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MEASUREMENT OF TOTAL NEUTRON CROSS SECTION FOR SOME ORGANIC MATERIALS IN THERMAL ENERGY REGION

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Abstract: The total cross sections of some organic moderators were measured for the thermal neutron energy range. The samples measured were normal dodecane, tri-butyl phosphate (TBP), and the mixture of 70 v/o of normal dodecane and 30 v/o of TBP which is typically used in a reprocessing plant. The cross section was measured as the transmission of thermal neutrons through samples using the chopper and T-O-F facility installed at the Musashi reactor TRIGA-II, (100 kW). It was found that although the measured cross section is almost the same for dodecane and TBP, they both show different behavior compared with that of water. We propose to use the new spectral densities to calculate the cross sections for the above moderators within the framework of Nelkin's formalism for water.

(total cross section, thermal neutron, dodecane, TBP, time-of-flight, chopper, TRIGA-II.)

Introduction

The mixture solution of normal dodecane and TBP is used in reprocessing plants during the chemical extraction process. This solution functions, on some occasions, as moderator for neutrons and can potentially cause, when combined with fissile material, critical accidents. However, the mixture solutions neutron cross sections have to the author's knowledge not yet been measured. Although it is desirable to measure the scattering law to generate the complete scattering kernel for the analysis of reactor physics problems, due to the lack of a strong neutron source the total neutron cross section is measured in thermal neutron energy. It is known that the most of the detailed properties of molecules which affect the neutron scattering are lost in the total cross section. None the less we constructed the spectral density for a hydrogen atom in the organic moderator which can reproduce the measured cross section.

Measurement of Total Neutron cross Section with T-O-F Method

T-O-F Facility(1)

The experiment was performed with the chopper and time-of-flight (T-O-F) facility of the Musashi reactor (TRIGA-II, 100 kW). The outline of the experimental arrangement is shown in Fig. 1. A Si rod consisting of a large 52 cm long nearly-perfect single crystal was used as a

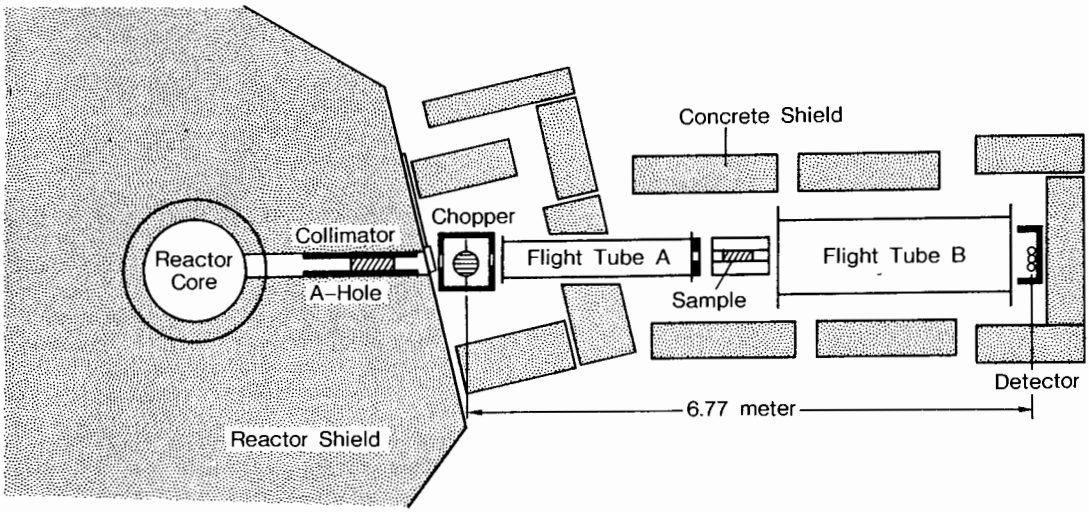

Fig. 1 Arrangement of Chopper and Time-of-Flight Spectrometer

very effective thermal neutron bandpass filter.

The BN (boron nitride) rotor of the chopper was 12 cm in diameter and 12 cm in height with nine slits of 0.4 × 7.0 cm^2 and was connected with a driving motor. In this experiment the rotation frequency was 2000 rpm and the neutron burst width (FWHM) was 275 μsec. The neutron flight path length was 6.77 m with an energy resolution of 5.5% for 0.002 eV neutrons. The neutron beam was collimated to 2 cm diameter in front of the samples. Three BF_3 detectors were set at the end of the flight path.

