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Attenuation of Thermal Neutron Through Graphite

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Abstract

Calculation of the nuclear capture, thermal diffuse and Bragg scattering cross-sections as a function of graphite temperature and crystalline from for neutron energies from 1meV<E<10 eV were carried out. Computer programs have been developed which allow calculation for the graphite hexagonal closed-pack structure in its polycrystalline form and pyrolytic one.

The calculated total cross-section for polycrystalline graphite were compared with the experimental values. An overall agreement is indicated between the calculated values and experimental ones. Agreement was also obtained for neutron cross-section measured for oriented pyrolytic graphite at room and liquid nitrogen temperatures.

A feasibility study for use of graphite in powdered form as a cold neutron filter is details. The calculated attenuation of thermal neutrons through large mosaic pyrolytic graphite show that such crystals can be used effectively as second order filter of thermal neutron beams and that cooling improve their effectiveness.

Introduction

A beam of monochromatic neutrons, selected from the spectrum of a nuclear reactor by means of diffraction by a monochromator crystal, will in general be contaminated with higher-order components. When a primary wavelength larger than 2Å is selected, the second-order component is so important that a suitable filter becomes indispensable. In this respect, the use of highly oriented pyrolytic graphite has led to a considerable improvement in neutron-diffraction techniques.

Pyrolytic graphite (PG) has been in use for about 30 years as a filter. The principle of operation is that for neutrons travelling along the c-axis of this material, with certain neutron wavelength (λ) cannot be Bragg scattered, while second-order neutrons ($\lambda/2$) are strongly scattered. This differs considerably from polycrystalline graphite in which all neutron energies above the lowest Bragg cut-off at approximately 0.0018eV are strongly scattered.

It was shown by Bergsma and Van Dijk[1] that for first-order neutrons in the energy range between 10 and 15meV (λ between 2.33 and 2.86 Å) pyrolytic graphite is more effective as second-order filter than a quartz single crystal. By applying pyrolytic graphite as second-order filter in neutron powder diffractometry, Loopstra[2] and Pinto[3] demonstrated its high efficiency for first-order neutrons with λ=2.60 Å. Recently the efficiency of pyrolytic graphite filters could be improved even more by a reduction of the mosaic spread of the oriented graphite filter from 5° to values as low as 0.4°[4,5].

In these contexts, the dependence of the total cross-section on graphite parameters must be known, it being acknowledged that the thermal diffuse scattering (TDS) and neutron capture cross-section represents dominant contributions to the total cross-section at thermal-neutron energies.

Freund[6] has reported a semi-empirical formula, which permits the calculation of TDS as a function of material constants, temperature and neutron energy, neglecting the contribution of Bragg scattering to neutron transmission through a large imperfect single crystal. Recently, Naguib and Adib[7,8] have reported a general formula which allows calculation of the total thermal-neutron cross-section for an imperfect single crystal, as a function of crystal mosaic spread and orientation of the plane at which the crystal was cut.

Adib et al[9], adapted this formula, carrying out a feasibility study for use of poly-and mono-crystalline lead as cold and thermal-neutron filters, respectively. The low-Debye temperature, θ_D of lead and the difficulty in producing a single crystal with well-defined mosaic spread were found to limit its use as thermal neutron filter.

The present work concerns a feasibility study for use of polycrystalline graphite as a cold neutron filter, graphite is considered to be a better choice than lead. Its Dybe temperature being higher and its capture cross-section lower than lather. We also present below a detailed study of the characteristics of pyrolytic graphite as second order filter in neutron scattering experiments.

THE THEORETICAL TREATMENT

The total cross section determining the attenuation of neutrons by crystalline solid is given by

$$\sigma = \sigma_{abs} + \sigma_{tds} + \sigma_{Bragg} \tag{1}$$

where the neutron capture cross-section (i.e. absorption) σ_{abs} for most elements obeys the $1/\sqrt{E}$ law, and can be written as

$$\sigma_{abs} = C_1 E^{-1/2} \tag{2}$$

with E the neutron energy and C_1 a constant which can be calculated from values provided by Dunford and Rose [8].

The second contribution σ_{tds} , as shown by Freund[6] can be split into two parts, σ_{sph} and σ_{mph} depending on neutron energy. The single-phonon-scattering cross section σ_{sph} concerns the energy range $E \ll K_B \theta_D$, where K_B is Boltzmann' s constant and θ_D is the characteristic Debye temperature.

