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THERMAL NEUTRON DIFFUSION PARAMETERS OF HEAVY WATER

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Abstract—The decay of the asymptotic thermal neutron distribution was measured by the pulsed technique in D_2O (99·82 mol per cent, $\vartheta = 22°C$) within a rather large range of geometrical buckling ($0{\cdot}0013\ cm^{-2} < B^2 < 0{\cdot}0470\ cm^{-2}$). In the lower buckling region ($B^2 < 0{\cdot}0060\ cm^{-2}$) we employed a modal analysis method outlined in a previous paper (MEISTER, 1963). From the experimental decay constant α plotted against B^2 we obtained the diffusion constant $D_0 = (2{\cdot}000 \pm 0{\cdot}009) \times 10^5\ cm^2/sec$ and the diffusion cooling constant $C = (5{\cdot}25 \pm 0{\cdot}25) \times 10^5\ cm^4/sec$ in a fairly good agreement with earlier experimental data (SJÖSTRAND, 1958; GANGULY and WALTNER, 1961; WALTNER and WESTFALL, 1962).

1. INTRODUCTION

AFTER a burst of fast neutrons has been injected into a non-multiplying medium, the asymptotic thermal neutron flux dies out with a decay constant α given by

$$\alpha = v_0\Sigma_a + D_0B^2 - CB^4 + FB^6 + \ldots, \qquad (1)$$

where the buckling B^2 of the flux distribution is determined by the geometrical size of the medium [equation (2)]. From measurements of α on samples of different B^2, in principle, the absorption cross section Σ_a, the diffusion constant D_0, and the diffusion cooling constant C can be determined by means of equation (1).

In the case of heavy water, pulsed experiments have been performed so far (SJÖSTRAND 1958; GANGULY and WALTNER 1961; WALTNER and WESTFALL 1962) only on relatively small samples with buckling $B^2 = 0{\cdot}0300\ cm^{-2}$ up to $0{\cdot}12\ cm^{-2}$ having linear dimensions smaller than 10 transport mean free paths. In these experiments, corrections for the extrapolated boundary of the sample are required which are higher than 15 per cent of B^2, so that systematic errors may occur if the extrapolation distance deviates from the assumed value. For a rather accurate determination of D_0 it is therefore reasonable to carry out pulsed experiments on appreciably larger D_2O geometries in a buckling region where additional higher terms than $-CB^4$ in equation (1) are not very important.

2. EXPERIMENTS ON SMALLER D_2O-GEOMETRIES

In the higher buckling region, $0{\cdot}0080\ cm^{-2} < B^2 < 0{\cdot}0470\ cm^{-2}$, an aluminium tank of 64·9 cm dia. and 70 cm height was used as a D_2O container. The 2-mm tank walls were clad with 0·5 mm cadmium sheet. In addition, the whole system was shielded by B_2O_3 and paraffin for reduction of background intensity which was mainly due to neutrons scattered back from the floor. During experiments with a lower D_2O level ($B^2 > 0{\cdot}0280\ cm^{-2}$), a cadmium disk was fixed inside the tank some centimeters above the D_2O level in order to exclude effects of backscattering from the more distant container walls (LOPEZ and BEYSTER, 1962). The accuracy of the D_2O-level adjustment was better than 0·5 mm.

The target of the pulsed d–t accelerator was placed above the top lid of the container on the central axis in order to prevent excitation of higher azimuthal flux modes. The neutron detector (BF_3 counter, 20th Century 31 EB 70 G) was arranged diametrically below the bottom of the tank so as to eliminate the 2nd radial flux mode; at higher D_2O level ($B^2 = 0{\cdot}0083\ cm^{-2}$), however, the detector was symmetrically fixed at half the water height at the casing of the tank.

The thermal neutron flux decay following each neutron burst (pulse length $\approx 25\ \mu sec$) of the accelerator was recorded by a 256-channel time analyser (TMC Model CN-110, pulsed-neutron unit) using channel widths from 10 to 40 μsec and a repetition rate of 100 to 360 sec^{-1}. The experimental data were corrected for counting-losses on the basis of a 1·7 μsec overall dead time (maximum correction $\approx$ 3·7 per cent) and for a constant background contribution given by the background channel of the analyser. Actually, the background was composed of an initial decaying part (decay constant $\approx 8000\ sec^{-1}$) followed by an almost constant part as it was found by an empty-container experiment.

