

# Diffusion Length of Thermal Neutrons in Water

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The thermal-neutron diffusion length in a hydrogenous medium can be simply determined by measuring the variation of the thermal-neutron density at large radii from the center of a photoneutron source. The necessary requirement is that the relaxation length of the source in a medium be less than the diffusion length, so that diffusion dominates at large radii. Using a Ra-Be photoneutron source, the diffusion length of thermal neutrons was determined by indium-foil measurements at large radii. A value of  $2.763 \pm 0.015$  cm was obtained. This result was confirmed in a second experiment with an  $\text{Sb}^{124}$ -Be source.

## 1. Introduction

In connection with the National Bureau of Standards program of absolute calibration of a standard-neutron source, the diffusion length of thermal neutrons in water was measured. A wide spread of values has been published, and the present experiment is compared with some of the earlier results.

For a high-energy neutron source the thermal-neutron density in water at large radii from the source decreases in accordance with the relaxation length defined below. If a source having only low-energy neutrons is employed so that the rate of neutron thermalization per unit volume falls off much more rapidly with increasing radius than the thermal-neutron density from a hypothetical thermal-point source, the thermal-neutron density at large radii will vary as a function of the diffusion length and radius only (as in the case of the thermal source). Photoneutron sources have sufficiently low maximum neutron energies for this purpose, and two sources having different neutron-energy maxima were selected for measurements. The general diffusion equation is

$$dn/dt = q - a + D\nabla^2 n, \quad (1)$$

where  $n$  is the thermal-neutron density,  $q$  is the rate of thermal-neutron production per unit volume (thermal-neutron source-strength density), and  $a$  is the absorption rate per unit volume.

$$a = nv/\lambda_a; \quad D = \lambda_{tr}v/3,$$

where  $\lambda_a$  and  $\lambda_{tr}$  are the thermal-neutron absorption and transport mean free paths, respectively, for the average Maxwellian velocity,  $v$ , and  $D$  is the diffusion constant. The steady-state equation is

$$\nabla^2 n - n/L^2 + q/D = 0, \quad \text{with } L^2 = \lambda_a \lambda_{tr} / 3, \quad (2)$$

where  $L$  is the diffusion length. This equation has been amply treated by Fermi and others in standard texts.

With water as the medium and at large distances from the source, one can assume with sufficient accuracy that

$$q = Ke^{-r/b}/r^2, \quad (3)$$

where  $K$  is a constant,  $r$  is the distance from the source center, and  $b$  is the relaxation length.

This relation is very nearly obeyed even for a source with a broad spectral energy distribution such as is generated by  $\text{Ra}=\text{Be}(\alpha, n)$ . At large  $r$ ,  $b$  is usually evaluated by measuring resonance activities at energies near thermal with Cd-covered foils. Then at large  $r$

$$\nabla^2 n - \frac{n}{L^2} = \frac{K}{Dr^2} e^{-r/b}. \quad (4)$$

Since the distribution has spherical symmetry, let  $n = u/r$ . Then

$$\nabla^2 n = \frac{1}{r} \left( \frac{d^2 u}{dr^2} \right)$$

and

$$\frac{d^2 u}{dr^2} - \frac{u}{L^2} = \frac{K}{Dr} e^{-r/b}.$$

The solution to the equation satisfying the boundary condition  $u \rightarrow 0$  as  $r \rightarrow \infty$  is (derivation in appendix):

$$u = nr = \frac{KL}{2D} e^{-r/L} \left[ C - \int_{\alpha_1 r}^{\infty} \frac{e^{-x}}{x} dx + e^{2r/L} \int_{\alpha_2 r}^{\infty} \frac{e^{-x}}{x} dx \right], \quad (5)$$

where  $\alpha_1 = 1/b - 1/L$  and  $\alpha_2 = 1/b + 1/L$ . The exponential integrals  $\int_{\alpha r}^{\infty} (e^{-x}/x) dx = EI(-\alpha r)$  may be evaluated from "Tables of sine, cosine and exponential integrals," NBS Mathematical Tables MT5 and 6 (U. S. Government Printing Office, Washington, D. C., 1947).

