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A METHOD TO MEASURE THE DIFFUSION COEFFICIENT BY NEUTRON WAVE PROPAGATION FOR LIMITED SAMPLES

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Abstract

A study has been made of the use of the neutron wave and pulse propagation method for measurement of thermal neutron diffusion parameters. Earlier works an homogeneous and heterogeneous media are reviewed. A new method is sketched for the determination of the diffusion coefficient for samples of limited size. The principle is to place a relatively thin slab of the material between two blocks of a medium with known properties. The advantages and disadvantges of the method are discussed.

1. INTRODUCTION

The existence of neutron waves was discussed in 1948 by Weinberg and Schweinler [1]. The first experimental work was reported in 1955 by Raievsky and Horowitz [2] who measured the diffusion coefficient of graphite and heavy water by the neutron wave technique. A number of experimental and theoretical studies on neutron wave propagation in multiplying and non-multiplying media have been published in the sixties. The possibility to investigate the neutron thermalization process and measuring the physical constants of thermalization was pointed out by Perez and Uhrig [3] and verified experimentally for a graphite system by Perez and Booth [4]. By the wave method it is possible to investigate the diffusion and thermalization process and the neutron spectra which are important in determining the behavior and properties of nuclear systems. A merit of the wave experiments lies in the possibility of obtaining information on space as well as on time dependency.

2.BASIC THEORY

Diffusion of monoenergetic neutrons from a harmonically modulated source in an infinite medium can be described by diffusion theory to obtain a physical background of the phenomenon. The space and time distribution of the neutrons in a medium in which there is a sinusoidally modulated source is obtained from the time dependent diffusion equation:

$$\frac{1}{v}\frac{\partial\Phi(x,t)}{\partial t} = D\frac{\partial^2\Phi(x,t)}{\partial x^2} - \Sigma_a \Phi(x,t) \tag{1}$$

for the plane modulated source in x = 0 with the total strength:

$$S = S_0 + \delta S\, e^{i\omega t} \tag{2}$$

We seek a solution in the form of the sum of the stationary and modulated parts of the neutron flux

$$\Phi(x,t) = \Phi_0(x) + \delta\Phi(x)\, e^{i\omega t} \tag{3}$$

For the stationary part $\Phi_0(x)$ we have :

$$\frac{d^2\Phi_0(x)}{dx^2} - \frac{\Sigma_a}{D}\Phi_0(x) = 0 \tag{4}$$

For x > 0 and a boundary condition $\lim_{x\to 0} j(x) = S_0/2$ we have the solution:

$$\Phi_o(x) = \frac{S_o L}{2D} e^{-x/L} \tag{5}$$

where

$$L^2 = \frac{D}{\Sigma_a} \tag{6}$$

is the diffusion length of the medium. For the wave part $\delta\Phi(x)$ we get:

$$\frac{d^2[\delta\Phi(x))]}{dx^2} - \frac{\Sigma_a + \frac{i\omega}{v}}{D} \delta\Phi(x) = 0 \tag{7}$$

and for similar conditions as described above we have the solution:

$$\delta\Phi(x) = \frac{\delta S L_\omega}{2D} e^{-x/L_\omega} \tag{8}$$

Here, L_ω is a complex diffusion length given by:

$$\frac{1}{L_\omega^2} = \frac{\Sigma_a + \frac{i\omega}{v}}{D} = \frac{1}{L^2} + \frac{i\omega}{vD} \tag{9}$$

or:

$$\frac{1}{L_\omega} = \alpha + i\beta \tag{10}$$

The neutron flux

$$\Phi(x,t) = \frac{S_o L}{2D} e^{-x/L} + \frac{\delta S}{2D} e^{-\alpha x} e^{i(\omega t - \beta x)} \tag{11}$$

is therefore propagated for small frequencies as an attenuated wave with the depth of penetration

$$\frac{1}{\alpha} \approx \sqrt{\frac{2 D v}{\omega}} \tag{12}$$

the wavelength

$$\frac{2\pi}{\beta} \approx 2\pi \sqrt{\frac{2 D v}{\omega}} \tag{13}$$

and the phase velocity

$$\frac{\omega}{\beta} \approx \sqrt{2 D v \omega} \tag{14}$$

The propagation velocity depends on the frequency, becoming larger as the frequency increases. It means that the medium in which the neutron waves propagate is dispersive.

3. MEASUREMENTS IN HOMOGENEOUS MEDIA

To obtain useful information on the neutron transport properties of a medium from neutron wave propagation experiments one usually observes the dispersion of the neutron waves to determine the propagation constant $1/L_\omega = \alpha + i\beta$. This is a complex function of the modulation frequency ω. A simple investigation using the diffusion approximation shows that the criteria for materials suitable for such experiments are: sufficient diffusion length L of neutrons in the medium (from the spatial viewpiont) and – in terms of time – small absorption $\Sigma_a = D_o / L^2$. This is the reason why only materials like graphite, beryllium and heavy water have been investigated and are of future interest in wave experiments.

TABLE 1

Thermal neutron properties of different media

	Graphite	Heavy water	Beryllium	Lead	Light water
L [cm]	50	100	22	13	2.8
$v\Sigma_a [s^{-1}]$	80	20	230	1260	4900

In Figure 1 is shown the space propagation of the modulated part of the neutron flux for two media with strongly different diffusion properties (see Table 1). The effect of modulation disappears at a distance of about two diffusion lengths from the source.