The second part of TDS is predominate in the range $E \geq K_B T$ where also down scattering and multiple phonon process occur. Freund[6] reported a semi-empirical formula which permits to calculate the TDS as a function of materials constants, temperature and neutron energy.

The contribution of Bragg scattering σ_{Bragg} to the total cross section takes into account the reflections from different (*hkl*) planes. In case of poly- crystalline material the reflections are from all planes having $d_{hkl} \geq \lambda/2$. As shown by Bacon[11] , σ_{Bragg} per atom for a fine powder material (polycrystalline material) can be given as:

$$\sigma_{Bragg} = \frac{N_o \lambda^2}{2m} \sum_{\lambda=\lambda/2} F_{hkl}^2 d_{hkl} e^{-2w} \tag{3}$$

where N_0 is the number of unit cells per cubic centimeter, m number of atoms per unit cell, F_{hkl} is the structure factor of the unit cell and e^{-2w} it is Debye Waller factor.

In pyrolytic graphite the crystallites are aligned to a high degree with their hexagonal c-axes parallel, whereas the a-axes are oriented at random. In the case of perfect alignment of the c-axes, the lattice planes *(hkl)* are tangent to a cone with its axis along the c-direction and an apex angle θ_{hkl} determined by :

$$\sin\theta_{hkl} = \frac{1}{c} d_{hkl} \qquad (4)$$

The interplanar distance d_{hkl} is given by the relation:

$$\frac{1}{d_{hkl}} = \left\{ \frac{4}{3a^2}(h^2 + k^2 + hk) + \frac{l^2}{c^2} \right\}^{\frac{1}{2}} \qquad (5)$$

when a pyrolytic graphite plate is oriented with the c-direction parallel to the incident neutron beam, a strong attenuation due to coherent elastic scattering by the *(hkl)* planes will occur if neutron wavelength satisfies the Bragg condition:

$$\lambda = 2\, d_{hkl} \sin\theta_{hkl} \qquad (6)$$

However as shown by Frikkee[12] it is possible to tune pyrolytic graphite plates for optimum scattering of second-order neutrons in a continuous wavelength range by varying the angle between the c-direction and the incident neutron beam. If this angle denoted by ψ, and if the mosaic spread is negligible in comparison in with ψ, the lattice planes *(hkl)* will scatter neutrons in the following wavelength intervals:

$$2d_{hkl}\sin(\theta_{hkl} - \psi) \le \lambda \le 2d_{hkl}\sin(\theta_{hkl} + \psi), \quad \text{for } \theta_{hkl} \ge \psi \ ,$$

$$0 \le \lambda \le 2d_{hkl}\sin(\theta_{hkl} + \psi), \qquad \text{for } \theta_{hkl} \le \psi \ ,$$

The plaes *(00l)*, on the other hand, scatter neutrons with a discrete wavelength

$\lambda = 2\, d_{00l} \cos\psi$.

The possibility to tune a pyrolytic graphite filter is a consequence of the fact that the scattering cross-section due to the *(hkl)* planes reaches pronounced maxima at the boundaries in the (λ, ψ) plane given b:

$$\lambda^{\pm}_{hkl} = 2d_{hkl}\sin|\theta_{hkl} \pm \psi| \tag{7}$$

It was shown by Frikkee[12] that the number of crystallites $N_C(\lambda\,; hkl)$ is given by $\lim_{\lambda\to\lambda^{\pm}_{hkl}} N(\lambda; hkl) = C\left\{4d_{hkl}\sin\psi\cos\theta_{hkl}\left|\lambda-\lambda^{\pm}_{hkl}\right|\right\}^{-\frac{1}{2}}$ (8)

Where C is a constant. Apparently the function $N_C(\lambda\,; hkl)$ diverges at the boundary values according to the asymptotic form $N(\lambda;hkl)\propto\left|\lambda-\lambda^{\pm}_{hkl}\right|^{-\frac{1}{2}}$. The Bragg scattering cross-section of PG crystal due to reflection from *(ool)* planes is given by

$$\sigma_{\mathrm{Bragg}} = -\frac{1}{Nt_o}\ln\prod_{hkl}\left(1-P^{\theta}_{ool}\right) \tag{9}$$

where N is the number of atoms/cm^3, t_o is the effective thickness of the crystal in cm and $\mathbf{P}^{\theta}_{ool}$ is the reflecting power of the *(ool)* plane inclined by an angle θ_{hkl} to the incident beam direction where $\mathbf{P}^{\theta}_{ool}$ is given by

