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Decay of the pulsed thermal neutron flux in polyethylene samples. Experiment and MCNP simulation

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Abstract

The decay of the pulsed thermal neutron flux in polyethylene, an important hydrogenous neutron moderator, has been investigated in various ways. The fundamental decay constant depends on the neutron absorption and scattering properties of the material and on the size of the sample. For two cubic polyethylene samples the decay constant has been obtained experimentally and evaluated from a theory, from results of another experiment, and from the MCNP simulations of two types. The results are compared and the observed delicate discrepancies are discussed.

1. Introduction

Thermal neutron scattering properties of materials containing hydrogen bound in molecular structures were recently considered from a point of view of the thermal neutron transport in a macroscopic scale, *e.g*. [1], [2], [3], [4]. Granada's [5] synthetic scattering model was developed and applied to a theoretical consideration of the thermal neutron diffusion parameters of various hydrogenous organic materials used as the neutron moderators, [6], [7].

A physical experiment is the final check of the theoretical description of the transport of thermal neutrons through a medium. In this respect, the pulsed neutron experiment is a fruitful and accurate test. In the so-called variable buckling experiment, the time decay of the thermal neutron flux after the neutron burst can be detected with a high accuracy and the neutron diffusion parameters of the medium (the absorption rate, the diffusion constant and the diffusion cooling coefficient) can be evaluated. As they are theoretically expressed by the macroscopic absorption and transport cross-sections (including also the scattering kernel), a comparison between the theoretical and experimental data is available. Another way to investigate the problem is to use Monte Carlo methods.

Polyethylene is considered in the paper. Its thermal neutron diffusion parameters can be known:

– from the buckling experiment,

– from a theoretical calculation using basic dependencies for the microscopic absorption and scattering cross-sections,

– from Monte-Carlo simulations.

Preliminary results of comparison between data obtained from the three mentioned sources are discussed to show some discrepancies between them. Two „buckling points" have been obtained using polyethylene cubic samples and the fundamental decay constants have been measured in the pulsed experiment. The same data have been evaluated using the parameters determined theoretically from Granada's model and from a Monte-Carlo simulation. The MCNP code has been used in which a special procedure is present for the transport of thermal neutrons through media containing hydrogen. The thermal neutron pulsed flux has been directly simulated in two cubic samples. Independently, the decay constants have been

evaluated using the thermal neutron pulsed parameters obtained from a former simulation of the complete buckling experiment in spherical geometry [8].

The aim of this preliminary research is to investigate whether all the mentioned results differ to each other on a significant level and whether the experimental method is sensitive enough to verify these results.

2. Decay of the pulsed thermal neutron flux

In the pulsed neutron experiment, a sample being investigated is irradiated by a burst of neutrons. After the neutron burst the resulting thermal neutron flux $\varphi(t)$ in the sample decays exponentially in time according to

$$\varphi(t) = a\, e^{-\lambda t} + \sum_k a_k\, e^{-\lambda_k t} \quad , \tag{1}$$

where λ is the fundamental mode decay constant, and k enumerates higher modes. Such a spectrum is registered and the time decay constant λ is mathematically isolated. From a theory of the decay of the thermal neutron pulse [9] the decay constant λ is expressed by the following formula:

$$\lambda = \langle v\Sigma_a \rangle + D_0 B^2 - CB^4 + FB^6 - \mathrm{O}(B^8) \ , \tag{2}$$

where:

B^2 – the geometric buckling, (a function of shape and size of the sample),

and the thermal neutron diffusion parameters:

$\langle v\Sigma_a \rangle$ – the absorption rate,

D_0 – the diffusion constant,

C – the diffusion cooling coefficient,

F – a correction to the diffusion cooling coefficient.

The above dependence gives a base to determine experimentally the thermal neutron diffusion parameters of various media. In the so-called buckling experiment the decay constant λ is measured for a series of samples which are different in size (*i.e.* they have different geometric bucklings). Then the thermal neutron diffusion parameters are obtained from a fitting

procedure, using Eq. (2). On the other hand, knowledge of the thermal neutron diffusion parameters of a given medium enables us to calculate according to Eq. (2) the decay constant λ of the flux $\varphi(t)$ in a sample of this material.