Many papers concerning neutron wave investigations have been published. In the great majority of the published papers the authors have investigated the dependence of the wave length and the

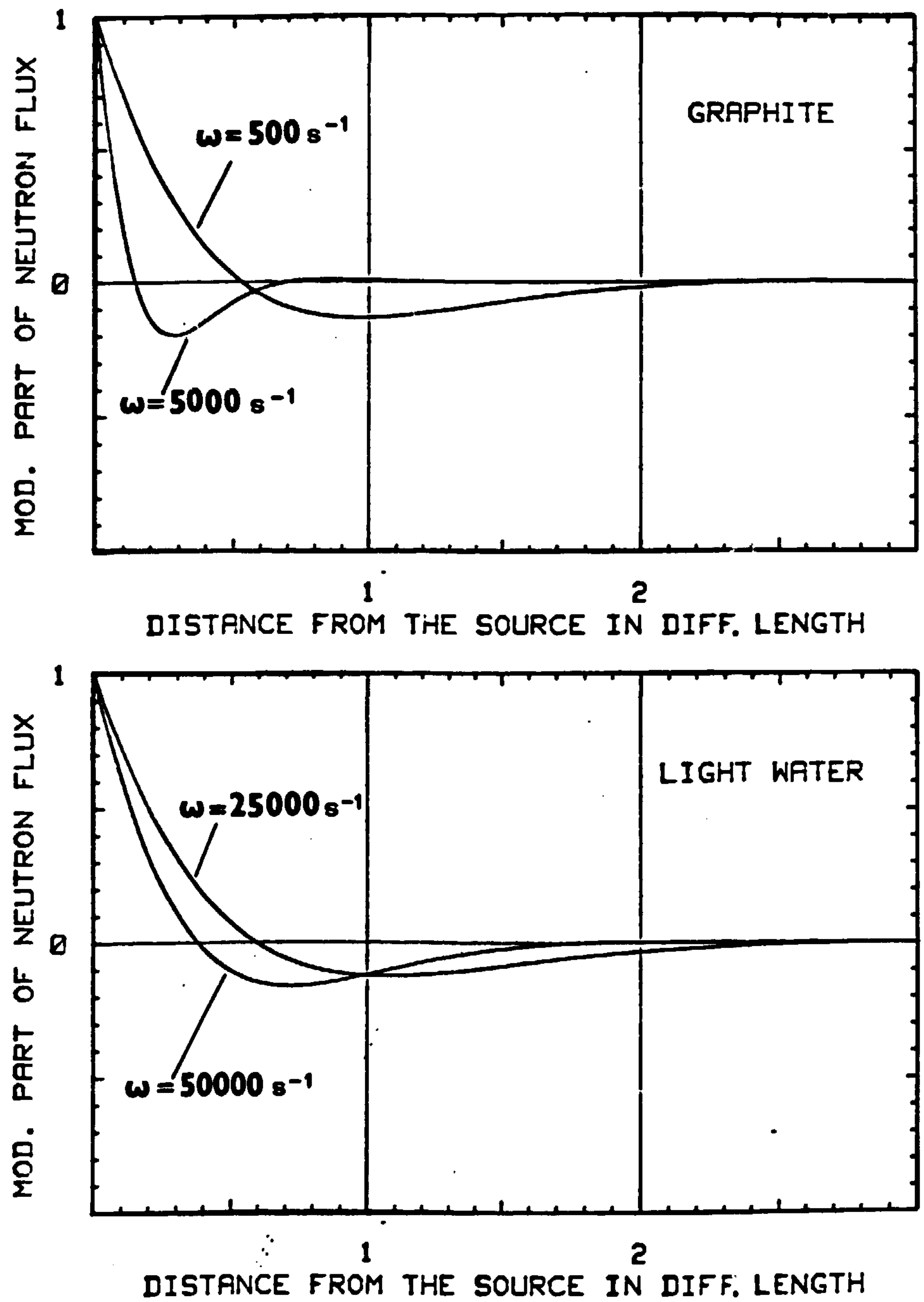


Figure 1. Space propagation of the modulated part of the neutron flux for graphite (L = 50 cm) and for light water (L = 2.8 cm).

attenuation distance of the neutron wave on the diffusion parameters for different frequencies of the sinusoidal source. Several approximations have been used and the neutron energy spectrum also has been taken into account [5 - 10] . Moore [11] first points out that the thermal neutron pulse propagation experiment could supply the same information as the thermal neutron wave propagation if a Fourier transformation is performed on the pulse responses. The pulsed propagation experiments have been made with good success for different media such as graphite [12], heavy water [13], lead [14] and beryllium [15 - 17] assemblies. A full description of the experimental procedure both for pulse and wave propagation has been given by Booth, Hartley and Perez [18] and by Dunlap and Perez [13]. This same experimental set up was recently used by Perez, Meade and Ohanian [19] for investigation of a