During the actual experiments a proper delay time of at least 330 μsec was employed in order to allow

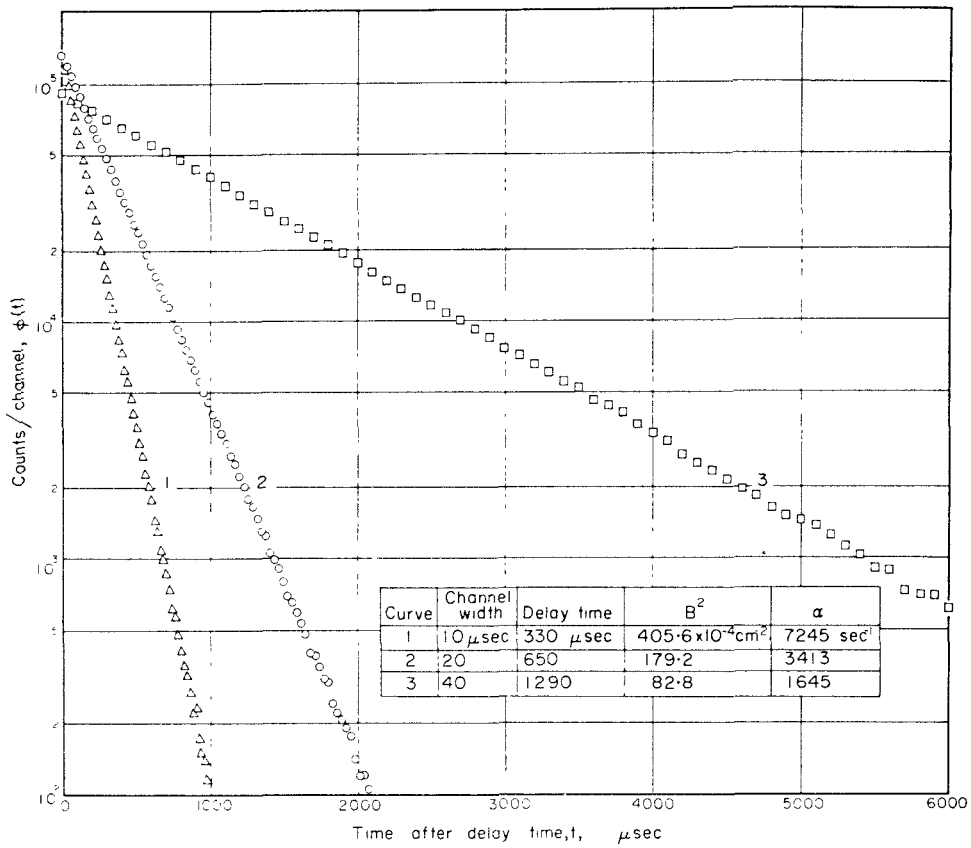

FIG. 1.—Decay of thermal neutron flux $\phi(t)$ (higher buckling experiments, tank radius $R = 32{\cdot}45$ cm); divide abscissa units by 2 for curve 3.

a sufficient decay of higher spatial and energy modes before the registration of the flux decay was started. The experimental decay curves show a strict exponential behaviour within about 3 flux decades as is shown in Fig. 1. From these curves the fundamental mode decay constants α were determined by graphical interpolation.

Table 1 lists the α-values obtained which were corrected for deviation of D_2O temperature from the mean value $\vartheta = 22°C$ assuming $\alpha \sim D_0$ and a temperature dependence of D_0 given by BAUMANN *et al.* (1962). The geometrical buckling B^2 was calculated from the inner tank radius $R = 32{\cdot}45 \pm 0{\cdot}01$ cm and the indicated D_2O level height H according to

$$B^2 = \left(\frac{\pi}{H + 2d}\right)^2 + \left(\frac{2{\cdot}405}{R + d}\right)^2. \qquad (2)$$

The extrapolation distance d was assumed to be given by the formula

$$d = 0{\cdot}71\, \lambda_{tr} \qquad (3)$$

adopting the conventional (Maxwellian average) transport mean free path $\lambda_{tr} = 3D_0/\bar{v} = 2{\cdot}42$ cm (cf. Section 4). However, the question arises whether equation (3) derived for a half-space geometry will be valid also in the case of a time-dependent neutron flux within a sample of linear dimensions as small as several transport mean free paths. One-velocity S_8-calculations have been carried out which indicated that the value of d obtained from equation (3) is

