Neutron thermalization in heavy water–Measurement and calculation of spectra[1]

By M. Abbate and J. Lolich

Comisión Nacional de Energía Atómica Centro Atómico Bariloche 8400 San Carlos de Bariloche, (R.N.), Argentina

Abstract

Spatially dependent neutron spectra have been measured in non-poisoned heavy water (99.53 mol %), using the time-of-flight technique. From a critical review of the available scattering kernels, the ENDF/GASKET model was selected; this model in conjunction with a modified version of the DTF-IV transport code in S_8 and P_1 approximations, proved to be satisfactory for the cases studied. Also calculations were performed by the THERMOS and NYRESE codes. For the spectra studied the largest discrepancies between theory and measurement were 15%, a substantial improvement over prior studies.

Zusammenfassung

Neutronenthermalisierung in Schwerwasser – Messungen und Rechnungen von Spektren

Es wurden Messungen von ortsabhängigen Neutronenspektren in reinem Schwerwasser (99.53 mol. %) mit der Standard-Flugzeitmethode durchgeführt. Es zeigt sich, daß die modifizierte Version des DTF-IV-Codes zuverlässige Ergebnisse liefert, wenn die anisotrope Streuung in der P_1-Nöherung und die Winkelabhängigkeit des Neutronenflusses in der S_8-Näherung benutzt werden. Es wurden auch Rechnungen mit dem THERMOS- und NYRESE-Code gemacht. Die Gruppenkonstontensätze wurden mit dem Streuungsmodell ENDF/GASKET erholten, welches auf Grund einer kritischen Untersuchung der bekannten Modelle ausgewöhlt wurde. In den behandelten Fällen war der größte Unterschied zwischen gemessenen und gerechneten Ergebnissen 15%, was einen wesentlichen Fortschritt im Vergleich zu vorhergehenden Untersuchungen bedeutet.

INIS DESCRIPTORS

NEUTRON SLOWING-DOWN THERORY
HEAVY WATER
NEUTRON SPECTRA
TIME-OF-FLIGHT METHOD
NEUTRON TRANSPORT THEORY
COMPUTER CALCULATIONS
D CODES
T CODES
N CODES

Introduction

An extensive study of the different molecular models formulated by many authors to obtain the scattering kernel for the description of the neutron thermalization in D_2O has not been made. Experimentally, there are no measurements of the angular thermal neutron spectrum in non-poisoned or lightly poisoned D_2O.

Wydler [1], has measured spectra in different D_2O solutions poisoned with cadmium (equivalent concentrations of 0.35, 0.81 and 1.20 b/at. D). The calculations were made with the free gas, GAKER (or *Nelkin-Honeck*) [2], and *Haywood* [3] models. Discrepancies between theory and experiment were higher than 25% in the $1/E$ energy region.

Beyster [4], has studied solutions of D_2O poisoned with boron (2.1 b/ot. D), and cadmium (2.4 b/at. D). The calculations were made with the GAKER molecular model for D_2O and the program GAPLSN [5]. Good agreement was achieved in the central region of the studied geometry; close to the boundary the discrepancies were of the order of +20% to −5% for the first solution and +15% to −10% for the second one. He has also studied the neutron spectrum close to a fuel rod; the discrepancies obtained lie between +15% and −30%, for the *Koppel* [6] bound deuterium model and the 1-DF program (a version of the DTF-IV code [7]).

The motivation for the present work was to measure carefully the angular spectra as a function of position in non-poisoned heavy water (99.53 mol %). Understanding a simple clean geometry experiment could assist us in the study of planned heterogeneous measurements including heavy water.

This research is part of the program of the Argentine Comisión Nacional de Energía Atómica (CNEA), to evaluate the various models, codes, and data sets used to calculote the spectra in D_2O moderated lattices and to analyze the sensitivity of cell parameters to the scattering low.

Molecular model of D_2O

From a review of the bibliography of the different models for D_2O, 12 different ones were identified. A careful study of the experiments and results previously obtained was also made.

