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PYROLYTIC GRAPHITE AS A SELECTIVE NEUTRON FILTER

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The transmission of neutrons through pyrolytic graphite (PG) crystals, set at different angles with respect to incident beam, were calculated using an additive formula. A computer program HOPG was developed to provide the required calculation. An overall agreement between the calculated neutron transmissions through a slab of 1,85 mm thick PG crystal with an angular spread of c-axes of 0,4°, set at different angles to the incident beam, and the available experimental ones in the wavelength range from (0,02 to 1,4) nm were obtained. A feasibility study for use of PG crystal as an efficient second-order neutron filter is detailed in terms of crystal thickness, angular spread of c-axes and its orientation with respect to the neutron beam. It was shown that a PG crystal with an angular spread of 0,8° is sufficient for optimum scattering of second-order neutrons in the wavelength band (0,384 - 0,183) nm, by adjusting the filter in an appropriate orientation.

Introduction

The filtering characteristics of PG were first discussed by Brockhouse and Diefendorf [1]. Its use as an effective filter for low energy (less than 15 meV) neutron beam has been further advocated by Loopstra [2]. Good filtering characteristics were reported for oriented graphite crystals [2] with mosaic spread of 5°.

Last years, a significant advance has been made in producing high quality PG crystals. Graphite crystals are now available with mosaic spreads between 0,4 to 3,5°. It was shown by Riste and Otnes [3], that these graphite crystals are exceptional good monochromators for neutron scattering experiment. Highly oriented pyrolytic graphite (HOPG) is shown by Shirane and Minkiewicz [4] to be an extremely efficient $\lambda/2$ filter in the energy range between 13 - 15 meV.

Recently Adib et al. [5] calculated the nuclear capture, thermal diffuse and Bragg scattering cross-sections as a function of graphite temperature and crystalline form for neutron energies from 1 meV < < E < 10 eV. A computer program PG has been developed [5] which allow calculation for the graphite hexagonal close-packed structure in its pyrolytic form. The calculated attenuation of thermal neutrons through PG (mosaic spread > 2°) crystals with c-axis parallel to incident neutron beam showed that such crystals can be used effectively as second order filter within energy intervals (4 - 7) and (10 - 15) meV.

As shown by Frikkee [6] that PG crystal with perfect alignment of its c-axes may be tuned for optimum scattering of second-order neutrons in the wavelength range between 0,112 and 0,425 nm, by adjusting the filter in an appropriate orientation. However, Frikkee [6] neglected the effect of PG mosaic spread value on the width of the wavelength tuning interval. Moreover, the attenuation factor of second-order neutrons as a function of PG crystal thickness is not also given by Frikee [6].

The recent measurements, reported by Mildner et al. [7], of neutron transmission through highly oriented 1,85 mm thick PG (mosaic 0,4°) crystal set at different angles to the incident beam justifies the existence of the tuned intervals reported by Frikkee [6].

Therefore the present work concerns a feasibility study for use of PG crystal to tune for optimum scattering of second-order neutrons in the wavelength range between 0,183 and 0,384 nm by adjusting the filter crystal in appropriate orientation. The neutron transmission through PG crystals were calculated as a function of both their mosaic spread value and thickness for efficiently removing of the second-order neutrons.

Theoretical treatment

The graphite absorption cross-section due to nuclear capture is very small (≈ 3 mb at $E_n = 0{,}025$ eV). Therefore the total cross-section determining the attenuation of neutrons by a PG crystal is given by the sum:

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

where σ_{tds} is the thermal diffuse scattering and σ_{Bragg} correspond to Bragg scattering cross-section due to reflection from (hkl) planes.

As shown by Freund [8] σ_{tds} can be split into σ_{mph} (multiple phonon) and σ_{sph} (single phonon) depending on neutron energy.

