Measurement of the Transport Mean Free Path of Thermal Neutrons in D_2O by a Boron Poisoning Method*

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The transport mean free path of thermal neutrons in D_2O was experimentally determined by measuring the change in the relaxation length upon the introduction of known amounts of B_2O_3. The value found for 99.7 percent pure D_2O was 2.49 ± 0.04 cm. Upon correcting for the small hydrogen impurity, a value of 2.52 ± 0.04 cm was obtained for λ_{tr} in pure D_2O.

INTRODUCTION

THE transport mean free path λ_{tr} of thermal neutrons in D_2O was measured by an extrapolation method at the Montreal Laboratory of the Natural Research Council of Canada in 1946.[1] In this experiment the thermal neutron flux in the neighborhood of a plane cadmium boundary was measured and the distance d beyond the boundary at which the flux extrapolated to zero was determined. From the relationship $\lambda_{tr}=0.71d$, a value 2.31 ± 0.09 cm for the transport mean free path of 99.4 percent pure D_2O was obtained. Applying a correction for the mass spectrographically measured hydrogen impurity gave a value of $\lambda_{tr}(D_2O)=2.4\pm0.1$ cm for 100 percent pure D_2O. Because of the importance of $\lambda_{tr}(D_2O)$ in reactor calculations, it was desirable to measure this value by a different method. Accordingly, the relaxation length of thermal neutrons in a cylindrical column of D_2O was measured for various amounts of boron poisoning. From the variation of the relaxation length with boron concentration, the effective value of λ_{tr} and the hydrogen impurity content were determined for the particular D_2O used. The effective value was then corrected for the hydrogen impurity to give a value of 2.52 ± 0.04 cm for the transport mean free path of 100 percent pure D_2O.

THEORY

In a weakly absorbing, homogeneous medium, the diffusion length L, the macroscopic total cross section Σ, and the macroscopic absorption cross section Σ_a are related by

$$1/L^2=3\Sigma(1-\bar{\mu})\Sigma_a(1-k\Sigma_a/\Sigma), \tag{1}$$

where μ is the average value of the cosine of the angle through which a neutron is scattered in a single collision, and $k=0.80-\bar{\mu}(1-\bar{\mu})^{-1}$. In terms of the macroscopic scattering cross section Σ_s, Eq. (1) may be written as

$$1/L^2=3\Sigma_s(1-\bar{\mu})\Sigma_a+3\Sigma_a^2(1-\bar{\mu})(1-k). \tag{2}$$

The second term on the right side of this equation may often be neglected. In this experiment this term is at most about 0.1 percent of the first term. The first term may be written as $3\Sigma_a/\lambda_{tr}$, where

$$\lambda_{tr}=1/\Sigma_{tr}=1/[\Sigma_s(1-\bar{\mu})] \tag{3}$$

defines the transport mean free path for the medium.

For a cylindrical tank, with a neutron source at the bottom and bounded by cadmium on all the other sides, the thermal neutron flux is represented by

$$\phi=AJ_0(\mu r)\sinh\nu(h-z), \tag{4}$$

where z is measured from the bottom of the tank. The constants μ (not to be confused with $\bar{\mu}$ above) and ν will be referred to as the radial buckling and the reciprocal of the relaxation length, respectively. They are related to the diffusion length by

$$1/L^2=\nu^2-\mu^2. \tag{5}$$

Now ν^2 and μ^2 can be determined from the flux distribution, so if Σ_a is known, λ_{tr} can be determined. For D_2O, however, Σ_a is small and is very sensitive to impurities, in particular hydrogen, so that an accurate determination of λ_{tr} in this case is difficult. If a strong absorber such as boron is added, Σ_a may be considerably increased without measurably affecting λ_{tr}. Combining Eqs. (2), (3), and (5), and putting $\Sigma_a=\Sigma_a(D_2O)+\Sigma_a(B)$ gives

$$\nu^2-3\Sigma_a^2(1-\bar{\mu})(1-k)=3\Sigma_{tr}\Sigma_a(B)+3\Sigma_{tr}\Sigma_a(D_2O)+\mu^2. \tag{6}$$

The μ^2 term is determined by the radius of the tank and by λ_{tr} and so is essentially constant. Thus λ_{tr} can be determined from the slope of the straight line obtained by plotting the left side of Eq. (6) against $\Sigma_a(B)$.