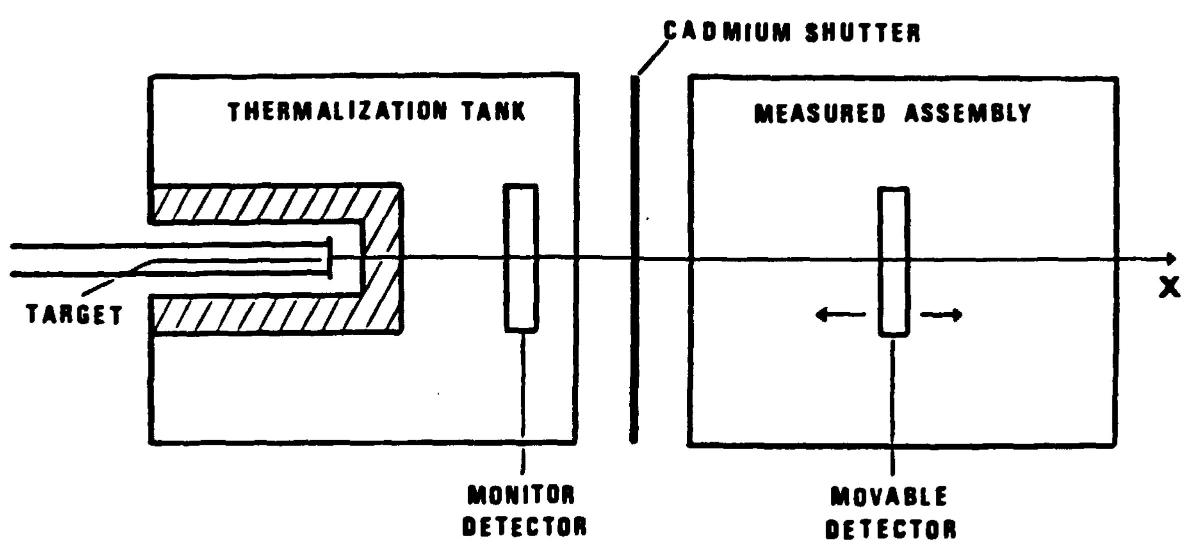


Figure 2. Measurement geometry for the neutron wave experiments

neutron wave experiment in small graphite blocks in order to study the transverse wave propagation and the asymptotic behavior of the neutron waves. A time profile of the thermal neutron pulse as a function of position in the medium under investigation is measured (in the case of a pulse source) and as a function of frequency (in the case of a sinusoidally modulated source). A thermal neutron source on the surface of the measured medium must be realized by a special thermalizing tank. Neutrons from a fast neutron source must be thermalized with the maximum thermal to fast neutron ratio and with the minimum loss in thermal neutron intensity. The typical thermalizing tank is constructed from steel, water and graphite. The steel section surrounding the neutron source takes advantage of the high inelastic scattering cross-section of iron. Next, graphite is used effectively to continue the thermalization by elastic

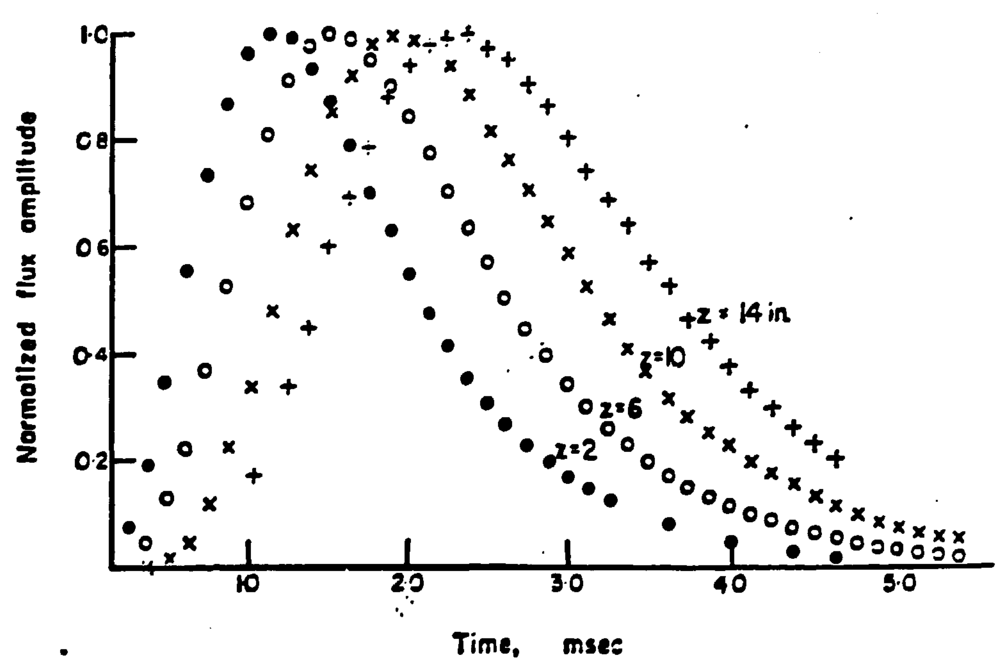


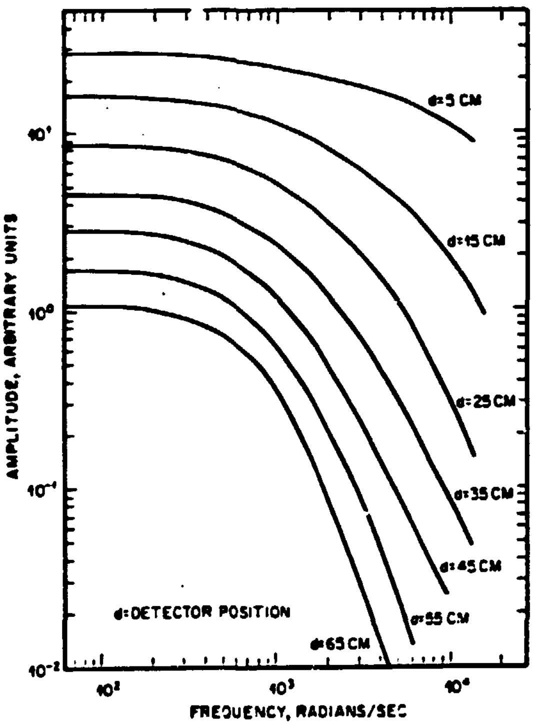
Figure 3. Typical thermal pulse at different z positions of the detector for the pulse propagation experiment in beryllium [15].

scattering with minimum absorption. The water or paraffin section is used to optimize the thermal to fast neutron ratio. The scheme of apparatus is shown in Figure 2.

