.01283(24)**	**1.00352(24)**✓	**1.00456(24)**✓
LEU-MET-THERM-002-004	1.00000(1410)	**1.00041(23)**	**0.99460(24)**×	**0.99687(23)**×
LEU-MET-THERM-002-005	1.00000(1410)	1.01568(22)	**1.00768(24)**✓	**1.00877(23)**✓
LEU-MET-THERM-002-006	1.00000(1460)	**0.99473(23)**	**0.99246(22)**×	**0.99420(22)**×
LEU-MET-THERM-002-007	1.00000(1410)	**1.00666(21)**	**1.00053(21)**✓	**1.00255(22)**✓
LEU-MET-THERM-002-008	1.00000(1410)	**1.00004(24)**	**0.99335(24)**×	**0.99530(24)**×
LEU-MET-THERM-002-009	1.00000(1410)	**1.00425(24)**	**0.99637(23)**✓	**0.99796(24)**✓
LEU-MET-THERM-002-010	1.00000(1410)	**1.00301(22)**	**0.99867(22)**✓	**0.99980(22)**✓
LEU-MET-THERM-002-011	1.00000(1410)	**1.00910(21)**	**1.00379(21)**✓	**1.00489(21)**✓
LEU-MET-THERM-002-012	1.00000(1410)	**1.00910(21)**	**1.00379(21)**✓	**1.00170(21)**✓
LEU-MET-THERM-015-001	1.00000(510)	**1.00149(23)**	**0.99801(23)**×	**1.00000(23)**✓
LEU-MET-THERM-015-002	1.00000(510)	0.99489(23)	0.99098(23)×	0.99390(24)×
LEU-MET-THERM-015-003	1.00000(510)	**1.00276(23)**	**0.99953(24)**✓	**1.00192(23)**✓
LEU-MET-THERM-015-004	1.00000(510)	**1.00090(23)**	**0.99671(23)**×	**0.99910(23)**×
LEU-MET-THERM-015-005	1.00000(510)	**1.00059(22)**	**0.99666(23)**×	**0.99899(23)**×
LEU-MET-THERM-015-006	1.00000(510)	**0.99952(23)**	**0.99635(23)**×	**0.99898(23)**×
LEU-MET-THERM-015-007	1.00000(510)	**0.99996(23)**	**0.99611(22)**×	**0.99838(23)**×
LEU-MET-THERM-015-008	1.00000(510)	1.00783(22)	**1.00052(23)**✓	**1.00188(22)**✓
LEU-MET-THERM-015-009	1.00000(510)	1.00870(22)	**1.00063(22)**✓	**1.00178(22)**✓
LEU-MET-THERM-015-010	1.00000(510)	1.00832(22)	**1.00073(22)**✓	**1.00222(23)**✓
LEU-MET-THERM-015-011	1.00000(510)	1.00797(23)	**1.00083(23)**✓	**1.00233(22)**✓
LEU-MET-THERM-015-012	1.00000(510)	1.00832(21)	**1.00105(22)**✓	**1.00213(22)**✓
LEU-MET-THERM-015-013	1.00000(510)	1.00863(22)	**1.00081(23)**✓	**1.00172(22)**✓
LEU-MET-THERM-015-014	1.00000(510)	1.00820(23)	**1.00055(22)**✓	**1.00152(23)**✓
LEU-MET-THERM-015-015	1.00000(510)	1.00799(23)	**0.99872(24)**✓	**1.00022(23)**✓
LEU-MET-THERM-015-016	1.00000(510)	1.00826(23)	**0.99968(24)**✓	**1.00000(23)**✓
LEU-MET-THERM-015-017	1.00000(510)	1.00842(23)	**0.99879(23)**✓	**0.99981(23)**✓
LEU-MET-THERM-015-018	1.00000(510)	1.00822(23)	**0.99914(23)**✓	**1.00025(23)**✓
LEU-MET-THERM-015-019	1.00000(510)	1.00756(23)	**0.99868(24)**✓	**0.99978(23)**✓
LEU-MET-THERM-015-020	1.00000(510)	1.00813(23)	**0.99944(23)**✓	**1.00026(24)**✓
LEU-MET-THERM-015-021	1.00000(510)	1.00820(22)	**0.99937(24)**✓	**1.00024(23)**✓
LEU-MET-THERM-015-022	1.00000(510)	1.00831(23)	**0.99902(23)**✓	**0.99997(23)**✓
U233-COMP-THERM-004-001	1.00170(180)	0.99640(12)	0.99704(11)✓	0.99817(12)✓

−1159 and 36 pcm. With the exception of the case corresponding to the ^{233}U–^{232}Th fueled system (U233-COMP-THERM-004), all the remaining cases show a decrease in the multiplication factor. As the calculations using the ENDF/B-VII library tend to overestimate the multiplication factor of heavy water moderated systems (van der Marck, 2006), this reduction implies an improvement (*C*/*E* ratio closer to unity) in 39 of the 65 cases (60%).