If the D_2O solution contains a volume fraction r of H_2O, the effective value of the transport mean free

TABLE I. Summary of data.[a]

B_2O_3 concentration (mg/l)	Average temp. (°C)	$\Sigma_a(B)$ $\times10^3$ (cm^{-1})	Average $\mu\times10^2$ (cm^{-1})	Average $\nu\times10^2$ (cm^{-1})	$(\nu^2-\Sigma_a^2)$ $\times10^3$ (cm^{-2})
0	23.6	0	3.107 ± 0.016	3.291 ± 0.018	1.083 ± 0.012
46.9 ± 0.2	22.9	0.536 ± 0.008	3.100 ± 0.012	4.163 ± 0.018	1.733 ± 0.015
72.3 ± 0.1	23.3	0.827 ± 0.011	···	4.561 ± 0.022	2.079 ± 0.020
97.8 ± 0.2	23.3	1.119 ± 0.015	3.101 ± 0.010	4.938 ± 0.029	2.437 ± 0.029
123.5 ± 0.5	23.4	1.413 ± 0.020	3.082 ± 0.020	5.270 ± 0.019	2.775 ± 0.020
146.8 ± 0.5	26.1	1.671 ± 0.023	3.086 ± 0.016	5.573 ± 0.013	3.103 ± 0.014

[a] The errors listed are standard deviations.

* This report is based upon studies conducted for the U. S. Atomic Energy Commission.

[1] Auger, Munn, and Pontecorvo, Can. J. Research **A25**, 143 (1947).

path is given by

$$\frac{1}{\lambda_{tr}(\text{eff})}=\frac{1-r}{\lambda_{tr}(D_2O)}+\frac{r}{\lambda_{tr}(H_2O)}, \quad (7)$$

where $\lambda_{tr}(D_2O)$ and $\lambda_{tr}(H_2O)$ refer to 100 percent D_2O and 100 percent H_2O, respectively. This is easily verified by substituting in Eq. (3):

$$\Sigma_s=(1-r)\Sigma_s(D_2O)+r\Sigma_s(H_2O),$$

and

$$\bar{\mu}=(1/\Sigma_s)[(1-r)\Sigma_s(D_2O)\bar{\mu}(D_2O)+r\Sigma_s(H_2O)\bar{\mu}(H_2O)].$$

EXPERIMENTAL PROCEDURE

The measurements were made in the Atomic Energy Research Laboratory at North American Aviation, Inc. The D_2O was contained in a cadmium covered aluminum tank, 5 ft in diameter and 6 ft high. The tank was supported by a 5-ft diameter, 6-ft high graphite pedestal containing a water boiler reactor at its center. The reactor was operated at a power level of approximately $\frac{1}{2}$ watt and provided a flux of about 10^6 thermal neutrons per square centimeter per second at the base of the D_2O tank. Flux measurements were made along the axis of the tank and along a central diameter with indium foils 1 cm square and 0.005 cm thick (95 to 100 mg/cm^2). Bare and cadmium covered foil measurements were taken, and saturated foil activities were obtained using standard counting techniques. The foils were spaced at 5-cm intervals along the axis and at 4.9-cm intervals along the diameter.

Successive amounts of B_2O_3 were added to the D_2O. After each addition, the solution was thoroughly stirred in the aluminum tank, and samples were removed for analysis from at least four different parts of the tank. Flux measurements were then made for each B_2O_3 concentration. Six different concentrations were used, ranging from zero to approximately 150 mg of B_2O_3 per liter of solution. For each concentration a minimum of four radial and eight axial sets of foil exposures were made.

CALCULATIONS

1. Effective Values of λ_{tr} and $\Sigma_a(D_2O)$

A value of 750±10 barns was taken for the boron absorption cross section for 2200-m/sec neutrons.[2] Assuming the neutrons to have a Maxwellian velocity distribution at the temperature T of the D_2O solution, the effective absorption cross section was determined to be $\sigma(\text{eff})=(750\pm10)\times(\pi^{\frac{1}{2}}/2)\times(293/T)^{\frac{1}{2}}$ barns. For each boron concentration the macroscopic absorption cross section $\Sigma_a(B)$ was determined by multiplying $\sigma(\text{eff})$ by the average measured density of boron nuclei.

