S N Ishmaev, I P Sadikov, A A Chernyshov

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Neutron moderation in ice and polyethylene at low temperature

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I.V. Kurchatov Institute of Atomic Energy

S.N. Ishmaev, I.P. Sadikov,
A.A. Chernyshov

The Choice and Optimisation of a Moderator for a Pulsed Slow-Neutron Source

Summary

The neutron physics requirements are considered for a moderator intended for the production of intense pulsed fluxes of slow neutrons using the linear accelerator with a booster now being built at the IAE. In this paper the results are presented of the experimental investigations carried out with the object of optimising hydrogenous moderators. The time dependence of the neutron flux for different energies and for various moderator compositions based on polyethylene and water was measured by means of a crystal spectrometer on the linear accelerator. The effect of the geometry, the thickness and the temperature of the moderator, the addition of absorbers and use of reflector-filters on the intensity and pulse width of monoenergetic neutrons was studied.

1. INTRODUCTION

A powerful pulsed neutron source based on a high-current linear electron accelerator (1) and subcritical breeding system using fast neutrons, ie a booster whose reactivity is modulated with time by a mechanical method (2), is being set up at the Kurchatov Institute of Atomic Energy for carrying out experiments in solid state and nuclear physics using slow neutrons. Certain characteristics of this pulsed fast neutron source are given in Table I.

The fast neutron source must be surrounded by a moderator since neutrons in the energy range from 0.001 to 1 eV are required for a broad class of experiments. It will be seen that the moderator must satisfy a number of specific conditions, the main one of which is the demand for maximum neutron intensity in the energy range of interest with the required pulse width. The use of a moderator of the usual type leads to serious losses in the intensity.

Thus results of multi-group calculations (2) show that the thermal neutron intensity ($E < 0.5$ eV) from the surface of a water moderator 5 cm thick, doped with boron to obtain a pulse length of about 18 μsec, is only a few per cent of the total flux of fast neutrons falling on the moderator. A large proportion of all neutrons leaks away in the moderation process and is absorbed in the moderator and the booster reflector. In principle considerable scope exists for enhancing the intensity of the thermal neutron source by increasing the efficiency of the moderator. To do this methods must be sought for reducing the neutron leakage during the moderation process but increasing it within the low energy range.

The problem of moderator optimisation has become particularly urgent recently since projects for high intensity pulsed neutron sources are being considered in various countries, using both linear accelerators with boosters and pulsed fast reactors (3). The effect of various factors on the integral yield of thermal and cold neutrons from moderators has been investigated in a number of papers, which are in the main experimental. But the available information is first incomplete, and secondly for the most part gives no information whatever about the pulse time-width as a function of neutron energy. It is important to know the time behaviour of neutrons escaping from the moderator ($\phi(E,t)$) for experiments to be carried out on a pulsed source. It is extremely difficult to obtain this dependence by calculation since a nonstationary kinetic equation with spatial dependence must be solved. At the same time the differential scattering cross-sections must be known for a broad neutron energy range. Still greater difficulties arise when multiregion systems are considered.

A crystal spectrometer using time-of-flight analysis is the most suitable instrument for the experimental solution of the problem of moderator optimisation. For a pulsed source this method allows the function $\phi_E(t)$ to be measured directly for several fixed neutron energies. Not long ago such methods were used to investigate the time dependence of neutron thermalisation in water (4), as well as for moderator optimisation for pulsed neutron sources (5, 6).

In this paper we first consider the requirements imposed on the neutron physics characteristics of the moderator from the point of view of experiments which can be set up on the pulsed source which is being built. Based on our experimental data, we then discuss the various ways of improving the moderator efficiency.

It should be noted that the problems of radiation damage of the moderator materials, problems of safety engineering, heat release, heat removal, etc., are not considered here. These require a separate analysis after a target design has been proposed which is optimum from the point of view of the neutron physics parameters.

2. REQUIREMENTS IMPOSED ON A PULSED SLOW-NEUTRON SOURCE

We characterise the slow neutrons pulse, $\phi_E(t)$, produced by the source using two parameters: the magnitude of the intensity at its maximum, ϕ_E^{max}, and its effective duration τ, defined as the ratio of the total neutron flux in the pulse to its maximum value:

$$\tau = \frac{\int_0^\infty \phi_E(t)dt}{\phi_E^{max}} , \qquad (1)$$

Thus all possible experiments may be divided approximately into three groups which are distinguished from one another by their requirements for these parameters.

In a majority of experiments to be carried out on pulsed sources the neutron energy analysis is performed by time-of-flight techniques. For example, traditional measurements of total cross sections (see Fig. 1a), structural investigations by the neutron time-of-flight method (Fig. 1b), measurements of inelastic scattering of neutrons by means of a phased chopper (Fig. 1c), or a crystal spectrometer in "inverse geometry" (Fig. 1d), etc., belong to this group. In such experiments $\phi_E(t)$ plays the part of the initial burst.

It is not difficult to show that the total number of neutrons N_E of given energy E incident within a time-of-flight interval Δt_K on a detector (or the sample) located at a distance L from the source is given by the following relation:

$$N_E \sim \frac{\int \phi_E(t)\,dt}{L^3} \Delta t_K \, \nu = \frac{\phi_E^{max} \, \tau \, \Delta t_K}{(\tau^2 + \Delta t_K^2)^{3/2}} \nu R^3 \qquad (2)$$

where ν is the pulse repetition rate, $R = \frac{\sqrt{\tau^2 + \Delta t_K^2}}{L}$ is the time-of-flight resolution, Δt_K is the channel width of the time analyser in cases a, b and d or the open time of the mechanical chopper in case c (see Fig. 1). We note that the quantity Δt_K is not a characteristic of the pulsed source, but is chosen by considering the particular experiment. But from the expression(2) it follows that for a given resolution N_E will be a maximum if $\Delta t_K = \frac{\tau}{\sqrt{2}}$. Then

$$N_E \sim \frac{\phi_E^{max}}{\tau} \nu R^3 \tag{3}$$

Consequently, for experiments of this group we must aim to achieve the maximum value of the ratio ϕ_E^{max}/τ which gives a figure-of-merit for the pulsed source.

However, independent methods can be used for monochromatisation and time selection of the neutron beam, such as a single crystal and a mechanical chopper (Fig. 1e), a rotating crystal (Fig. 1f) and so forth. In this case the largest possible value of ϕ_E^{max} is required from the source, while the pulse width τ must not be less than the burst width of the auxiliary system for shortening the pulse. In certain cases the maximum time-integrated flux, $\phi_E^{max}.\tau$, may be required from the pulsed source; for example, in experiments to store ultra-cold neutrons.

Thus it is necessary in all cases to endeavour to obtain the maximum value ϕ_E^{max}. In addition in a majority of cases, a reduction in the pulse width τ of the burst is required. However, the quantities ϕ_E^{max} and τ are found to be related with one another. To establish their relationship and to determine the requirements imposed on the moderator, we can use the following simple analysis.