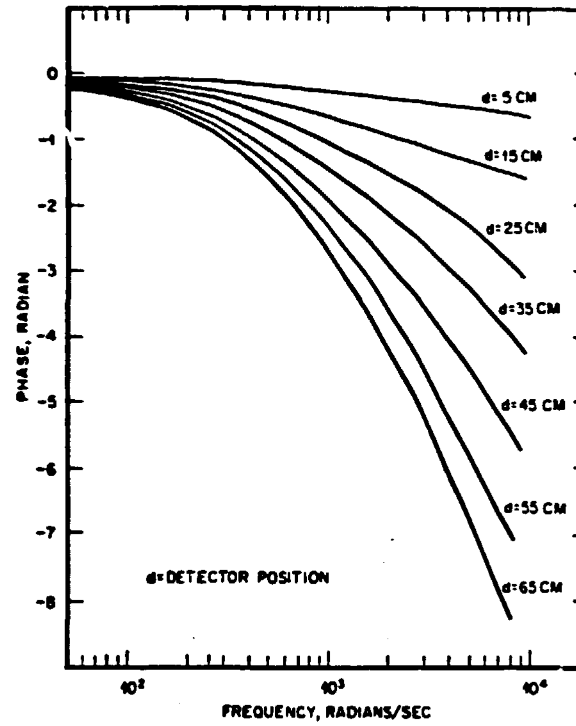
To subtract out the detector counts due to epicadmium neutrons escaping from the thermalization tank it is necessary to take two runs of measurement, with and without a cadmium shutter, respectively. The monitoring detector is used to monitor the source strength and normalize data taken at different x positions of the movable detector. A typical thermal pulse in several points of the medium is shown in Figure 3. The data analysis for the thermal neutron pulse consists of taking the Fourier transformation of the experimental data determining the amplitude and phase angle of Fourier coefficients for each detector loction. In Figures 4a) and 4b) is shown the frequency response of the neutron pulse for several detector positions and in Figures 4c) and 4d) are shown the amplitude and phase angle vs. distance from the source. The slope of these curves determines the real and imaginary parts (α and β) of the inverse complex diffusion length. The curves presented in Figure 4 may be obtained experimentally if a sinusoidally modulated source is used at different frequencies. All details about data collection and analysis are reported in [12, 13, 18].

4.MEASUREMENTS IN HETEROGENEOUS MEDIA

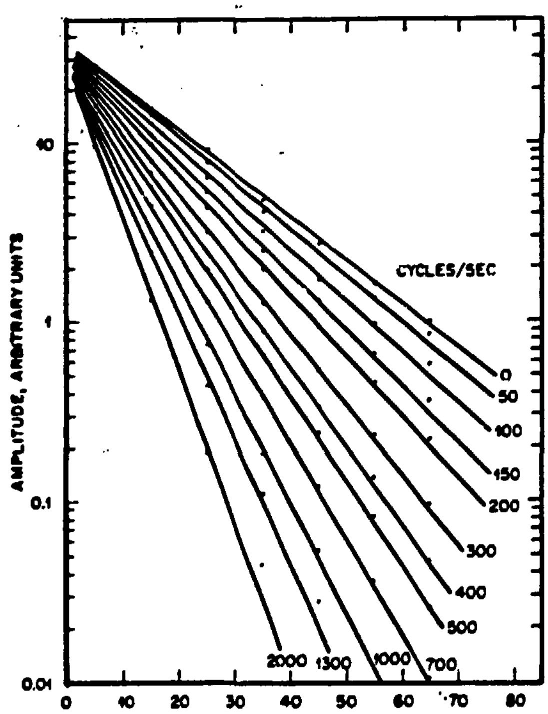
Besides the mentioned kind of investigation the neutron wave propagation across two adjacent media has also been taken into studies [20 -25]. The waves propagate as described above until they



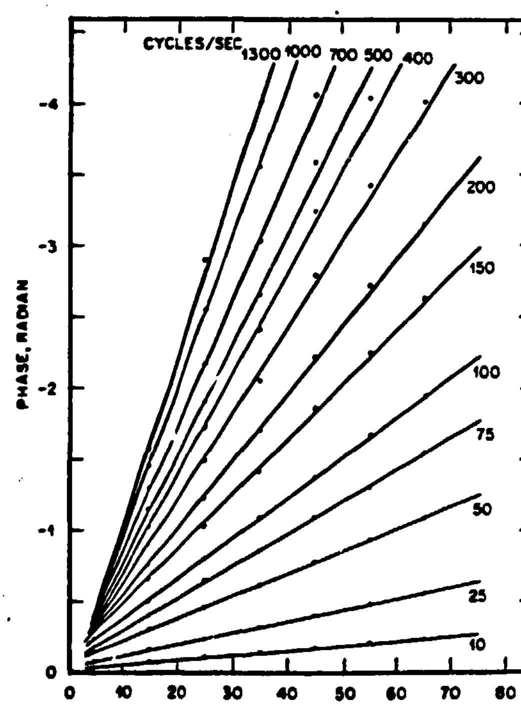
a) —*Frequency response of neutron-pulse propagation in graphite as a function of amplitude (corrected for input pulse shape).*



b) —*Frequency response of neutron-pulse propagation in graphite as a function of phase lag (corrected for input pulse shape).*



c) —*Amplitude vs. distance from source for several frequencies.*



d) —*Phase lag vs. distance from source for several frequencies.*

Figure 4. Fourier analysis of experimental results for a pulse propagation experiment in graphite [12].

reach the interface. At the interface there is a component which is reflected, whereas another component is transmitted. The solution of the diffusion equation must include terms of the incident, reflected and transmitted components. The magnitude of the reflected wave depends upon the properties of the medium reflecting it. Therefore, there is the possibility, by making measurements in one medium adjacent to another, to deduce parameters of the second medium (knowing the properties of the first one) [26]. The interface effect through reflection is negligible after a few mean free paths and decreases with the frequency. This effect is described by Baldonado [26] and Sadhwani [25]. The reflection effect makes this idea difficult to realize experimentally.