The radial buckling was determined by fitting each radial set of foil exposures to a zeroth-order Bessel function. Similarly, the relaxation length was determined by fitting each axial set of foil exposures to a hyperbolic sine function. The respective values of μ and ν thus obtained for each boron concentration were then averaged.

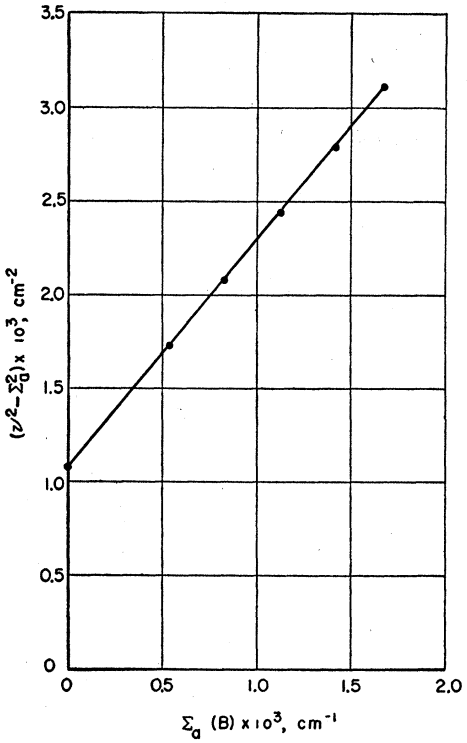

FIG. 1. Relaxation length as a function of the boron absorption. The slope of the line $(\nu^2-\Sigma_a^2)$ *vs* $\Sigma_a(B)$ determines the effective value of λ_{tr} for the D_2O. The effective absorption of the D_2O before the addition of any boron is obtained by subtracting μ^2 from the y intercept and dividing by $3\Sigma_{tr}$.

A summary of the measurements is presented in Table I. The first column gives the average value of the B_2O_3 concentration in milligrams per liter of solution. The second column gives the average temperature of the $D_2O-B_2O_3$ solution while the flux measurements were being made. The third column presents the computed macroscopic cross section of the B_2O_3 impurity; the absorption of the oxygen was neglected. The next two columns give the average values for the radial buckling and the reciprocal of the relaxation length. Note that the value of μ is constant within the accuracy of the measurements. The value of μ averaged over all concentrations is $0.03095+0.00010$ cm^{-1}. In the last column the values of $\nu^2-3\Sigma_a^2(1-\bar{\mu})(1-k)$ are tabulated. Since the term involving Σ_a^2 is small compared to ν^2, accuracy in $\bar{\mu}$ is not required here. With $\bar{\mu}=0.15$ (see below), $3(1-\bar{\mu})(1-k)=1.0$.

The values of $\nu^2-\Sigma_a^2$ are plotted against $\Sigma_a(B)$ in Fig. 1. From the slope and y intercept and the average value of μ the following effective values were deter-

[2] U. S. Atomic Energy Commission Report AECU-2040, May 15, 1952 (unpublished).

mined:

$$\lambda_{tr}(\text{eff}) = 2.49 \pm 0.04 \text{ cm},$$
$$\Sigma_a(D_2O) = (1.04 \pm 0.10) \times 10^{-4} \text{ cm}^{-1}.$$

2. Value of λ_{tr} for Pure D_2O

From the value of $\Sigma_a(D_2O)$ we can estimate the amount of hydrogen impurity in the D_2O and correct λ_{tr} accordingly. Converting the effective value of $\Sigma_a(D_2O)$ to the value for 2200 m/sec neutrons, we have

$$\Sigma_a(D_2O, 2200) = (1.2 \pm 0.1) \times 10^{-4} \text{ cm}^{-1}.$$

Of this the D_2O itself contributes about $0.4 \times 10^{-4} \text{cm}^{-1}$.[3] Attributing the remaining 0.8×10^{-4} cm^{-1} to H_2O requires a hydrogen to deuterium ratio of 0.0030 ± 0.0005.[4] This value is about twice the value normally obtained by some mass spectrograph analyses. However, since the effective absorption of the D_2O is very sensitive to the hydrogen impurity, the ratio of H to D computed by the above means should be more reliable. The contribution of other impurities in the D_2O to the macroscopic absorption is less than 0.01×10^{-4} cm^{-1} and has accordingly been neglected. The other impurities consist mainly of oxides of Si, Al, and Fe, and amount to less than 50 mg per liter of solution.