3. Measurement of the fundamental decay constants λ in polyethylene cubes

An experiment for two polyethylene samples has been made at the 14 MeV pulsed neutron generator. Polyethylene of the mass density $\rho_E = 0.947$ g cm^{-3} has been used. Two samples have been made as cubes of the sides $a_1 = 8.90$ cm and $a_2 = 5.94$ cm. Their surfaces have been covered with a 2-mm cadmium shields which assure the vacuum boundary conditions for the thermal neutron flux. Two independent detection lines are used in the experimental set-up to register the time behaviour of the flux $\varphi(t)$. Two ^{3}He detectors are placed symmetrically (at windows in the cadmium cover) in the middle of two faces of the cubic sample. The pulsed fast neutron source is placed in the middle of a third face. A scheme of the measurement geometry is shown in Fig. 1.

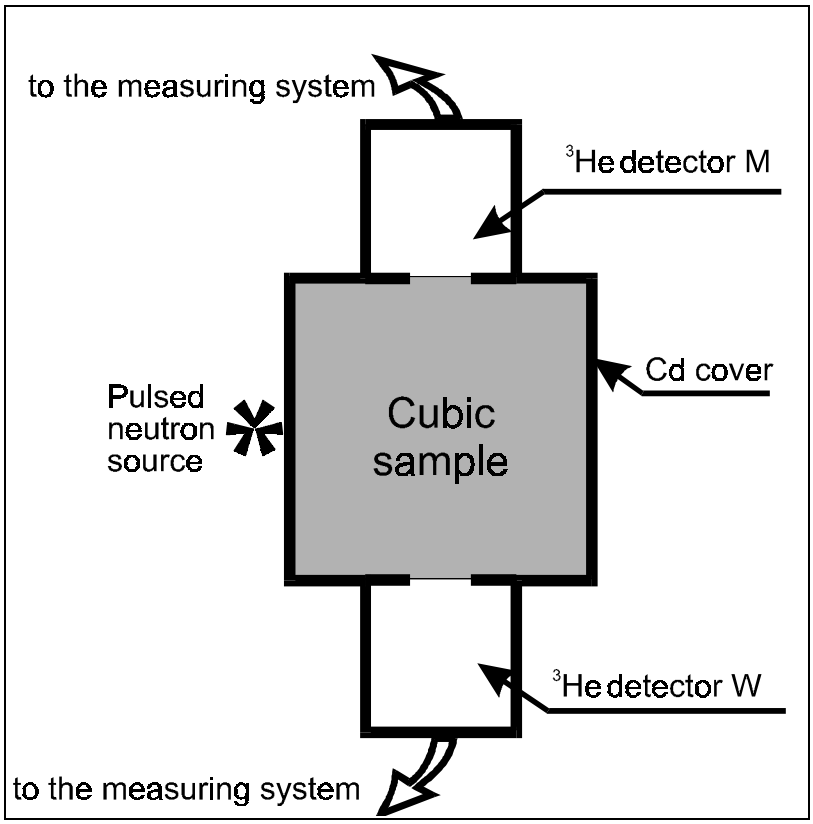

Fig. 1. Scheme of the measurement geometry.

Other details of the arrangement and the instrumentation system can be found in [10] and [11]. The fundamental decay constants λ have been isolated from the stored time spectra using the method given in [12] which contains also suggestions how to adjust the pulsing and data collecting parameters of the experiment. The experimental conditions and the results are specified in Table 1.

Table 1. The decay constants λ of the thermal neutron flux in polyethylene samples (ρ_E = 0.947 g cm^{-3}) measured at the temperature (20 ± 0.4) °C.