TABLE 1.—EXPERIMENTAL DECAY CONSTANTS α

Tank radius	Buckling	Decay constant
R (cm)	B^2 (10^{-4} cm^{-2})	α (sec^{-1})
70·3	13·05	280·4 ± 1·0
70·3	17·1	362 ± 1·8
70·3	23·9	491 ± 3
70·3	33·3	675 ± 5
70·3	45·5	913 ± 10
32·45	82·8	1643 ± 8
32·45	134·2	2623 ± 12
32·45	175·9	3371 ± 25
32·45	226·0	4298 ± 40
32·45	283·3	5308 ± 35
32·45	349·2	6400 ± 60
32·45	405·6	7239 ± 50
32·45	452·5	7955 ± 60
12·85	459·8	8093 ± 90
32·45	465·8	8173 ± 100

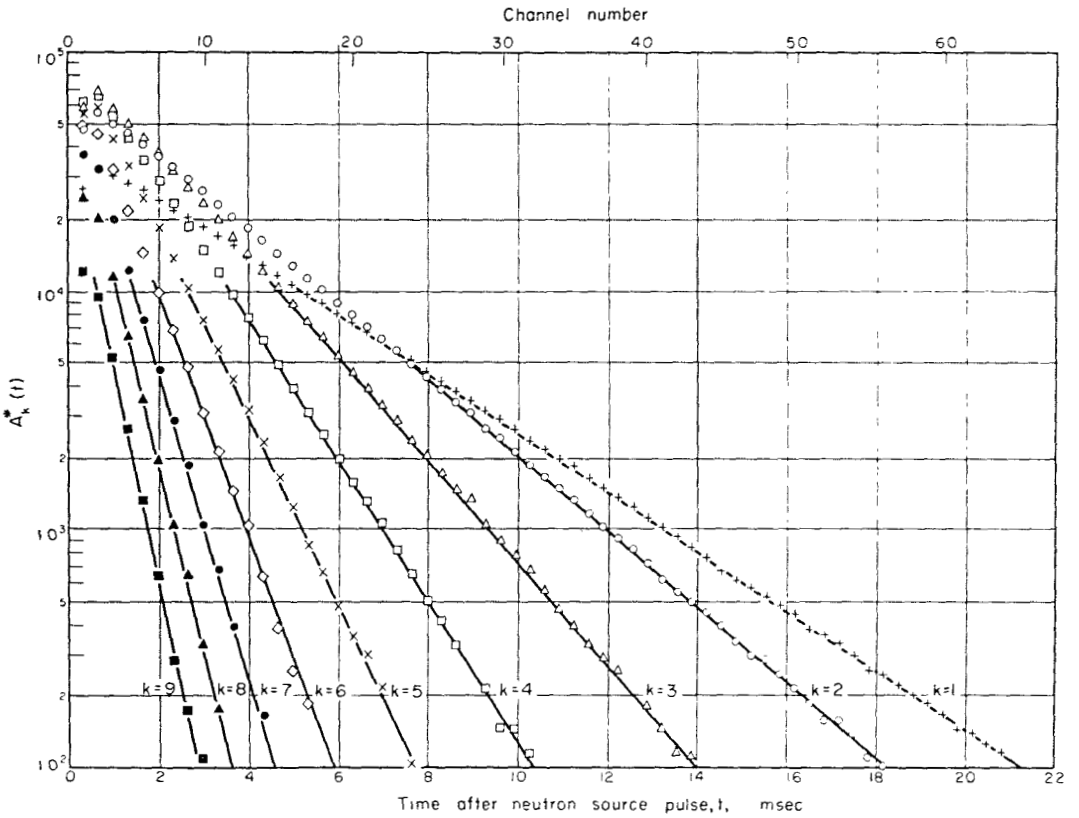

FIG. 2.—Decay of axial modes of various index k (lower buckling experiments, tank radius $R = 68{\cdot}0$ cm).