A pulsed slow-neutron source is a combination of three independent systems: the target of the electron accelerator, the booster and the moderator. Therefore the resulting form of the slow-neutron pulse $\phi_E(t)$ is the convolution of three functions:

$$\phi_E(t) = \int_0^t g_3(t-t')dt' \int_0^{t'} g_2(t'-t'')\, g_1(t'')dt'' \tag{4}$$

where $g_1(t)$ is the pulse of the fast neutrons from the accelerator

$$g_1(t) = \begin{cases} q, & 0 \leq t \leq \tau_1, \\ 0, & t > \tau_1, \end{cases} \tag{5}$$

$g_2(t)$ characterises the "response" of the subcritical multiplying system to a neutron from the external source:

$$g_2(t) = \frac{1}{l} e^{-t/Kl} \frac{K_\infty - K_{eff}}{K_\infty} \quad (6)$$

$g_3(t)$ is the moderator efficiency, which describes the probability of emission from the moderator of a neutron with energy E per incident fast neutron. The form of the function $g_3(t)$ can be obtained by measuring the neutron flux time dependence after the moderator is irradiated by short pulses of fast neutrons.

The integral of the convolution of the three functions is given by

$$\int_0^\infty \phi_E(t)\,dt = \int_0^\infty g_1(t)\,dt \int_0^\infty g_2(t)\,dt \int_0^\infty g_3(t)\,dt =$$

$$= \phi_E^{max} \cdot \tau = g_1^{max} \cdot \tau_1 \cdot g_2^{max} \cdot \tau_2 \cdot g_3^{max} \cdot \tau_E \quad (7)$$

$$= \frac{q}{l} \frac{K_\infty - K_{eff}}{K_\infty} \phi_E^{max} \tau_1 \tau_2 \tau_E ,$$

where τ_1, τ_2, τ_E are the effective pulse widths of the accelerator, booster and moderator respectively, given by relationships analogous to (1). For the sake of simplicity, we assume that the quantities τ_i are equivalent to the variances of the functions $g_i(t)$. Then

$$\tau^2 = \tau_1^2 + \tau_2^2 + \tau_E^2 \quad (8)$$

Substituting the relations (7) and (8) into the expression, we find that for experiments of the first group

$$N_E \sim \frac{\tau_1 \tau_2 \tau_E}{\tau_1^2 + \tau_2^2 + \tau_E^2} \frac{q}{l} \frac{K_\infty - K_{eff}}{K_\infty} \phi_E^{max} \nu R^3 \quad (9)$$

If we can vary the moderator pulse width τ_E without altering ϕ_E^{max} substantially (see Section IV), then the figure of merit for the source for a fixed resolution R is given only by the relation between τ_1, τ_2 and τ_E. It is easy to see that expression (9) has, in principle, no optimum value with respect to τ_1, τ_2 and τ_E, and N_E increases with all three pulse widths although their mutual relationships are found to be different. For example:

$$
\begin{aligned}
&1) \quad \tau_1 = \tau_2 = \tau_E, \longrightarrow N_E \sim 1/3\,\tau_1 \\
&2) \quad \tau_1 \ll \tau_2 = \tau_E, \longrightarrow N_E \sim 1/2\,\tau_1 \\
&3) \quad \tau_1 \ll \tau_2 \ll \tau_E, \longrightarrow N_E \sim \frac{\tau_2}{\tau_E}\,\tau_1
\end{aligned}
\qquad (10)
$$

However, the two quantities — the accelerator pulse width τ_1 and the booster pulse width $\tau_2 = K\ell$ — cannot be made arbitrarily large, since there is a limit to the average power of the fast-neutron source. Therefore for a fixed resolution the maximum value of N_E is obtained when τ_1 and τ_2 are maximum, while the moderator pulse width τ_E is determined by the condition

$$\left.\frac{\partial N}{\partial \tau_E}\right|_{\tau_1, \tau_2} = 0$$, which gives

$$\tau_E = \sqrt{\tau_1^2 + \tau_2^2} \qquad (11)$$

Using the data from Table I and the relations (11) and (8), we obtain the following parameters for the source:

$$\tau_1 = 5.5\ \mu\text{sec},\ \tau_2 = 19.5\ \mu\text{sec},\ \tau_E = 20\ \mu\text{sec},\quad \tau = 28\ \mu\text{sec} \qquad (12)$$

Such working parameters are optimal if we can vary the flight distances within wide limits in order to obtain the required resolution $R_E = \frac{2\tau v}{L}$ for various neutron energies. Usually L is limited either by the frame overlap $L \leqslant \frac{v}{\nu}$, or by the value of the maximum possible flight path. Our arrangement has $L_{max} < 15$ m, and Table II gives the best values of the resolution R_E for this arrangement. They have been obtained if the source operates with the parameters (12) which are optimum from an intensity point of view.

The following conclusion can be drawn from this table. If the resolution $R_E \sim 1\%$ is considered to be adequate, then within the energy range $E < 0.1$ eV the fast neutron source must operate with maximum values of the parameters, while the moderator pulse width τ_E must be ~ 20 μsec. To obtain such a resolution for $E > 0.1$ eV we must reduce τ, which may be achieved by reducing the gain factor of the booster. Here, in accordance with the relation (11), τ_E also must be reduced to values of a few microseconds. It is obvious that such requirements for the pulse width can be satisfied only by a hydrogenous moderator.

It must be emphasised that such small values of τ_E required for the moderator are associated with the comparatively short burst of the fast-neutron source under consideration. For pulse reactors of the IBR [7] and SORA [8] types which produce a longer flash ($\gtrsim 50$ μsec) the requirements on τ_E may be less stringent.

3. EXPERIMENTAL METHODS

A series of experiments was carried out with the object of optimising the moderator for the pulse source being built. The linear accelerator of the IAE was used as a fast-neutron source with a short (0.6 μsec) burst. The time dependence $\phi_E(t)$ of the neutron flux leaking from various hydrogenous compositions was measured by means of a crystal spectrometer. The parameters of the experimental installation, the methods of measurement and the data processing are described in detail in [4].

The diagram of the experiment is shown in Fig. 2. Containers with various moderating compositions based on water and polyethylene were placed in a cryostat with the fast-neutron source located close to its rear wall. A set of flat aluminium containers was used so that the moderator thickness could be varied from 1 to 12 cms in steps of 1 cm. The area of the polyethylene plate was 29 x 24.5 cm^2, while that of water was 30 x 30 cm^2. The area of the moderator surface viewed by the monochromator crystal remained constant throughout and was 5 x 10 cm^2.

The measurements were performed at two temperatures, 300^o and 77^oK. The cooling of the system was carried out by liquid nitrogen which was fed by remote control to the cryostat from an external reservoir, and the temperature of the moderator was controlled by 3 copper-constantan thermocouples. In order to reduce the results of the measurements of the various systems to the same fast-neutron source intensity the cryostat containing the moderator under investigation could be drawn from the beam periodically by remote control in the course of a measurement. Consequently the accelerator target played the part of a standard source for calibration measurements. This enabled the data to be normalised with accuracy which was not worse than 3%.

