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Measurement of the total cross section of heavy water in the 0.1 meV–1 eV energy range at 20 and 50 °C

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Summary. — Despite the importance of heavy water as a neutron moderator, there are few measurements of its total neutron cross section for cold and thermal energies, and none of them covers the range of temperature (40–70 °C) used in moderator and reflector tanks in research reactors, and in CANDU nuclear power plants. To cover this deficit, we measured the total cross section of liquid heavy water at 20 °C and 50 °C using the SANS beamline at the LENS facility at Indiana University. The time-of-flight technique was used, in a sample-in/sample-out measurement. The use of the solid methane cold neutron source at LENS allowed measuring in a broad range in energy, from 0.1 meV to 1 eV. In this paper we present details of the measurement and processing of the data, and comparison with previous experimental measurements and calculation models. This work is included in the Action Plan of the IAEA Coordinated Research Project "Advanced Moderators for Intense Cold Neutron Beams in Materials Research".

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1. – Introduction

Heavy water is one of the most widely used neutron moderators, and is used as a neutron reflector and as a moderator/coolant in both research and power reactors (table I). Despite this, there are few measurements of its total cross section in the cold and thermal energy range and none of them covers the temperature range used in nuclear reactors as moderator or reflector (40–70 °C).

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TABLE I. – *Total cross section measurements for heavy water available in EXFOR.*

Data set	EXFOR #	Year	Temperature [°C]
Kropff	30283002	1974	20
Dritsa	20038001	1971	22, 200
Meyers	11019001	1953	N/A (room temperature)
Rainwater	11145001	1948	N/A (room temperature)

This lack of data is particularly problematic because the model developed by Keinert and Mattes [1] and currently used for the calculation of the thermal neutron scattering cross section in heavy water in the major evaluated nuclear data libraries predicts an anomalous behavior of the total cross section when the temperature changes. Figure 1 shows the total cross section calculated from ENDF/B-VII data at 293.6 K and 350 K [2]. At energies around 3 meV this model predicts a reduction of the total cross section with increasing temperature. Although possible, this behavior would constitute an anomaly in the expected increase of the total cross section caused by the increased upscattering with temperature.

2. – Experimental method

The total cross section was measured using the time-of-flight technique using the SANS beamline of the Low Energy Neutron Source (LENS) at the Center for the Exploration of Energy and Matter in Indiana University [3]. The transmission set up was prepared in the sample area of the SANS beamline (fig. 2), using a six position linear sample changer and a 1″, 50 kPa ^{3}He detector. The detector was located inside a boron

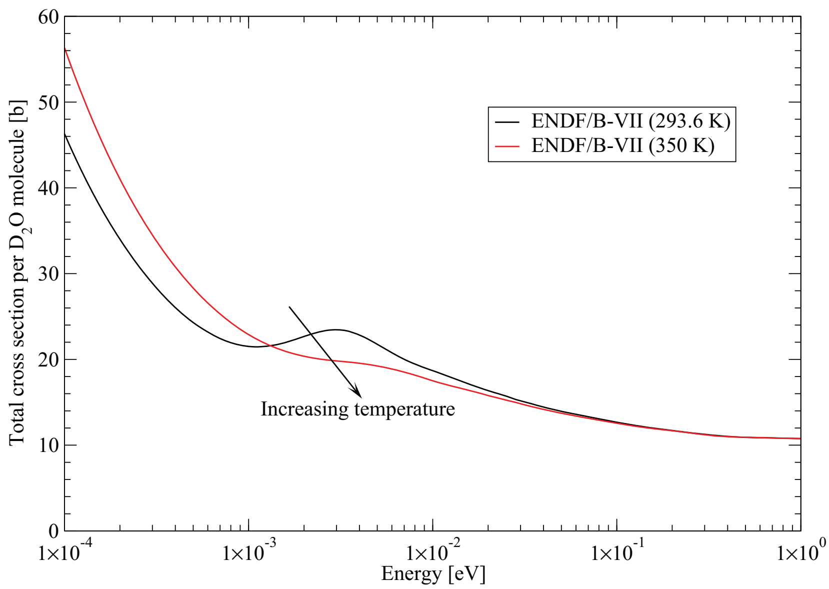

Fig. 1. – Total cross section of heavy water at 293 and 350 K, according to the Keinert and Mattes model.