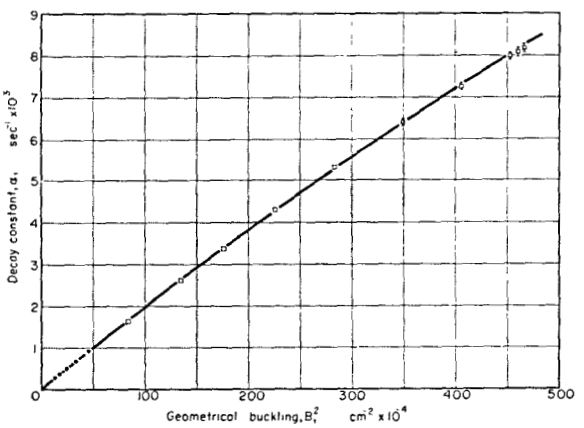

FIG. 3.—Decay constant α as a function of geometrical buckling B^2.
● Modal analysis ($R = 68{\cdot}0$ cm, $H = 270$ cm).
T
□ Higher buckling experiments ($R = 32{\cdot}45$ cm).
⊥
—— Least squares fit of equation (1) to experimental data.

correct better than 2 per cent for slab geometries as thin as $2\,\lambda_{tr}$, if the conventional recipe $\phi(z_0 + d) = 0$ is used rather than the boundary condition

$$\left[d\frac{d\phi}{dz} + \phi\right]_{z_0} = 0.$$

Moreover, there may be an additional effect on d due to the energy-dependent λ_{tr} of D_2O which has not been investigated here.

In order to verify the extrapolation length d assumed in (3) pulsed experiments were made at $B^2 = 0{\cdot}04525$ cm^{-2} on an approximate slab geometry ($R = 32{\cdot}45$ cm, $H = 12{\cdot}25$ cm) as well as an approximate cubic system ($R = 12{\cdot}85 \pm 0{\cdot}03$ cm, $H = 19{\cdot}60 \pm 0{\cdot}08$ cm, $B^2 = 0{\cdot}04598$ cm^{-2}) obtained with a smaller aluminium container; results corrected to $B^2 = 0{\cdot}04525$ cm^{-2} are:

$$\alpha = 7955 \pm 60 \text{ sec}^{-1} \quad \text{('slab' geometry),}$$

$$\alpha = 7957 \pm 90 \text{ sec}^{-1} \quad \text{('cubic' geometry).}$$

The very good agreement of α indicates, that the extrapolation length has been properly taken into account.

The experimental decay constants α given in Table 1 are plotted as a function of B^2 as seen in Fig. 3; some results of lower buckling experiments (Section 3) are included as well. The curve obtained by the method of least squares is seen to represent the experimental data within the limits of error. The curvature shows that the diffusion cooling effect is important.

The experimental errors adopted in Table 1 were derived from the scattering of α in various runs and the $\pm 0{\cdot}5$ mm inaccuracy of D_2O level height H, assuming the B^2 values to be correct. Accordingly, the relative error of α increases from 0·5 per cent at $B^2 = 0{\cdot}0080\ \text{cm}^{-2}$ up to 1·2 per cent at $B^2 = 0{\cdot}0470\ \text{cm}^{-2}$.

3. EXPERIMENTS AT LOWER BUCKLING

For pulsed experiments in the lower buckling region, $B^2 < 0{\cdot}0060\ \text{cm}^{-2}$, we made use of the experimental tank of a subcritical assembly (MEISTER, 1961) 136 cm in diameter, which was filled with D_2O. The target of the pulsed source (EYRICH, 1962) was located below the bottom of the tank, the BF_3 counter used for measuring the flux decay was positioned in a thin aluminium tube which was mounted on the central axis of the tank inside the D_2O. The flux perturbation caused by the BF_3 counter and the resulting correction of experimental data were found to be very small.

The experimental technique was the same as in the case of pulsed experiments on D_2O lattices reported in a previous paper (MEISTER, 1963). In order to determine the decay constant α as a function of B^2 we employed a modal analysis method: The neutron flux decay $\phi(z_i, t)$ was measured at $N-1$ equidistant axial detector positions $z_i = i \,.\, H/N$ (H = effective height, $i = 1, 2 \ldots N-1$), and the Fourier coefficients of the axial flux distribution

$$\phi(z, t) = \sum_{k=1}^{\infty} A_k(t) \sin \frac{k\pi z}{H}\,, \quad A_k(t) = A_{k0} e^{-\alpha_k t}$$