It is impossible to discuss in this paper all the dato studied. Therefore only the chosen model, the ENDF/GASKET [5] model, according to the description of *Koppel* and *Houston,* is treoted in detail.

The ENDF/GASKET molecular model for D_2O includes the incoherent approximation and free translotion for the molecule, the scattering by the oxygen atoms is token into account by adding to the data for $D(D_2O)$ at 296 K, the $S(\alpha,\beta)$ for a free gas of mass 16. Thus two important processes are not treated, since the oxygen contribution to $\varrho(\beta)$ is not negligible and the deuterium is lorgely coherent. The scattering functions were calculoted by means of the computer code GASKET [8] and were summed according to *Jarvis* [9], to obtain the $S(\alpha,\beta)$ for D_2O. The contribution of oxygen was found to be significant only for $\beta<1$ and $0.4>\alpha>5$.

The $S(\alpha,\beta)$ obtained was compared with the experimental results of *Egelstaff* [10], *Haywood* [11], *Harlind* [12] and *Whittemore* [13] and the computed results using the model of *McMurry* [12; 13]. In general our $S(\alpha,\beta)$ gives lower values.

From the experimental results obtained by other authors, the accuracy of the discrete frequency distribution used, could be verified. However, it seems necessory to include another oscillator at $\beta\simeq 0.7$ which implies improvement in the description of the hindered translation of the molecule.

To improve the continuous frequency distribution of the model, new measurements similar to the one of *Whittemore* for lower values of α are necessary.

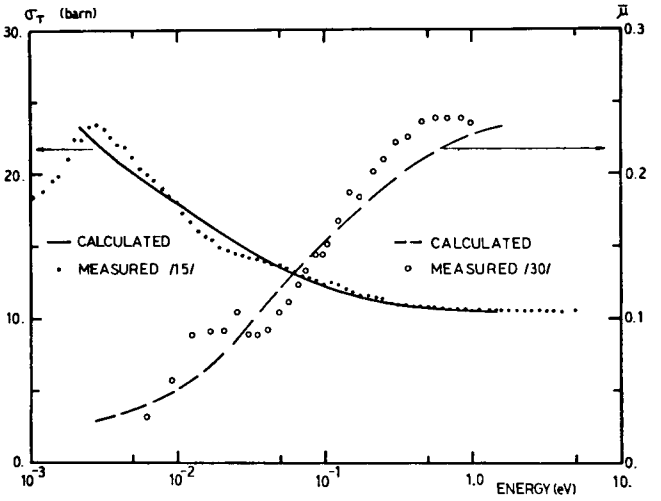

Fig. 1: Average cosine of the scattering angle and total cross section for D_2O

[1] The present work was carried out with the aid of the Organization of the American States.

In Fig. 1 the calculated values of the total cross section are compared with those measured by *Kropff* [15]. The agreement is fairly good. The discrepancies for energies lower than 0.006 eV could be attributed to coherent effects. The average cosine of the scattering angle $\bar{\mu}$ (E_0), calculated from $\sigma_1(E_0)$ and $\sigma_0(E_0)$ showed the same effect.
The agreement obtained for the decay constant compared with previous results [16] is reasonable good. Our values for α_0, D_0, C and F have been calculated from the α vs B^2 function computed with the aid of the code CAGE [17].

For D_2O (100 %) we obtained:

$$\alpha_0 = 9.66\,\text{sec}^{-1}$$
$$D_0 = 2.02\times10^5\,\text{cm}^2\,\text{sec}^{-1}$$
$$C = 3.62\times10^5\,\text{cm}^4\,\text{sec}^{-1}$$
$$F = 1.95\times10^5\,\text{cm}^6\,\text{sec}^{-1}$$

Description of the experiments

The experiments consisted basically of measuring the angular neutron spectra as a function of the position in D_2O at 23 °C using the time-of-flight technique. The facility used is detailed fully in Refs. [18] and [19]. The experimental arrangement for these measurements is illustrated in Fig. 2.