Preparation of Samples and Sample Holders

The measurements were performed for three samples; normal dodecane, tri-butyl phosphate (TBP) and the mixture of 70 v/o of normal dodecane and 30 v/o of TBP. The purity of the normal dodecane was 99.3 w/o, and it contained 0.9 w/o of normal $C_{11}H_{22}$ and 0.3 w/o of $C_{13}H_{28}$. The measured density was 0.750 g/cc at 20°C. The purity of the TBP was 99.8 w/o with density of 0.982 g/cc at 20°C. The most important impurity was 0.2 ppm of Cl.

The sample holder was a cylindrical tank of 99.6% pure Al, with dimensions of 160 mm dia and 11 mm thickness. The central part of the sample holder was 5 mm thick and covered with two 3 mm plates.

Treatment of Data and Calibration of the System

Treatment of Data

Conceptual flow of data treatment is shown in Fig. 2. The total cross section can be obtained by dividing the open beam spectrum by the transmitted neutron spectrum.

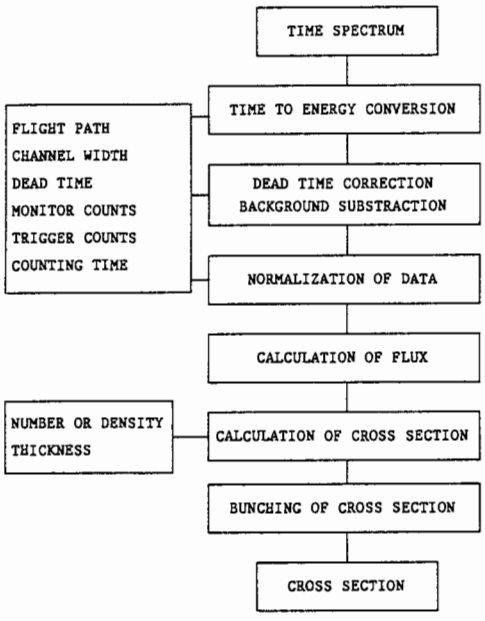

Fig. 2 Flow Chart of Data Treatment

$$\sigma(E) = \frac{1}{Nd} \ln \frac{\phi_0(E)}{\phi_s(E)},$$

where N is the molecular number density, and d is the thickness of the sample. The neutron spectrum transmitted through the sample $\phi_s(E)$ is corrected by deadtime of the counting system, the monitored reactor power, and the background neutron count. The background was measured by covering the sample holder with a Cd plate of 3 mm thickness. The time spectra of $\phi_s(E)$ and its background is shown in Fig. 3. In this figure, water is used as the sample. The same correction is applied to the neutron spectrum $\phi_0(E)$ through the vacant sample holder. The time spectra of neutrons without the sample and its background are shown in Fig. 4.

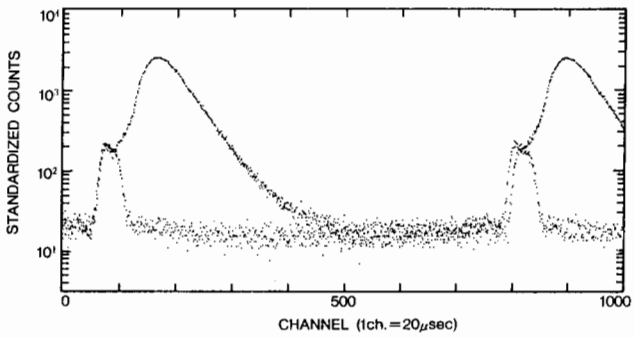

Fig. 3 Time Spectrum of Water and BACKGROUOND

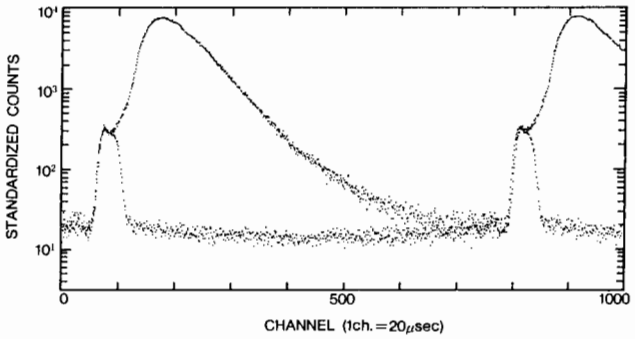

Fig. 4 Time Spectrum of OPEN-RUN and BACKGROUND

Estimation of Error

The relation between the energy resolution and time resolution is,

$$\frac{\Delta E}{E} = 2 \frac{\Delta t}{t},$$

where E is the energy corresponding to the flight time t. In the present experimental condition Δt was measured to be 275 μsec. Therefore the energy resolution of the experiment can easily be estimated.