In each experiment the function $\phi_E(t)$ was measured simultaneously for the following neutron energies; 0.005, 0.020, 0.045, 0.080 and 0.125 eV, which correspond to 5 orders of reflection of neutrons from single crystal of iron silicide. The time resolution depended on the neutron energy and ranged from 6 to 2 μsec. Corrections for neutron reflectivity by the single crystal, and for detector efficiency (see (4)) were made for each energy. In addition we determined from the measured function $\phi_E(t)$, the amplitude, ϕ_E^{max}, the total intensity, $J_E = \int_0^\infty \phi_E(t)dt$, and the effective pulse width of neutrons at each energy, $\tau_E = J_E/\phi_E^{max}$. These parameters for all the moderator compositions investigated are presented in Table III.

4. WAYS OF INCREASING THE EFFICIENCY OF THE MODERATOR

The shape of the pulse, $\phi_E(t)$, is determined by the time dependence of the neutron energy spectrum, and by the absorption and leakage of neutrons. The neutron spectrum is characterized by the processes of moderation, thermalisation and diffusion, and is determined by the type of moderator, the chemical bond of the atoms and the temperature of the medium. The effect of absorption and leakage depends on the composition, the dimensions and geometry of the moderating system.

We shall consider the effect of these factors separately, with the object of finding ways of obtaining the maximum pulse amplitude for slow neutrons leaking from the moderator, with the requirement that the pulse width must be of the order 20 μsec.

1. Effect of Temperature and Chemical Bond

The results of an investigation into the time dependence of neutron thermalisation in water at T = 318 and 77°K (4), can serve as an illustration of the effect of temperature. The function $\phi_E(t)$ presented in Fig. 3 was measured with high resolution for several neutron energies in the range from 0.005 to 0.247 eV.

The rapid growth in intensity characteristic for small time values is caused by the moderation and thermalisation process. For large time values the intensity for neutrons of all energies falls exponentially with the time, with the exponent corresponding to the neutron lifetime for an equilibrium spectrum in the given system.

For $E \gg kT$, the contribution of the diffusion coefficient is negligibly small, and the time taken to reach maximum intensity corresponds to the thermalisation time τ_E^o for the given energy. This time is also the principal factor determining the pulse width. τ_E^o depends on the density and the features of the chemical bonding of the hydrogen atoms, and increases as the neutron energy decreases.

For $E \lesssim kT$, the contribution of the diffusion coefficient is large and the pulse width for all energies is determined in practice by the lifetime of neutrons of the equilibrium spectrum

$$\tau_E \simeq \tau_D = \frac{1}{\langle \Sigma_a v \rangle + \langle D v \rangle_T B^2} \quad \text{for } E \lesssim kT \qquad (13)$$

Here $\langle \Sigma_a v \rangle$ and $\langle Dv \rangle_T B^2$ are the rates of absorption and leakage of neutrons, averaged over the equilibrium spectrum. We note that for hydrogenous moderators, such as water, polyethylene, zirconium hydride, the diffusion coefficient $\langle Dv \rangle_T$ decreases as the temperature is lowered; this leads to an increase in τ_D.

It is important to note the following. For neutrons of low energies ($E \lesssim kT$), the thermal motion is a factor which prevents the obtaining of the maximum possible amplitude ϕ_E^{max}. This is connected with the fact that a considerable proportion of neutrons are scattered with energy gain into a higher energy state. Cooling of the medium down to a low temperature, such that the condition $kT \ll E$ is fulfilled, allows not only the maximum intensity to be increased, but also the pulse width to be reduced considerably, down to the value τ_E^o. It is obvious that for a given moderator τ_E^o characterises the minimum pulse width which can be attained without reducing ϕ_E^{max}.

Figure 4 shows the dependence of the moderation time on neutron energy for three hydrogenous moderators. For H_2O and $ZrH_{1.9}$ the data of our experiments (4), (9) was used, while for liquid hydrogen the results of the calculation of Beyster and Russell (10) are given.

It can be seen that the behaviour of τ_E^o in the region of thermal neutrons is determined essentially by the chemical bonding of the hydrogen atom. A sharp reduction in the rate of moderation is observed after the neutron energy becomes less than a certain excitation energy characteristic to the given moderator, either rotary (H_2 - 0.015 eV, H_2O - 0.060 eV) or oscillatory (ZrH_2 - 0.13 eV) degrees of freedom. Since we require a pulse width $\tau_E \sim 20$ μsec, the use of H_2O as a moderator can be effective only for neutrons with energies $E \gtrsim 0.020$ eV. To obtain neutrons of lower energies ($E < 0.020$ eV) with the short pulse width, liquid hydrogen is the only effective moderator. Organic moderators, such as liquid methane, for example, are not considered here, since their practical use is limited on account of their high radiation damage.

2. Effect of Geometry and Dimensions of Moderator

The geometry and dimensions of the moderator are important factors in the determination of the intensity and pulse width of a pulse of slow neutrons. If the thermal neutron mean free path is small in comparison with the moderator thickness, then the intensity must depend essentially on whether neutrons are extracted from the surface or from within the moderator using a re-entrant hole. If it is assumed that there is a requirement that the source must operate with a maximum number of experimental beams, then various means by which neutrons may escape from the surface must be considered.

a. Neutron extraction dependence on moderator thickness

In this situation the quantity ϕ_E^{max} is proportional to the product of the probability of avoiding leakage and absorption in the moderation process, P, and the probability of emission from the moderator of neutrons of the given energy. If we consider that in the process of moderation an equilibrium spatial distribution is established in the system, then the neutron flux from the surface of the moderator can be described approximately by

$$\phi_E^{max} \sim \lambda_{tr}(E)\, v\, P / d_\ell \qquad (14)$$

where d_{ℓ} is the thickness of the moderator taking into account an extrapolation length and $\lambda_{tr}(E)$ is the transport mean free path.

An increase in the dimensions of the system reduces the neutron loss through leakage in the moderation process, but at the same time it reduces also the intensity gradient on the surface, which determines the magnitude of thermal neutron leakage. Consequently, an optimum exists for ϕ_E^{max} for some value of the moderator thickness.