This first analysis takes into consideration only the change of a single aspect in the microscopic nuclear data (the thermal scattering kernel of heavy water) in a complex calculation of heterogeneous critical systems. Considering this, modifying only one part of the cross sections might not necessarily lead to a visible improvement in the k_{eff} results, with discrepancies due to other imperfections of the nuclear data used for the modeling. In particular, some of the heavy water benchmarks considered in this work are known to be calculated better using older nuclear data libraries, which can be seen in the work by van der Merck van der Marck (2006) and Taylor and Hollenbach Taylor and Hollenbach (2013). For example, 8 of the 11 cases from HEU-SOL-THERM-004 and HEU-SOL-THERM-020 benchmarks analyzed by Taylor and Hollenbach are among the 22 which, at first, are not improved by using our new thermal scattering library. However, by substituting specific isotopes in the reference nuclear data library, it was found that most of the differences are attributable to the deuterium evaluation in ENDF/B-VII Taylor and Hollenbach (2013); Kozier et al. (2011).

One possible solution to these discrepancies is to replace the ENDF/B-VII evaluation of deuterium by the evaluation included in the ROSFOND-2010 library (Zabrodskaya et al., 2007). This evaluation has a slightly modified free atom scattering cross section for deuterium ($\sigma_s^{\mathrm{free}} = 3.390$ b against $\sigma_s^{\mathrm{free}} = 3.395$ b in the ENDF/B-VII library, but the differences in the elastic neutron scattering are within the uncertainty of σ_s^{free} (Mughabghab, 2006)). There are also small differences in the neutron capture (*e.g.*, $\sigma_{\mathrm{th}}(n,\gamma) = 0.519$ mb against 0.506 mb in the ENDF/B-VII library), and noticeable differences in the angular distributions of the elastic scattering at high neutron energies ($10\ \mathrm{keV} < E < 3.2\ \mathrm{MeV}$). Although the differences at the low neutron energies are small, if the new thermal scattering library is combined with this evaluation for deuterium, the observed effect in the fissile solution systems is reverted, resulting in 48 of 65 benchmark cases (74%) improving over the baseline ENDF/B-VII-based calculation. For comparison, replacing only the deuterium evaluation for the one in ROSFOND-2010 but not using the new thermal scattering library improves the results of the HEU-SOL-THERM-004 and HEU-SOL-THERM-020 benchmarks (highly enriched uranium fluoride solutions in D_2O), but affects negatively other benchmarks, and overall only 11 of the 65 benchmark cases yield better results using this combination (see Fig. 3).

Thus we found that if these changes are applied (*i.e.*, ROSFOND-2010 ^{2}H evaluation + CAB Model for D and O in D_2O), the number of benchmarks that can be calculated within the uncertainty of $\pm 1\sigma$ is increased from 27 in 65 (42%) to 53 in 65 (82%); see Table 2.

4. Conclusions

The new thermal scattering law files for heavy water represent an improvement over existing scattering law files available in the modern evaluated nuclear data libraries. Part of these improvements are in the cold neutron energy range, which have little impact on the thermal critical systems, but other improvements – related to a better representation of the neutron scattering in oxygen – are in the region where the neutron spectrum is important in the heavy water moderated systems.

When the new thermal scattering libraries are applied to the calculation of neutron criticality benchmarks, we find a significant (up to 1100 pcm) difference in the results of multiplication factors, improving the calculation in 60% of the cases. The average change in the multiplication factor is 543 pcm.

If this new thermal scattering library is combined with the evaluation of deuterium cross sections distributed in the ROSFOND-2010 library, then 74% of the calculations yield better results than those of the baseline benchmark set obtained with the ENDF/B-VII.0 based nuclear data library.

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