The error of the total cross section is determined by the following factors: density of the sample solution, thickness of the sample solution and statistical error of neutron counting. We added the error estimated by repeated measurements.

Calibration by Measurement of Water Cross Section

This T-O-F facility has been used for measurement of the total cross section of solid samples. However, this was the first time that this facility was utilized to measure the cross section of solution samples. To establish the experimental procedure for solution samples the measurement was conducted for light water and the measured cross section compared with the one in BNL-325. It was found that agreement between the present experiment and the published data is satisfactory. Through this comparison it can be concluded that the whole experimental procedure is valid and properly calibrated.

Experimental Results

The measured cross sections are shown in Figs. 5 and 6. The total cross section for water is also shown in Fig. 5. As is seen from Fig. 5, the total cross section for normal dodecane and TBP is virtually the same when they are compared as the cross section per hydrogen atom. The cross section for organic moderator, however, is larger than that of water for the energy range of 0.01 to 0.1 eV. This could be understood that the motion of an organic molecule is hindered to a greater degree than water.

In Fig. 6 the total cross section of the mixture of 70 v/o normal dodecane and 30 v/o TBP is shown and compared with the calculated value from each cross section (dashed line). The directly measured cross section agrees well with the composed cross section.

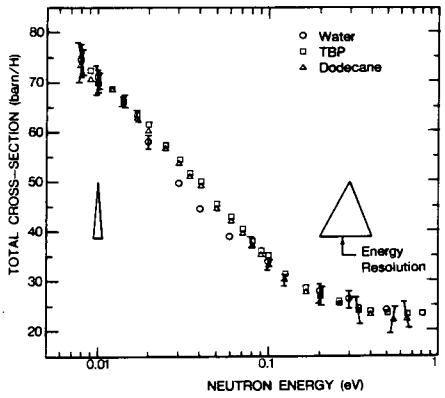

Fig. 5 Total Cross Section Divided by Number of Hydrogen Atome in Oraganic Compounds and Water.

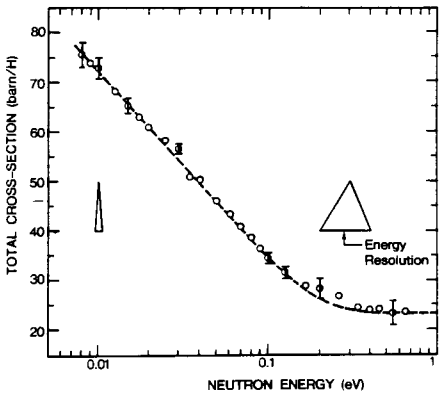

Fig. 6 Total Cross Section Divided by Number of Hydrogen Atome in Organic Compound (70% Dodecane and 30% TBP).
———; Synthesized from Each Cross Section

Analysis of Experiment

Within the incoherent approximation the thermal neutron scattering process is governed completely by the so called spectral density or frequency distribution which is the Fourier transform of the velocity auto-correlation function of the atom in a moderator molecule[2]. The spectral density is reduced from the scattering law experimentally.

However, the measurement of the scattering law, because it require a very strong thermal neutron source, is one of the most difficult experiments to perform. There is no way to determine the spectral density from the total cross section, because the total cross section can be obtained by integration of spectral density by several times.

Since the molecular structure of normal dodecane is similar to polyethylene, and it is found that there is no distinct difference of the total cross section between normal dodecane and TBP, we empirically determined the spectral density for a hydrogen atom starting from that of polyethylene[3]. Needless to say, the proposed spectral density can only reproduce the measured cross section. In the future we plan to demonstrate independently the effectiveness of the proposed spectral density by analysing critical experiments conducted with organic moderator and fuel pins[4].

Spectral Density

In the present analysis the combination of the delta functions is assumed for the spectral density. This formalism is proposed by Nelkin[5] and programmed as the GAKER code[6]. As mentioned above, starting from the spectral density of polyethylene, the spectral density is sought to fit the measured total cross section on a trial and error basis.

