Diffusion Heating and Cooling of Thermal Neutrons in Water[1]

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The diffusion heating effect of thermal neutrons in light water has been investigated by measuring the diffusion length as a function of boric acid concentration. Diffusion lengths were obtained by a negative source technique while the boric acid concentrations were determined by both pulsed neutron methods and by direct chemical analysis. From these measurements the diffusion parameters of thermal neutrons in water have been obtained. Preliminary results for the microscopic absorption cross section of hydrogen σ_a, the diffusion coefficient $\overline{D}_o$, and the diffusion heating coefficient C of light water are 0.323 ± 0.032 barn, 37,135 ± 1,196 cm^2 sec^{-1}, and 2,836 ± 598 cm^4 sec^{-1} respectively. Pulsed neutron techniques have also been used to obtain independent data for these same diffusion parameters with the results $\sigma_a = 0.321 \pm 0.005$ barn, $\overline{D}_o = 37{,}651 \pm 545$ cm^2 sec^{-1}, and $C = 5{,}843 \pm 887$ cm^4 sec^{-1}. Both sets of data are in favorable agreement with recently published results.

Introduction

For a large weakly absorbing system the energy distribution of neutrons in thermal equilibrium with the atoms (or molecules) of the medium is given by the Maxwell-Boltzmann distribution law,

$$N(E)\,dE = 2\pi\,(\pi kT)^{-3/2}\,E^{1/2}\,e^{-E/kT}\,dE \qquad (1)$$

where N(E) dE is the fraction of thermal neutrons with energies between E and E+dE, k is the Boltzmann constant, and T is the absolute temperature of the medium. In many practical situations N(E) deviates significantly from the ideal Maxwellian spectrum because of the combined effects of neutron absorption and neutron leakage from the system.

In steady state situations, where the number of thermal neutrons per unit volume is not a function of time, N(E) progressively deviates from the Maxwell-Boltzmann distribution as the absorption within the medium increases. To maintain steady state conditions in a large medium with negligible leakage, neutrons must diffuse into a given volume element at the same rate as they are lost by absorption. Since most thermal neutron absorption cross sections vary as 1/v, where v is the speed of the neutron, the average energy at which neutrons are absorbed is less than the average energy of those compensating neutrons diffusing into the volume element. As a result, the neutron distribution function N(E) has a deficiency of low energy neutrons so that the average neutron energy is greater than that given by the Maxwell-Boltzmann distribution. According to kinetic theory the average energy is directly proportional to the absolute temperature. Therefore, the temperature characterizing the thermal neutron energy distribution for the case of

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1/v absorption is above that of the medium. For this reason the effect of neutron absorption on N(E) is called diffusion heating.

In a small weakly absorbing medium the main effect which removes neutrons is leakage. Since a fast neutron has a greater probability of escaping from the system than a slow one, the medium continuously loses neutrons whose average energy is greater than that of the neutrons remaining in the system. This effect, which becomes more pronounced as the leakage rate increases, shifts the peak of the neutron energy spectrum into the direction of smaller energies thereby characterizing N(E) with a temperature lower than that of the medium. Thus, the effect of leakage on the neutron energy spectrum is just the opposite of diffusion heating and is called diffusion cooling.

The distortion of the neutron energy spectrum from the ideal Maxwell-Boltzmann distribution due to absorption (diffusion heating) and leakage (diffusion cooling) effects is summarized in Fig. 1. One

