Properties of Organic
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Neutron Properties of Organic Moderators and Coolants

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Abstract: *A comparison of the pulsed neutron technique and the stationary method of measuring diffusion parameters of thermal neutrons is made. The measurements presently made on Santowax-R and Dowtherm-A by these methods are compared with the results of the theoretical neutron scattering model developed by the author. The model was further tested by measuring the cold neutron cross sections of benzene, diphenyl, Dowtherm-A, m-, o-, and p-terphenyl molecules.*

§1.   Introduction

One of the more imaginative concepts in reactor design involves the use of the organic liquids as coolants or coolant-moderators. The advantages of such a system are many. Firstly, compared with the use of water as coolant, where the reactor pressure must be high (about 1000 psi for boiling-water and 2000 psi for pressurized-water reactors), an organic coolant with low vapour pressure may be chosen. Thus high-temperature low-pressure operation, resulting in high thermal efficiency as well as lighter and less costly pressure vessel and components is possible. Secondly, in general, the organics have low corrosion characteristics with most materials and are compatible with most fuels, cladding, and structural materials. A direct result of this is the ability to use conventional materials, such as carbon or low-alloy steels, instead of stainless steels, in the pressure vessels, pumps piping, etc., resulting in significant savings in capital costs. Thirdly, the induced radioactivity in pure organic

liquids is very low, resulting in reduced shielding requirements of the primary loop and therefore further reduction in capital costs. Fourthly, organic liquids in general have well-known and well-defined physical, chemical, and handling characteristics.

The search for organic liquids that are resistant to high temperature and radiations led to the choice of polyphenyls as the most suitable organics in this respect.

§2.   Polyphenyls

When a phenyl radical joins a benzene ring, it replaces one of the hydrogen atoms of the ring, so that these are essentially two phenyl radicals, each C_6H_5, joined together. This new molecule is called diphenyl, biphenyl, or phenyl-benzene, and has a chemical formula $C_{12}H_{10}$. Dowtherm-A ($C_{12}H_{10}O_{0.713}$) is a eutectic mixture containing 26.5% diphenyl and 73.5% diphenyl oxide by weight. Santowax-R ($C_{18}H_{14}$) is a mixture of o-,m-,p-, and quar-terphenyls in

the appropriate proportions of 9 : 52 : 30 : 8 respectively. Terphenyls are the molecules of three joined benzene rings.

§3. Stationary method versus pulsed experiment

The present experiments applied the stationary method for Dowtherm-A and pulsed method for Santowax-R in order to measure the thermal neutron diffusion parameters of these compounds. Each of the methods has many advantages over another but also suffers from some disadvantages too.

The pulsed method suffers from at least two problems. The first is the exact determination of the fundamental mode decay constant λ, and the second the exact calculation of the buckling B^2. Although assuming that the experimenter has an ideal neutron detection equipment, i.e., no dead time effects exist which may affect λ measurements considerably, there still remains other causes for errors in the λ - determination such as background neutrons from the source, backscattering of neutrons, the presence of high spatial modes, and the presence of higher energy modes. In addition, there may be physical effects which prevent the establishment of the pure fundamental mode.

In order to determine the geometrical buckling of the moderator blocks, most researchers increase the linear dimensions by twice the extrapolation length. This is certainly correct as long as the moderator is large, so that errors in the extrapolation length do not effect B^2.

Due to the energy dependency of the mean free paths, space-energy transients may arise near the surfaces which lead to a different 'effective extrapolation length'. The extrapolation length may also depend on the shape of the moderator.

The stationary method of this experiment suffers from the difficulties encountered in the boron analysis of the poison mixed with polyphenyls.[1]   In an experiment on Dowtherm-A, trihexylene glycol biborate [tri-(2 methyl-2:4-pentanediol) biborate], commercially known as *Borester*-7 was used to mix with Dowtherm-A at various concentrations. Unfortunately, the experimental arrangement did not allow the direct determination of boron concentrations by the addi-

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tion of known amount of *Borester*-7. The neutronic measurements were accurate to about $\frac{1}{2}\%$, but some difficulties exist in determining the boron concentration to a similar accuracy.