obtained from

$$A_k^*(t) = \frac{2}{H} \sum_{i=1}^{N-1} \phi(z_i, t) \sin \frac{ik\pi}{N} \tag{4}$$

were plotted against the time t. Figure 2 shows a typical plot of the amplitude of the k-th mode $A_k^*(t)$ vs. t for the special case $H = 270$ cm, $N = 10$. In the region $t \gtrsim 4$ msec, $A_k^*(t)$ shows an undisturbed exponential decrease from which the decay constant α_k of the corresponding mode was easily obtained. With the index k of the mode, however, there is an increase of the statistical error of $A_k^*(t)$ caused by differences of comparable numbers occurring in equation (4).

Experiments were carried out at three different effective water heights, $H = 210$, 240, and 270 cm, using either $N = 6$ or 10 detector intervals. The resulting decay constants α_k are listed in Table 2 together with errors derived from the scattering of experimental data assuming an 0·5 per cent inaccuracy of the normalization of $\phi(z_i, t)$ in equation (4). In addition, Table 2 lists the geometrical buckling B^2 of each mode given by the equation

$$B^2 = \mu_r^2 + k^2\pi^2/H^2 \tag{5}$$

with a radial buckling $\mu_r^2 = (11{\cdot}70 \pm 0{\cdot}15) \times 10^{-4}$

TABLE 2.—DECAY CONSTANTS α_k FROM MODAL ANALYSIS COMPARED WITH INTERPOLATION FUNCTION EQUATION (1)

Water height	Index	Buckling	Experimental α_k		Calculated α	$\varepsilon = (\alpha_k - \alpha)/\alpha$	
H (cm)	k	B^2 (10^{-4} cm^{-2})	α_k(sec^{-1}) ($N = 6$)	α_k(sec^{-1}) ($N = 10$)	α(sec^{-1})	ε ($N = 6$)	ε ($N = 10$)
210	1	13·94	296 ± 1·5		296·8	−0·003	
	2	20·65	429 ± 2·5		429·8	−0·002	
	3	31·84	657 ± 5		650·5	+0·010	
	4	47·5	988		957·1	+0·032	
	5	67·6	1265		1347	−0·061	
240	1	13·41	286 ± 1·5	286 ± 1·2	286·3	−0·001	−0·001
	2	18·55	388 ± 2·5	388 ± 2·0	388·2	−0·001	−0·001
	3	27·12	559 ± 5	555 ± 4	557·5	+0·003	−0·004
	4	39·12	784	786 ± 7	793·4	−0·012	−0·009
	5	54·5	1041	1069 ± 14	1093·4	−0·048	−0·022
	6	73·5		1369	1460		−0·062
	7	95·7		1748	1885		−0·073
	8	121·5		2160	2372		−0·089
	9	150·4		2586	2980		−0·132
270	1	13·05		280 ± 1·0	279·1		+0·003
	2	17·10		362 ± 1·3	359·5		+0·007
	3	23·88		491 ± 3	493·6		−0·005
	4	33·3		675 ± 5	679·2		−0·006
	5	45·5		913 ± 10	918·1		−0·005
	6	60·4		1197	1208		−0·009
	7	78·0		1483	1547		−0·041
	8	98·3		1790	1934		−0·075
	9	121·2		2115	2366		−0·106

cm^{-2} which was obtained in earlier experiments (MEISTER, 1961) by measuring the radial flux distribution within the experimental tank filled with D_2O.

In Table 2, a comparison is made between the α_k values of various modes and α calculated from equations (1) and (5) on the basis of the diffusion parameters obtained in Section 4 by the method of least squares. For the effective water height $H = 240$ cm, $N = 6$ as well as $N = 10$ detector intervals were chosen for comparison.

In general, there is an agreement better than 1 per cent between experimental and calculated values of α, if the index k of the mode is not too high. In the region $k \geqslant N/2$, α_k turns out to be systematically lower than the calculated value. This may be ascribed to an influence of the axial mode of index $2N - k$ which contributes to the $A_k^*(t) = A_k(t) - A_{2N-k}(t) + A_{2N+k}(t) - \ldots$ given by equation (4). If $k \to N$, the decay constants of A_k and A_{2N-k} are approaching each other, so that a separation of these modes will become difficult (MEISTER, 1963). As is seen from Table 2, however, practically no systematic deviations occur if only modes with $k \leqslant N/2$ are considered. For this reason, modes having $k > N/2$ were discarded in the final analysis of the experimental data.