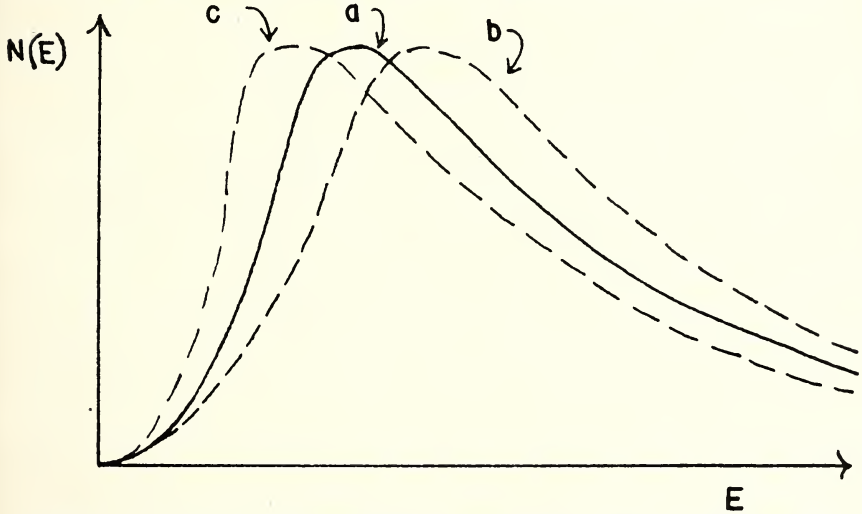

Figure 1. Distortion of thermal neutron energy spectrum due to absorption and leakage effects. a) Maxwell-Boltzmann distribution, b) distribution with 1/v absorption, no leakage, c) distribution with leakage, no absorption.

may investigate the diffusion heating effect quantitatively by measuring the thermal neutron diffusion length as a function of the concentration of a strong 1/v neutron absorber added to the system. By measuring the time rate of decay of thermal neutrons in a weakly absorbing medium as a function of the size (which determines the leakage rate) of the system one may determine the quantitative effects of diffusion cooling.

Theory of Measurement—Diffusion Heating

The steady state, source-free thermal neutron diffusion equation (3) is

$$D_o(E)\ \nabla^2 n(\vec{r},E) - v\ \Sigma_a(E)\ n(\vec{r},E) = 0 \qquad (2)$$

where $n(\vec{r},E)$ is the number of thermal neutrons per unit volume per unit energy at position $\vec{r}$ and having energy E. $D_o(E)$ is the energy-

dependent diffusion coefficient and $\Sigma_a(E)$ is the macroscopic absorption cross section. For a medium whose thermal neutron absorption cross section varies as 1/v this equation may be written as

$$\nabla^2 n(\vec{r}) - \kappa^2 n(\vec{r}) = 0 \tag{3}$$

where $\Sigma_a = \lambda/v$, $\kappa^2 = \lambda/\overline{D}_o$, and where

$$\overline{D}_o = \frac{\int_o^\infty D_o(E)\, N(E)\, dE}{\int_o^\infty N(E)\, dE}$$

For $\kappa^2 = 0$, N(E) is given by the Maxwell-Boltzman distribution (Eq. 1). Now $\overline{D}_o$ may be thought of as a function of κ^2 and so can be expanded in a Taylor series about the point $\kappa^2 = 0$, corresponding to an absorption-less medium. Then,

$$\overline{D}_o(\kappa^2) = \overline{D}_o(0) + \kappa^2 d\overline{D}_o/d\kappa^2 + \cdots .$$

Hence,

$$\lambda = v\,\Sigma_a = \overline{D}_o(\kappa^2)\,\kappa^2 = \overline{D}_o(0)\,\kappa^2 + C_h\,\kappa^4 + \cdots \tag{4}$$

where the diffusion heating coefficient C_h is defined as

$$C_h = d\overline{D}_o/d\kappa^2\Big|_{\kappa^2 = 0}. \tag{5}$$

In order to simplify the notation $\overline{D}_o(0)$ shall hereafter be written as $\overline{D}_o$.