We have carried out measurements of the intensity $\phi_E(t)$ for various thicknesses of a hydrogenous moderator slab. The geometry of the experiment and the results of the measurements for polyethylene and water are shown in Fig. 5. The errors presented in this figure include both the statistical accuracy of the measurements and the uncertainty of the normalisation. A comparison of the absolute neutron yields from polyethylene and water must take into account the difference in the transverse dimensions of the moderators, and check measurements have shown that the data for polyethylene must be increased by 25%. Qualitatively the results for water and polyethylene do not differ much. Both moderators are practically equivalent from the point of view of thermal neutron yield. It can be seen that the maximum of the integral neutron yield is fairly broad; for water it occurs at a thickness d = 5-6 cm and for polyethylene at d = 4.5-5.5 cm. From the point of view of pulse amplitude the optimum is obtained for d = 4-5 cm. It should be noted that although the shape of the distributions ϕ_E^{max} as a function of d differ slightly from one another, the value of d_{opt} for practical purposes does not depend on the energy of thermal neutrons, and corresponds approximately to the moderation length.

The results given have been obtained under conditions differing from those which will exist in the booster. Under real conditions the moderator will be irradiated by a flux of neutrons from the tungsten reflector surrounding the active core. At the same time the source will have large dimensions, the spectrum of fast neutrons will be somewhat softer, and furthermore, the leakage conditions of neutrons from the moderator will be different. In order to determine how strongly these conditions can effect the value of d_{opt}, measurements were carried out with an arrangement created by placing a tungsten wall 3.4 cm thick between the lead target of the accelerator and the water moderator. It was found that the total intensity J_E varied only slightly as a function of d. The curve of J_E (d) for E = 0.020 eV is shown as a broken line in Fig. 5 as an example. We see that the value of d_{opt} remains practically the same. Although the conditions of these measurements are not completely equivalent to the real situation, we can expect that the values of d_{opt} will not differ much, since the maxima in the distributions $J_E(d)$ and $\phi_E^{max}(d)$ are fairly broad. The calculations (2) carried out for the real conditions for the booster show that the integral yield of thermal neutrons is a maximum for d = 5 cm — this is not at variance with the results of these measurements.

In Fig. 6 the pulse shapes as a function of neutron energy are presented for the optimum (from the point of view of ϕ_E^{max}) thickness of water (d = 4 cm) at T = 300 and 77°K. The pulse parameters J_E, ϕ_E^{max} and τ_E are presented in Table III.

The pulse width τ_E increases monotonically as the thickness of the moderator increases. This is on account of the increase in the thermal neutron lifetime (13). As can be seen in Fig. 5, the values of τ_E are considerably greater than 20 μsec for a thickness of water d_{opt} = 4 cm. Since a simple reduction of the moderator thickness leads to an appreciable drop in the pulse amplitude, other methods of reducing the pulse width must be adopted; viz the introduction of additional absorption, or deep ($kT \ll E$) cooling of the moderator.

b. Moderator with holes

If a large surface area of a moderator is used for a given experiment, the neutron yield in a given direction can be increased by means of a system of narrow channels which allow neutrons to escape from inside the moderator. It is obvious, however, that in this situation the slow-neutron pulse width must increase also because of uncertainty in the escape coordinate. In addition this has been shown by the results obtained on IBR by Baiorek et al (11) who measured the time distribution and integral neutron yield for such a moderator with holes.

Measurements were carried out on a polyethylene moderator with a total thickness of 6.6 cm, containing holes with a diameter of 0.5 cm and a depth of 4.4 cm. The ratio of the hole area to the total surface area was ∿ 50%. Thus, with regard to the total number of hydrogen atoms, such a moderator was equivalent to a solid layer of polyethylene having the optimal thickness d = 4.4 cm. A comparison of the relations $\phi_E(t)$ obtained for three neutron energy values is presented in Fig. 7. From this figure and Table III we can see that the use of a moderator with holes is effective from the point of view of increasing the integral yield. This is particularly noticeable for neutrons of low energies for which the mean free path is small. Thus, for E = 0.005 eV, $J_{0.005\ eV}$ increases by a factor of 2.4. This gain, however, is achieved at the cost of the pulse width which is increased considerably (∿ 1.5 times). On account of the uncertainty in the escape coordinate we observe a less sharp leading edge of the pulse and a shift of the pulse maximum towards longer times. It can be concluded that the use of a moderator with holes is justified only for experiments where the integral intensity of thermal neutrons must be increased.

3. Use of Reflector-Filters

The optimal thickness of a hydrogenous moderator is not large, as is seen from Section 2, and as a result, many neutrons are lost through leakage in the moderation process. The intensity of neutrons emitted from the surface of the moderator can be increased, if we place downstream a medium possessing a large scattering cross-section in the high-energy range, but a small cross-section for thermal neutrons. Such a medium can be regarded as a reflector reducing the leakage in the moderation process, and as a filter transmitting slow neutrons. The basic requirements imposed on such a reflector-filter are that the ratio of the total scattering cross-sections in the epithermal and thermal ranges is large, and that the absorption cross-section is small.

A threshold curve of $\Sigma_s(E)$ and a small Σ_a are possessed by the polycrystalline moderators Be, BeO and C, for example. However, the limiting energy caused by the last Bragg cut-off is low, and the use of these materials in the role of reflector-filters can be advisable only for the range of cold neutrons.

a. Beryllium

The results of measurements for the H_2O + Be system at T = 77°K are presented in Fig. 8 and Table III. The thickness of ice was optimum (d = 4 cm), while the thickness of the beryllium wall varied from 0 to 4 cm.

As was to be expected, beryllium reduces the amplitude noticeably and extends the pulse width of neutron energy above the Bragg cut-off. However, a considerable gain in the intensity is observed for the energy E = 0.005 eV. A layer of beryllium having a total thickness of 2 cm enables the amplitude to be increased 1.7 times, and it should be noted that the pulse width for this energy is not altered. The data obtained show that from the point of view of the pulse amplitude the optimum thickness of the beryllium reflector-filter is between 2 and 4 cm. A further increase in the thickness leads only to a growth in the integral intensity. For d_{Be} = 4 cm the integral yield of cold neutrons is increased 2.2 times in comparison with ice having an optimum thickness. Measurements of integral spectra in this system at T = 300^{o}K showed an increase in the yield of cold neutrons due to beryllium of the same amount. This is associated with the fact that the difference in the cross-sections before and after the Bragg cut-off is sufficiently large in Be even at room temperature.

b. Single crystal quartz

Perfect single crystals can be used as the reflector-filters for thermal neutrons. It is known that the total scattering cross-section in an ideal single crystal is reduced sharply in the thermal energy range. The drop in the cross-section is the greater, the smaller the number of sources of incoherent scattering and the smaller the cross-section for inelastic scattering. Therefore cooled perfect single crystals with a high Debye temperature must be chosen and crystalline quartz SiO_2 is one such possible material. Special investigations were carried out by us into the effectiveness of such a reflector-filter.

The results of measurements of the energy and temperature dependence of the total cross-section of SiO_2 are presented in Fig. 9. In the single crystal at T = 294^{o}K, the cross-section in the thermal energy range is found to be 6 times smaller than for high neutron energies, while for T = 87^{o}K this ratio approaches $\sim$20. The cross-section of molten quartz with its amorphous structure is given in the same figure for the purpose of comparison.