$$P^{\theta}_{ool} = \frac{a}{1+a+(1+2a)^{1/2}\coth[b(1+2a)^{1/2}]} \tag{10}$$

$$b = \mu t_o / \gamma_{ool}$$

$$a = Q_{ool}W(\Delta)/\mu,$$

$$\gamma_{ool} = \cos\psi,$$

Where μ is the linear absorption coefficient γ_{ool} is the direction cosines of the diffracted beams relative to inward normal to the crystal surface. $W(\Delta)$ is the Gaussian distribution of the crystal mosaic blocks and given by

$$W(\Delta) = \frac{1}{\eta\sqrt{(2\pi)}}e^{-\Delta^2/2\eta^2} \tag{11}$$

Where η is the standard deviation of the mosaic blocks.

While the Bragg scattering cross-section from a plane with Miller indices *hkl* where $h \neq 0$ or $k \neq 0$ close to boundaries can be given as :

$$\sigma_{Bragg} = N_c \sum_{\lambda=\lambda/2} F_{hkl}^2 d_{hkl} e^{-2w} W(\Delta) \qquad (12)$$

Where $N_C(\lambda\ ;\ hkl)$ is the number of crystallites with the pro per orientation for (*hkl*) Bragg reflection of neutrons in the interval between λ and λ+dλ.

Two computer codes PolyCrystalline Graphite (PCG) and (HOPG) High Oriented Pyrolytic Graphite have been developed in order to calculate the total cross-section and the transmission of neutrons of energy range from 0.1meV to 10eV incident an polycrystalline graphite and an mosaic pyrolytic crystals respectively.

These codes are based on the calculation of the nuclear absorption cross-section and TDS cross-section σ_{tds} contribution in a similar way to that given by M.Adib[9]. In the PCG code the Brag scattering term σ_{Bragg} is calculated using Eq.(3) for all (hkl) planes and their higher orders having a non-zero structure factor. While in, HOPG code σ_{Bragg} is calculated using Eq.(9) for *00l* planes having non-zero crystallographic Q-value otherwise using Eq.(12).

For comparison of the experimental neutron cross-section data with the calculated values, the programs take into consideration the effects of both neutron wavelength resolution and incident neutron beam divergence.

Comparison with Experimental Results and Discussion

The formula given by Eq.(1) was fitted for poly and pyrolytic graphite crystals. The main graphite physical parameters required in these calculations are listed in Table 1.

Table 1 : Physical Parameters of graphite

Average Atomic Weight	12
Crystal Structure	HCP
Space Group	$P6_3/mmc$ (192)
Lattice Parameters	a =b= 0.2464nm, c = 0.6736nm
Number of Unit Cells/m3	2.89 E+28
Coherent Scattering Length (m)	0.661 E-14
Absorption Cross-Section for Thermal Neutrons σ(E=0.0225eV) (barn)	0.0031
Total Scattering Cross-Section σ_{bat} (barn)	5.555

Poly crystalline graphite

Using PCG code, the total cross-section of graphite at temperatures of 300K and 478K were calculated for neutron in the energy range from 0.1meV up to 1eV the results of calculation are displayed in Fig.(1) as sold line. For comparison, the experimental data presented are almost in agreement with experimental values for the fitted parameters C_2=20 and θ_D=1050K. From Fig.1(1) one can observe that the graphite total cross-section beyond the cut-off wavelength $\lambda c=2d_{oo2}$ (at E< 1.8meV) is about ... barn. This value is less than the free atomic cross-section 4 at neutron energies higher than 1eV.

