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Cross-Section of Single-Crystal Materials used as Thermal Neutron Filters

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ABSTRACT

Transmission properties of several single crystal materials important for neutron scattering instrumentation are presented. A computer codes are developed which permit the calculation of thermal diffuse and Bragg-scattering cross-sections of silicon., and sapphire as a function of material's constants, temperature and neutron energy, E, in the range 0.1 meV .A discussion of the use of their single-crystal as a thermal neutron filter in terms of the optimum crystal thickness, mosaic spread, temperature, cutting plane and tuning for efficient transmission of thermal-reactor neutrons is given.

Keywords: Thermal Neutron Filters,Sapphire,Silicon

INTRODUCTION

The use of large, perfect single-crystals of various materials as filters for thermal neutron beams has long been known [1]. Several materials such as quartz (SiO_2) [2], bismuth [3], silicon [4], lead [5] and sapphire (Al_2O_3) [6-9] have been suggested as most successful filter materials. At high neutron energies, greater than about 1eV, the total neutron cross section σt of each of the above mentioned materials is in the range of a few barns, but at lower thermal energies, less than 0.1eV, where much of the coherent Bragg scattering is disallowed the effective cross-section for single-crystal specimens is much reduced. That is also due to the decrease of the thermal diffuse (TDS) or inelastic scattering cross-section with the decrease of neutron energy. "Freund [10] has reviewed a variety of single-crystal filter materials and calculated the total neutron cross-sections from transmission measurements. He uses a simple model to determine the single-phonon, multiple-phonon and absorption cross-sections as functions of the neutron wavelength and fits the data to a general formula with two adjustable parameters. There is good agreement between the calculated neutron cross-sections and the experimentally determined cross-sections for several materials, e.g. single-crystal silicon at both 300 and 77 K. There are, however, substantial differences between his results for sapphire and the experimental results of Nieman et al. [6], especially at neutron energies >100 meV. Freund [10] suggests that improvements in the quality of sapphire single-crystals will reduce these differences and thereby decrease the problems associated with elastic scattering of the higher-energy thermal neutrons. Born et al. [11] have examined the intensity transmitted by a single-crystal of Al2O3 over a range of orientation angles within ±2º and have observed no significant changes. They have found that a 90 mm-long sapphire-crystal filter has a transmission of about 0.8 for wavelengths in the range 0.12-0.24 nm and 0.07 for epithermal neutrons.

These transmission results are in good agreement with those obtained by Nieman et al. [6], who suggest that the effective attenuation coefficient can be minimized by fine-tuning the crystal orientation. Born et al. [11] have found that there is no need to tune the crystals for every wavelength. They find that the beam intensity transmitted by their crystals does not depend on the crystal orientation." reported by Mildner et al.[7] .

The aim of the work is to study the filtering characteristics of sapphire and silicon crystals and to show that they are a better choice when they used as a thermal neutron filter. The physical parameters of some most promising elements are listed in Table1.

Table1: Physical Parameters of Most Promissing Elements

Single crystal	σ_{free} -b	σ_t –b at 0.02eV	σ_t –b at 1 meV	θ_D -K	t-cm	Attenuation factor		
						E>1 eV	0.02 eV	1meV
Silicon	2.032	0.4	1.0	420	22.71	10	1.57	3.11
Bismuth	9.054	2.0	0.7	300	7.87	10	1.66	1.19
Lead	11.052	7.0	2.4	280	6.45	10	4.30	1.65
Sapphire	14.25	0.6	1.0	1040	6.9	10	1.09	1.16

Here, if it is required to attenuate neutrons with E > 1 eV by a factor of 10, thus the corresponding crystal thickness can be deduced as well as the attenuation factors at low neutron energies. The choice of the element to be used as a good thermal neutron filter depend upon how low are the values of such attenuation factors at low neutron energies.

From the Table1 it is evident that silicon and sapphire single crystals are a better choice as a thermal neutron filter.