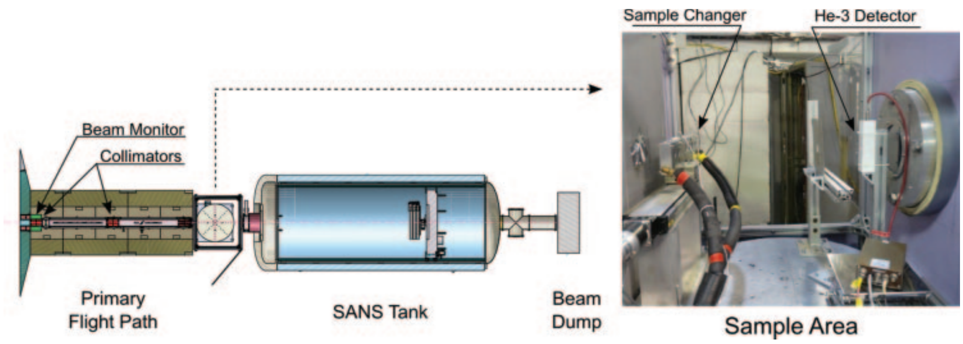

Fig. 2. – SANS beamline (left). Detail of sample area with transmission set up (right).

nitride shield with a 15×10 mm (H $\times$ W) aperture facing the beam shield to reduce background. A 20×20 cm Cd sheet with a 2 cm hole was located between the sample changer and the detector to reduce sample dependent background by in-scattering. The total flight path from the source was 8.80 m.

A D_2O (99.99% purity) sample was sealed inside a 5.00 mm 120-QS Hellma quartz cuvette, and different cuvette was used for sample-out. The cuvettes were cleaned with heavy water and blown dry with N_2 before filling. Heat conduction between the sample holder and the cuvettes was improved by applying silver paste on the sides. Sample temperature was controlled within $\pm 0.1\,^{\circ}$C.

Measurements were performed with the sample-in/sample-out technique, alternating a position with an empty clean cell with the 5.00 mm sample. A background rate of ~ 0.2 counts/min was determined before the measurements; this count rate, compared with a sample-in count rate of $\sim 10^4$ counts/min for the 10 Hz, 150 μs pulse-width proton beam, is sufficiently low that measurement of the background spectrum is rendered unnecessary.

Three sample-in measurements and three sample out measurements were considered for the processing at 20 °C (six sample-in and six sample-out at 20 °C), each consisting in 2×10^6 monitor counts (~ 1 hr of beam time). The time-of-flight spectra was computed with logarithmic binning (20 bins/dec), and corrected for mean emission time [4] and dead time, using $\tau = 4\,\mu$s. The first 300 μs of the spectra were discarded, and the measurements were normalized by monitor counts, resulting in two quantities: I_{SI}, and I_{SO} the beam intensity in the sample in and sample out positions, respectively.

With these quantities, a first approximation to the total cross section was computed:

$$\sigma_t = \frac{-1}{N\Delta x} \log\left(\frac{I_{\mathrm{SI}}}{I_{\mathrm{SO}}}\right),$$

with $N = \frac{\rho N_{\mathrm{A}}}{M}$ ($\rho_{20\,^{\circ}\mathrm{C}} = 1.105\,\mathrm{g/cm^3}$, $\rho_{50\,^{\circ}\mathrm{C}} = 1.096\,\mathrm{g/cm^3}$, $M = 20.0276\,\mathrm{g/mol}$ [5]). This first approximation resulted in a 3% difference with the data from Kropff, a systematic discrepancy that could be traced to differences in the optical thickness of the different cells, and which is usually solved by normalization to the free gas cross section which is reached assymptotically at high energies. The data obtained in our measurement had poor statistics in the epithermal region to adjust with the free gas cross section; therefore, we decided to use the data by Kropff as a reference, renormalizing the data sets with a constant $C = 1.03$.