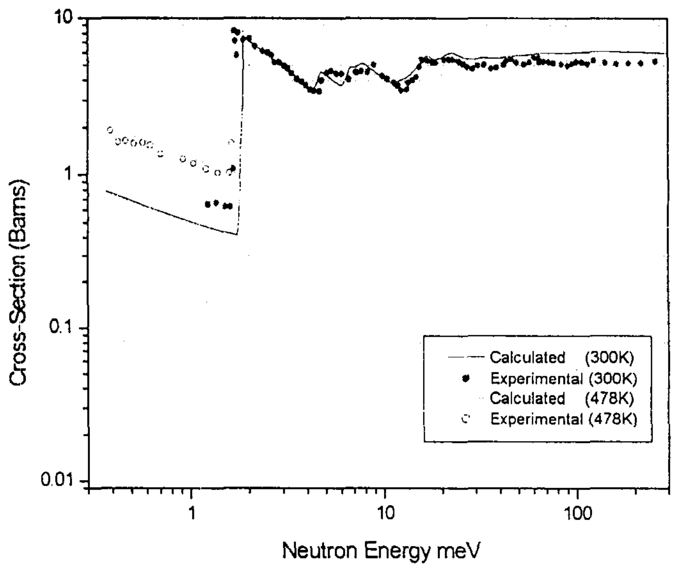

Fig .1 The Total Neutron Cross-Section For Polycrystalline Graphite .

To show the effect of both thickness and temperature of poly-crystalline graphite on its filtering features, the calculation were performed at room and Liquid Nitrogen (LN2) temperatures, for neutron wavelength in the energy range 10^{-3} - 1nm the results of these calculations are displayed in Fig. 2a and b, for room and LN2 temperatures respectively. The indication is that a 10cm thick poly-crystalline graphite cooled at LN2 temperature, has a better effect-to-noise ratio for neutrons with wavelengths ≥ 0.671nm

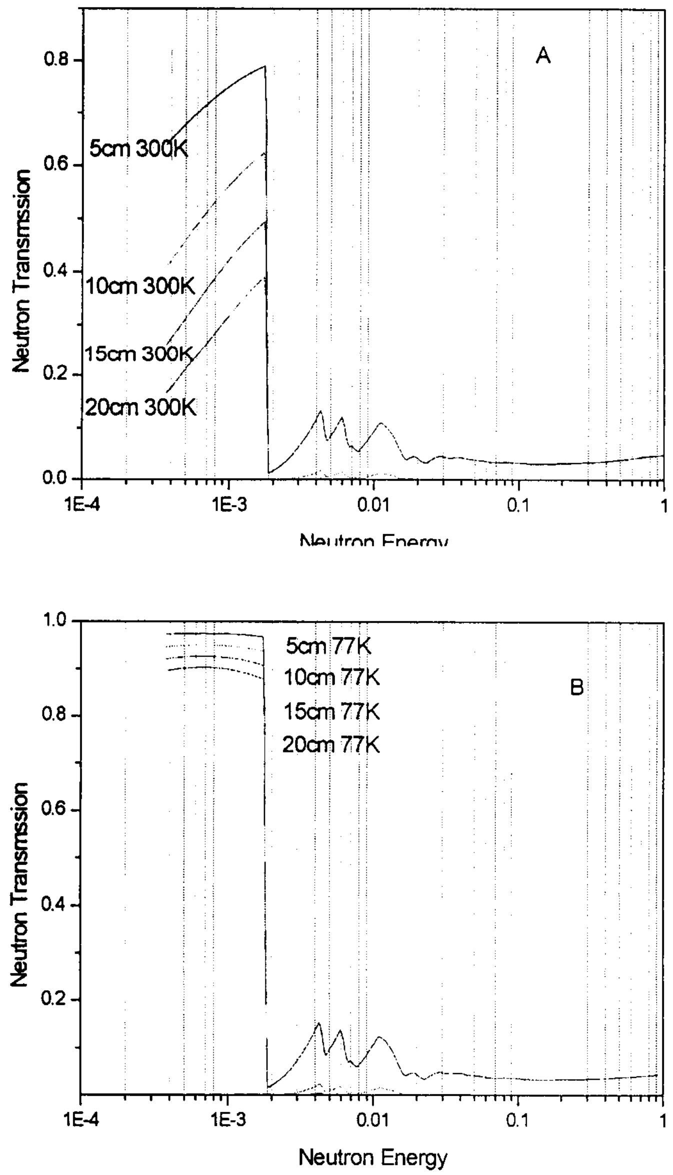

Fig.2 Neutron Transmission of Polycrystalline Graphite.

The calculation cold neutron flux, for which there is a Maxwellian distribution, for a neutron gas temperature close to liquid hydrogen, is displayed in Fig.3 before and after transmission through a 10cm thick poly crystalline graphite cooled to 77K. Its is observed that 10cm of graphite transmits about.٦. %, for neutrons with wavelength < 0.67nm. One can conclude that poly-crystalline graphite under these conditions is sufficient to efficiently remove the epithermal and fast neutrons less than ... %, while providing a high intensity of cold neutrons.