A plane layer of crystalline quartz having a thickness of 4 cm was used as a reflector-filter screening the surface of a polyethylene moderator whose thickness was close to the optimum. The distribution $\phi_E(t)$ was compared for systems with and without a reflector and the results are shown in Figs. 10 and 11. It can be seen that the use of a quartz reflector indeed increases the flux of thermal neutrons. The gain in the pulse amplitude increases as the temperature falls and the energy of neutrons decreases, and it reaches the maximum value $\sim$ 1.7 for E = 0.005 eV, (T = 77^{o}K). Cooling allows the energy range in which this gain is observed to be increased considerably. This is

Heterogeneous threshold absorption

In this case the moderator is divided into two parts by means of a threshold neutron absorber such as cadmium. Then the conditions regarding leakage and absorptions for fast neutrons remain practically the same, while the thermal neutron lifetime is determined by a system which has much smaller dimensions. This is equivalent to increasing B^2 for thermal neutrons.

Fig. 12 shows how homogeneous and heterogeneous absorptions influence the pulse shape for neutrons of high and low energies. In the first case the neutron flux from the surface of a water moderator of optimum thickness (d = 4 cm) was measured. In the second case the moderator was divided into two equal parts by means of a cadmium sheet. The broken lines are the calculated curves corresponding to homogeneous doping of a pure moderator (d = 4 cm) by a quantity of $\frac{1}{v}$ - absorber, such that the same neutron lifetime is obtained as in the heterogeneous case.

The following can be said. The introduction of the absorber for neutrons of high energy leads to only a small reduction ($\sim$ 16%) in the pulse amplitude, but it enables its pulse width to be reduced considerably. For low energy neutrons a more noticeable reduction in the maximum intensity is observed. For example, an attempt to reduce the lifetime by a factor of 2.5 leads to a reduction of 30% in $\phi^{max}_{0.005\ eV}$ for heterogeneous doping, and 40% for homogeneous doping. We can see that both methods do not differ much, and are sufficiently effective for higher energy neutrons.

It must be emphasised that for the reduction of the slow-neutron pulse width it is better to add absorbers to a moderator of optimum thickness than to simply reduce the dimensions of the system. For example, to obtain τ_D = 23 μsec we must reduce the thickness of H_2O from 4 to 2 cm. This leads to a reduction of $\phi^{max}_{0.02\ eV}$ by a factor of 1.8 in comparison with the case of additional absorption.

5. Cold Neutron Source

The creation of an effective cold neutron pulsed source with the required pulse width gives rise to the greatest difficulties. This is associated with the low rate of neutron thermalisation at low temperatures for a majority of moderators. From this point of view liquid hydrogen is the best moderator for obtaining neutrons with energies $E < 0.020$ eV. Calculations based on a model of free H_2 gas give very small neutron moderation times (< 20 μsec) down to such low energies. Such assessments of course are valid because liquid hydrogen is chemically very weakly bonded.

The important factor which determines the cold neutron yield is the fact that for neutrons with energy $E < 0.015$ eV the mean free path in liquid parahydrogen sharply increases (approximately by a factor of 7) in comparison with the length for higher energies. This means that for sufficiently thick moderators, the loss of neutrons with energy $E > 0.015$ eV by leakage in the moderation process is not large, but at the same time cold neutrons must be emitted effectively from a fairly large moderator volume. On the other hand, the effect of reducing the scattering cross-section of parahydrogen must play a negative role when we endeavour to obtain neutrons with very small energies within the limited volume of the moderator. In addition, an appreciable broadening of the pulse for neutrons with $E < 0.015$ eV can take place as a result of the uncertainty in the escape coordinate. Unfortunately as yet there are no experimental data regarding the time-dependence of neutron thermalisation in liquid orthohydrogen and parahydrogen.

The major advantage of liquid hydrogen is its high radiation resistance in comparison with other hydrogenous moderators. However, the practical use of H_2 is associated, as we know, with serious problems of safety engineering and heat removal. With this in mind it is desirable to have the minimum volume of such a moderator. Therefore a design must be considered in which the neutron spectrum is first thermalised by another moderator, for example water held at the usual temperature, and then incident on a cold source with a small amount of H_2.

To test this idea, measurements were carried out for a two-zone system consisting of water (d = 3 cm) at $T = 300^{\circ}K$ and a thin layer (1 cm) of ice cooled down to $77^{\circ}K$, and the neutron intensity from the surface of the latter was measured. A heat insulating gap with the thickness 2 cm existed between the moderators. The geometry of the experiment is shown in Fig 13 and the results are compared in Table III for the two-zone system and a continuous moderator of optimum thickness at T = 300 and $77^{\circ}K$. It can be seen that the presence of a gap between the moderators leads to a considerable reduction in the amplitude and an increase in the thermal neutron pulse length. At the same time the integral intensity is not altered so much.

The use of the two-zone system enables the integral yield of cold neutrons to be doubled in comparison with water at T = 300°K. However, this quantity was less by about the same factor than for the case of a block of ice cooled down to 77°K. Check measurements at room temperature showed that as a result of the gap $J_{0.005\ eV}$ decreases only by 20% in comparison with a continuous moderator. We can conclude therefore that a layer of ice having the thickness 1 cm is insufficient for total thermalisation of neutrons.

The results obtained here cannot be transferred directly to the case of a liquid hydrogen source, since the neutron physics properties of ice and hydrogen differ much from one another. It is clear, however, that the unavoidable constructional gap between the warm and cold moderators, as well as the influence of the warm moderator itself, can lead to considerable worsening of the neutron pulse parameters.

5. CONCLUSIONS

Moderators such as water and liquid hydrogen must be used on the pulsed source being built to obtain intense slow neutron fluxes of small pulse width. The experimental investigations carried out allow certain conclusions to be drawn regarding the effectiveness of various moderating compositions based on water. The pulse characteristics for all the systems investigated for various neutron energies are presented in Table III. For ease of comparison the integral intensities and pulse amplitudes are normalized to their values for water and polyethylene of optimum thickness at T = 300°K. The actual values of J_E and ϕ_E^{max} for water (30 x 30 x 4) cm^3 and polyethylene (29 x 24.5 x 4.4) cm^3 at T = 300°K are shown in brackets.

On the basis of the analysis of the results obtained we propose the following variants of moderators for the various neutron energy ranges.

E > 0.050 eV

a) H_2O (d = 4 cm) + absorber, T = 300°K

or

b) H_2O (d = 4 cm) , T = 77°K.