Cube	T_{imp} [μs]	T_{rep} [ms]	Δt [μs]	$\lambda \pm \sigma(\lambda)$ [s^{-1}]	
a = 8.90 cm	100	1.3	2	Detector M	14 523 ± 33
				Detector W	14 549 ± 40
a = 5.94 cm	70	0.8	1	Detector M	23 667 ± 87
				Detector W	23 684 ± 49

where: T_{imp} – width of the neutron burst, Δt – time channel of the multiscaler,
T_{rep} – burst repetition time, total number of channels used: 512

4. Pulsed thermal neutron parameters for polyethylene

The experimental results may be compared with those evaluated from Eq. (2) using the thermal neutron diffusion parameters known from various sources. Polyethylene, $(CH_2)_n$, is produced as a polymer of various densities (approx. from 0.91 to 0.965 g cm^{-3}). Since different authors obtained the thermal neutron diffusion parameters for polyethylene at slightly different densities it has been necessary to re-calculate their values for the density ρ_E of the polyethylene samples used in the experiment. The method described in [8] and [13] has been used.

Sjöstrand's *at al.* [14] experimental results at the polyethylene density 0.918 g cm^{-3} were: $\langle v\Sigma_a \rangle = 5900 \pm 90$ s^{-1}, $D_0 = 26\,500 \pm 600$ cm^{2}s^{-1}, $C = 2600 \pm 800$ cm^{4}s^{-1}. Their analogues at our experimental density ρ_E are given in Table 2.

We have theoretically evaluated the thermal neutron diffusion parameters for polyethylene of the mass density ρ_E. The absorption rate has been obtained from the elemental composition of polyethylene using relevant microscopic cross-sections [15]. The diffusion constant D_0 and the diffusion cooling coefficient C have been calculated using Granada's model for slow-neutron scattering in molecular gases [5], according to a summary presented in [7]. The results are given in Table 2.

The thermal neutron diffusion parameters for polyethylene of the density ρ_E have been also calculated using the thermal neutron density-removed diffusion parameters which were obtained by Monte Carlo simulations of the variable buckling experiments in spherical geometry [8]. They are also given in Table 2.

Table 2. The thermal neutron diffusion parameters of polyethylene re-evaluated to the density $\rho_E = 0.947$ g cm^{-3}.

Primary data from	$\langle v\Sigma_a \rangle$ [s^{-1}]	D_0 [cm^2 s^{-1}]	C [cm^4 s^{-1}]	F [cm^6 s^{-1}]
experiment [14]	6086	25 688	2443	–
theory [5], [7]	5981	25 796	2674	–
MCNP simulations [8]	6051	27 161	1926	139

5. Monte Carlo simulation of the experiment with two polyethylene cubes

The performed experiment with the two polyethylene cubes has been simulated using the MCNP4B code [16]. Cross-section tables were taken from the ”rmccs” and ”tmccs” MCNP standard libraries. In order to simulate a square neutron pulse, neutrons were generated within

the 100 μs in width time interval with a constant probability. They were generated uniformly inside the volume of the sample. The neutron source was isotropic. Initial energies of neutrons were sampled from the Maxwellian distribution at the room temperature. The neutron flux in the sample was scored in 900 time bins, 1 μs in width. As a scoring function a tally F4 – the track length estimate of cell flux – was used. The number of histories (NPS) was of the order 10^8. Next, the decay constant λ was calculated like in a real experiment [12]. Its relative error was not greater than 0.5 %. The resulting time decay constants for the two polyethylene cubes are given in Table 3.

Table 3. The decay constants λ of the thermal neutron flux in two cubic samples by different methods.

	a_1 = 8.90 cm		a_2 = 5.94 cm	
	λ [s^{-1}]	ε [%]	λ [s^{-1}]	ε [%]
Present experiment	14 536	–	23 676	–
Experiment-based evaluation *) a)	14 480	–0.4	23 359	–1.3
Theory-based evaluation *) b)	14 373	–1.1	23 163	–2.2
MCNP-based evaluation *) c)	14 948	2.8	24 517	3.6
MCNP direct simulation	14 968	3.0	24 676	4.2

*) Using in Eq. (2) the thermal neutron diffusion parameters specified in Table 2, based on a) [14], b) [5], [7], c) [8].