5.PROPOSED METHOD OF MEASUREMENT

In the present paper we suggest another method of measurement of the diffusion parameters of a sample by utilizing the wave propagation properties. The sample under investigation is placed between two adjacent layers of moderator. The plane modulated source is placed in the point $x = 0$ on the outerface of the moderator. The geometry is shown in Figure 5. The modulated neutron flux travels through the first "1" , second "2" and third "3" medium and is reflected by the two interfaces at $x = d$ and $x = a + d$. The attenuation and phase shift of the transmitted wave, which travels after the sample (in medium "3") , depend upon the neutron properties of both media. We look for the neutron properties of the sample if the properties of the moderator are known. It has been found numerically that for a fixed value of the frequency ω^* the phase shift of the neutron wave in medium "3" is the same (or differs by $2\pi/\omega$) as for the homogeneous moderator with Σ_{a1} and D_1 as parameters. Next it has been found that a dependence between the frequency ω^* and the diffusion coefficient of the sample exists (Figure 7). It may be recognized as a first step to elaborate an experimental method for measurement of the diffusion coefficient for relatively small samples.

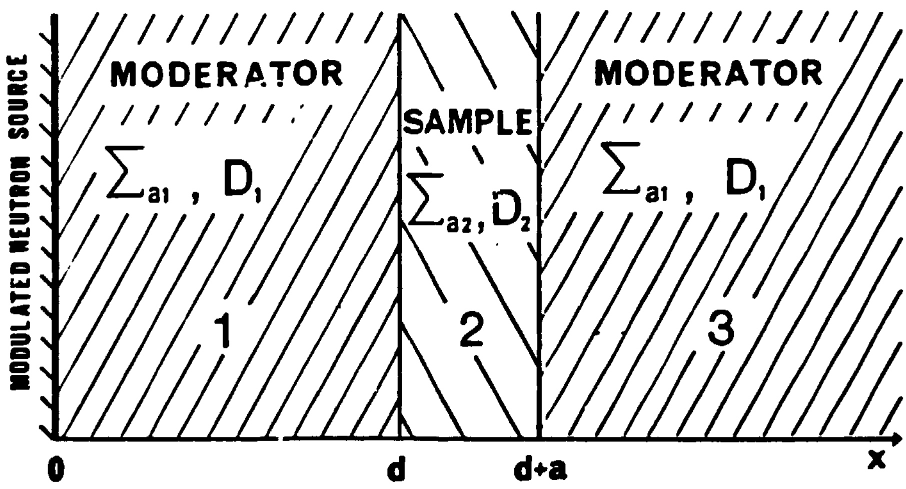


Figure 5. Calculation geometry. The sample "2" is placed between two layers of moderator "1" and "3" which extend over $0 \leq x \leq d$ and $a + d \leq x \leq +\infty$.

The diffusion equation (1) has been solved for the geometry presented in Figure 5 with the boundary and interface conditions:

$$\lim_{x \to 0} \left(-D_1 \frac{d\Phi_1(x)}{dx}\right) = \frac{S}{2} \tag{15}$$

$$\lim_{x \to \infty} \Phi_3(x) = 0 \tag{16}$$

$$\Phi_1(x) = \Phi_2(x) \qquad \text{for} \quad x = d \quad \text{and} \quad x = a + d \tag{17}$$

$$D_1 \frac{d\Phi_1(x)}{dx} = D_2 \frac{d\Phi_2(x)}{dx} \quad \text{for} \quad x = d \quad \text{and} \quad x = a + d \tag{18}$$

We seek for the modulated part of the flux the general solution of the form:

$$\delta\Phi_1(x) = A_1' \, e^{-x/L_{\omega 1}} + B_1' \, e^{x/L_{\omega 1}} \tag{19}$$

$$\delta\Phi_2(x) = A_2' \, e^{-x/L_{\omega 2}} + B_2' \, e^{x/L_{\omega 2}} \tag{20}$$

$$\delta\Phi_3(x) = A_3' \, e^{-x/L_{\omega 1}} \tag{21}$$

where $\delta\Phi_n(x)$ denotes the modulated part of the neutron flux in medium "1" , "2" and "3" , respectively. The terms with A_n' coefficients represent the transmitted parts of the fluxes and the terms with B_n' coefficients the reflected ones. According to the boundary conditions (16) - (18) we have:

$$\delta\Phi_1(x,t) = [\, e^{-x/L_{\omega 1}} + B_1 \, e^{(x-2d)/L_{\omega 1}}] \, e^{i\omega t} \tag{22}$$

$$\delta\Phi_2(x,t) = [\, A_2 \, e^{-(x-d)/L_{\omega 2}} + B_2 \, e^{(x-d)/L_{\omega 2}}] \, e^{i\omega t} \tag{23}$$

$$\delta\Phi_3(x,t) = B_3 \, e^{-(x-a)/L_{\omega 1}} \, e^{i\omega t} \tag{24}$$

where

$$B_1 = 2\frac{D_1}{L_{\omega 1}}\frac{1}{K_1}\frac{\frac{K_1}{K_2}e^{2a/L_{\omega 1}}+1}{\left(\frac{K_2}{K_1}\right)^2 e^{2a/L_{\omega 1}}-1} - 1$$

$$A_2 = 2\frac{D_1}{L_{\omega 1}}\frac{K_2}{K_1^2}\frac{e^{2a/L_{\omega 2}}\,e^{-d/L_{\omega 1}}}{\left(\frac{K_2}{K_1}\right)^2 e^{2a/L_{\omega 1}}-1}$$