Some of the hydrogen will be in the form of H_2O, but most of it will be in the form of HDO. However, it may be shown that the transport cross section of HDO is equal to the simple average of the transport cross sections of H_2O and D_2O. Hence, we may take all the hydrogen to be in the form of H_2O and all the deuterium in the form of D_2O. Substituting in Eq. (7), the values $\lambda_{tr}(\text{eff}) = 2.49 \times 0.04$ cm, $\lambda_{tr}(H_2O) = 0.48 \times 0.01$ cm (see reference 4), and $r = 0.0030 \times 0.0005$, gives finally

$$\lambda_{tr}(100 \text{ percent pure } D_2O) = 2.52 \pm 0.04 \text{ cm}.$$

3. Value of $\bar{\mu}$

For D_2O we may take $\sigma_s = \sigma_t = (10.0 \pm 0.72E^{-\frac{1}{2}} \pm 0.3)$ barns, where the energy E is in electron volts.[5] Averaging over a Maxwellian collision distribution gives an effective $\sigma_s = 14.0 \pm 0.3$ barns, and so $\Sigma_s(D_2O) = 0.467 \pm 0.010$ cm^{-1}. Putting this value of Σ_s and the above value of λ_{tr} into Eq. (3) gives

$$\bar{\mu} = 0.15 \pm 0.03.$$

4. Comparison of the Two Measurements of λ_{tr}

The determination of λ_{tr} by the boron poisoning method has some definite advantages over its determination by the extrapolation method. The extrapolation method, while offering a more direct measurement of λ_{tr}, suffers from the fact that near the boundary the flux is small and the corrections for the depression of the flux by the detectors are neither negligible nor constant. In the boron impurity experiment such corrections are unimportant. Since in this case none of the foils need be less than several mean free paths from the D_2O surface, the depression of the flux at a foil due to the foil itself is essentially the same for all the foils. The depression of the flux at a foil caused by neighboring foils can be made small by adequately separating the foils; in any case, the exponential nature of the flux along the axis of the tank makes this depression the same constant factor for each foil. In the boron poisoning experiment the major source of error is caused by the uncertainty in the boron cross section.

Two small corrections should be applied to the value of λ_{tr} quoted in the extrapolation experiments of Auger *et al.* before comparing it with the value of this experiment. First, using a sinh function instead of a linear function and using transport theory instead of diffusion theory increases λ_{tr} by 0.25 cm. Second, using 0.48 cm for the transport mean free path of light water instead of 0.32 cm makes the hydrogen impurity correction only 0.07 cm. With these changes the value of λ_{tr} (100 percent D_2O) from the extrapolation experiment is 2.63 ± 0.12 cm. This is in reasonable agreement with the value obtained in the present experiment.

DISCUSSION

The scattering cross sections of boron and oxygen are approximately 4 barns each. At the maximum B_2O_3 concentration, the B_2O_3 macroscopic scattering cross section is approximately 3×10^{-5} cm^{-1}. Even with a factor of 2 or 3 for molecular effects, this is very small compared with Σ_{tr} which is approximately 0.4 cm^{-1}. Hence, the change in λ_{tr} upon the introduction of the B_2O_3 into the D_2O is negligible. A similar argument applies to the other nonhydrogen impurities present in the D_2O solution.

No measurable change in relaxation length with height was detected. The higher B_2O_3 concentrations showed what appeared to be hardening effects. However, this was traced to the scattering in of fast neutrons from the water-boiler reactor radiation shield and was completely eliminated by the addition of paraffin slabs across the top of the shield. The constancy of the relaxation length with height lent experimental justification to the assumption of a Maxwellian velocity distribution for the neutrons.

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[3] $\sigma_a(D_2O)$ was taken equal to 1.2 ± 0.1 millibarns; see Kaplan, Ringo, and Wilzbach, Phys. Rev. **87**, 785 (1952).

[4] F. J. Sisk, Oak Ridge National Laboratory Report ORNL-933, March 15, 1951 (unpublished). This unclassified report lists the following effective values for H_2O: $\sigma_a = 0.68 \pm 0.01$ barn, $\lambda_{tr} = 0.48 \pm 0.01$ cm.

[5] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948).