We note that in this neutron energy range both types are practically equivalent. The use of additional absorption to obtain $\tau_E \sim 20$ μsec leads only to a small (> 20%) reduction in the pulse amplitude in comparison with the case of cooling. Apparently preference should be given for the first variant, since technically it is simpler, particularly when a heterogeneous threshold absorber such as cadmium is used.

$$0.020 < E < 0.050 \text{ eV}$$

This energy range is most important for a majority of experiments. Here are three moderator combinations possible:

a) H_2O (d = 4 cm) plus absorber, T = 300°K

b) H_2O (d = 4 cm) plus absorber plus SiO_2 (d = 4 cm), T $\lesssim$ 77°K

c) H_2O (d ∿ 2 cm) T_1 = 300°K plus H_2 (d ∿ 3 cm) T_2 = 20°K

Case a) is simpler, but intensity losses can be considerable in comparison with case b), where cooling and use of a quartz reflector-filter enables the pulse amplitude to be increased by a factor of not less than 2. It must be borne in mind, however, that the use of cooled ice in an intense source involves serious problems of heat release and dangerous accumulation of energy through radiation absorption by the frozen radicals. A calculation for an actual design must answer the question as to whether the second variant can be realised on the booster.

The use of liquid hydrogen (of any ortho-para ratio) appears to have scope for this neutron energy range for the following reasons. The temperature of the moderator (∿ 0.002 eV) lies substantially below the energy range of neutrons under consideration. This means that the pulse width is determined only by the moderation time, which is small. Therefore here no additional absorption need be introduced. If we consider that the optimum thickness depends approximately to the length of the moderator, then for hydrogen it is found to be 1.5 times larger than for water because of their difference in density. Thus, a layer of liquid hydrogen with a thickness of 6 cm and lying immediately adjacent to the reflector of a target may be the most effective moderator for this neutron energy range. We note that hydrogen has the best radiation resistance of all hydrogenous moderators and, in addition, is in a liquid state at low temperatures. Consequently it is estimated that the probability for the danger of occurrence of thermal shock caused by the recombination of radicals in hydrogen is much less than in ice. However, the heat release caused by radiation absorption in such a source can be unacceptibly large. For example, an estimate shows that power of $\sim$ 1kW can be released as a result of the slowing down of neutrons. An obvious solution of this problem is to use a two-zone H_2O-H_2 system (case c), but in order not to make the parameters of neutron pulse much worse, the layer of water and the constructional gap between the warm and cold moderators must be chosen as small as possible.

$E < 0.020$ eV

The only possible form for a moderator for this neutron energy range is a composition of the type

$$H_2O\ (d \sim 2\ cm)\ 300^oK\ \text{plus para-}H_2\ (d \sim 6\ cm)\ 20^oK$$

Here an increase in the H_2 layer can be justified because of the drop in the cross-section of parahydrogen for $E < 0.020$ eV. Of course, it is not possible to obtain a pulse length $\sim$ 20 μsec for low energy neutrons without substantial losses in intensity. This is associated with both the large lifetime and the time broadening of the pulse on account of the uncertainty in the escape coordinate. Therefore here we must endeavour to increase ϕ_E^{max}, but if the resolution is to be improved, then we must use additional experimental devices for shortening the pulse.

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TABLE I

Pulse repetition rate	$\nu < 150$ Hz
Electron accelerator pulse width	$\tau_1 \leqslant 5.5$ µsec
Total number of fast neutrons produced on Pu target/sec	$q.\nu.\tau = 3.4 \times 10^{14}$ n/sec
Lifetime of prompt neutrons in booster	$\ell = 65 \times 10^{-3}$ µsec
Maximum booster amplification factor	$K = \frac{1}{1 - K_{eff} + \beta} = 300$
Maximum resulting fast neutron pulse width	20 µsec
Average fast neutron flux escaping from the surface of the W reflector of the booster	0.35×10^{14} n/cm^2 sec

TABLE II

$\nu = 150$ Hz ; $L_{max} = v/\nu \lesssim 15$ m ; $\tau = 28$ μsec						
E (eV)	0.005	0.025	0.050	0.100	0.500	1.000
L_{max} (m)	6.7	15	15	15	15	15
R_E (%)	0.8	0.8	1.2	1.6	3.7	5.2

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I. V. Kurchatov Institute of Atomic Energy

S. N. Ishmaev, I. P. Sadikov,
A. A. Chernyshov

Neutron Moderation in Ice and Polyethylene at Low Temperatures

Abstract

In order to study the problem of the optimisation of a pulse source of slow neutrons on the IAE linear accelerator, an investigation has been made of the effect of deep cooling of hydrogenous moderators (H_2O and $(CH_2)_n$ at T = 300, 77 and 4.2°K) on the pulse shape of thermalized neutrons of various energies in a range from 0.0018 to 0.090 eV. At low temperatures polyethylene turns out to be a better moderator than ice; this is explicable by the greater normal oscillation level density in the low-energy part of the spectrum. Cooling $(CH_2)_n$ to T = 4.2°K makes it possible to increase by 7 times the pulse amplitude of neutrons with an energy of 0.0018 eV, and the total neutron output by 16 times in comparison with T = 300°K. For the energy range 0.1 - 0.01 eV, moderator cooling allows the neutron pulse width to be reduced by 2-3 times without loss of peak intensity. This improves significantly the conditions for spectroscopic time-of-flight resolution in this energy range.

1. INTRODUCTION

Research into the moderation of neutrons from a pulse source in deeply cooled moderators is of interest for two main reasons. When the environment is at a low temperature the processes of neutron scattering with energy gain are reduced significantly. Under such conditions it is possible to distinguish more precisely between the influence of the atomic chemical binding and the effects of thermal motion, and to use more effectively the experimental data on time-dependent neutron spectra for checking moderator models. On the other hand, information on the effect of the moderator temperature on the neutron output at various energies and their pulse time widths is important for the creation of an efficient source of slow neutrons for physical research on an accelerator or a pulse reactor. The present work is concerned principally with the second problem, and is a supplement to experiments carried out previously by the authors(1) on the optimisation of hydrogenous moderators.

It has been noted that in order to obtain cold neutrons on a high-powered pulse source, the most suitable moderator for neutron output, as well as for radiation stability and heat release, is liquid hydrogen. However, the creation of such a source presents a serious engineering problem, complicated by the question of ensuring safety. For this reason it is sensible to examine a simpler variant - a solid hydrogenous moderator, cooled to the temperature of liquid nitrogen or helium. In addition, such a moderator may also prove to be preferable from the point of view of higher energy neutron output. In this present work ice and polyethylene were studied since these moderators possess a high hydrogen atom density at temperatures of 300, 77 and 4.2°K.

2. CONDITIONS OF MEASUREMENT

The IAE linear accelerator serves as the pulse source for fast neutrons. The time-dependence of the neutron flux $\phi_E(t)$ transmitted from the surface of the moderator under study was measured by time-of-flight techniques on an apparatus with a crystal spectrometer in indirect geometry[(2)]. The fluxes for 7 chosen energies in a range between 0.0018 and 0.090 eV were measured simultaneously using as a monochromator a pyrolytic graphite crystal (8 cm in diameter and 0.34 cm in thickness) with a mosaic structure of 1.5°. The reflection of neutrons from the (002) plane at a Bragg angle of 87.5° was used. The time resolution in the measured energy range varied from 30 to 5 μsec respectively.