As  $r \rightarrow \infty$ , the solution may be expressed as

$$n = \frac{c_1 e^{-r/L}}{r} - \frac{b^2 L^2 k}{L^2 - b^2} e^{-r/b}. \quad (6)$$

If  $L > b$ , the first term will dominate at sufficiently large radii (the necessary requirement for evaluation of  $L$ ).

Since the second term acts as a perturbation, measurements with sources having different values of  $b$  serve as a check on the effect of the perturbation at the radii where measurements are possible.

For a  $\text{Ra-Be}(\gamma, n)$  source the maximum neutron energy emitted is only 0.7 Mev (compared to 13 Mev

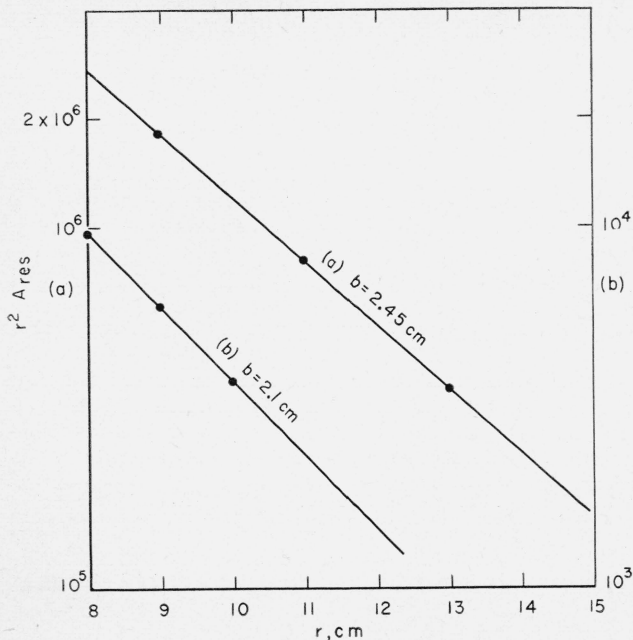


FIGURE 1. Resonance data for the determination of the relaxation length.

a, Ra-Be ( $\gamma, n$ ) source, indium foils; b,  $Sb^{124}$ -Be source, gold foils.

for an  $\alpha, n$  source), and the slowing-down length is short. Beyond 8 cm from the center of the National Bureau of Standards photoneutron standard [1]<sup>1</sup> the indium resonance activity decreases in accordance with eq 3, with a relaxation length  $b=2.45$  cm (fig. 1, a).

## 2. Experimental Arrangement

The Bureau photoneutron source was placed in the center of a 42-in. diameter stainless-steel tank filled with distilled water. Four indium foils, 2.0 by 3.4  $cm^2$  in area and 76  $mg/cm^2$  thick, held in light aluminum frames, were suspended by wires from a metal framework attached to the top of the tank. The foils were centered in the horizontal plane passing through the source at positions 90 deg apart. A cathetometer mounted horizontally above the tank made it possible to position the foils accurately to within a fraction of a millimeter at the radii employed. The source, a 4-cm-diameter Be sphere enclosing a 1-curie  $RaBr_2$  capsule, was pin-mounted on a vertical stainless-steel rod extending from the tank floor.

Periodic measurements of the water temperature gave a mean value of 22° C, with maximum deviations of 2 deg.

## 3. Counting Techniques

The foils were activated to saturation at radii 14, 16, 18, 20, and 22 cm from the origin with and without 0.020-in. Cd covers. All four foils were simultane-

ously counted with "thin" mica end-window Geiger counters, the outputs of which were fed to a mixing circuit, which permitted the foils to be counted as a unit by a single scaler. The sides of the foils facing the source also faced the counter windows when placed in their respective holders inside the lead Geiger-counter shields. Uranium standards were used to monitor the Geiger counters, and repetitive cycles were made of standard, background, and foils, with counting intervals of 5, 10, and 20 minutes, respectively, during the decay of the foils.