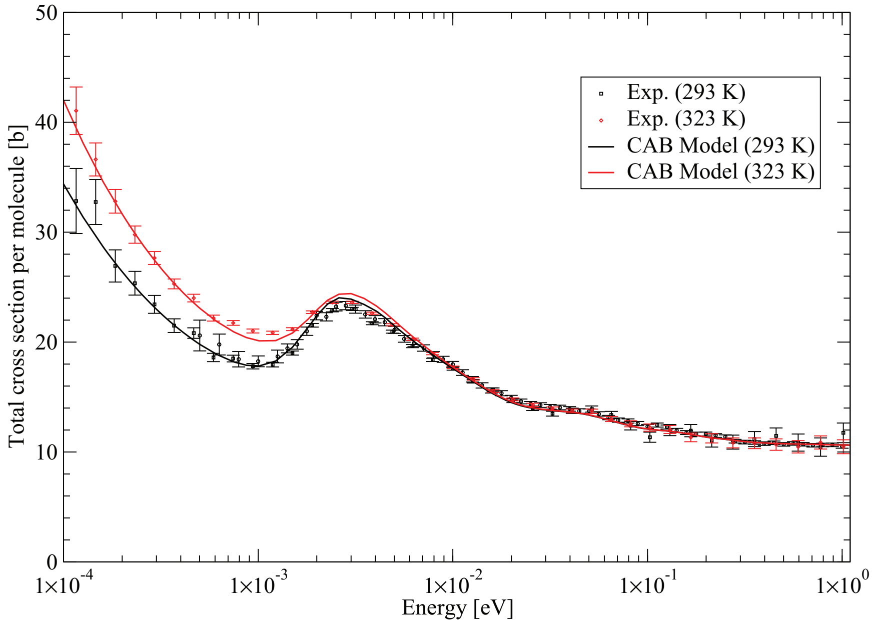

Fig. 3. – Measured total cross section of heavy water at 293 and 323 K, compared with CAB model calculations.

3. – Results and conclusions

The total cross section obtained at 20 and 50 °C (293 and 323 K) after the renormalization explained in the previous section is shown in fig. 3. The data is compared with the calculations with the CAB Model [6] at the same temperatures.

The results do not show the abnormal temperature behaviour predicted by the model developed by Keinert and Mattes: on the contrary, there is an increment of the total cross section with temperature for all thermal energies. It is also worth to note that the structural features present in the total cross section of heavy water at room temperature (*i.e.* the dips present at $E \approx 1\,\text{meV}$ and at $E \approx 20\,\text{meV}$, and the shoulder at $E \approx 3\,\text{meV}$) are also present in the cross section at 50 °C. This is evidence of the strong effect of structure of water above room temperature, which is consistent with diffraction measurements [7], and only considered in part in the Keinert and Mattes model.

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REFERENCES

[1] Mattes M. and Keinert J., *Thermal neutron scattering data for the moderator materials H2O, D2O and ZrHx in ENDF-6 format and as ACE library for MCNP (X) codes.*, Technical Report INDC (NDS)-0470 (IAEA) 2005.
[2] Chadwick M. B., Herman H. *et al.*, *Nucl. Data Sheets*, **112** (2011) 2887.
[3] Baxter D. V., Cameron J. M. *et al.*, *Nucl. Instrum. Methods B*, **241** (2005) 209.
[4] Lavelle C. M., Baxter D. V. *et al.*, *Nucl. Instrum. Methods A*, **587** (2008) 324.
[5] Hill P. G., MacMillan R. D. and Lee V., *Tables of thermodynamic properties of heavy water in SI units.*, Technical Report (Atomic Enery of Canada Ltd.) 1981.
[6] Marquez Damian J. I., Malaspina D. C. and Granada J. R., *Ann. Nucl. Energy*, **65** (2014) 280.
[7] Soper A. K., *Chem. Phys.*, **258** (2) 121.