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Perturbation scheme for estimating uncertainties in thermal scattering cross sections of water

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ABSTRACT

Neutron-moderator scattering interactions in the thermal energy range are often treated with specialised cross section data derived from the Thermal Scattering Law S(α,β). This scattering law is calculated based on theoretical and experimental models that describe the structure and dynamics of the principle moderator molecule. In this work, a perturbation scheme based on the Total Monte Carlo method is described and the uncertainties of the parameters used to calculate S(α,β) for H in H_2O and D and O in D_2O from the ENDF/B-VIII.0 library were estimated and propagated through to the cross sections and to an integral criticality scenario in the OPAL Reactor, Sydney Australia, using the transport code Serpent. The calculated uncertainties in the total cross sections are in reasonable agreement with experimental data and provide a basis for future model refinement; uncertainties in several parameters used in the LEAPR of NJOY were identified as critical to specific energy regions and behaviours of the total cross sections. The effect on criticality of these uncertainties was found to be 48 pcm and 41 pcm for H_2O and D_2O, respectively, within the OPAL Reactor during a low-power configuration.

1. Introduction

Neutron moderators are an important material in thermal fission reactors, providing the vast majority of the neutron thermalisation required to realise a critical system. The most typical moderators used today are overwhelmingly light (H_2O) and heavy water (D_2O), often serving as both a coolant and moderator. At lower incident energies, a neutron can interact with the molecular structure of a material as well as the nucleus of a given atom and as such, requires a specialised treatment of interaction probability; momentum and energy exchange between the water molecule and an incident neutron can take place via translation, rotation, libration and vibration of the water molecule. Using nuclear data processing codes, such as NJOY, the thermal scattering cross section of such materials is calculated from the thermal scattering law S(α,β).

Up until recently, the thermal scattering data used in the major nuclear data libraries was derived from two models: from GA model (MacFarlane, 1994) and the IKE model (Mattes, 2005) based on experimental data compiled by Koppel and Houston (1978). Now, the new evaluation ENDF/B-VIII.0 library utilises a new model for H_2O and D_2O: the Centro Atomico Bariloche (CAB) model for water (Brown et al., 2018) (Márquez Damián et al., 2014). The new JEFF-3.3 library also uses the D and O in D_2O obtained from the CAB model. This model is based on molecular dynamics (MD) data obtained using the TIP4P-2005f flexible model for water (González and Abascal, 2011) used for calculation of the continuous frequency spectrum, vibrational modes and partial structure factors (for D_2O), and experimental data from Novikov used in calculation of the diffusion parameters (Novikov et al., 1990).

While the validation of the CAB models for water look promising, an estimation of the uncertainty of the data is important. In this work we present a methodology for parameterisation of the H_2O and D_2O models and a perturbation scheme based on the Fast Total Monte Carlo (TMC) method (Rochman et al., 2014). This will allow estimates of uncertainties on successive calculated quantities using these thermal scattering cross sections as well as offer a basis for future model refinement.

2. CAB model

2.1. Key parameters of thermal scattering law models

The expression for the double differential thermal scattering cross section is given by (Parks, 1970):

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

where E and E' are the incident and secondary neutron energies, σ_b is the bound scattering cross section, kT is the temperature in eV, α is the dimensionless momentum transfer, β is the dimensionless energy transfer and $S(\alpha,\beta)$ is the symmetric form of the scattering law. The bound scattering cross section which is related to the free atom cross section by:

$$\sigma_b = \frac{(A+1)^2}{A^2}\sigma_{free} \quad (2)$$

$S(\alpha,\beta)$ can be calculated using parameters derived from a physical model of a material, typically using a nuclear data processing code such as LEAPR in the NJOY package (Muir et al., 2012). Prior to 2017, the major evaluations (e.g. ENDF and JEFF) were produced largely based on two models: the General Atomics (GA) model (up to ENDF/B-VI.8) and the IKE model (JEFF-3.2, ENDF/B-VII.1) both building primarily off the work conducted by Nelkin (1960) and Haywood and Thorson (1962). Presently, new models, called the CAB models for water, based largely on MD simulations and newer experimental data are being used in ENDF/B-VIII.0 and JEFF-3.3 (JEFF-3.3 uses only D and O in D_2O from the CAB model). Full details of the CAB models can be found in (Márquez Damián et al., 2014). Table 1 contains the key scalar parameters for these models.