$$B_2 = 2\frac{D_1}{L_{\omega 1}}\frac{1}{K_1}\frac{e^{-d/L_{\omega 1}}}{\left(\frac{K_2}{K_1}\right)^2 e^{2a/L_{\omega 1}}-1}$$

$$B_3 = 4\frac{D_1}{L_{\omega 1}}\frac{D_2}{L_{\omega 2}}\frac{e^{a/L_{\omega 2}}}{K_2^2 e^{2a/L_{\omega 2}} - K_1^2}$$

$$K_1 = \frac{D_2}{L_{\omega 2}} - \frac{D_1}{L_{\omega 1}}$$

$$K_2 = \frac{D_2}{L_{\omega 2}} + \frac{D_1}{L_{\omega 1}}$$

The transmitted flux $\delta\Phi_3(x,t)$ can be represented as a function:

$$\delta\Phi_3(x,t) = \rho\, e^{-x\alpha_1}\, e^{i(\omega t - \beta_1 + \phi)} \qquad (25)$$

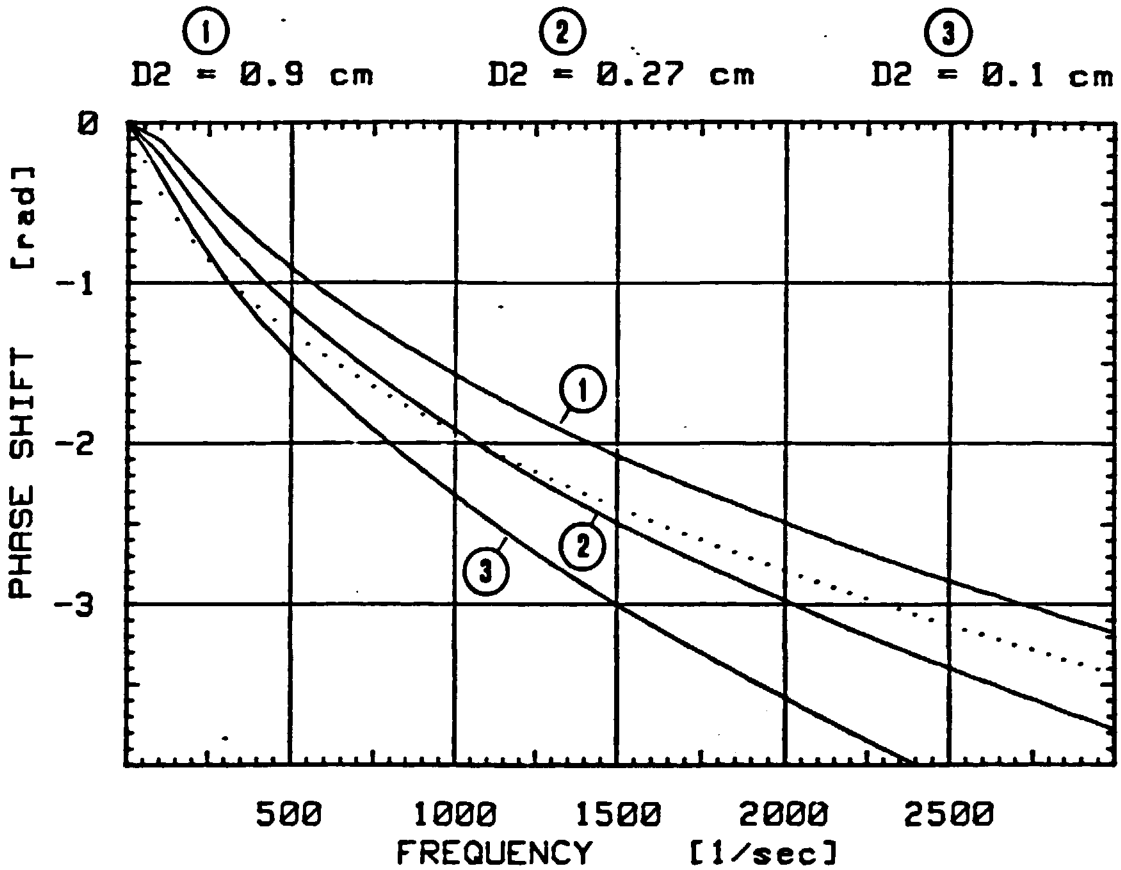
where $\rho\, e^{-x\alpha_1}$ is the attenuation factor of the neutron wave in the medium "3" and ($\phi - \beta_1$) the total phase shift. The values α_1 and β_1 are defined from equation (10) for the moderator:

$$\alpha_1^2 = \frac{\Sigma_{a1}}{2\,D_1} + \frac{1}{2v\,D_1}\sqrt{v^2\,\Sigma_{a1}^2 + \omega^2} \qquad (26)$$

$$\beta_1^2 = -\frac{\Sigma_{a1}}{2\,D_1} + \frac{1}{2v\,D_1}\sqrt{v^2\,\Sigma_{a1}^2 + \omega^2} \qquad (27)$$

The relation between the phase shift of the transmitted wave $\Phi_3(x,t)$ and the frequency for different parameters of the sample has been found numerically. It is presented in Figure 6. In the figure the relation between the phase shift and the frequency is drawn for the infinite medium, too, with the neutron parameters Σ_{a1} and D_1. The curve for the homogeneous system cross the other curves , which means that the phase shift for both cases is the same or differs by $2\pi/\omega$ in this point. The intersection of the curves can be explained on the basis of the phase shift determination equation (23). The phase shift increases when the diffusion coefficient decreases or when the absorption cross-section decreases.The dependence upon the cross-section is stronger for lower frequencies and weaker when the frequency increases. For higher frequencies the dependence upon the diffusion coefficient increases.