The proposed spectral density is shown in Fig. 7 with that of water of Nelkin[5] and polyethylene of Koppel et al.[3]

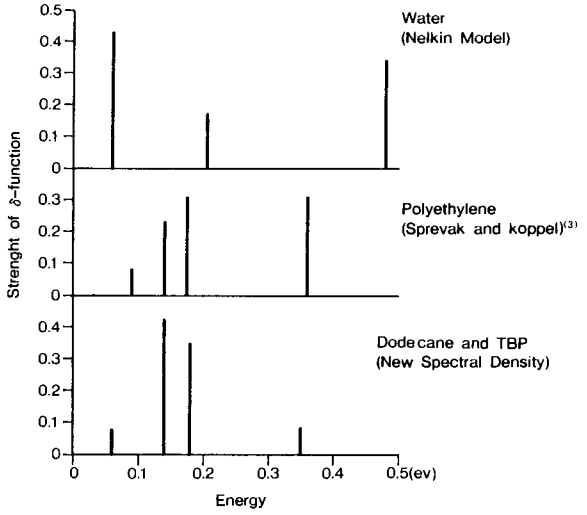

Fig. 7 Spectral Density for a Hydrogen Atom in Various Moderators

Comparison with Experiment

The total scattering cross section for a hydrogen atom was calculated with the GAKER code and the above spectral density. Assuming the calculated cross section is the one for normal dodecane, we added the contribution from C to the calculated cross section. In Fig. 8 this calculation is compared with the experimental results. Agreement of these two cross sections is satisfactory except for the low energy region.

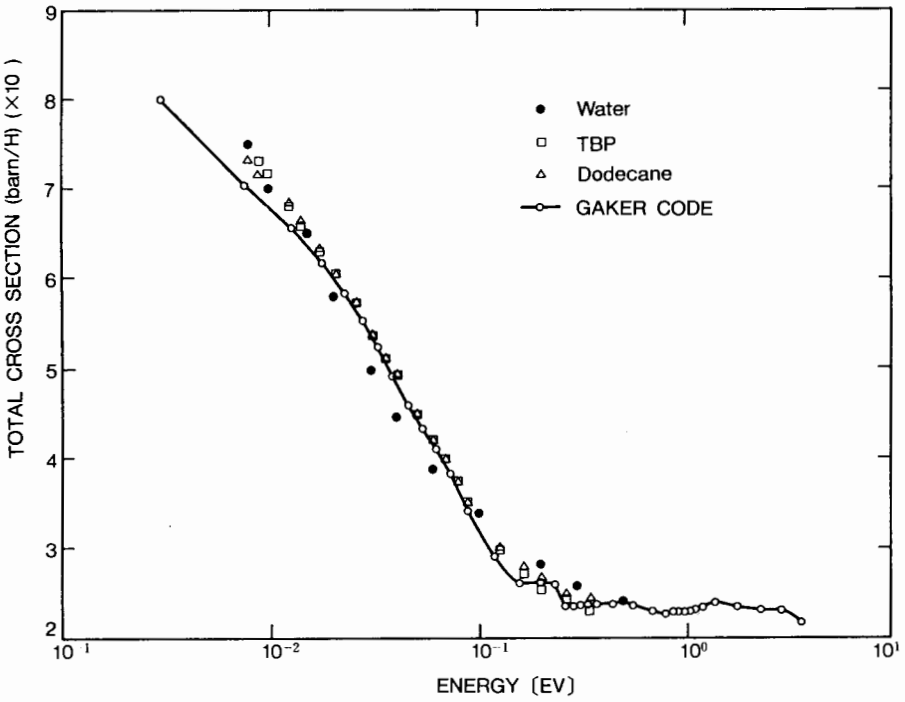

Fig. 8 Total Cross Section Divided by Number of Hydrogen Atom in Organic Compound and Water: Comparison with Experiment and Calculation

Conclusion

The total neutron cross sections in the energy range of 0.008 0.5 eV for organic moderators were successfully measured with the chopper and T-O-F facility installed at the Musashi reactor. The cross sections of normal dodecane and TBP were found to be almost identical when compared per hydrogen atom. The spectral density, which reproduces the measured total cross section, was proposed for the hydrogen atom in these two molecules.

Reference

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(6) Integral Neutron Thermalization Annual Summary Report, GA-5798, p155 (1964).