§4. Experiment on Dowtherm-A by stationary method

The thermal neutron diffusion length in Dowtherm-A has been measured at 20, 50, 80, 110, 140, 170, 200, and 220°C. By varying the neutron absorption cross sections by the addition of trihexylene glycol biborate, the neutron diffusion constant and diffusion heating constant were determined at 20, 80, 140, and 200°C. The measurement of the boron concentrations was carried out by a double-beam infrared absorption method using the characteristics absorption frequency of the B-O-B bond at 1412 cm^{-1}.

The experiment essentially consisted in measuring the relaxation length of thermal neutrons from the reactor thermal column in $100 \times 100 \times 75$ cm.^{3} aluminium tank.[2] The negative source technique was applied using a U^{235} fission chamber as the detector. The gas-tight tank was heated externally by heating blankets and temperature was controlled to within 0.25°C. The uncertainly in the extrapolation distance for the worst case caused an error for 0.15% in diffusion length.

§5. Experiment on santowax-R by the pulsed neutron technique

Neutrons were produced for the experiment by a 14 MeV neutron generator using the deuterium-tritium reaction producing 3×10^{11} neutrons per sec. during the pulse. The pulse duration was 20 μs with a repetition rate of 100 pulses/sec. The containers consisted of three rectangular aluminum boxes of approximately $40 \times 40 \times 60$ cm.^{3}., $20 \times 20 \times 35$ cm.^{3}., and $12 \times 12 \times 20$ cm.^{3} in dimensions covered with cadmium sheets and insulated by 5 cm. thick rockwool thermal insulation.[3] The whole system was further shielded by about 15-20 cm. of boric acid blocks.

The temperature was measured by a set of thermocouples within 0.2°C. Depending on the size of the container, either 1 cm or 2, 5 cm. dia. BF$_3$ detector was used.

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§6. Cold neutron scattering from polyphenyls

The cold neutron cross sections of benzene, diphenyl, Dowtherm-A, m-, o-, and p- terphenyl molecules have been measured at room temperature in wavelength range of 6-20 Å by transmission method using the cold neutron time-of-flight facility.[4] A chopper using two 32 cm. dia. discs rotating at 1350 rpm on the same shaft was used to cut off neutrons with wavelengths of 6 Å and less.[5] The discs were made of 0.2 mm. cadmium sandwiched between 2 mm. thick aluminium plates with the slots phased at the distance of 50 cm. from each other. A flight path of 1.6 meter kept in a He atmosphere was used. The sample was shifted in and out of the beam automatically every two min. Beam monitoring was used but with frequent sample cycling no intensity correction was necessary. The size of the collimated incident neutron beam was 5×9 mm.^{2} The 2.5 cm. dia. BF$_3$ detector was placed at 70 mm. from the samples. Spectra were accumulated in 2×256 channels of a multichannel time analyzer.

The solid samples were cast in 40×70 mm.^{2} aluminium frame sandwiched tightly between two 0.3 mm. parallel aluminium plates. Two 1-mm O.D. pipes circulated the liquid into and out of the sample holder thus assuring that no voids in the sample can occur due to air or gas as a result of filling or evaporation.

§7. The theoretical model

In order to predict the thermal spectra, a model has been developed for neutron scattering by protons bound in polyphenyls, analogous to that of Nelkin for water. The physical model[6] comprises a free translator of mass 78, a hindered rotational oscillator 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 model assumes that the benzene molecule constitutes the basic dynamical unit and the differences in various polyphenyls in the reactor calculations are important only in the fine structures. The sensitivity of the predictions of the model to the variations of mode parameters is investigated.[7]

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

$$\left[\Sigma_T (E) - \chi\mu \right] \phi (E,\mu) = \int_{-1}^{+1} d\mu' \int_{0}^{\infty} dE'$$
$$\Sigma s (E', E, \mu_o) \phi (E',\mu') \qquad \ldots (1)$$

by the method described by Honeck.[8] In this equation $\Sigma_T (E)$ is the total macroscopic cross section at energy E and μ_o is the cosine of the angle between the directions corresponding to the cosines μ and μ'.

The scattering kernel, $\Sigma_s (E' E, \mu)$, was calculated by the scattering model for polyphenyls as suggested above. The fundamental space eigenvalue, χ, was calculated by the B$_L$ method, but the diffusion constant and the diffusion cooling constants were calculated by a power series expansion. The flux and the scattering kernel were both expanded in the Legendre polynomials and the fundamental space eigenvalue was expressed by the relation;

$$\chi^2 = \sum_{J=1}^{\infty} (-1)^{j+1} \alpha j\, a^{j} \qquad \ldots (2),$$

where in an $1/V$ absorber $a = \Sigma a$ (KT).