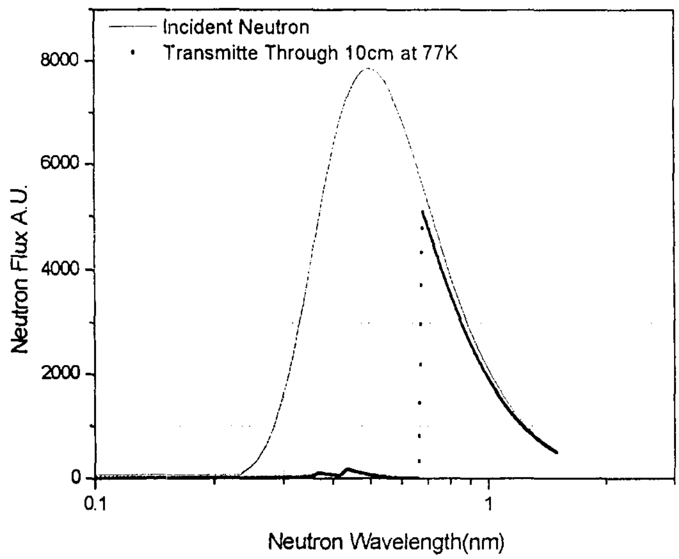

Fig.3 Transmitted Neutron Flux through Polycrystalline graphite .

Pyrolytic Graphite

The total cross-section data [2] of pyrolytic graphite for neutrons incident along the average direction of alignment of the C-axis is displayed in Fig.4 as close a circles. The PG used in this investigation consisted of plates of 2"x 2"x3" in which the C-axes were aligned to written 5° from the normal of the largest face. The calculation values using HOPG code are displayed as sold lines, assuming that PG has standard deviation of mosaic spread ≈ 2° and values of C_2 and θ_D that are the same as that for poly-crystalline case.

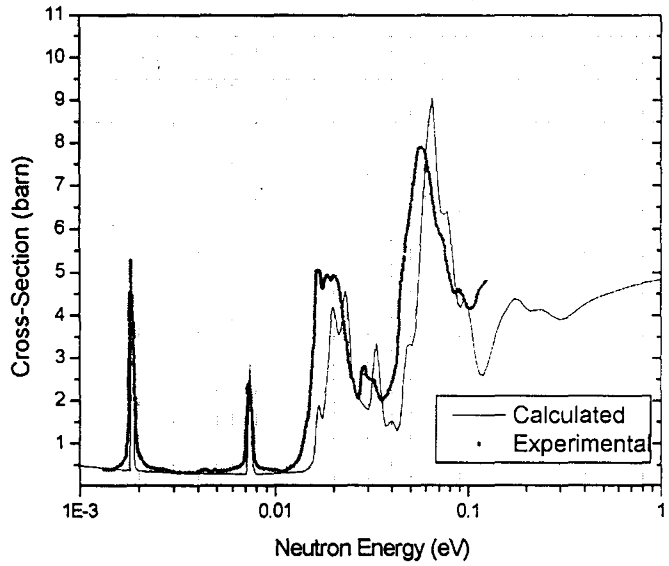

Fig.4 **Neutron Transmission Through Graphite Single Crystal.**

The calculation values are in reasonable agreement with measurement values the observed disagreement is due to the divergence of N_c at boundaries $\lambda^{\pm}_{hkl}$.

Table 2 list σ_{4E} and σ_E for E running from 10 to 15meV. For λ=0.26nm the intensity ratio between the first and second order can be improved by a factor of 100 at fist order transmission of 70%, with a filter thickness of 8cm. Thus the filtering losses can be kept quite moderator when a wavelength close to 0.26 nm is selected. It is found that for the stoll higher than second order neutrons the filter is always of sufficient size when it is sufficient for the second order.

Table 2 Total cross-section and their ratio of PG

E(meV)	λ(nm)	σ_E barn	σ_{4E} barn	σ_{4E}/σ_E	σ_{4E}/σ_E [2]
10.0	0.2845	0.46197	2.9413	6.366864	7.3
11.0	0.2725	0.48284	1.6434	3.403612	11.5
12.0	0.2605	0.50735	5.3675	10.57948	14.9
12.5	0.2545	0.52117	2.7555	5.287142	14.3
13.0	0.2505	0.53103	2.6875	5.060919	12.7
14.0	0.2405	0.55879	4.9049	8.777716	8.1
15.0	0.2325	0.59176	5.53	9.345005	4.3

However the 2° PG is quite expensive, particularly enlarge quantities necessary for filter purposes. Yelon etal[5] studied some lower quality PG crystals. The measured cross sections represented by Yelon etal[5] of 8° and 15° mosaic PG crystals at both room temperature(RT) and Liquid nitrogen(LN2) are displayed in Fig.5, while the calculation values are displayed in the same figure as solid lines.