6. Comparison of the time decay constants obtained in different ways. Conclusions.

The experimental values of the time decay constants of the two polyethylene cubes may be compared with those obtained by the Monte Carlo simulation of the pulsed neutron experiment and those evaluated from Eq. (2) using different sets of the thermal neutron diffusion

parameters from Table 2. The geometric buckling of the cube is defined [17] as

$$B^2 = 3\left(\frac{\pi}{a+2d}\right)^2 \tag{3}$$

where a is a side of the cube and d is the extrapolation length. It is defined in the Milne problem and for the hydrogenous medium is given ([18], [19]) by $d = 0.76\langle l_{tr}(E)\rangle = 2.28\,\langle 1/v\rangle D_0$ where l_{tr} is the thermal neutron transport mean free path and $\langle x\rangle$ denotes the average of x weighted with the neutron energy distribution. For the Maxwellian distribution of the thermal neutron energy flux, $\langle 1/v\rangle = \sqrt{\pi}/(2v_T)$ where v_T corresponds to the neutron energy $E_T = k_B T$ and T is the temperature of the system ($v_T = 2200$ m s^{-1} at the temperature of 20.4 ^{o}C).

When the buckling B^2 is determined from Eq. (3) the corresponding decay constant λ is obtained from Eq. (2) using the thermal neutron parameters, $\langle v\Sigma_a\rangle$, D_0, C, (and F, if known). For the two polyethylene cubes the decay constants have been evaluated using the re-calculated experimental parameters, the theoretical values and the parameters from the simulated MCNP experiment, according to the list given in Table 2. One should notice here that the geometric buckling for a given cube is not always the same. It is defined not only by the geometric dimension a but is also slightly dependent on the diffusion constant D_0 through the extrapolation length d. Therefore, a use of the different values of the thermal neutron diffusion constant changes slightly the value of the geometric buckling.

The obtained decay constants are given in Table 3. The obtained deviations $\varepsilon = (\lambda - \lambda_E)/\lambda_E$ have been defined as relative to the λ_E data from the present experiment.

A general observation is that the differences between the results obtained by different methods are small. They attain about 6 % for the most distant values in the case of the smaller cube. However, taking into account a high accuracy of the present experimental results $\sigma(\lambda_E)/\lambda_E \approx 0.3$ %, further conclusions can be drawn. The results from the old [14] and present experiment are in a good agreement although the decay constants bases on [14] are underestimated a little. The theoretical results, compared to the present experiment, are underestimated $1 \div 2$ %. Greater differences which are observed at λ's for the smaller cube

could come partly from a possible uncertainty of the buckling value. Due to the influence of the extrapolation distance this uncertainty is higher at a greater buckling (*i.e.* at a smaller size of the sample).

Such a doubt does not exist when the results λ of the present experiment and the MCNP direct simulation are compared. In this case, the geometric buckling is not used and the results are defined at the pure geometric size. The MCNP simulation overestimates the results by $3 \div 4$ %. Moreover, this simulation is in a good agreement with the evaluation based on the simulations in spherical geometry [8] although those decay constants were obtained using the geometric buckling and a re-calculation from another mass density.

The present and previous MCNP simulations were done using available libraries of the thermal cross-sections, dedicated to the code. The observed behaviour of the results might suggest that new, higher-accuracy thermal cross-section libraries are desired for the MCNP code. It seems that a library which would be created based on the synthetic scattering model ([5], [7]), used in the theoretical calculation in the present work could give a better agreement with experiment. Here we must also remark that the application of the MCNP code for calculations of thermal neutron transport is limited, because the relevant cross-sections for hydrogenous media are available only for a few substances and temperatures. In this respect the synthetic scattering model would also help since it has been developed for a number of hydrogenous materials [6].

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