The diffusion constants and cooling constants were related to the coefficients :

$$Do = V_T /\alpha_1 \qquad \ldots (3)$$
and
$$C = V_T\, \alpha_2/\alpha_1^{3} \qquad \ldots (4)$$

where V_T is the neutron speed corresponding to an energy KT.

§8. Conclusions

The results of the measurements[9] on the thermal neutron diffusion parameters of Dowtherm-A by the stationary methods and of Santowax-R by the pulsed neutron technique at various temperatures along with the results due to the proposed scattering model for polyphenyls are presented in Table 1. A good agreement between the theory and the experiment is observed.

The results on the slope of the total neutron cross section curve as a function of energy in the 6-20 Å range for various polyphenyl molecules are presented in Table 2. Although these measurements show differences between the molecules due to their differences in rotational degree of freedom the diffusion parameters are insensitive to such differences. A comparison of the results clearly indicates the adequacy of the proposed model for reactor calculations.

Table 1.—THE THERMAL NEUTRON DIFFUSION PARAMETERS OF VARIOUS POLYPHENYLS

T (oC)	(g/cm³)	L (cm)	Da (10³ cm²/ sec.)	H (10³ cm⁴/ sec.)	Compound	Reference
20	1.063	4.166	47.50	18.05	Dowtherm-A	Theoretical model [2,6]
80	1.014	4.778	59.52	24.89		
140	0.963	5.438	73.25	39.8		
200	0.908	6.168	89.00	53.13		
24	1.066	3.903	44.94	15.14	Diphenyl	
85	0.98	4.649	58.52	23.19		
105	0.97	4.826	62.41	29.49		
171	0.991	5.439	75.84	37.77	Santowax-R	
201	0.967	5.752	82.89	42.55		
207	0.962	5.830	84.74	44.26		
232	0.943	6.102	90.79	48.06		
300	0.89	6.842	108.41	69.59		
20	1.063	4.199 ± 0.016	48.6 ± 0.6	17.2 ± 3.5	Dowtherm-A	Experiment [2]
80	1.014	4.792 ± .019	60.4 ± 0.7	27.6 ± 4.6		
140	0.963	5.422 ± 0.038	74.6 ± 0.8	34.6 ± 7.8		
200	0.908	6.005 ± 0.055	86.2 ± 1.6	56.4 ± 18.9		
170		5.54 ± 0.12	80.4 ± 1.6	38.9 ± 8.3	Santowax-R	Experiment [3]
200		5.84 ± 0.13	85.4 ± 2.2	31.6 ± 1.2		
230		6.41 ± 0.17	96.6 ± 2.5	53.9 ± 1.4		

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Dr. Sefidvash obtained first position in B.Sc. (Mech. Eng.) from the University of West Virginia, USA, in 1961. After obtaining his M.Sc. (Nucl. Eng.) from North Carolina State University in 1968, Dr. Sefidvash finally secured his Ph.D. degree from the Imperial College of Science and Technology, London. He has also qualified as a Fall-out Shelter Analyst and Environmental Engineer, having been awarded certificates for both by the US Department of Civil Defence.

Dr. Sefidvash has also done research work in the field of low-energy neutron physics (1967-69) at the Reactor Laboratory of the Technical University of Helsinki, Finland, as a guest scientist. He has contributed original articles to Analytical Chemistry, Journal of Nuclear Energy, Transactions of American Nuclear Society, etc.

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Table 2—SLOPE OF THE TOTAL CROSS SECTION CURVE AS A FUNCTION OF NEUTRON ENERGY

Compound	Slope (barn/Å)
Benzene (C_6H_6)	$6.45-0.24$
Diphenyl ($C_{12}H_{10}$)	2.97 ± 0.24
Dowtherm-A ($C_{12}H_{10}O0.715$)	4.06 ± 0.12
Terphenyl-o ($C_{18}H_{14}$)	$3.84-0.28$
Terphenyl-p ($C_{81}H_{14}$)	3.40 ± 0.24
Terphenyl-m ($C_{18}H_{14}$)	2.62 ± 0.36
Sefidvash model	4.1

§9. References

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