The single phonon scattering cross-section, concerns the energy range $E \ll K_B\theta_D$, where K_B is Boltzmann's constant and θ_D is the Debye temperature characteristic of the graphite. The second part of TDS is predominant in the range $E \geq K_BT$ where down scattering and multiphonon processes occur. As shown by Freund [8], the predicted empirical equation for σ_{mph} fits the experimental results rather well except for graphite.

However, using the static incoherent approximation Cassels [9] has estimated the short-wavelength elastic cross-section. Hence the multiphonon scattering cross-section term given by Freund [8] in the range E >> $K_B\theta$ can be replaced by:

$$\sigma_{mph} = \sigma_{free}\left\{1-\left(\lambda^2\big/2w\right)\left[1-\exp\left(-2w\big/\lambda^2\right)\right]\right\}, \quad (2)$$

where e^{-w} is the Debye-Waller factor [10] and σ_{free} is the free atom cross-section given as:

$$\sigma_{free} = \sigma_{bat}\, A^2\big/\left(A+1\right)^2, \quad (3)$$

where σ_{bat} is the sum of the coherent and incoherent scattering cross-sections of the bound atom, and A is the atomic mass number.

Following Frikkee [6], in PG 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:

$$\sin\theta_{hkl} = \frac{1}{c} d_{hkl}.$$

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

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

When a PG 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 the neutron wavelength satisfies the Bragg condition:

$$\lambda = 2d_{hkl}\sin\theta_{hkl}. \quad (4)$$

However, it is possible to tune the PG 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 is denoted by ψ, and if the mosaic spread is negligible in comparison with ψ, the lattice planes (hkl) will scatter neutrons in the following wavelength intervals:

$$\begin{gathered} 2d_{hkl}\sin\left(\theta_{hkl}-\psi\right) \le \lambda \le 2d_{hkl}\sin\left(\theta_{hkl}+\psi\right) \\ \text{for } \theta_{hkl} \ge \psi, \\ 0 \le \lambda \le 2d_{hkl}\sin\left(\theta_{hkl}+\psi\right) \\ \text{for } \theta_{hkl} \le \psi. \end{gathered} \quad (5)$$

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

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

where θ is the glancing angle.

The contribution of Bragg scattering arises from coherent elastic scattering due to reflections from different (hkl) planes is given by K. Naguib and M. Adib [11]. In case of PG the Bragg scattering cross-section due to reflection from $(00l)$ planes is given by [5].

$$\sigma_{Bragg}(00l) = -\frac{1}{Nt_o}\ln\left(1-P_{00l}\right), \quad (6)$$

where N is the number of unit cell/cm^3, t_o is the effective thickness and P_{00l} is the reflecting power of the $(00l)$ plane when the neutron beam inclined by an angle ψ to the c-direction.

The reflecting power P_{00l} for zero absorption of imperfect graphite crystal is given by [10].

$$P_{00l} = \left[\left(Qt_0/\gamma_0\right)W(\Delta)/\left\{1+\left(Qt_0/\gamma_0\right)W(\Delta)\right\}\right], \quad (7)$$

where Q is the well-know crystallographic quantity given by [10]:

$$Q = \frac{\lambda^3 N^2}{\sin 2\theta} \quad F_{hkl}{}^2,$$

where F_{hkl} is the amplitude of the diffracted neutron beam for the *hkl* reflection, γ_{ool} is the direction cosine of the diffracted beam relative to inward normal to the crystal surface, $W(\varDelta)$ is the Gaussian distribution of the graphite mosaic blocks and given by:

$$W(\varDelta) = \frac{1}{\eta(2\pi)}\exp\left[-\varDelta^2\big/2\eta^2\right], \quad (8)$$

where η is the standard deviation on the mosaic blocks.