LOWER FREQUENCIES		HIGHER FREQUENCIES	
$\beta \uparrow$	when $\Sigma_a \downarrow$	$\beta \Uparrow$	when $\Sigma_a \downarrow$
$\beta \Uparrow$	when $D_1 \downarrow$	$\beta \uparrow$	when $D_1 \downarrow$



SIGMA A1 = 0.00036 1/cm SIGMA A2 = 0.01 1/cm
D1 = 0.9 cm
d = 20 cm a = 10 cm

Figure 6. The relation between the phase shift of the transmitted flux $\Phi_3(x,t)$ and the frequency for different values of the diffusion coefficient D_2 of the sample (moderator - graphite). The dashed line - for homogeneous graphite moderator.

Now one can explain the behaviour of the curves in Figure 6. Before the intersection point (for lower frequencies) the curve for the moderator is situated below the curve for the heterogeneous system because $\Sigma_{a1} \ll \Sigma_{a2}$. Above the frequency ω^* the dependence upon the diffusion coefficient is stronger and the curve for the moderator is situated above heterogeneous one because $D_2 < D_1$. The dependence between the frequency for the intersection point ω^* and the diffusion coefficient of samples has been found and is presented in Figure 7.

At last the possibility of a measurement from the point of view of neutron yield was checked. We assumed a sinusoidally modulated neutron source with the amplitude 10^6 n/sec and idealized measurement channel width Δt (see Figure 8). The perpendicular size of the investigated system has been assumed to be (b * c). The effective diffusion length is changed by the transverse buckling of the medium:

$$\frac{1}{L_\omega^2} = \frac{1}{L_{\omega\,inf}^2} + \left(\frac{\pi}{b}\right)^2 + \left(\frac{\pi}{c}\right)^2 \qquad (28)$$

The number of counts in one channel Δt (after one period) as a function of the distance from the source was evaluated. The results are presented in Table 2. It seems that the effect is measurable. In Table 3 is shown the time dependence of the counts per channel in a fixed point at $x = 10$ cm and $x = 85$ cm in the

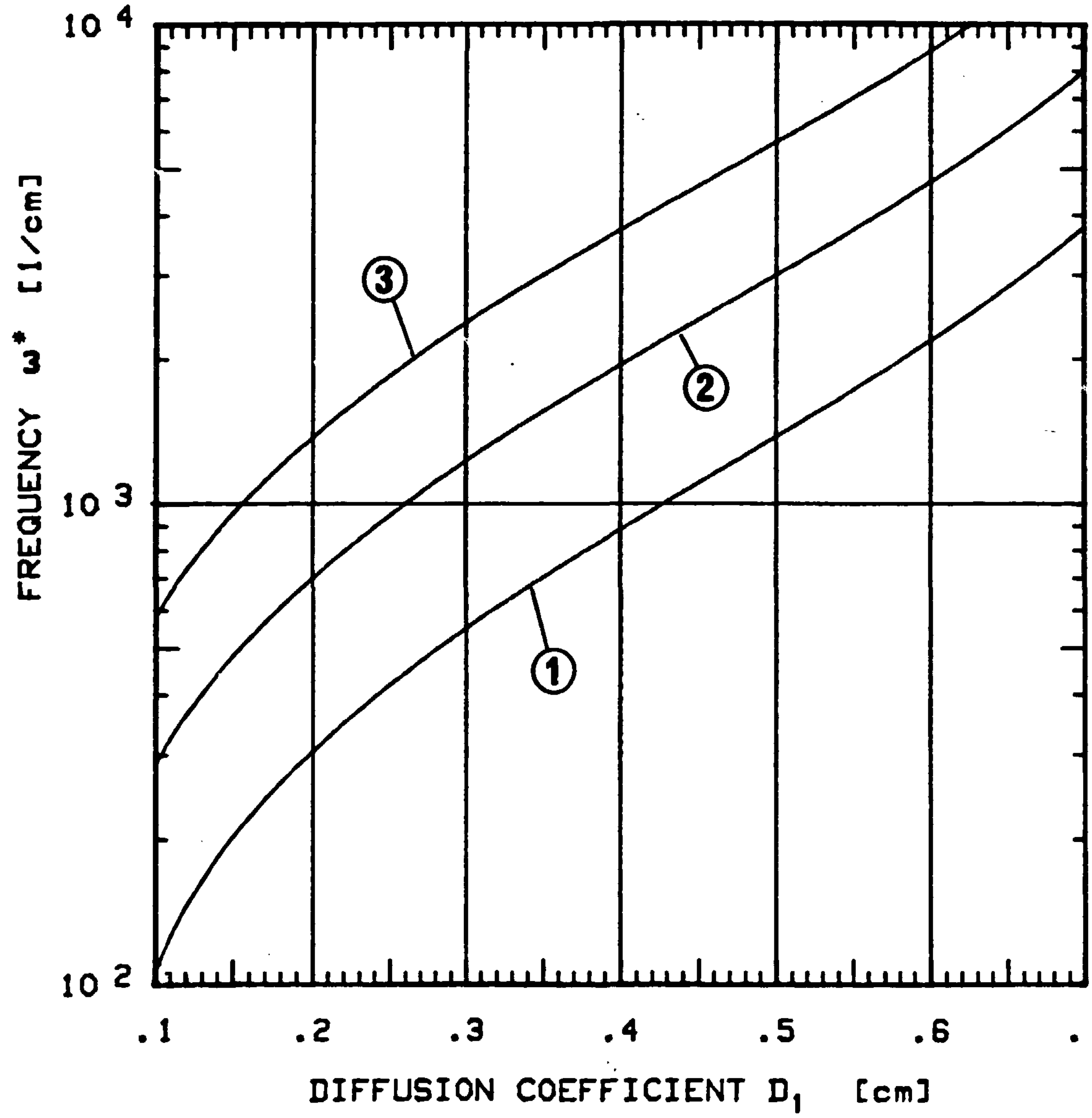
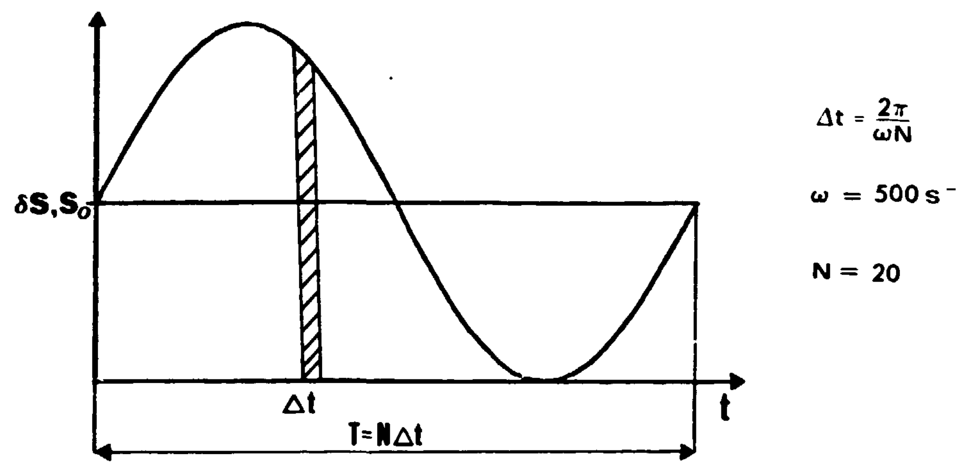