Now suppose boric acid, which is a 1/v absorber, is dissolved in a large volume of water. Eq. (4) then becomes

$$-v\,\Sigma_a(H_3BO_3) = v\,\Sigma_a(H_2O) - \overline{D}_o\,\kappa^2 - C_h\,\kappa^4 \tag{6}$$

where higher order terms in κ^2 have been neglected. One may measure κ^2, which is just the square of the reciprocal of the thermal neutron diffusion length, as a function of boric acid concentration by a negative source technique (1). Briefly, this method consists of measuring the neutron density function $n(\vec{r})$ along the axis of a large cylindrical moderator with a Pu-Be neutron source located on the base of the cylinder. Two axial distributions are needed. One is taken with a cadmium plate, having the same dimensions as the base of the cylinder, covering the neutron source while the second distribution is measured with the cadmium plate removed. For regions away from the top of the cylinder the point by point difference of the two axial distributions can be fitted to the equation

$$n(z) = A\, e^{-\gamma z} \tag{7}$$

where A is a constant and where

$$\kappa^2 = \gamma^2 - (2.405/R)^2. \tag{8}$$

R is the effective radius of the cylinder. Thus, the measured distributions determine γ from which κ^2 is calculated. Now $v\,\Sigma_a$ (H_3BO_3) may be measured directly by pulsed neutron techniques (5) or calculated from the known 1/v absorption cross section of boron (5) and the measured concentration of H_3BO_3. This concentration may be determined by a standard chemical analysis involving boric acid titration with mannitol.

As seen from Eq. (6), a least squares fit of the data to a parabola in κ^2 determines the diffusion parameters $v\,\Sigma_a\,(H_2O)$, $\overline{D}_o$ and C_h.

Theory of Measurement—Diffusion Cooling

Following a neutron burst, which may result from a pulse of deuterons striking a tritium target, the thermal neutron distribution in a source-free homogeneous medium is governed by the time-dependent diffusion equation

$$\overline{D}_o\ \nabla^2 n(\vec{r},t) - v\,\Sigma_a n(\vec{r},t) = \frac{\partial}{\partial t} n(\vec{r},t). \tag{9}$$

Eq. (9) may be solved by the method of separation of variables. After long times following each neutron burst the neutron distribution is well represented by the fundamental mode. For a pulsed neutron source located on the axis but beneath a cylindrical water moderator, the thermal neutron distribution corresponding to this fundamental mode is

$$n(r,z,t) = A\,J_o(2.405\,r/R)\,\sin(\pi z/H)\,e^{-\alpha t} \tag{10}$$

with $$\alpha = v\,\Sigma_a(H_2O) + \overline{D}_o\,B^2 \tag{11}$$

and $$B^2 = (2.405/R)^2 + (\pi/H)^2. \tag{12}$$

R and H are the extrapolated radius and height of the cylinder while B^2 is called the geometric buckling of the system. Now $\overline{D}_o$ is averaged over N(E) which depends on the neutron leakage rate and so on the size and shape of the system. Therefore, $\overline{D}_o$ may be expanded in a Taylor series about $B^2 = 0$, which corresponds to an infinitely large system for which there is no leakage.

$$\overline{D}_o(B^2) = \overline{D}_o(0) + B^2\,d\overline{D}_o/dB^2 + \cdots \tag{13}$$

Eq. (11) now becomes

$$\alpha = v\,\Sigma_a(H_2O) + \overline{D}_o\,B^2 - C_c\,B^4 \tag{14}$$

where higher order terms in B^2 have been neglected and where now $\overline{D}_o$ has the meaning $\overline{D}_o(0)$. The diffusion cooling coefficient C_c is defined by the equation

$$C_c = -\,d\overline{D}_o/dB^2\Big|_{B^2=0}. \tag{15}$$

Measurement of the decay constant α by pulsed neutron techniques (2) for a variety of cylinder sizes yields data which may be fitted by the least squares process to a parabola in B^2 thereby determining the constants $v\,\Sigma_a(H_2O)$, $\overline{D}_o$ and C_c.