## 4. Results

The data are given in table 1. A correction of 6 percent must be made for the absorption of 1.44 ev in resonance neutrons by the Cd covers. Errors given are determined by the consistency of the data as well as the counting statistics. The activities given are the counting rates of the foils at saturation normalized to the same monitor reading. Figure 2 is a semilog plot of radius times thermal activity as a

TABLE 1. Data from NBS standard Ra-Be photoneutron source

Position ( $r$ in cm)	Activity without Cd (Counts per minute)	Resonance activity $\times$ 1.06 (Counts per minute)	Thermal activity ( $A_{th}$ ) (Counts per minute)	$rA_{th}$
14.0	7,657 $\pm 38$	190 $\pm 7$	7,467 $\pm 39$	104,540
16.0	3,546 $\pm 18$	64 $\pm 15$	3,482 $\pm 23$	55,710
18.0	1,533 $\pm 9$	* 22 $\pm 6$	1,511 $\pm 11$	27,200
20.0	655 $\pm 10$	* 8 $\pm 3$	647 $\pm 10$	12,940
22.0	292 $\pm 5$	* 3 $\pm 1$	289 $\pm 5$	6,360

\* Calculated.

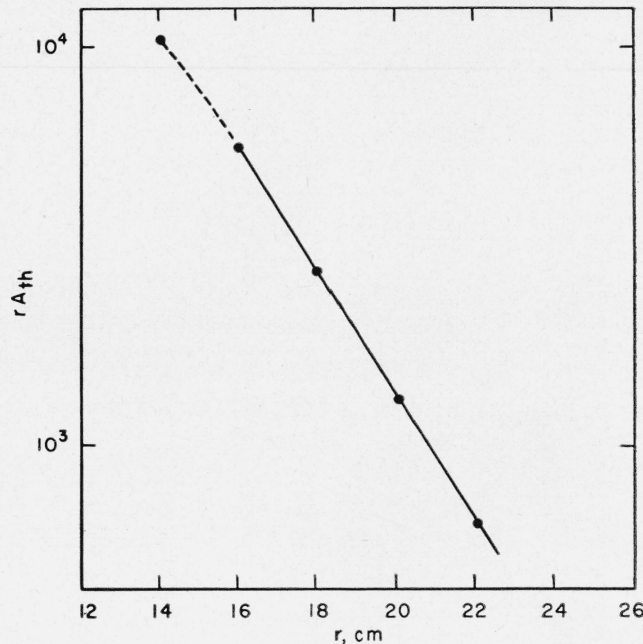


FIGURE 2. Indium foils in water.

Ra-Be ( $\gamma, n$ ) source.

<sup>1</sup> Figures in brackets indicate the literature references at the end of this paper.

function of  $r$ . Beyond 16 cm,  $rA_{th}$  decreases exponentially.

$$\frac{A_1 r_1}{A_2 r_2} (1 \pm \epsilon) = e^{\frac{r_2 - r_1}{L}},$$

where  $\epsilon$  is the experimental error of the ratio

$$\ln \left( \frac{A_1 r_1}{A_2 r_2} \right) \pm \epsilon = \frac{r_2 - r_1}{L}.$$

Using the data at 16 and 22 cm as an example,

$$\ln \frac{5571}{636} \pm \epsilon = \frac{6}{L}$$

$$2.170 \left( 1 \pm \frac{\epsilon}{2.170} \right) = \frac{6}{L}; \quad \epsilon = 0.018$$

$$L = 2.765 \pm 0.023 \text{ cm.}$$

The weighted least-squares fit of the data between 16 and 22 cm yield  $L = 2.763 \pm 0.015$  cm.