4. DETERMINATION OF DIFFUSION PARAMETERS

In order to derive $v_0\Sigma_a$, D_0, and C from the experimental data, α was assumed to be a function of B^2 given by equation (1) with negligible terms higher than B^4. In a least squares' fit to the data listed in Table 1 we took into account individual weighting factors $g_i \sim 1/\alpha^2$, assuming constant relative errors of α.

The diffusion parameters resulting from the fit are presented in Table 3; the errors listed were derived from the mean squared deviation of experimental points from the resulting interpolation curve [equation (1)]. The experimental data were evaluated separately in the regions $B^2 \lesssim 0{\cdot}0060$ cm^{-2} ($v_0\Sigma_a = 19{\cdot}0$ sec^{-1} was assumed in the region $B^2 > 0{\cdot}0060$, $C = 5{\cdot}25 \cdot 10^5$ cm^4/sec in the region $B^2 < 0{\cdot}0060$ cm^{-2}); the results are consistent within the limits of error, showing that the modal analysis method gives correct results if the number k of the mode is not too high ($k \leqslant N/2$).

TABLE 3.—DIFFUSION PARAMETERS OBTAINED BY METHOD OF LEAST SQUARES

Buckling range (10^{-4} cm^{-2})	$v_0\Sigma_a$ (sec^{-1})	D_0 (10^5 cm^2/sec)	C (10^5 cm^4/sec)
$13{\cdot}0 < B^2 < 60$	$21{\cdot}1 \pm 2{\cdot}0$	$1{\cdot}990 \pm 0{\cdot}010$	
$80 < B^2 < 470$		$2{\cdot}005 \pm 0{\cdot}010$	$5{\cdot}15 \pm 0{\cdot}29$
$13{\cdot}0 < B^2 < 470$	$19{\cdot}0 \pm 2{\cdot}5$	$2{\cdot}000 \pm 0{\cdot}009$	$5{\cdot}25 \pm 0{\cdot}25$

Combining the experimental α (B^2) data within the whole buckling range we obtain:

$$v_0\Sigma_a = (19{\cdot}0 \pm 2{\cdot}5)\ sec^{-1},$$
$$D_0 = (2{\cdot}000 \pm 0{\cdot}009) \times 10^5\ cm^2/sec,$$
$$C = (5{\cdot}25 \pm 0{\cdot}25) \times 10^5\ cm^4/sec.$$

As is seen in Fig. 3, the experimental α-values are well represented by the curve drawn which was calculated on the basis of these values. Therefore, a possible B^6 term seems to have only small influence in this range of B^2.

The value of $\alpha_0 = v_0\Sigma_a = 19{\cdot}0 \pm 2{\cdot}5$ sec^{-1} is consistent with the $\alpha_0 = 16{\cdot}5$ sec^{-1} calculated from nuclear cross sections for 99·82 mol per cent D_2O, but it is rather insignificant because of its large limits of error.

The diffusion constant D_0 turns out to be somewhat lower than the results of stationary experiments (BAUMANN, 1962) and is believed to be rather accurate. It is, however, in good agreement with earlier results obtained by pulsed experiments (GANGULY and WALTNER; WALTNER and WESTFALL) and by the modulated source technique (RAIEVSKY and HOROWITZ, 1954).

The diffusion cooling constant C is definitely greater than that obtained in earlier experiments of comparable accuracy (WALTNER and WESTFALL) at $\vartheta = 28°C$, since our α-values at high B^2 are somewhat lower than those of WALTNER and WESTFALL.

The value of $C = 5{\cdot}25 \times 10^5$ cm^4/sec resulting from our experiments is in good agreement with $C = 5{\cdot}13$ and $4{\cdot}85 \times 10^5$ cm^4/sec obtained by HONECK (1962) from calculations based on the scattering kernel of D_2O. The parameter C of the B^4 term includes just as well a contribution arising from one-velocity transport theory, which was given by SJÖSTRAND as $-\frac{1}{15}\frac{D_0}{\Sigma_{tot}^2}$; this term gives a rather large contribution of $-0{\cdot}753 \times 10^5$ cm^4/sec to the parameter C in the case of D_2O.

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