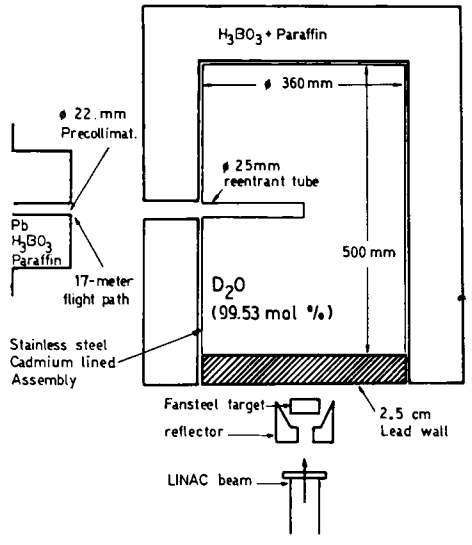

Fig. 2: Experimental set up to measure neutron spectrum in D_2O

The experimental geometry comprised o cylindrical stainless steel vessel 50 cm long and 36 cm in diameter, lined with a cadmium sheet and shielded with a boron mixture. The neutron beam wos extracted radially via o 25 mm-diameter reentrant hole perpendicular to the source assembly axis. The dimensions of the vessel used were based on previous studies, carried out in order to obtain a mean emission time not higher than 5 % with respect to the flight time.
The neutron beam from the reentrant tube was well defined by a collimator and shielding prior to its entrance to the 17. meter flight path. The neutron background was determined by inserting a B-10 filter in the reentrant hole. Neutrons were detected by means of a Li-6 glass scintillator.
The D_2O used (99.53 mol % determined by spectrophotometric analysis), was provided by the Atucha Nuclear Power Plant. It had an impurity equivalent to a boron concentration of 0.064 b/at. D., which was obtained from the pulsed-decay time measurement of the thermal neutron field as a function of time in different geometries. To prevent the contamination with H_2O, the D_2O was maintained and transferred under a nitrogen atmosphere.

Static measurements of the thermal and epithermal flux distributions were made using indium foils in conjunction with the cadmium difference method. From the radial distribution a check of the goodness of the shielding (Bessel Jo distribution) was obtained except in the last centimeter close to the boundary where there is perturbation originating from the structural material neccesary to connect the reentrant tube with the vessel. This measurement was also used to determine the spatial form of the moderation source to be used for input to the calculations.
From the distribution along the axis of the cylinder, the inverse relaxation constant, γ, was derived to get the local buckling. The result was:

$$\gamma^2 = 0.00153 \pm 0.00010\,\text{cm}^{-2}$$

Neutron spectra were measured at six positions along the radius of the tank. For $r = 0.0$ cm, 10.5 cm and 14.0 cm the measurements were carried out at 0° and 180°. For $r = 16$ cm, 17.0 cm and 17.8 cm, at 0°, but these last two results were not closely comparable to the calculation because of the perturbation cited before; therefore they are not included in this report. The neutron beam intensity was monitored with two U-235 miniature fission counters located inside the vessel. These monitors agreed within 1 % during all measurements; thus it can be considered that the different experimental neutron spectra ore normalized within this error.
Running time for each experiment were chosen to ensure good statistical accuracy. For all the reported spectra, the standard deviation due to statistics was less than 5 % over all the energy range. The mean temperature of D_2O for all the measurements was 23.3 °C; it was monitored with an iron-constantan thermocouple. The measured decay constant was 220 ± 11 μsec. The mean emission time was obtained in the usual way [19]; it amounted to less than 5.3 % of the flight time for the energy range studied.
The computer code NYRTOF [20], was used for the data reduction. The poorest energy resolution was 11 %, but only in the energy range from 0.02 to 0.27 eV did the resolution exceed 5 %. The spectra obtained were corrected by the time resolution function according to the prescription of *Beckurts* [21].
For the worst case encountered, the maximum correction to the experimental flux was 1.5 %. The measured spectra are shown in Fig. 3 and 4, from them the adequacy of the spatial resolution is evident.