THEORETICAL TREATMENTS

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

$$\sigma = \sigma_{abs} + \sigma_{tds} + \sigma_{Bragg} \quad \ldots\ldots \quad \textbf{(1)}$$

where σ_{abs} the absorption cross-section due to nuclear capture processes, σ_{tds} is the thermal diffuse scattering (TDS) or inelastic scattering cross-section and σ_{Bragg} corresponds to elastic or Bragg scattering. The first contribution σ_{abs} for sapphire is simply proportional to the neutron wavelength λ and energy in the range 10-4< E <10 eV. Thus, σ_{abs} can be written as $\sigma_{abs} = C_1 E^{-1/2}$ with C_1 a constant which can be calculated from values provided by Sears [12].

As shown by Freund [10], the second contribution σ_{tds} can be split in two parts, σ_{mph} (multiple phonon) and σ_{sph} (single phonon), depending on neutron energy

$$\sigma_{tds} = [A/(A+1)]^2 \sigma_{bat}\left[1 - e^{-WC_2E}\right] + E^{-1/2}\left[C_1 + \frac{\theta_D^{1/2}\sigma_{bat}}{36A}\begin{cases} R\ldots\ldots\ldots\ldots\ldots X \le 6 \\ 3.3X^{-7/2}\ldots X > 6 \end{cases}\right] \quad \ldots\ldots \quad \textbf{(2)}$$

where e^{-W} is the Debye-Waller factor [10], C_2 is a constant which is dependent on the scattering material and given by equation C2 =4.27exp[A/61] Freund [10] , $X = \theta_D / T$ (T is the sample temperature),σ_{bat} is the sum of coherent and incoherent scattering cross-sections of the bound atom), A in case of compounds is the average atomic mass number, and the series R is given by $R = \sum_{n=0}^{22} B_n X^{n-1} / [n!(n+5/2)]$, with B_n being the Bernoulli numbers [13].

The single phonon scattering cross-section, concerns the energy range E<<$k_B\theta_D$, where k_B is Boltzmann's constant and θ_D is the Debye temperature characteristic of the material. It is determined by phonon annihilation processes. The second part of TDS is predominant in the range E $\geq k_B T$ where also down scattering and multi-phonon processes occur.

However, using the static incoherent approximation, Cassels [14] has estimated the short-wavelength elastic cross-section, which is extinct for perfect single crystals. Hence the multi-phonon scattering cross-section in the range E>> $k_B\theta$, given by the first term of Eq.(2), can be replaced by:

$$\sigma_{mph} = \sigma_{free}\left\{1-\left(\lambda^2/2W\right)\left[1-\exp\left(-2W/\lambda^2\right)\right]\right\} \quad \ldots\ldots \quad \textbf{(3)}$$

The contribution of Bragg scattering σ_{Bragg} to the total attenuation arises from coherent elastic scattering due to reflections from different (hkl) planes. In the case of mono-crystalline material, the Bragg scattering cross-section is given by:

$$\sigma_{Bragg} = \frac{1}{Nt_0}\ln\left(\frac{1}{\prod\limits_{hkl}\left(1-P_{hkl}^{\theta}\right)}\right) \quad \ldots\ldots \quad \textbf{(4)}$$

where N is the atom number density , t0 is the thickness of the crystal in the beam direction , and P_{hkl}^{θ} is the reflecting power of the (hkl) plane inclined by an angle θ_{hkl} to the incident beam direction. As shown by Naguib and Adib [15], the reflecting power P_{hkl}^{θ} for an imperfect single crystal depends upon its mosaic spread, the direction cosine of the incident beam γ_0 relative to the inward normal to the crystal surface cutting along the (h_c k_c l_c) plane, the direction cosine of the diffracted beam γ_{hkl} and the inclination of the (hkl) plane to the crystal surface α_{hkl}. For simplicity, P_{hkl}^{θ} for sapphire - rhombohedral structure was deduced from its equivalent trigonal structure with unit cell parameter a_o=0.4759 nm and c_o = 1.299 nm. Usually sapphire single-crystals are cut along either the c-axis i.e. along (001) plane or the a-axis (100).

