

DIFFUSION PARAMETERS OF THERMAL NEUTRONS IN SANTOWAX-R

S. SALMENHAARA and F. SEFIDVASH*

Reactor Laboratory, Technical University of Helsinki, Otaniemi, Finland

(Received 17 June 1970)

Abstract—The diffusion parameters of Santowax-R have been measured at the temperatures 170°, 200°, and 230°C using the pulsed neutron technique, and over a buckling range of 0.014–0.206 cm⁻². A Philips neutron generator was used to generate the neutron pulses. The decay constants of the neutrons were determined by means of a BF₃ counter operating with a 256-channel time-analyser. The measured diffusion constants and diffusion cooling constants are in good agreement with the results of the scattering model developed by one of us (FS) for polyphenyls.

INTRODUCTION

AMONG organic compounds, Santowax-R, a mixture of various terphenyl isomers, appears to be most resistant to the decomposing effects of the combination of high temperatures and radiations present in a nuclear reactor. Interest has been shown in using Santowax-R as a coolant in heavy water moderated organic cooled reactors, and therefore an accurate knowledge of the diffusion parameters of thermal neutrons in Santowax-R is of practical interest. On the other hand, these results can be used to test the model proposed for polyphenyls (SEFIDVASH *et al.* 1969).

Prior to this experiment, according to our knowledge, no measurements have been made on the diffusion parameters of thermal neutrons in Santowax-R by the use of the pulsed neutron technique. The diffusion length of thermal neutrons, using the stationary method, was measured by HONKI *et al.* (1965) and GODDARD and TREMPER (1969) over a temperature range of 171–300°C.

For measuring the diffusion parameters of thermal neutrons in organic moderators, the use of pulsed neutron technique is more advantageous than the stationary method. This is due to the difficulties that exist in the precise measurement of the boron concentration in polyphenyls (SEFIDVASH 1968b). In the stationary method a 1/v absorber such as boron is required, which is then mixed, at various concentrations, with the moderator, the diffusion lengths of thermal neutrons then being measured in these media.

THEORY

The theoretical values of the diffusion parameters for thermal neutrons were obtained by solving the Boltzmann equation

$$[\Sigma_T(E) - \kappa\mu]\phi(E, \mu) = \int_{-1}^{+1} d\mu' \int_0^\infty dE' \Sigma_s(E' \rightarrow E, \mu_0)\phi(E', \mu')$$

by a method described by HONECK (1962). In this equation $\Sigma_T(E)$ is the total macroscopic cross-section at energy E , and μ_0 is the cosine of the angle between the directions corresponding to the cosines μ and μ' .

The scattering kernel, $\Sigma_s(E', E, \mu_0)$, was calculated by the scattering model for polyphenyls previously developed (SEFIDVASH 1968a; SEFIDVASH *et al.* 1969). The physical model comprises a free translator of mass 78, a hindered rotational oscillator

* Present address: Arya-Mehr University, Tehran, Iran.

of effective mass 21.3 at an energy of 0.02 eV, and two vibrational oscillators at energies 0.146 and 0.38 eV with equal masses of 2.128.

The fundamental space eigenvalue, κ , was calculated by the B_L method, but the diffusion constant and diffusion cooling constants were calculated by a power series expansion. The flux and the scattering kernel were both expanded in Legendre polynomials and the fundamental space eigenvalue was expressed by the relation

$$\kappa^2 = \sum_{j=1}^{\infty} (-1)^{j+1} a_j \sigma^j$$

where, for a $1/v$ absorber, $a = \Sigma_a(kT)$.

The diffusion constants and cooling constants were related to the coefficients

$$D_0 = V_T/x_1$$

$$C = V_T x_2/x_1^2$$

where V_T is the neutron speed corresponding to an energy, kT .

EXPERIMENT

Neutrons were produced for the experiment by a Philips neutron generator using the deuterium-tritium reaction producing 3×10^{11} neutrons/sec during the pulse. For these measurements, the pulse duration was 20 μ sec with a repetition rate of 100 pulses/sec. This pulsing mode provided adequate counting rates, as well as a sufficient time for the neutrons from one pulse to decay before the next pulse.

The containers consisted of three rectangular aluminium boxes of approximate dimensions $40 \times 40 \times 60$ cm³, $20 \times 20 \times 35$ cm³, and $12 \times 12 \times 20$ cm³, covered with cadmium sheets and insulated by 5 cm thick rockwool thermal insulation. The whole system was further shielded by about 15–20 cm of boric acid blocks.