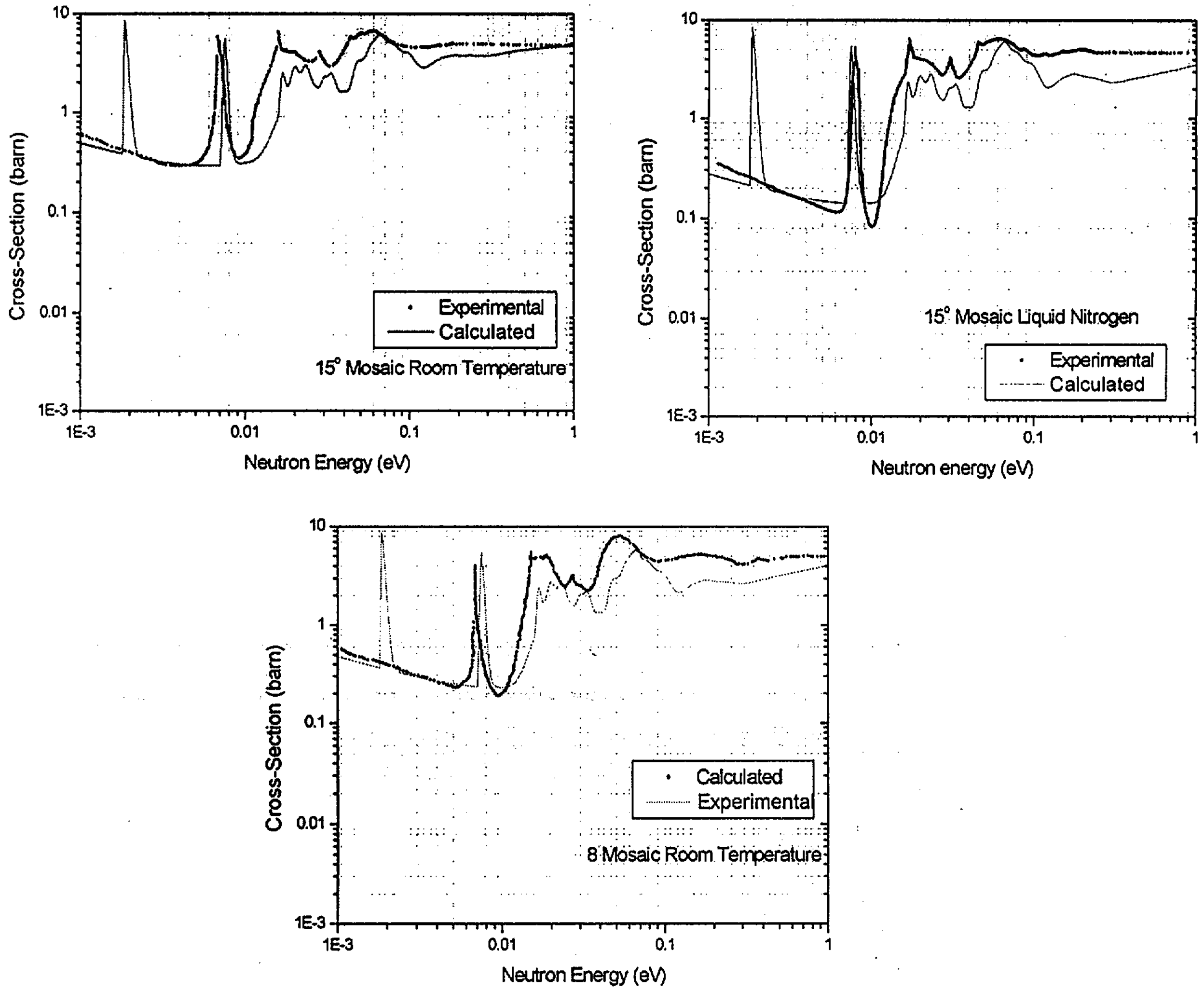

Fig.5 **Neutron Transmission Through Graphite Single Crystal at Different Mosaic.**

The peak in the cross-section at about 7.5mev is broadened by the mosaic and makes it feasible to use this as a filter passing 1.9mev first order while removing 7.5meV second order neutrons. This is not normally done with 2° PG. When the 15° mosaic PG cooled to liquid nitrogen (LN2) and 10meV is seen to decrease. Such decrease is clearly helpful for use at the very largest wavelengths.