Therefore, following Naguib and Adib [16], if the angle between the neutron beam direction and the direction of the cutting plane is ψ, then the direction cosine of the diffracted beam γ_{hkl} for cutting along (001) plane can be expressed as:

$$\gamma_{hkl} = d_{hkl}\left\{\frac{l}{c_0}\cos\Psi + \left[\frac{2}{\sqrt{3}a_0}\left(\frac{h}{2}+k\right)\right]\sin\Psi\right\} \quad \ldots\ldots \quad \textbf{(5)}$$

and the inclination α_{hkl} angle can be described by:

$$\cos\alpha_{hkl} = d_{hkl}/c_0$$

While in case of the cutting plane (100)

$$\gamma_{hkl} = d_{hkl}\left\{\sqrt{\frac{4}{3a_0^2}\left(h+\frac{k}{2}\right)}\cos\Psi + \left[\frac{2}{3a_0c_0}\left(h+\frac{k}{2}-1\right)\right]\sin\Psi\right\} \quad \ldots\ldots \quad \textbf{(6)}$$

and $\cos\alpha_{hkl} = \sqrt{\frac{4}{3a_0^2}}d_{hkl}\left(h+\frac{k}{2}\right)$,

where $$d_{hkl} = \frac{1}{\sqrt{\frac{4}{3a_0^2}\left(h^2+k^2+hk\right)+\frac{l^2}{c_0^2}}} \quad \ldots\ldots \quad \textbf{(7)}$$

A computer codes have been developed in order to calculate the total cross-section and transmission of neutrons of energy range from 10-4 to 10 eV incident on mono-crystals.

RESULTS AND DISCUSSIONS

Sapphire Single crystal

The formula given by Eq.(1) was calculated and compared with the experimental results for mono-crystalline sapphire. The main physical parameters required in the calculation are listed in Table (2), where the crystallographic specifications of α-Al_2O_3 (sapphire) are taken from Ref.[17, 18].

Table (2): Physical parameters of sapphire

Average at. Wt.	20.4
Density	3.965 gm/cm^3
Crystal structure	Rhombohedral (n=2), or Trigonal (n=6)
Lattice parameters	Rhombohedral: a_o=0.5128nm=b=c; and α=55.28°=β=γ Trigonal: a_o=0.4759nm and c_o = 1.299nm
Atomic positions (2 atoms/unit cell)	Al: w, w, w; W, W, W; $1/2+w, 1/2+w, 1/2+w$; $1/2-w$, $1/2-w$, $1/2-w$; and w=0.352 O: u, $1/2-u$, 1/4; $1/2-u$, 1/4, u; 1/4, u, $1/2-u$; $\bar{u}$, $1/2+u$, -1/4; $1/2+u$, -1/4, $\bar{u}$; -1/4, $\bar{u}$, $1/2+u$; and $u = 0.556$
Coherent scattering length	$b_{Aluminum}$= 3.449 fm, b_{Oxygen}= 5.805 fm
Absorption cross-section (σ_a)	0.231 barn at λ=0.1798 nm
Bound atom scattering cross-section (σ_{bat})	15.7 barn
Debye temperature θ_D	1040 K
Melting point	2040°C

To decrease the fluctuations in the transmission, which are due to Bragg reflections, an optimum choice of the crystal mosaic spread is essential. Neutron transmission in the a-axis direction through 8 cm thick sapphire crystal, cooled to the liquid N2 temperature for different values of mosaic spread were calculated and displayed in Fig.(1).