However, it was shown by Frikkee [6] that the scattering cross-section due to non-$00l$ planes reaches pronounced maximum at the boundaries in the $(\lambda;\psi)$ plane given by:

$$\lambda^{\pm} = 2d_{hkl}\sin\left|\theta_{hkl} \pm \psi\right| . \qquad (9)$$

It is also shown by Frikkee [6]; that number of crystallites $N(\lambda;hkl)$ with the proper orientation for (hkl) Bragg reflection of the neutrons in the interval between λ and $\lambda + d\lambda$, diverges at the values $\lambda^{\pm}_{hkl}$ according to the asymptotic form

$$N_c(\lambda;hkl) \propto \left|\lambda - \lambda^{\pm}_{hkl}\right|^{-\frac{1}{2}} . \qquad (10)$$

Consequently the Bragg scattering cross-section due to reflection from non-$00l$ planes of a PG crystal with mosaic η and set at angle ψ, at wavelength λ in the interval between λ^- and λ^+, can be given as

$$\sigma^{non-00l}_{Bragg} = \frac{N_o\,\lambda^3\,F^2_{hkl}\,e^{-2w}}{4d_{hkl}\,\sin\psi\,\cos\theta_{hkl}\left|\lambda - \lambda^{\pm}_{hkl}\right|^{\frac{1}{2}}} . \qquad (11)$$

While at boundaries the Bragg scattering cross-section is broaden by mosaic spread and can be expressed as

$$\sigma^{non-00l}_{Bragg} = \frac{N_o\,\lambda^3\,F^2_{hkl}\,e^{-2w}W(\Delta)}{4d_{hkl}\,\sin\psi\,\cos\theta_{hkl}\,(\delta\lambda)^{\frac{1}{2}}} ,$$

where $\delta\lambda$ is the wavelength spread.

Consequently, the Bragg scattering of PG crystal set at angle ψ versus wavelength due to reflections from (hkl) planes can be given as:

$$\sigma_{Bragg} = \sigma^{00l}_{Bragg} + \sum_{hkl} \sigma^{non-00l}_{Bragg} , \qquad (12)$$

where summation is taken over all non-$(00l)$ planes satisfying the inequalities given by (5).

A computer code HOPG has been developed in order to calculate the transmission of neutrons in the wavelength range between 0,02 and 1,4 nm incident on PG at different angles. The HOPG code is an adapted version of the PG code [5]. The adapted version can additional provide the following calculations:

1. The σ_{TDS} term using both Freund's [8] and Cassels's [9] formula each in its given energy range.

2. The Bragg scattering cross-section term due to reflections from both $00l$ planes and non-$00l$ planes when the crystal is set at different angles w.r.t. the incident neutron beam.

3. For comparison the experimental neutron transmission data with the calculated values, the program takes into consideration effect of both wavelength resolution and beam divergence in either constant $\Delta\lambda$ mode of experimental set up or constant resolution $\Delta\lambda/\lambda$ one.

Comparison with experimental results and discussions

The main graphite physical parameters required in calculations are listed in Table 1.

Table 1. **Physical parameters of graphite**

Atomic weight	12
Crystal structure	HCP
Space group	P63/mmc (Nr. 94)
Lattice parameters	a = 0,2456 nm c = 0,6696 nm
Atomic positions	4 atoms / unit cell 0,0,0 ; 0,0,½; ⅔,⅓,0 ; ⅓,⅔,½
Number of unit cells / m^3	0,284 E29
Coherent scattering length b_c	6,61 fm
Absorption cross-section σ_a (E = 0,025)	0,0035 barn
Total scattering cross-section (σ_{bat})	5,551 barn
Debye temperature	1050 K
Boiling point	4827 °C

The values of wavelengths for various reflections (hkl) having non-zero structure factor and for different PG crystal setting angle θ relative to the incident beam are calculated using HOPG code. The result of calculations is listed in Table 2 along with the calculated values reported by Mildner [7]. Where the angle θ is the complementary angle of ψ .