Calculational methods

The neutron spectra were calculated utilizing a variety of computer codes described in Ref. [17].
The scattering kernel based on the ENDF/GASKET model [6], for bound deuterium in D_2O and assuming free translation for the oxygen was generated by means of the GASKET code [8]. Group cross sections, P_0, and P_1 for 30 energy groups were calculated using these scattering kernels with the NYR190 [22] and NYR081 [23] codes. A distributed source, P_0 and P_1, in one dimension was calculated by the NYR238 [24] code, using the measured transverse spatial distribution of epithermal neutrons.
The transport theory calcuoltions to obtain the angular neutron spectra were calculated by the NYR230 [7] code (a modified version of the one-dimensional DTF-IV code), in 34 spatial intervals, using the 30 group cross sections and the distributed source. NYR230 code was used in the S_8, P_0 and P_1 approximations.
Also, calculations were performed by the NYR342 [25] (a modified version of the THERMOS code [26]), and NYRESE [27]. The NYR342 code accepted the group constants computed with the ENDF/GASKET model. Calculations with NYRESE were carried out with the Cadilhac G and H functions [28], and also with the functions computed from the ENDF/GASKET model.

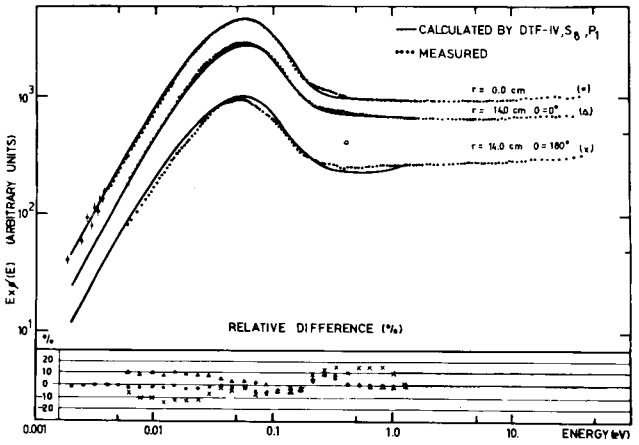

Fig. 3: Measured neutron spectrum in D_2O compared to calculation using P_1 approximation for $r = 0$ cm ($\Theta = 0°$) and $r = 14.0$ cm ($\Theta = 0°$ and $\Theta = 180°$)

The results of the calculated spectra are shown in Figs. 3, 4 and 5, together with the experimental data. It is important to point out that only the calculated central spectrum ($r = 0.0$ cm) was normalized at 1.35 eV to the experimental data for that position and energy. The calculated spectra at the other positions agrees reasonably well with experiment.

As an additional check, the effective neutron temperature was obtained from the central experiment spectrum. We obtained

$$T/T_0 = 1.10 \pm 0.01$$

which is in reasonable agreement (4 %) with the value computed with the expression af *De Sobrino* and *Clark* [29], which takes into account the effects of absorption and leokage.

Discussion of results

From the cases studied, the modified version of the DTF-IV transport code in P_1 approximation, proved to give best agreement than the P_0 approximations.

The results for the central spectra, together with the O° and 180° spectra for $r = 14.0$ cm are shown in Fig. 3. For the latter the angular dependence can clearly be observed. A maximum deviation of 15 % is observed for the most unfavorable case ($r = 14.0$ cm, 180 %), due to the low intensity of the source. For $r = 10.5$ cm (Fig. 4), the deviation has the same sign with a magnitude of only 5 %. In the same figure, the spectrum close to the surface is shown. A good spatial resolution can be observed. The agreement between theory and experiment is satisfactory.