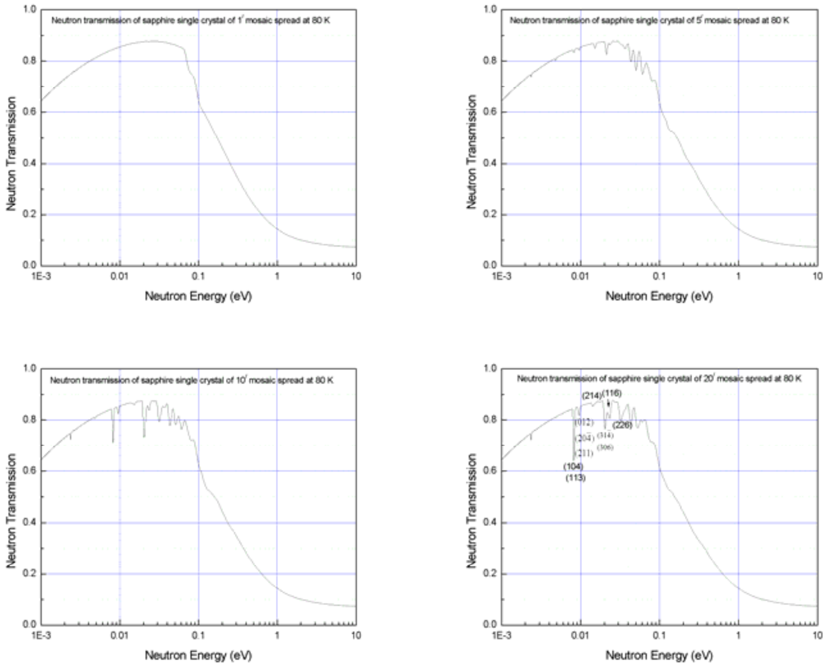


Fig. (1) Neutron transmission along the a-axis for different mosaic spreads.

For comparison, Fig.(2) shows similar calculations for the transmission in the c-axis direction for a 8 cm thick sapphire crystal under the same conditions .One can observe, that the Bragg dips in the neutron transmission through high-quality sapphire single crystal (FWHM) ≈ 1') are drastically reduced.. Such crystals are expensive to produce. " The γ -ray diffraction measurements carried out by Born et al [11] indicate the crystalline perfection and homogeneity of the high-quality sapphire crystals that are now available. The mosaic spreads (5-15") are very much lower than typical angular divergences for thermal-neutron beam tubes". Mildner et al [7]. It is obvious that the neutron transmission through 8 cm sapphire single crystal with mosaic spread of 5' along the a-axis is less disturbed by parasitic Bragg reflections than along the c-axis. Since the strong reflections from (119) and (113) planes disturb the neutron transmission along the c-axis $E \cong 0.02$ eV . As can be also observed parasitic Bragg reflections can limit the use of sapphire as a thermal neutron filter for mosaic spreads > 5'.

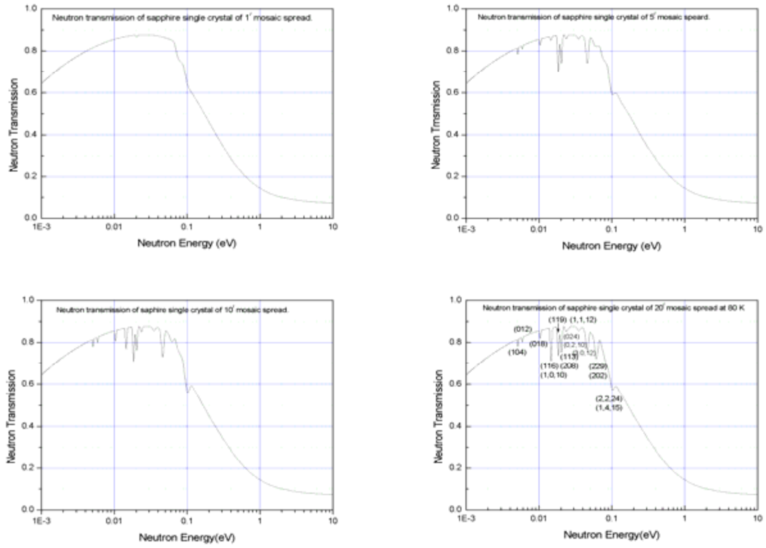
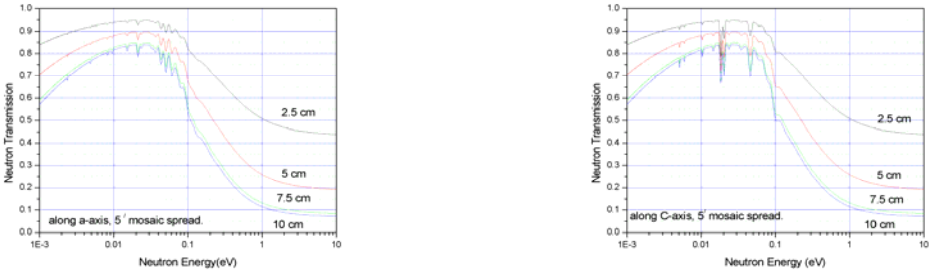


Fig. (2) Neutron transmission along the c-axis at different mosaic spreads.

Therefore high quality sapphire single crystals along the a-axis is a better choice than that along the c-axis. Such conclusion is in agreement with the experimental results reported by Mook et al [9].