The similarity between Eqs. (6) and (14) and between Eqs. (5) and (15) should be noted. κ^2 is equivalent to a negative geometric buckling B^2 while the decay constant α may be correlated with the negative poison $-v\,\Sigma_a(H_3BO_3)$. Thus, the diffusion heating and cooling coefficients are identical.

$$C_h = C_v = C \tag{16}$$

Experimental Arrangement

The experimental arrangement for the determination of α by pulsed neutron methods has been discussed previously (2) and will not be repeated here. A 15 gallon drum 38 cm in diameter, 55 cm deep, and lined with a cadmium sheet 0.020 inch thick was used for the diffusion heating measurements. Five Pu-Be neutron sources having a combined source strength of 8.0×10^6 neutrons per second were symmetrically

arranged around the axis of the tank on its base. Neutrons were detected by means on a neutron-sensitive scintillation probe manufactured by the Nuclear Chicago Corporation (Model DS8-10). The 96 percent enriched B^{10}, Zn(Ag) activated crystal had a diameter of ¾ inch, a thickness of $\frac{3}{16}$ inch, and was mounted at the end of a lucite light pipe 42 inches long. Both chemical and pulsed neutron techniques were used to determine $v\,\Sigma_a(H_3BO_3)$. Deionized water having a resistivity of about 1×10^7 ohm - cm and a temperature of 24 ± 1° C was used for all the measurements.

Experimental Results

A summary of the experimental data for both the diffusion heating and diffusion cooling measurements is shown in Table I. The data was

TABLE I. Experimental Data

Diffusion Heating		Diffusion Cooling	
κ^2 (cm^{-2})	$v\,\Sigma_a$ (H_3BO_3) - sec^{-1}	B^2 (cm^{-2})	α (sec^{-1})
0.1316 ± 0.0018	0.000	0.000	(4.68 ± 0.04)10^3
0.3303 ± 0.0075	(7.65 ± 0.12)10^3	0.050	(6.58 ± 0.06)10^3
0.4146 ± 0.0052	(11.65 ± 0.06)10^3	0.075	(7.54 ± 0.01)10^3
0.6350 ± 0.0089	(19.60 ± 0.11)10^3	0.100	(8.41 ± 0.02)10^3
0.7471 ± 0.0090	(25.06 ± 0.14)10^3	0.150	(10.25 ± 0.03)10^3
0.9305 ± 0.0180	(32.12 ± 0.14)10^3	0.200	(11.98 ± 0.02)10^3
1.1211 ± 0.0505	(41.91 ± 0.17)10^3	0.250	(13.82 ± 0.10)10^3
1.4218 ± 0.0757	(54.01 ± 0.18)10^3	0.300	(15.46 ± 0.13)10^3
1.4900 ± 0.0134	(56.29 ± 0.60)10^3	0.350	(17.34 ± 0.10)10^3
1.8166 ± 0.0295	(72.31 ± 0.35)10^3	0.400	(18.57 ± 0.21)10^3
		0.450	(20.49 ± 0.23)10^3
		0.500	(22.13 ± 0.17)10^3
		0.550	(23.66 ± 0.27)10^3
		0.600	(25.21 ± 0.20)10^3

analyzed using an extrapolation distance of 0.32 cm. A plot of the decay constant versus the geometric buckling is shown in Fig. 2. Data for the diffusion cooling experiment is plotted in the first quadrant while the diffusion heating results are shown in the third quadrant. The departure of the data from a straight line is a clear indication of the diffusion cooling and diffusion heating effects.

The diffusion parameters $v\,\Sigma_a(H_2O)$, $\overline{D}_o$, and C are determined by using a least squares process to fit the data in Table I to Eqs. (6) and (14). The results are shown in Table II along with some recently published data. Since the absorption cross section of oxygen for thermal neutrons is completely negligible, the 1/v microscopic absorption cross section of hydrogen σ_a^H may be calculated from $v\,\Sigma_a(H_2O)$.