### 5. Sb<sup>124</sup>-Be Experiment

An antimony pellet that was irradiated in the Oak Ridge National Laboratory pile for 2 months was substituted for the RaBr<sub>2</sub> capsule in a 4-cm-diameter beryllium sphere. The 60-day Sb<sup>124</sup> has three  $\gamma$ -rays above the Be photoneutron threshold, the principal one having an energy of 1.7 Mev and giving rise to 24-keV neutrons [2]. As the neutron energies are considerably lower than those from the NBS standard source, moderation occurs closer to the source, and the relaxation length is shorter. A value of 2.1 cm

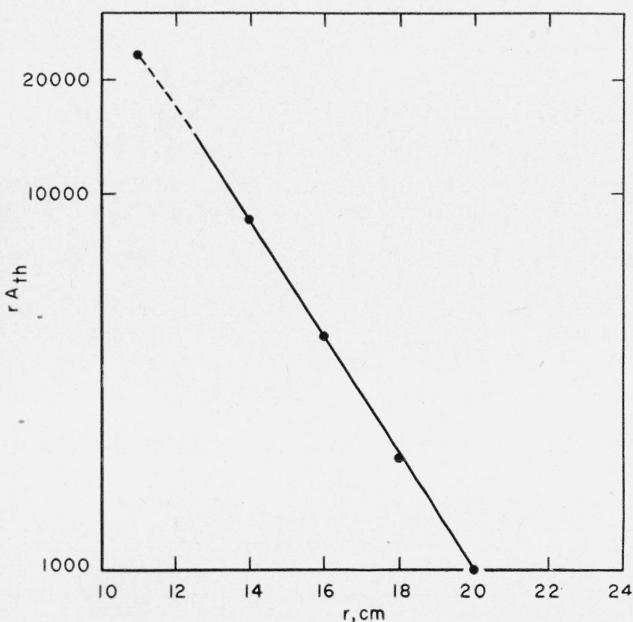


FIGURE 3. Indium foils in water.

Sb<sup>124</sup>-Be source.

TABLE 2. Data from Sb<sup>124</sup>-Be photoneutron source

Position ( $r$ in cm)	Activity without Cd (Counts per minute)	Resonance activity $\times$ 1.06 (Counts per minute)	Thermal activity ( $A_{th}$ ) (Counts per minute)	$rA_{th}$
14.0	625.8 $\pm 6$	12.4 $\pm 3.0$	613.4 $\pm 7$	8,588
16.0	262.3 $\pm 10$	<sup>a</sup> 1.5 $\pm 0.5$	260.8 $\pm 10$	4,173
18.0	110.7 $\pm 6$	<sup>a</sup> 0.5 $\pm 0.2$	110.2 $\pm 6$	1,984
20.0	50.6 $\pm 5$	<sup>a</sup> 0.2 $\pm 0.1$	50.4 $\pm 5$	1,008

<sup>a</sup> Calculated.

for  $b$  was obtained from the Sb-Be source in water, the measurements being made with Cd-covered indium and gold foils at radii beyond 8 cm from the source (fig. 1,b).

Indium foils that had been intercalibrated in the same slow neutron flux were simultaneously exposed to saturation at different radii from the source. The foils were successively removed and counted with the same end-window tube. For radii greater than 12 cm,  $rA_{th}$  decreases exponentially. At 14 cm the corrected cadmium reading is 1 percent of the thermal reading and decreases to 0.35 percent at 20 cm (calculated by using  $b=2.1$ ). The least-squares fit of the data (table 2) gives a value of  $L$  equal to  $2.76 \pm 0.07$  (fig. 3), which is in agreement with the Ra-Be result, even though the ratio of indium resonance to thermal-neutron counting rates is considerably lower for the Sb-Be over the range covered.