To find the optimum sapphire thickness, the neutron transmission through different crystal thickness, were calculated. Fig. 3a shows the result of calculation through a sapphire (100) crystal having mosaic spread of 5' while Fig.3b shows the situation through sapphire (001). It would appear that a 7.5 cm thick sapphire (100) is sufficient for removing neutrons with energies > 1 eV ($T_n < 8\%$) while providing high transmission ($T_n > 85\%$) for neutron energies < 0.02 eV . The fluctuations in the transmission curve which are due to Bragg reflections are less than 5%. These fluctuations can be smoothed by increasing the incident neutron beam divergence.



(a) **(b)**

Fig. (3) Neutron transmission through sapphire, for different crystal thickness.

Fig. 4 shows that 7 cm thick sapphire (100) crystal can be successfully used to transmit a thermal reactor flux having a Maxwellian distribution with neutron gas temperature close to 300 K , while significantly rejecting the accompanying slowing down flux (dE/E) ;with neutron energy E > 1 eV. Fig.4 also shows that there is a small increase ≈ 5% at neutron energies < 0.02 eV in the neutron transmission through the cooled sapphire crystal at LN2 temperature. Such small improvement as reported by Mildner et al [7] & [8] for many applications, this may be insufficient to warrant the expense and inconvenience of cryogenics.

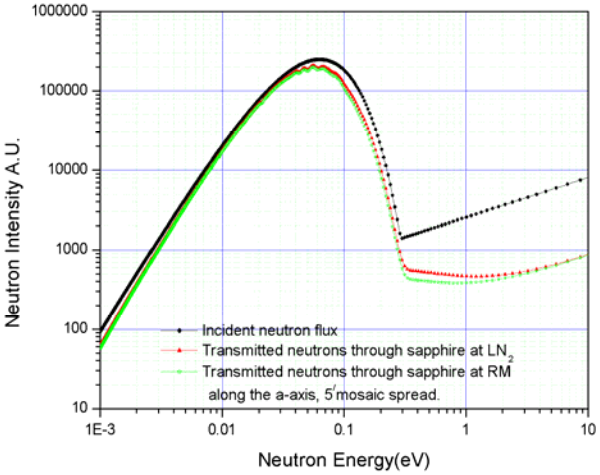


Fig. (4) Thermal –neutron flux transmitted through 7 cm sapphire (100) crystal.

Nieman et al [6] reported that low quality sapphire can be considerably more efficient thermal neutron filter than either silicon or quartz. For full realization of sapphire's potential., the user must prepared to fine tune the crystal orientation to minimize the attenuation coefficient for each energy of interest.

As conclusion one can notice that the calculated total neutron cross-section and effective attenuation coefficient for sapphire single crystals with rhombohedral structure were within an accuracy which is sufficient for determining its neutron transmission characteristics. Calculation shows that 7.5 cm thick sapphire single crystal, cut along a-axis and with a FWHM on a mosaic spread of 5 min of arc, is a good thermal neutron filter. A crystal cut along c-axis with a mosaic spread of about 1° is also an efficient thermal neutron filter, when it is fine-tuned to minimize the attenuation for each neutron energy of interest.

Silicon Single Crystal

In order to check the applicability of the deduced formula, the calculations were carried out for mono crystalline silicon crystals and compared with the experimental ones. The main silicon physical parameters used for calculations are listed inTable 4.

Table 3: The physical properties of Silicon

Atomic Weight	28.08
Crystal Structure	Diamond Structure
Lattice Constant	a_o = 054307 nm
Atomic Positions	0 0 0, ½ ½ 0, ½ 0 ½ , 0 ½ ½ , ¼ ¼ ¼, ¾ ¾ ¼ , ¾ ¼ ¾ , ¼ ¾ ¾
Number of unit cells/m^3	0.6243E+28
Debye Temperature	420K
Neutron capture cross-section at 0.025 eV	0.161 barns
σ_{bat}	2.180 barns
Coherent Scattering	4.2 fm

The total neutron cross-section data carried out by Brugger (1979) [4] at both room and liquid nitrogen temperatures are displayed as dots in Fig.5a & b respectively . The calculated values using DSIC are also displayed in Fig.5 as solid lines assuming that Si single crystal is perfect and the neutron incident perpendicular to the (111) plane . One can notice that the calculated values are in good agreement with the experimental ones. However as reported by Brugger(1979) the neutron Bragg reflections from the crystal were smoothed by slightly titling crystal around zero inclination angle during the measurements.