Table 2. **The calculated wavelengths (nm) for various reflections (hkl) and for different crystal setting**

hkl	$\psi = 0°$ $\Theta = 90°$		$\psi = 45°$ $\Theta = 45°$		$\psi = 58°$ $\Theta = 32°$		$\psi = 68°$ $\Theta = 22°$	
	λ, nm	Mildner λ, nm	λ, nm	Mildner λ, nm	λ, nm	Mildner λ, nm	λ, nm	Mildner λ, nm
002	0,6695	0,6696	0,473	0,4735	0,335	0,3548	0,251	0,2508
004	0,3348	0,3348	0,237	0,2367	0,177	0,1774	0,125	0,1254
006	0,2232	0,2232	0,158	0,1578	0,118	0,1183	0,084	0,0836
008	0,1674	0,1674	0,118	0,1184	0,089	0,0887	0,063	0,0627
100	-------	-------	0,3008	0,3008	0,3608	0,3608	0,3944	0,3944
101	0,1227	0,1227	0,3600	0,3600	0,3927	0,3927	0,4042	0,4043
102	0,1925	0,1925	0,3505	0,3505	0,3591	0,3591	0,3531	0,3531
103	0,3124	0,2125	0,3079	0,3079	0,3016	0,3017	0,2863	0,2863
104	0,2067	0,2060	0,2612	0,2612	0,2475	0,2475	0,2283	0,2283
105	0,1918	0,1918	0,2210	0,2210	0,2041	0,2041	0,1838	0,1838
106	0,1750	0,1750	0,1887	0,1887	0,1706	0,1706	0,1507	0,1507
110	------	-------	0,1737	0,1736	0,2083	0,2082	0,2277	0,2278
111	-------	0,0436	-------	0,1148	-------	0,1238	-------	0,1265
112	0,0794	0,0794	0,2092	0,2060	0,2257	0,2230	0,2305	0,2305

The good agreement obtained between both sets of calculations supports the application of HCP structure with four atoms per graphite unit cell. However the wavelength value at which the reflection from (111) plane occur is not included in our calculation since it has a zero structure factor.

Fig. 1 shows the transmission results reported by Mildner [7] for the graphite monochromator crystal set at nominal angles of 90, 45, 32, 22°, to the incident beam. Where the (0,4° mosaic) PG monochromator crystal, 50 mm high , 75 mm wide and 1,85 mm thick was placed in a sample position. The measurements were performed on the C3 beam line at the Intense Pulsed Neutron Sources at Argonne National laboratory. Mildner [7] reported that the positions of the dips due to (00l) reflections were used to determine the wavelength calibration of the data. The calibrated orientation angles θ are shown in Fig. 1.

For comparison, the calculated transmission versus neutron wavelength using HOPG code at calibrated angles θ and taking into consideration that, the measurements were set to a time resolution of 2 % or $\Delta\lambda/\lambda$ = 2 %, with experimental ones are also given in Fig. 1.

An overall agreement between the calculated neutron transmissions and measured ones at longer wavelengths (> 5 nm) for different angles. However, the slight disagreement observed may be due to the lake of the experimental transmission data through such thin PG crystal used by Mildner [7].Where the transmission at some wavelengths exceed one. Such inconsistent at = 30,3° is also observed by Mildner [7]. They reported that, they have no adequate explanation for this.

The calculated position and depth of the major Bragg dips in the transmission caused by various (hkl) reflections are found to be in good agreement with the measured ones. Such agreement supports the application of the HOPG code for calculations.

From Fig. 1 one can note that at shorter wavelengths, in the region where the non-$00l$ reflections are available, the transmission is reduced considerably. For small θ settings, the $10l$ reflections with their broad asymmetric dips in the transmission occur at wavelengths longer than the 002 reflection, whereas the opposite is true for the larger θ. Therefore, highly aligned graphite crystal may be tuned for optimum scattering of second-order neutrons, by adjusting the crystal in an appropriate orientation. Hence, as shown by Frikkee [6], one may expect to realize optimum scattering of neutrons by the (hkl) planes at the boundary curves $(hkl)^{\pm}$ in (λ, ψ) space defined by Eq. (9). Possible tuned positions of second-order neutrons are calculated and displayed in Fig. 2. On the basis of structure factors and multiplicities of Bragg reflections the best results may be expected at the curves $(11l)^{\pm}$ for even l, (002) and (006).