### 6. Discussion and Comparison of Results

For a purely thermal source located at the origin

$$n(r) = Ce^{-r/L}/r$$

and

$$\bar{r}^2 = \frac{\int_0^\infty n(r) r^4 dr}{\int_0^\infty n(r) r^2 dr} = 6L^2.$$

In analogy, the slowing-down distance,  $L_s$ , of neutrons from a high-energy source is usually estimated from the mean-square distance obtained from resonance-neutron measurements.  $\bar{r}_{res.}^2 = 6L_s^2$ , and the approximate relation

$$\bar{r}_{thermal}^2 = \bar{r}_{res.}^2 + 6L^2 \quad (7)$$

is used to calculate  $L^2$ .

Equation (7) treats the resultant thermal-neutron distribution as though the high-energy neutrons diffused from the source with a diffusion length equal to  $\sqrt{L_s^2 + L^2}$ . The shape of the radial thermal and resonance-neutron density distributions are actually Gaussian near the origin. As a result, the thermal and resonance  $\bar{r}^2$  values give somewhat overestimated diffusion lengths, and the value of  $L$  obtained by this method should represent an upper bound to the true figure.



Table 3 lists some of the early values obtained with RaBe( $\alpha, n$ ) sources, using eq 7.

TABLE 3.

Detector	$\bar{r}_{thermal}^2$	$\bar{r}_{res.}^2$	$L$	$b$	$L_0$
	<i>cm</i> <sup>2</sup>	<i>cm</i> <sup>2</sup>	<i>cm</i>	<i>cm</i>	<i>cm</i>
Rh [3]-----	327	277	2.85	9.4	6.8
In [4]-----	337	272	3.3	9.4	6.7
Dy+In [5]-----	330	278	2.94	10.1	6.8
In [6]-----	371	288	3.6	9.4	6.9
In [7]-----	363	313	2.88	9.8	7.2

Since eq 7 involves a small difference between two numbers, a 1 percent error in either  $\bar{r}^2$  term causes about a 3-percent error in  $L$ .

Berthelot, Cohen, and Reel [8] have determined  $L$  with a Ra-Be( $\alpha, n$ ) source in a water tank by a different technique. A thin spherical shell, which could be filled alternately with H<sub>2</sub>O (or H<sub>2</sub>O+H<sub>2</sub>SO<sub>4</sub>) and a CdSO<sub>4</sub> solution, surrounded the source. The difference in the neutron densities outside the sphere for the two cases arises from the diffusion of thermal neutrons present when the sphere is filled with water. Measurements with foils gave a best value of  $2.77 \pm 0.04$  cm for the thermal-diffusion length.

Larsson [9] recently has made extensive measurements of the same type at different CdSO<sub>4</sub> concentrations. An attempt was made to correct for the perturbation in the slow-neutron density in the immediate vicinity of the sphere and for the attenuation of high-energy neutrons by the cadmium. He obtained a diffusion length of  $3.25 \pm 0.13$  cm. However, a different interpretation of the data would give a lower value.

Perhaps the simplest method for measuring  $L$  is afforded by a nuclear reactor. If a water tank is placed on top of a thermal column, the neutron density, a few transport mean free paths from the boundary should decrease as  $e^{-z/L}$ , providing that the diameter and height of the tank are much greater than the diffusion length. Early reactor experiments gave a value of about 2.8 cm. Sisk [10] has recently obtained a value of  $2.67 \pm 0.02$  cm at 24° C, using the Oak Ridge pile and different boundary conditions.

Probably most of the variation in the values of  $L$  obtained by the different techniques is due to the failure to meet experimentally the rigorous boundary conditions required.

In the present experiment an upper limit to  $L$  is obtained unless the bracketed term in eq 5 can be determined. The exponential integrals have been evaluated, using  $L=2.76$ , and  $C$  was roughly determined for the radium source by calibrating the foils in a standard flux. A perturbation of less than 5 percent ( $C > 1.3$ ) in  $L$  should be caused by neglecting the exponential integrals. For the Sb-Be source, the exponential integrals are much smaller, and neglecting them should affect the calculation of  $L$  less. The Sb-Be( $\gamma, n$ ) source was fairly weak, and a stronger source would permit measurements beyond 20 cm, where the exponential integrals would have negligible effect.