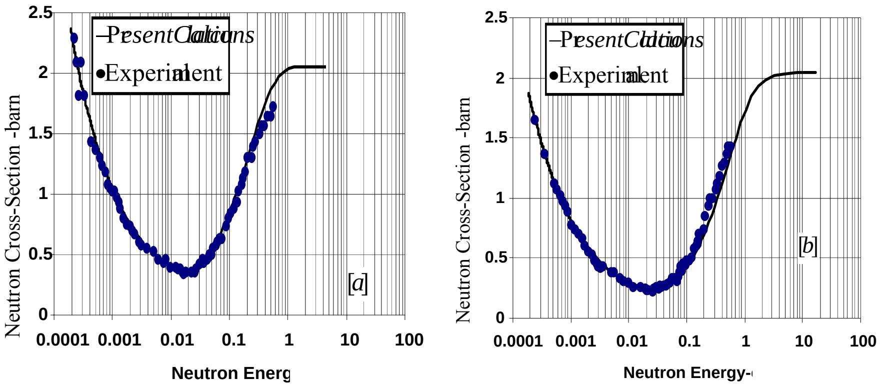


Fig.5 Neutron Cross-Section of a Perfect Si Single Crystal .

In order the check the effect of crystal mosaic spread on the neutron attenuation, the calculations were carried out assuming that 30cm Si single crystal is cut along (111) plane and cooled at liquid nitrogen temperature at different mosaic spread. Fig.6 displays the result of calculation. From the figure one can notice that the Bragg reflections can not be neglected at mosaic spread values higher than 0.5mRad.

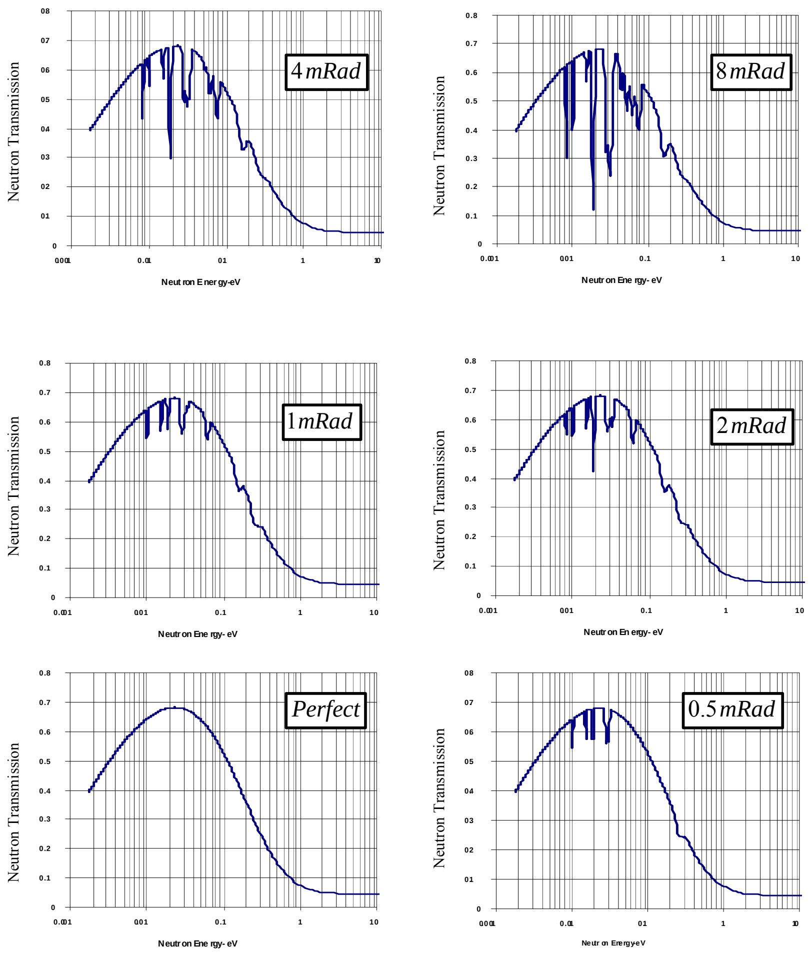


Fig.6. The Neutron transmission through 30cm of Si crystal for various mosaic spreads