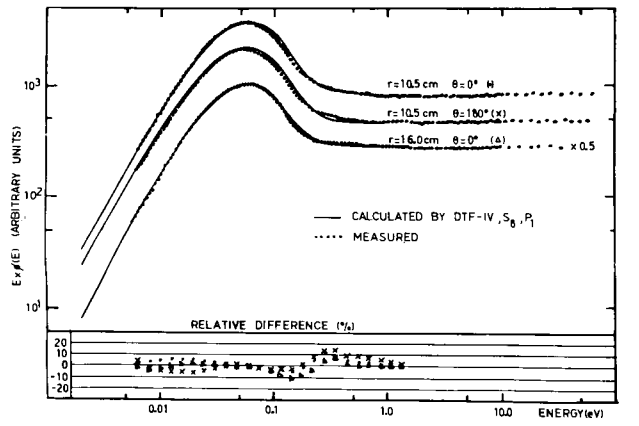

Fig. 4: Measured neutron spectrum in D_2O compared to calculation using P_1 approximation for $r = 10.5$ cm ($\Theta = 0°$ and $\Theta = 180°$), and $r = 16.0$ cm ($\Theta = 0°$)

For all the studied spectra, the lack of agreement in the energy interval from 0.1 to 0.4 eV may indicate that a more negative buckling must be used in the calculations. These discrepancies could also be partially attributed to perturbations of the reentrant tube.

In Fig. 5, the results obtained with the NYR342 code are shown, the agreement with the data for the central spectrum is good (+4.4 % to −7.7 %). Also the spectra computed with NYRESE is shown in this figure, their values are much less thermalized than the measured ones, the deviation grow up to 57 % at lower energies.

Conclusions

A systematic study of the neutron thermalization in heavy water was done. Spectrum measurements as functions of energy, angle and position were carried out for a valuable check of selected scattering law data. From the literature review done, it is clear that further studies of the coherent scattering and hindered translation of the D_2O molecule are desiroble. With respect to the model selected (ENDF/GASKET), it could be pointed out that it is necessary to refine it including another ascillator at $\beta = 0.7$, extending it to higher by 2 or 3 eV and treoting the oxygen in a more realistic way.

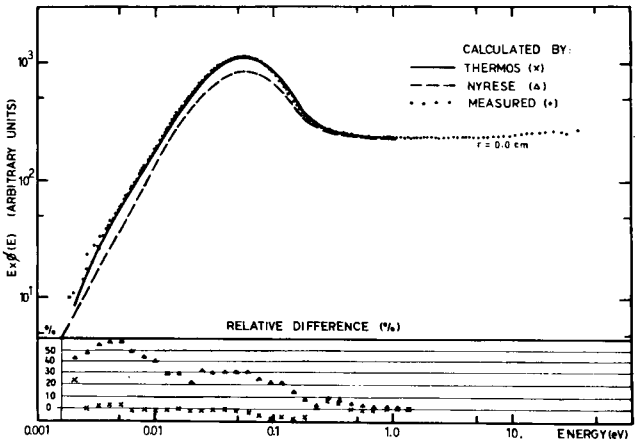

Fig. 5: Measured central neutron spectrum in D_2O compared to calculations with different codes

The necessity of considering the contaminating hydrogen in detail wos verified; its scattering kernel was calculated with the ENDF/GASKET model for $H(H_2O)$ in view of the lack of one for HDO.

As a check of the group canstants, the decay constant and diffusion parameters were calculated. The agreement with experimental values was excellent.

From the preceding "discussion of results" it is clear that for the studied cases, the best agreement with calculations were obtained using the transport theory code in S_8 and P_1 approximations. The discrepancies between theory and measurement were of the order of 10 %, but for the most unfavourable position ($r = 14.0$ cm, $\Theta = 180°$) a disagreement of 15 % was found. Within errors, this work appears to represent an improvement over prior measurement. It must be noted that there are no previous studies in non-poisoned heavy water.

It seems that the spectra computed by NYRESE trends to be insensitive to the molecular model used.

Finally, it must be pointed out, that the disagreement between THERMOS and DTF-IV (two well-known codes), were of the order of the discrepancies between theory and measurements.

The utility of the nuclear determination of the boron equivalent concentration from pulsed-lifetime experiments has been demonstrated.

From the obtained results, it is clear the necessity of refining the present models for the neutron scattering law of D_2O, developing a model far HDO. This work has also indicated the necessity of using P_1 approximation, to compute spatially dependent neutron spectra in small D_2O geometries.

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