To show the filtering efficiency of PG, the neutron transmissions through 1 cm thick PG crystal (0,4° mosaic) were calculated as a function of wave-

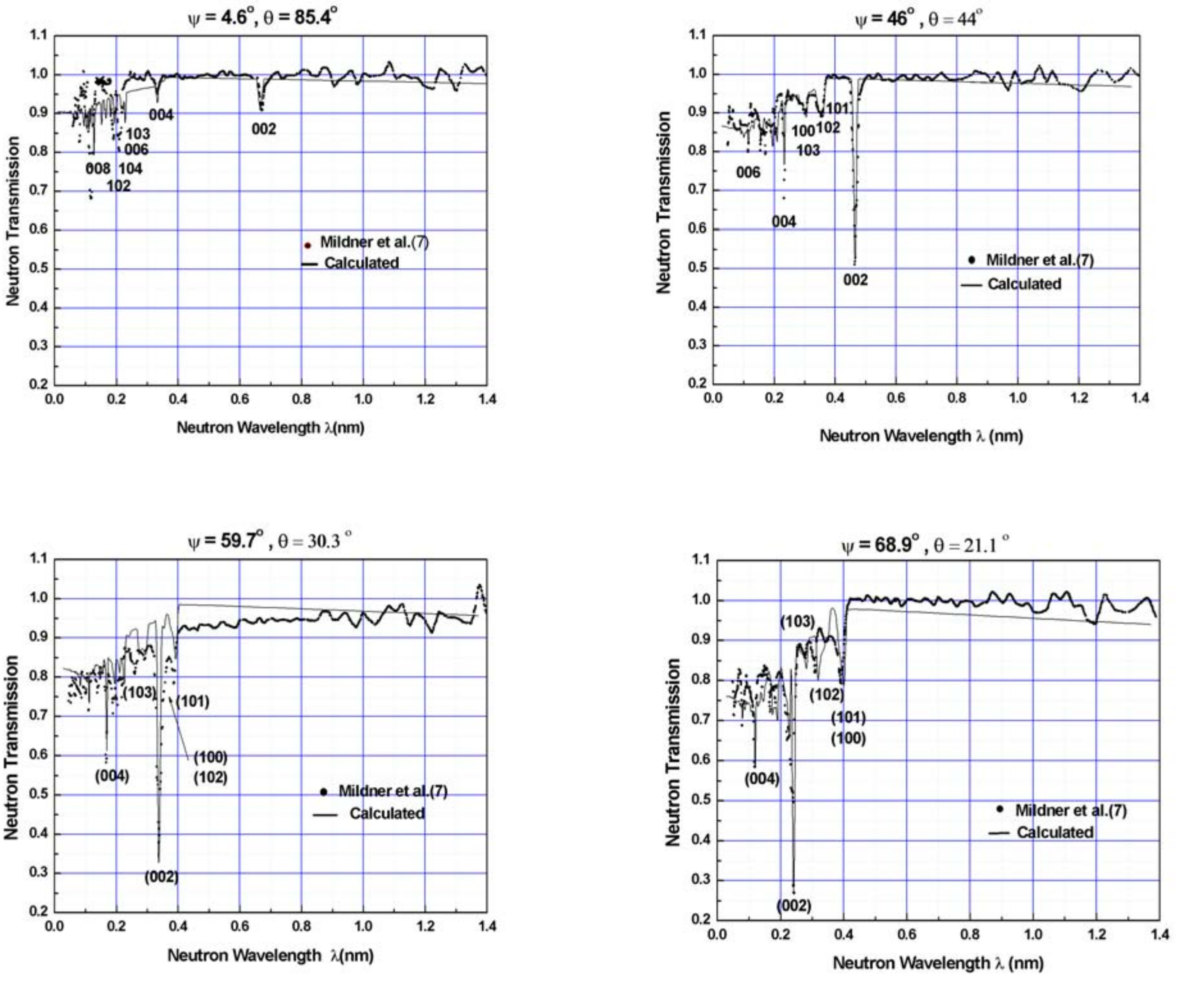


Fig. 1. Transmission results for PG set at different angles.