The moderator (a plate of 12 x 25 cm^2) was of the optimum thickness for thermal neutron output[(1)], 5 cm in the case of H_2O and 4.4 cm for $(CH_2)_n$. It was placed in a cylindrical cryostat with a capacity of 8 litres, filled with a coolant - liquid nitrogen or helium (see fig. 1). The total heat release under such conditions was about 1 watt, and continuous measurements at a temperature of T = 4.2°K (without the addition of helium) were limited to 3 hours. However, this time was sufficient to obtain information with reasonable statistical accuracy, thanks to the high reflectivity of the pyrolytic graphite crystal.

The results of all measurements were reduced to the same fast neutron source strength, and account was taken for corrections for the crystal reflectivity and the detector efficiency for the various neutron energies by the method described in paper [(2)].

3. THE RESULTS

In fig. 2 the results are given of the measurements of $\phi_E(t)$ for each neutron energy at the three temperatures of $(CH_2)_n$ and H_2O. On the basis of these data the degree of energy dependence of the full intensity $\phi(E) = \int_0^\infty \phi_E(t)dt$ and the neutron pulse width at half height was established (see fig. 3).

Let us consider the results obtained from the point of view of using ice and polyethylene as a source of slow neutrons for time-of-flight spectroscopy. It is known that the basic requirement for the pulse source is the realization of the maximum amplitude of neutron flux for the smallest possible pulse time width. When the neutrons are slowed to low energies they concentrate in a narrower energy range. However at this point there exist two factors which prevent the achievement of maximum neutron intensity; neutron scattering with energy gain and their absorption and leakage during the slowing down process.

The first factor plays a significant role when $E < kT$ where T is the environmental temperature. Its effect may be reduced by cooling the moderator to a temperature significantly lower than the energy range under study. This can be seen clearly from fig. 2. Moderator cooling to below 300^oK ($kT = 0.025$ eV) allows the neutron flux to be increased significantly in the region of $E < 0.030$ eV. In polyethylene at $T = 4.2^oK$, the total neutron output at $E = 0.0018$ eV increases 16 times, while the amplitude of the pulse increases approximately 7 times. For the region $E > 0.01$ eV the approximate ratio $E/kT > 1$, is shown to guarantee the achievement of the maximum possible neutron pulse amplitude in these moderators. The pulse time width is also dependent on the relationship between the neutron energy and the environmental temperature. When $E < kT$ the value $\Delta t_{\frac{1}{2}}$ is limited to a significant degree by an exponential decrease in the flux at large time values with a relaxation time constant, $<\Sigma_a v> + <Dv>B^2 - CB^4$, which characterises the neutron life-time in an equilibrium thermal spectrum for the given system. This is characteristic of the majority of curves for $T = 300^oK$, since the energy range being studied lies within the limits of the Maxwellian distribution at this temperature. When the ratio E/kT is increased, the contribution of the asymptotic component decreases, and $\Delta t_{\frac{1}{2}}$ will be determined by the thermalization time for a given energy. From the results obtained it follows that the cooling of ice and polyethylene to low temperatures allows the pulse time width for neutrons with energies of $E > 0.01$ eV to be reduced significantly without any intensity loss in the maximum amplitude. This improves the resolution conditions for time-of-flight experiments conducted in this energy range.

Neutron losses by absorption and leakage become especially significant when neutrons are slowed to low energies in a cooled moderator. Apart from the absorption cross-section, the diffusion coefficient and the moderator dimensions, these losses must depend to a considerable extent on the time needed for the neutrons to slow down to a given energy. With the reduction in energy this time increases, and its value will depend on the density of low-lying energy levels in the normal oscillation spectrum of the atoms in the moderator, on whose excitation the neutron might expend its energy. In this connection it is interesting to compare the results of measurements of $\phi_E(t)$ in water and polyethylene. At room temperature water and polyethylene are practically equivalent moderators both from the point of view of thermal neutron output and neutron pulse time width. However, at $T = 4.2^oK$ the intensity of the cold neutrons flux for polyethylene is found to be approximately twice that for water while the pulse width is less. This difference cannot be explained by a variation in the absorption and diffusion properties of these moderators. Evidently, it is a result of the higher density of levels in the low energy zone of the frequency spectrum of polyethylene compared with water. In particular this is indicated by the comparison in fig. 3 of data from the frequency spectrum $\rho(\epsilon)$ obtained from these moderators for $T = 295^oK$[3].

Thus the following conditions are necessary to achieve a high cold neutron intensity; the choice of a hydrogenous moderator which has a high density of levels in the low energy zone of the frequency spectrum; and its cooling to a temperature which is significantly lower than the energy range which is of interest. For the neutron energy range $E > 0.01$ eV a sufficiently effective and simple variant of the moderator source may be polyethylene, cooled by liquid nitrogen.

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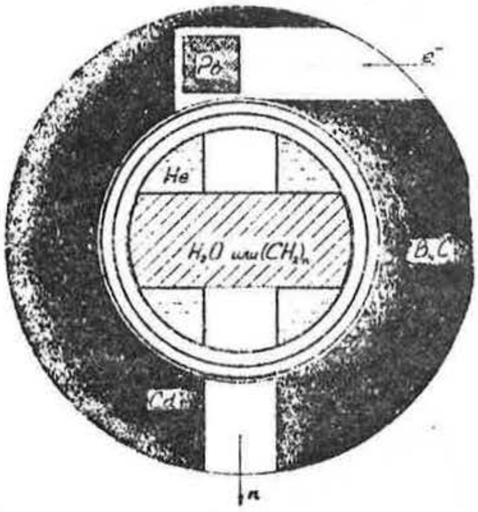

Figure 1 Diagram of the neutron source.