Figure 7. The relation between the diffusion coefficient of the sample and phase shift ω^* for three different values of the absorption cross-section of the sample. 1 — Σ_{a2} = 0.005 1/sec; 2 — Σ_{a2} = 0.01 1/sec; 3 — Σ_{a2} = 0.018 1/sec. Moderator — graphite.



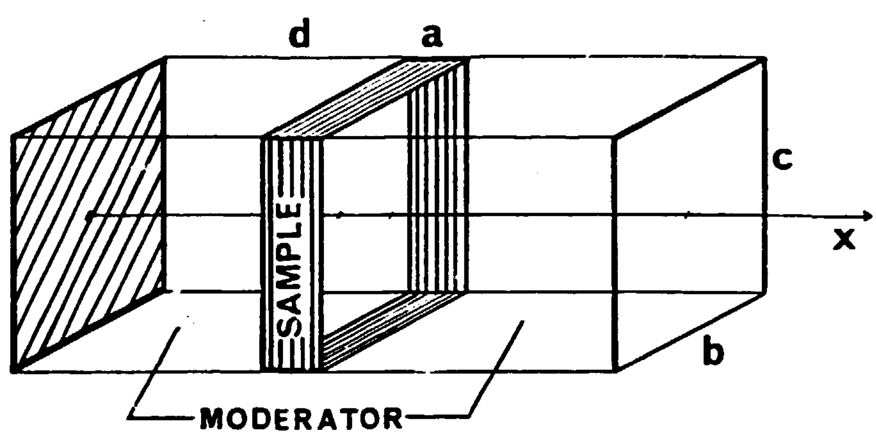


Figure 8. An example for calculation of neutron yield. The neutron source is modulated with a maximum depth of modulation. The size of the measured system is b * c = 100 * 100 cm^2 , d = 50 cm a = 20 cm. Neutron parameters of the moderator and of the sample are shown in Table 2.

TABLE 2

Variation of the number of counts per channel ($\Delta t = 0.63$ ms) with the distance from the modulated source (for the example shown in Figure 8).

Medium	Distance from source x [cm]	Number of counts
Moderator $\Sigma_{a1} = 0.00036$ [1/cm] $D_1 = 0.9$ [cm]	0	3 568
	10	3 957
	20	2 356
	30	1 414
	40	892
	50	205
Sample $\Sigma_{a2} = 0.001$ [1/cm] $D_2 = 0.1$ [cm]	51	196
	55	165
	60	135
	65	113
	69	105
Moderator (as above)	70	14
	75	11
	80	9
	85	7

Table 3

Variation of the number of counts with the time in a fixed point of the moderator (for the example shown in Figure 8).

Time after sin burst [ms]	Counts in one time channel Δt = 0.63 ms	
	x = 10 cm	x = 85 cm
0	3 947	7.1
1.3	3 798	7.0
2.5	3 053	8.3
3.8	1 997	10.7
5.0	1 032	13.1
6.3	528	14.7
7.5	676	14.8
8.8	1 421	13.5
10.0	2 478	11.1
11.3	3 442	8.7
12.6	3 947	7.1

moderator. The ratio of the maximum and minimum numbers of counts is about a factor of two in the point $x = 85$ cm (i.e. 10 cm after the sample).

The accuracy of the measurement of diffusion coefficient of the sample will depend upon the angle of the intersection of the curves in Figure 6 or upon the slope of the curve in Figure 7. But in this stage of evaluation of the problem it is too early to state anything definite about the accuracy of the method.

6. CONCLUSIONS

This first discussion of the problem shows that the method can give the diffusion coefficient from a limited sample size. If pulse propagation is used an ordinary pulsed neutron generator is a suitable neutron source. The detection and analysing system is relatively simple. There are some disadvantages of the method: a careful experimental design is needed for good result. The spectrum and transport effects must be taken into account, which may be difficult in the proposed geometry of measurement.

I want to acknowledge Prof Nils Göran Sjöstrand, head of the Department of Reactor Physics, for giving me the opportunity to make this work and for help and valuable comments. I want to thank Dr Gudmar Grosshoeg, Klaes-Håkan Beimer and Ryszard Rydz fot help during the preparation of the paper.

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