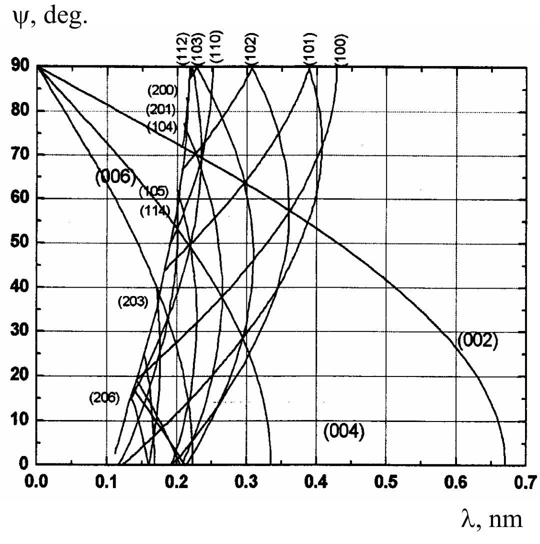


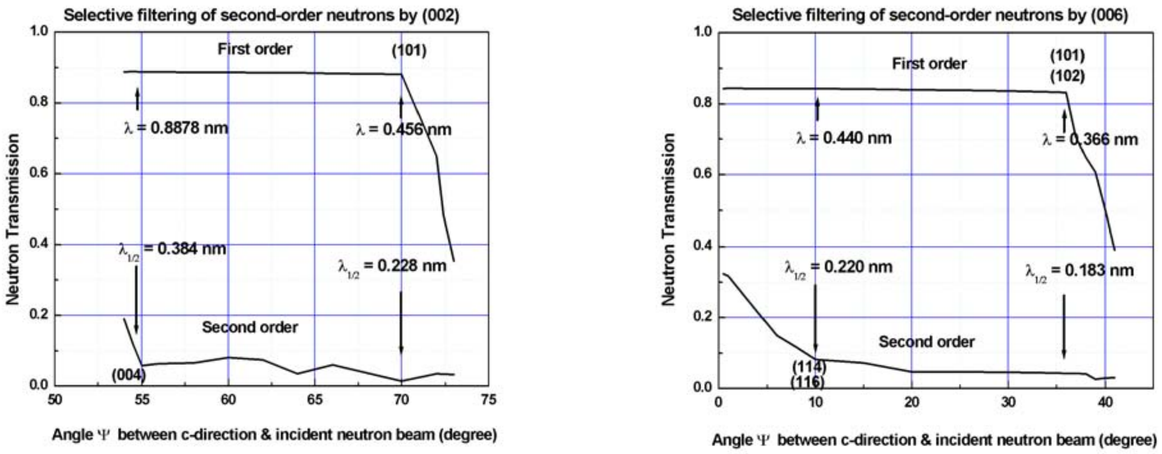
Fig. 2. Tuning diagram for a PG filter.

length at different setting angles ψ between ψ = 50 - 70° within steps of 1°. At each setting the transmission at boundary (002) reflection (i.e. at $\lambda_{1/2}$) is deduced along with the transmission at double (002) boundary (i.e. at λ). The result of these calculations is displayed in Fig. 3, *a* as a function of setting angle ψ. The indication is that such 1 cm thick PG crystal is an efficient second-order filter in the wavelength range (0,228 - 0,384) nm corresponding to setting angles ψ between 70 and 55° respectively.