In addition to the scalar parameters are three vector quantities, the alpha and beta grids and the continuous frequency spectrum (CFS). The specifics of the alpha and beta grids are not so important as to warrant comparison. The CAB model CFS for H in H_2O differs from the CFS used in previous evaluations in a few important ways: the diffusion component to be added back into the spectrum in LEAPR uses the Egelstaff-Schofield approximation (Egelstaff and Schofield, 1962), rather than the free-gas approximation; the presence of a translational mode around 6 meV represented in the CFS, experimentally observed in (Bellissent-Funel et al., 1995); a narrower rotational mode around 60 meV and an adjusted weight. Fig. 1 contains the CFS used in the last several evaluations for H in H_2O, adjusted to reflect the spectrum weights associated with each evaluation.

The CAB model for D in D_2O has similar differences with a narrower rotational band around 48 meV, the Egelstaff-Schofield diffusion model and increased weight. The IKE model used in JEFF-3.2 and ENDF/B-VII.1, however, also featured a translational vibrational mode in the CFS, in the form of a Debye distribution with a Debye temperature of ~20 meV. The CAB model for D in D_2O is used in ENDF/B-VIII.0 and JEFF-3.3. Fig. 2 contains the CFS used in the last several evaluations for D in D_2O.

The GA model assumes the scattering of D in D_2O as completely incoherent, as Koppel and Young (Koppel and Young, 1965) showed that although coherent scattering has a significant effect on the total cross section, its effect on thermalisation is of second order. The IKE model does include a structure factor for D in D_2O and uses the Sköld method to apply a coherent correction to the scattering law in LEAPR. This structure factor was obtained using a Lennard-Jones model.

In all the models discussed above, the oxygen atom in H_2O was treated as a free-gas scatterer with a mass of 16. The GA and IKE models also used a free-gas scatter for the oxygen atom in D_2O,

Table 1
Key LEAPR scalar parameters for H in H_2O and D in D_2O from the GA, IKE and CAB models.

Parameter	GA		IKE		CAB	
	H_2O	D_2O	H_2O	D_2O	H_2O	D_2O
Free-atom cross section σ_{free}	20.36b	3.37b	20.478b	3.395b	20.436b	3.395b
Diffusion weight w_d*	0.05556	0.05	0.02174	0.05	0.007918	0.01629
Oscillator 1 energy E_1	0.205 eV	0.142 eV	0.205 eV	0.145 eV	0.205 eV	0.15 eV
Oscillator 2 energy E_2	0.48 eV	0.305 eV	0.436 eV	0.338 eV	0.415 eV	0.205 eV
Oscillator 1 wt w_1*	0.1667	0.1667	0.1630	0.1667	0.1567	0.14293
Oscillator 2 wt w_2*	0.3333	0.3333	0.3261	0.3333	0.3133	0.2925
Continuous spectrum weight w_β*	0.4444	0.45	0.4891	0.45	0.5221	0.5449

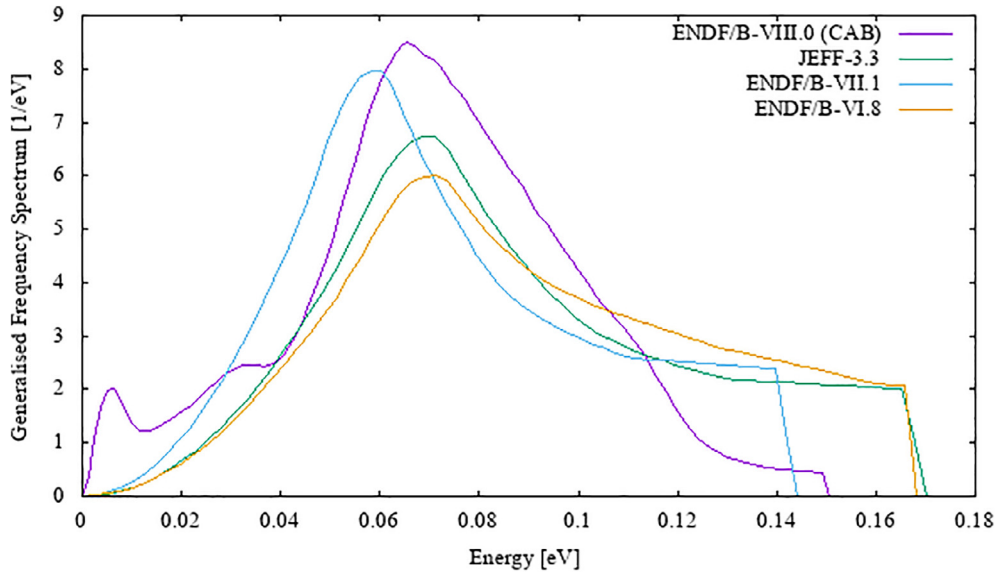

Fig. 1. CFS for H in H_2O at room temperature from previous evaluations up to the present. The CAB model is used in the ENDF/B-VII.0 evaluation.