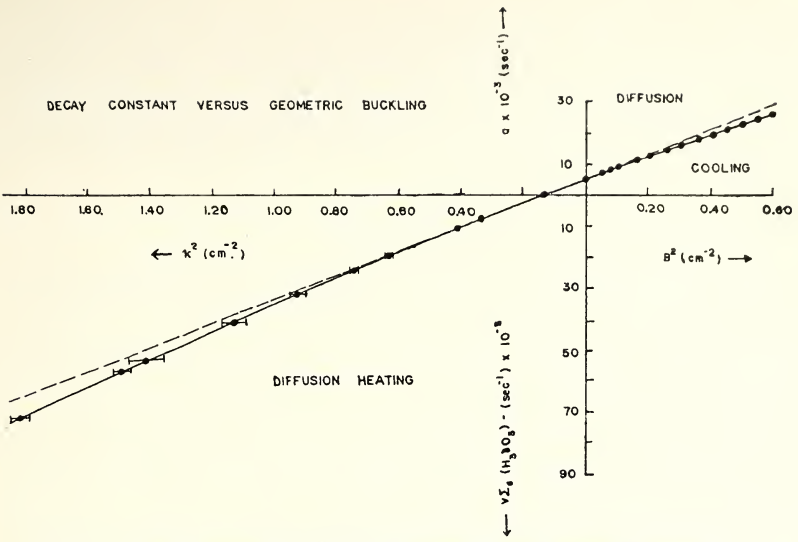

Figure 2. Least squares fit of decay constant versus geometric buckling.

TABLE II. Experimental Results
Thermal Neutron Diffusion Parameters For Water

Reference	$v\Sigma_a$ (sec^{-1})	σ_aH (mb)	$\overline{D}_o$ (cm^2 — sec^{-1})	C (cm^4 — sec^{-1})	Method
This work	4,748 ± 473	323 ± 32	37,135 ± 1,196	2,836 ± 598	H_3BO_3
This work	4,713 ± 67	321 ± 5	37,651 ± 545	5,843 ± 887	Pulsed
(4)	4,780 ± 27	325 ± 2	37,503 ± 366	5,116 ± 776	Pulsed
(6)	4,810 ± 24	327 ± 2	35,850 ± 100	2,900 ± 350	H_3BO_3
(6)			37,590	3,116	Theory

Summary

The diffusion parameters of thermal neutrons in water have been determined by two independent methods. Except for the diffusion cooling-heating coefficient, the results of the pulsed neutron measurement are in very favorable agreement with those obtained by the boron poisoning technique and likewise agree with recently published data. The two independently measured results for the diffusion heating coefficient are self-consistent and agree with the theoretical value. Although the measured values for the diffusion cooling coefficient are equal within experimental errors, they do not agree with the values obtained for the diffusion heating coefficient. Theoretically C_v and C_h are identical. One is inclined to place more confidence in the results from the boron poisoning measurements because the data covers a greater range of buckling values ($-\kappa^2$) than can be obtained in the pulsed neutron experiments. For small systems (large B^2) errors in the extrapolated dimensions in-

troduce relatively large errors in B^2. Because of the rapid rate at which neutrons leak out of a small system data may be taken before the fundamental mode has been achieved. For these reasons the diffusion heating experiment should yield a more reliable result for C than that obtained by pulsed neutron techniques. However, errors introduced by the neglect of the κ^6 and B^6 terms in Eqs. (6) and (14) may be significant and this matter needs more careful study .

Literature Cited

1. BRETSCHER, M. M. 1962. A Student Measurement of the Diffusion Length of Thermal Neutrons in Water by a Negative Source Technique. Amer. J. Phys. **30**:814.
2. BRETSCHER, M. M. 1962. Measurement of the Thermal Neutron Diffusion Parameters of Water by the Pulsed Neutron Method. Proc. Indiana Acad. Sci. **72**:249.
3. GLASSTONE, S. and M. C. EDLUND. 1952. The Elements of Nuclear Reactor Theory. D. Van Nostrand Co., Inc.
4. LOPEZ, W. M. and J. R. BEYSTER. 1962. Measurement of Neutron Diffusion Parameters in Water by the Pulsed Neutron Method. Nuc. Sci. and Eng. **12**:190.
5. MEADOWS, J. W. and J. F. WHALEN. 1961. Thermal Neutron Absorption Cross Sections by the Pulsed Neutron Method. Nuc. Sci. and Eng. **9**:132.
6. STARR, E. and J. KOPPEL. 1962. Determination of Diffusion Hardening in Water. Nuc. Sci. and Eng. **14**:224.