The use of PG as a selective neutron filter requires an optimum choice of the crystal mosaic spread is essential neutron transmission through a 8cm PG crystal cooled to the temperature of LN2 for different values of mosaic spread were calculated and are displayed in Fig.6.

As may be observed, for standard of deviation of mosaic spread >0.4° several wavelength bands which are feasible to use PG as a high efficiency second order

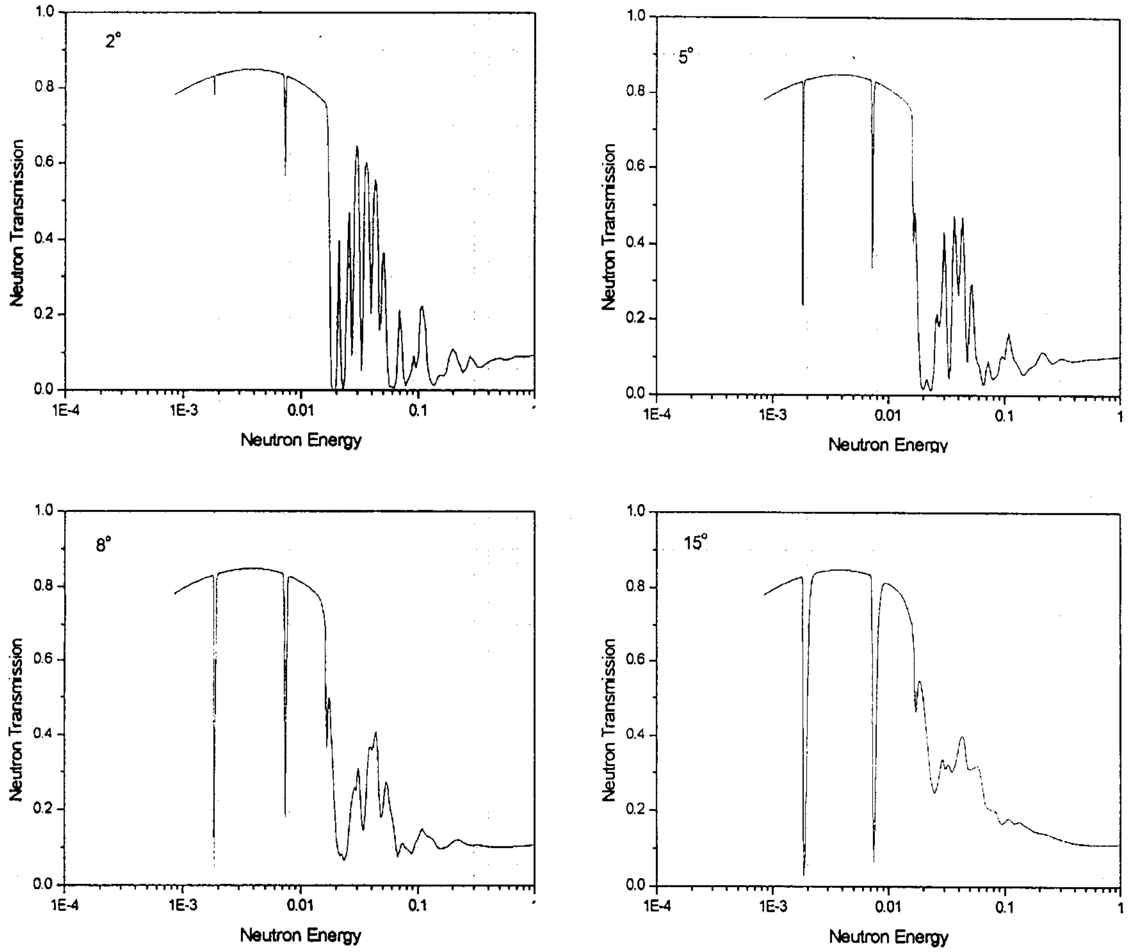

Fig.6. The Neutron transmission Graphite Single crystal for various mosaic spreads

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