The containers with Santowax-R were heated by 0.7–3 kW hot plates the temperature being measured by an iron-constantan thermocouple located 5 cm below the surface of the wax. The temperatures were recorded on a strip chart recorder and also visually observed using a digital voltmeter. The temperature was regulated by an independent thermocouple connected to an INDIN on-off type temperature controller, this being supplemented by a circuit which caused the controller to act as a proportional regulator. In this way, a temperature variation of less than 0.3°C was observed during each measurement. The temperature variation between different parts of each container was observed to be less than 0.2°C.

Depending on the size of the container, either a 1 cm or 2.5 cm diameter BF₃ detector was used. To minimize the higher harmonics effect, the detector and the source were positioned in the manner described by Joshi *et al.* (1965). The detector and the neutron generator were cooled by a room temperature air flow. The pulses from the detector were collected by a 256-channel time analyser in which the channel widths were 10 and 20 μ sec and the dead times were 20 and 25 μ sec respectively. After each neutron pulse there was a delay time of 40 . . . 400 μ sec before the analyser started counting. The measurements were made with nine bucklings ranging from 0.014 to 0.206 cm⁻² at the temperatures 170°, 200°, and 230°C \pm 0.5°C in each case. The measuring time varied from 3 to 8 hr during which more than 10⁶ counts were collected in the first channels.

ANALYSIS

The data from the multichannel time analyser was first corrected for dead time losses and for the small background. Typical curves of the decay of the neutron field are shown in Fig. 1. A weighted least squares analysis of the corrected data was used to obtain the value of the decay constant, λ , and its standard error. After one decay constant was computed from the given data, the two first channels were dropped and a second decay constant was computed, in this way the decay constant as a function of channels dropped was obtained. The region where the decay constant remains constant indicates freedom from higher harmonics. The mean value of the calculated λ 's in the harmonic free region was taken as the final decay constant.