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## 7. References

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## 8. Appendix

Solution of diffusion equation:

$$\frac{d^2u}{dr^2} - \frac{u}{L^2} = -\frac{K}{Dr} e^{-r/b}.$$

Let  $p = \frac{\partial}{\partial r}$

$$(p-1/L)(p+1/L)u = -\frac{K}{Dr} e^{-r/b}$$

$$u = \frac{-1}{(p-1/L)(p+1/L)} \frac{K}{Dr} e^{-r/b}.$$

Solving by partial fractions:

$$\frac{1}{(p-1/L)(p+1/L)} = \frac{S_1}{p-1/L} + \frac{S_2}{p+1/L}$$

$$S_1 = L/2, \quad S_2 = -L/2$$

$$u = LK/2D \left[ \frac{1}{p+1/L} - \frac{1}{p-1/L} \right] \frac{e^{-r/b}}{r}$$

$$u = C_1 e^{-r/L} + C_2 e^{+r/L} + KL/2D$$

$$\left[ e^{-r/L} \int \frac{e^{-r(1/b-1/L)}}{r} dr - e^{r/L} \int \frac{e^{-r(1/b+1/L)}}{r} dr \right].$$

Let  $\alpha_1 = 1/b - 1/L$ ,  $\alpha_2 = 1/b + 1/L$ .

The indefinite integrals may be expressed in terms of definite exponential integrals

$$\int \frac{e^{-\alpha x}}{x} dx = -\int_r^\infty \frac{e^{-\alpha y}}{y} dy = -\int_{\alpha r}^\infty \frac{e^{-x}}{x} dx.$$

To satisfy boundary conditions at infinity  $C_2=0$ , and setting  $C_1 = KLC/2D$ .

$$u = nr = \frac{KL}{2D} e^{-r/L} \left[ C - \int_{\alpha_1 r}^\infty \frac{e^{-x}}{x} dx + e^{2r/L} \int_{\alpha_2 r}^\infty \frac{e^{-x}}{x} dx \right]$$

To evaluate  $C$ ,  $K$  must be determined. Since less than 1 percent of the neutrons from either source are captured above thermal energies

$$Q = 4\pi \int_{r_0}^\infty q(r)r^2 dr = 4\pi \int_{r_0}^\infty -K e^{-r/b} dr,$$

where  $Q$  is the neutron emission rate of the source.

$$Q = 4\pi b K e^{-r_0/b}$$

The expression  $q = K e^{-r/b} / r^2$  does not hold down to the radius of the source, so a larger value of  $r_0$  than the source radius must be used. For the Ra-Be( $\gamma, n$ ) source  $r_0$  is estimated to be between 4.0 and 4.5 cm from previous data. Using  $K = Q e r_0 / b / 4\pi b$ , an absolute measurement of  $n$  would permit a rough determination of  $C$  as all other quantities are approximately known.

Exponential integrals for  $L = 2.76$ :

Source	$\alpha_1$	$\alpha_2$
Ra-Be.....	0.0458	0.7705
Sb-Be.....	.114	.8385

Ra-Be( $\gamma, n$ ) source		
$r$	$\int_{\alpha_1 r}^{\infty} e^{-z/x} dx$	$e^{2r/L} \int_{\alpha_2 r}^{\infty} e^{-z/x} dx$
<i>cm</i>		
16.....	0.351	0.037
18.....	.297	.030
20.....	.253	.024
22.....	.217	.019
Sb-Be( $\gamma, n$ ) source		
14.....	0.0868	0.0161
16.....	.0625	.0113
18.....	.0455	.0078
20.....	.0339	.0054

WASHINGTON, July 1, 1953.