Similar calculations of neutron transmissions at (006) boundary were carried out at setting angles ψ = 0 - 40°. Where the same parameters for calculations are used as in the previous case except that a 3 cm thick crystal was used. These calculations are displayed in Fig 3, *b*. Fig. 3, *b* shows that 3 cm thick PG crystal is also an efficient second-order filter in the wavelength range (0,183 - 0,228) nm.

The deduced wavelength intervals are narrower than that calculated by Frikkee [6]. Such discrepancy is due to the broadening effect of mosaic spread.

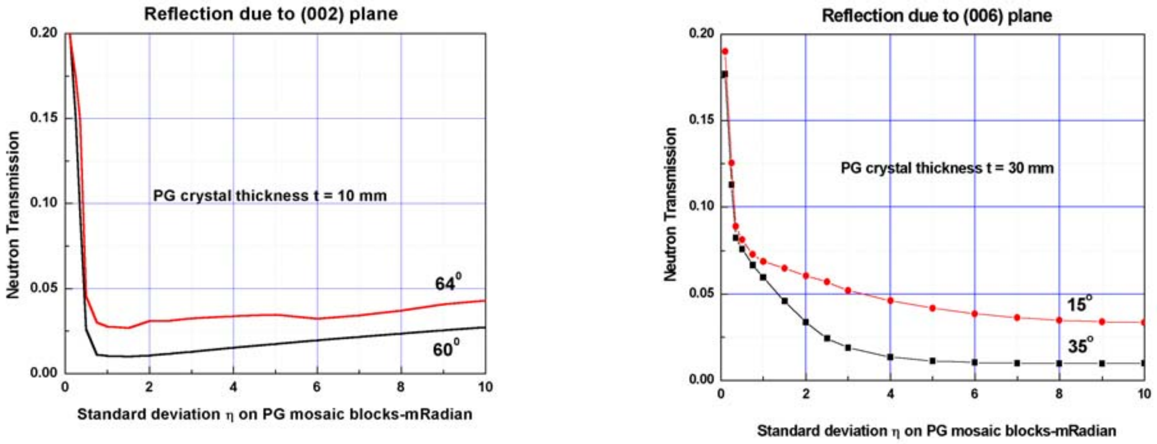
To obtain more wider wavelength interval where the filtering factor T(λ)/T($\lambda_{1/2}$) is constant, a highly



a *b*

Fig. 3. Selective filtering intervals of second-order neutrons.

oriented bulk PG crystals may be needed. On the other hand, the interference from different non-$00l$ planes due to the broadening edges at boundaries may provide that the filtering coefficient is not constant within the wavelength interval. Therefore an optimum choice of the crystal mosaic spread is essential. The neutron transmission at the boundary due to reflection from (002) plane for different values of standard deviation η were calculated. Fig. 4, *a* shows the result of calculation through PG crystal at setting angles ψ = 60 and 64°. As may be observed, the standard deviation $\eta \approx 6$ mradian, the neutron transmission of second-order is less than 2 %. Similar calculations of neutron transmission at boundary due to reflection from (006) were carried out at setting angles 15 and 35°. The result of calculation is displayed in Fig. 4, *b*. From Fig. 4 it seems that PG crystal with FWHM on mosaic spread 0,8° has a constant filtering factor within the whole wavelength interval.