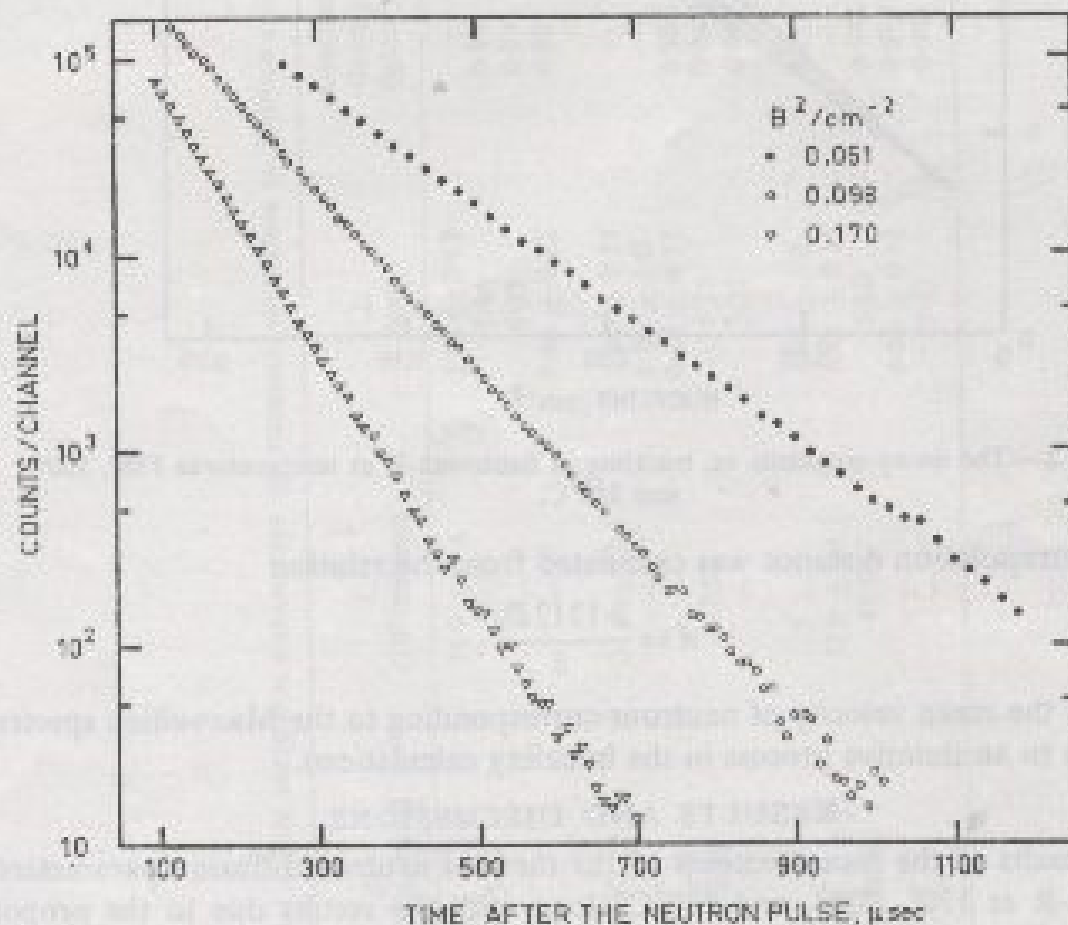


FIG. 1.—The examples of the decay curves for several bucklings at 200°C.

The thermal expansion of the aluminium containers was taken into account in the buckling calculations. As wax losses occurred during measurement, the height of the wax was determined in each case as an average value during a measuring time. A typical decrease of the height was about 0.5 per cent.

The decay constants, λ , and bucklings, B^2 were fitted by the weighted least squares method to

$$\lambda = \lambda_0 - D_0 B^2 - C B^4,$$

where $\lambda_0 = \overline{\nu \Sigma_a}$ is the macroscopic absorption probability, and D_0 and C are the diffusion constant and diffusion cooling constant respectively. The weighting function was the inverse square of the standard error of the decay constant. The λ vs. B^2 curves for Santowax-R at 170°C, 200°C, and 230°C are shown in Fig. 2.

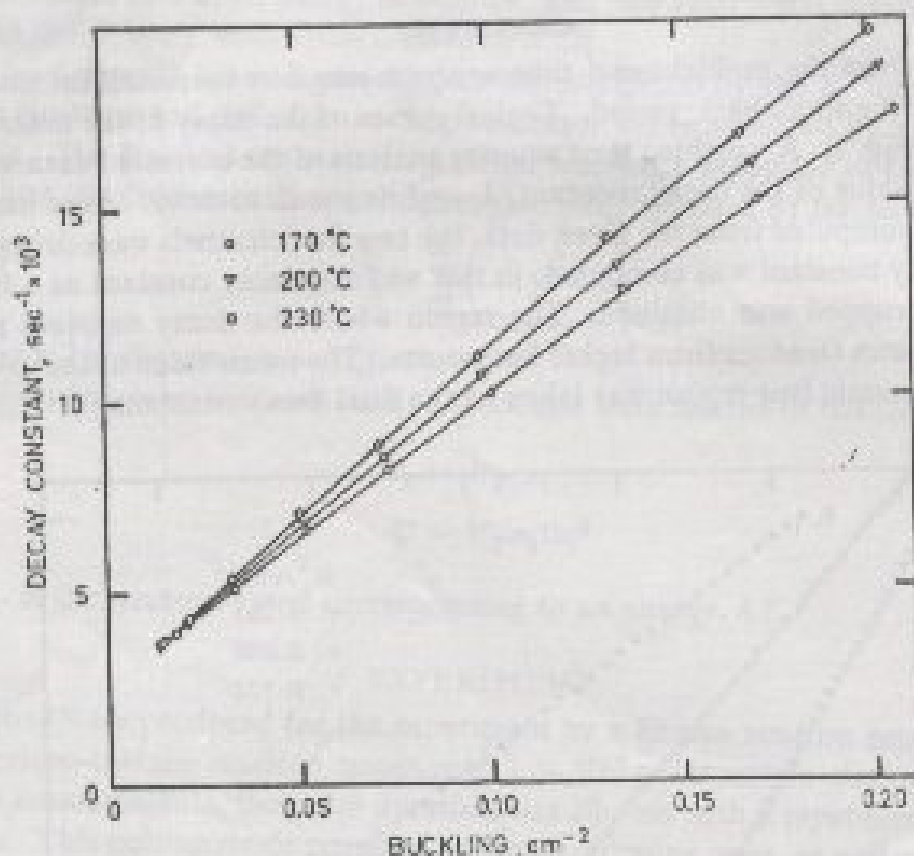


FIG. 2.—The decay constants vs. buckling in Santowax-R at temperatures 170°, 200° and 230°C.

The extrapolation distance was calculated from the relation

$$d = \frac{2.1312 D_0}{\bar{v}}$$

where \bar{v} is the mean velocity of neutrons corresponding to the Maxwellian spectrum. This leads to an iterative process in the buckling calculations.