To find the optimum thickness of silicon (111) crystal, the neutron transmission was calculated assuming that the mosaic spread has the value of 0.5mRad and 1mRad where the crystal was cooled at liquid nitrogen temperature. Fig.7a & b display the results of calculation for silicon single having mosaic 0.5mRad and 1mRad respectively. It seems that a 30 cm thick crystal with mosaic spread of 0.5mRad and cooled at liquid nitrogen temperature is free from parasitic Bragg reflection

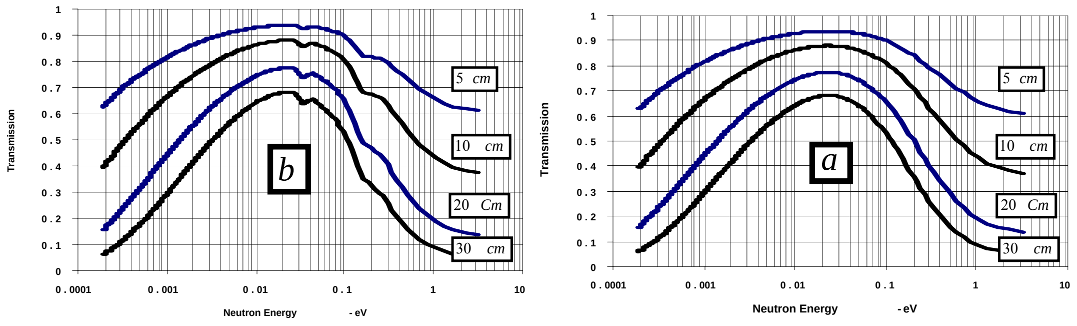


Fig.7 Neutron transmission through silicon crystals cut along (111) plane for different crystal thickness

To show that such silicon crystal can be successfully used as thermal neutron filter the calculation of the transmitted thermal neutron flux through 30 cm silicon was carried out . The result of calculation is displayed in Fig.8, where the thermal neutron flux was assumed to have a Maxwellian distribution with neutron gas temperature close to 300 K ,while the fast neutron one to have dE/E, where E is the neutron energy. Its seems that 30 cm thick single crystal cooled at liquid nitrogen it sufficient of almost removing epithermal neutron and transmits less than 1% of fast ones and less than 5% of γ-rays with average energy Eγ =2 MeV while, providing reasonable intensity of thermal neutrons .

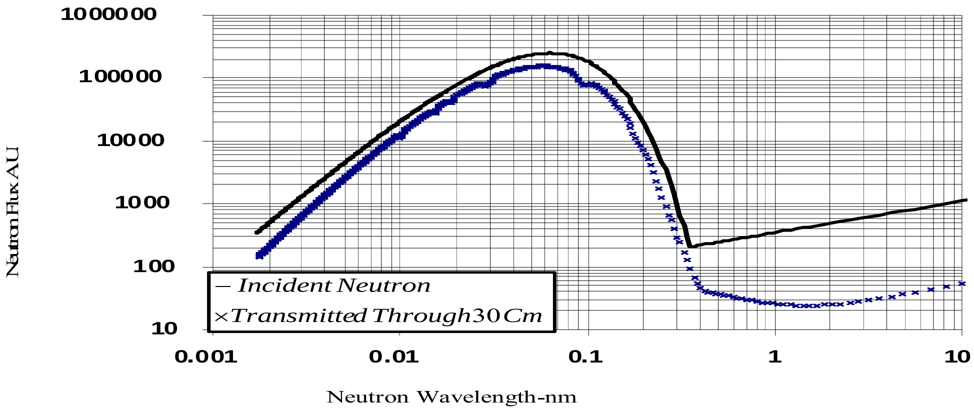


Fig..8 Transmitted thermal neutron flux through single silicon crystal cut along (111) plane.

The simple formula presented in this paper permits the calculation of the total cross-section of mono Si crystals having a diamond structure to be deduced within an accuracy which is sufficient for determining the validity of such a crystal when it used as a thermal neutron filter. Calculations showed that Si single crystal as a good thermal one at low γ -ray and fast neutron background.

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