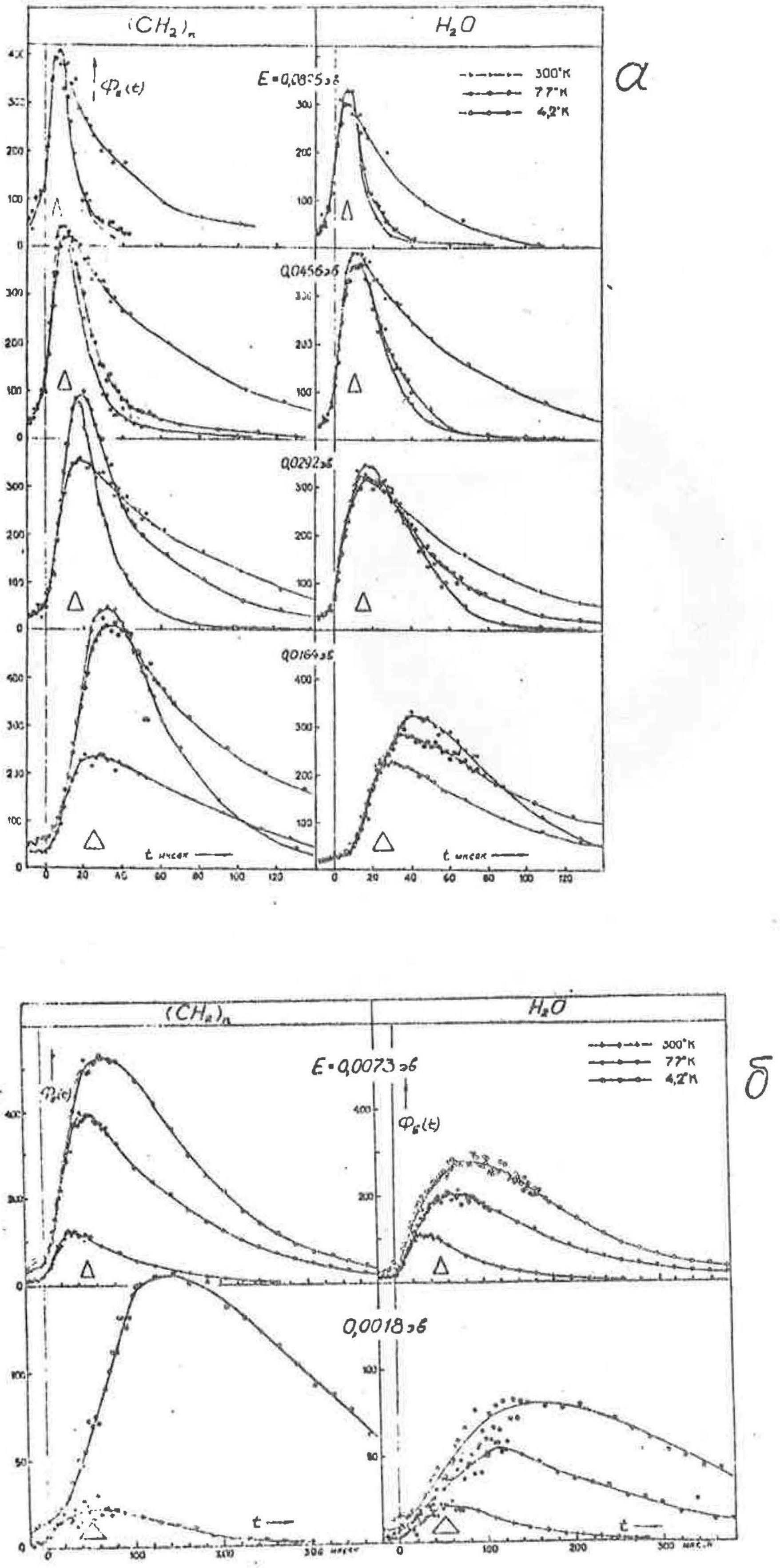

Figure 2 Caption: The effect of temperature on the shape of the neutron pulse at various energies in water and polyethylene.

1. t microsecs
2. eV

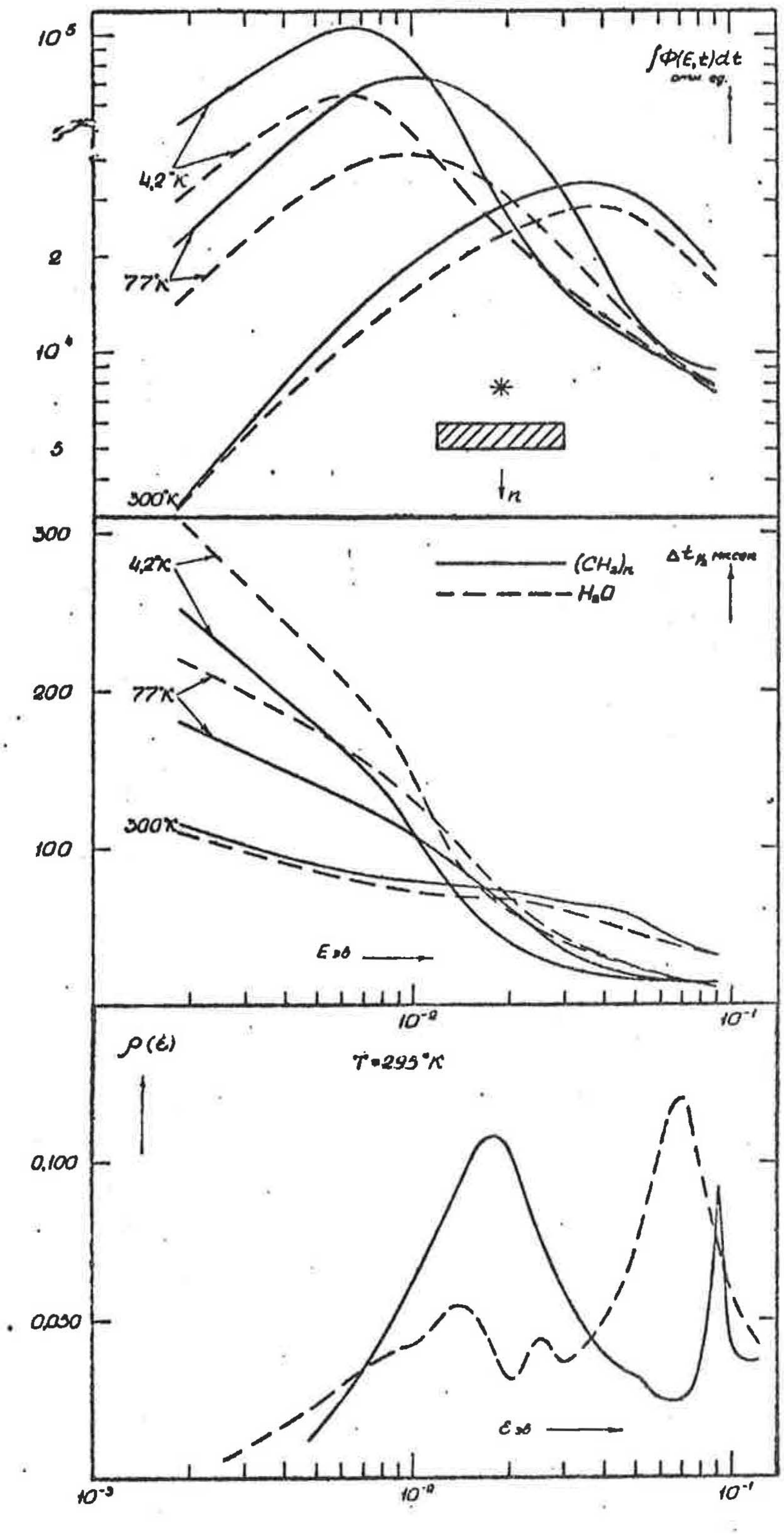

Figure 3 The dependence of the total intensity and the neutron pulse width on the neutron energy and temperature of water and polyethylene. The bottom figure shows the atomic normal oscillation spectra for water and polyethylene[3].