RESULTS AND DISCUSSIONS

The results of the measurements of the thermal neutron diffusion parameters of Santowax-R at 170°, 200°, and 230°C along with the results due to the proposed scattering model for polyphenyls are presented in Table I. A good agreement between the theory and experiment is observed. The experimental results of other workers are also presented in the same table. The results of HOHKI *et al.* (1965) suffer from low accuracy. The results of the present experiment fall between those of GODDARD and TREMER (1969) and the presented theory (SEFIDVASH 1968a, SEFIDVASH *et al.*, 1969).

Although a recent measurement of the total cross section for neutrons of wavelength in the 6–20 Å range with various Santowax-R constituents namely *ortho*-, *para*-, and *meta*-terphenyls shows that differences do exist among them due to the difference in their rotational degrees of freedom (SEFIDVASH, *in press*), the diffusion parameters are insensitive to such differences. The results of this experiment with those on diphenyl and Dowtherm-A (SEFIDVASH and GRANT, 1969), and their good agreement with the theory, clearly confirms that the neutron thermalization properties of polyphenyls are primarily determined by the benzene ring structure assumed in this theory.

TABLE 1.—THE DIFFUSION PARAMETERS OF THERMAL NEUTRONS IN SANTOWAX-R

T (°C)	λ_0 (sec ⁻¹)	D_0 (10 ⁶ cm ² /sec)	C (10 ⁶ cm ³ /sec)	L (cm)	Assumed density (g/cm ³)	Reference
170	2607 ± 60	80.4 ± 1.6	38.9 ± 8.3	5.55 ± 0.12	—	Present results
171	—	75.84	37.77	5.44	0.991	SEEDYASHI <i>et al.</i> (1969)
171	—	—	—	6.93	0.991	Free gas
171	—	—	—	5.88 ± 0.12	0.991	GONDAUD <i>et al.</i> (1969)
200	2510 ± 53	85.4 ± 2.2	31.6 ± 12.1	5.84 ± 0.13	—	Present results
200	—	—	—	6.86 ± 1.0	—	HOBBER <i>et al.</i> (1965)
201	—	—	—	6.16 ± 0.12	0.967	GOODARD <i>et al.</i> (1969)
201	—	82.89	42.55	5.75	0.967	SEEDYASHI <i>et al.</i> (1969)
201	—	—	—	7.21	0.967	Free gas
225	—	—	—	6.95 ± 1.2	—	HOBBER <i>et al.</i> (1965)
230	2347 ± 66	96.6 ± 2.5	53.9 ± 14.2	6.41 ± 0.17	—	Present results
232	—	90.79	48.06	6.10	0.943	SEEDYASHI <i>et al.</i> (1969)
232	—	—	—	6.48 ± 0.13	0.943	GOODARD <i>et al.</i> (1969)
232	—	—	—	7.51	0.943	Free gas

This model has also been tested against the rethermalization process in terphenyls. The model predicted a rethermalization length of 1.21 cm in terphenyl compared to 1.13 cm according to the experiment of RASTAS and SAASTAMOINEN (1969). The calculations were made using a P_2 -approximation assuming linearly anisotropic scattering.

The predictions of the proposed model and its good agreement with the present experiment raises the hope that it will find a useful application in the design of organic cooled heavy water moderated reactors.

Acknowledgement—The authors wish to thank Professor P. Jauho, the Director of the Laboratory, for his interest and encouragement in this work. The present work has been performed under the auspices of the Finnish Atomic Energy Commission.

REFERENCES

- GODDARD A. J. H. and TREMER G. E. B. (1969) *Nucl. Sci. Engng* 37, 167.
HOEKI S. *et al.* (1965) *Proc. 3rd Int. Conf. peaceful Uses atom. Energy, New York* 2, 316.
HONK H. C. (1962) BNL-719 4, 1186.
JOSEF B. V. *et al.* (1965) *J. nucl. Energy (Parts A/B Reactor Sci. Technol.)* 19, 515.
RASTAS A. and SAASTAMOINEN J. (1969) *Nucl. Sci. Engng* 36, 351.
SEFIDVASH F. (1968a) *Trans. Am. Nucl. Soc.* 11, 182.
SEFIDVASH F. (1968b) *Analyt. Chem.* 40, 1165.
SEFIDVASH F. and GRANT P. J. (1969) *J. nucl. Energy* 23, 225.
SEFIDVASH F. (In press) *Phys. Lett. B.*