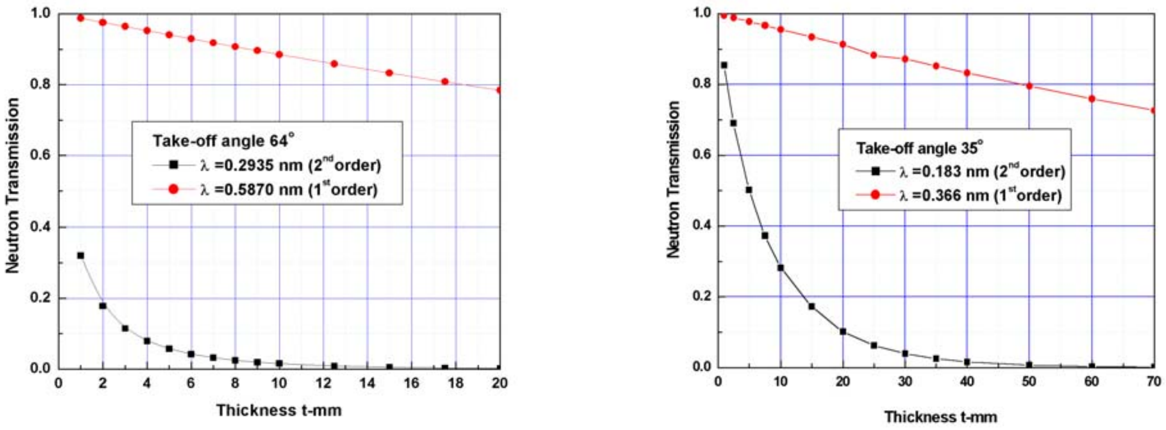


a *b*

Fig. 4. Neutron transmission versus mosaic spread.

To find the optimum PG thickness, the neutron transmission due to reflections from both (002) and (006) planes, were calculated and displayed in Fig. 5, *a* and Fig. 5, *b* respectively.

It would appear that, 1 cm thick PG crystal (0,8° FWHM on mosaic blocks) when it set at angle ψ = = 64° is sufficient for removing neutrons with wavelength $\lambda = 0{,}2935$ nm ($T \le 1{,}5$ %), while providing high transmission ($T \ge 88$ %) for neutrons with wavelength $\lambda = 0{,}587$ nm. Moreover, PG crystal (3 cm thick, 0,8° mosaic) set at angle ψ = = 35° is also sufficient for removing $\lambda_{1/2} = 0{,}183$ nm ($T \le 3$ %),while transmitting more than 87 % of first-order one. Almost the same filtering factors were obtained for neutrons with wavelengths within the interval (0,183 - 0,228) and (0,228 - - 0,384) nm due to reflections from (006) and (002) planes respectively.



a *b*

Fig. 5. Neutron transmission versus PG crystal thickness.

Conclusions

Use has been made of an addive formula determining the attenuation of neutrons by PG crystal, together with the HOPG code, which has been developed and presented in this paper.

Calculation shows that, 3 cm thick PG crystal (0,8° FWHM on mosaic spread) can be efficiently used as a second-order filter for neutrons within wavelengths (0,183 - 0,228) nm by adjusting the crystal in angles ψ from 37 to 8°. While only 1 cm thick PG is also a good second-order filter for neutrons within wavelengths (0,228 - 0,384) nm by adjusting the crystal in angles ψ from 55 to 70°.

More calculations are needed to search for favorable intervals for continuous tuning at shorter than 0,18 nm wavelengths where the PG crystal can be used as high efficient second-order filter. Such intervals are expected at the curves $(11l)^{\pm}$ for even l as predicated by Frikkee [6].

REFERENCES

1. *Brockhouse B.N., Diefendorf R.J.* // The Interaction of Radiation with Solides. - Amsterdam: North Holland Publ. Co., 1964.
2. *Loopstra B.O.* // Nucl. Instrum. and Methods. - 1966. - Vol. 44. - P. 181 - 186
3. *Riste T., Otnes K.* // Nucl. Instrum. and Methods. - 1969. - Vol. 75. - P. 197 - 202.
4. *Shirane G., Minkiewicz H.* // Nucl. Instrum. and Methods. - 1970. - Vol. 89. - P. 109 - 110
5. *Adib M., Ismaail H., Abbas Y. et al.* Attenuation of Thermal Neutron Through Graphite // Proc. 4th Conf. on Nuclear and Particle Physics (Cairo, Egyptian Nuclear Physics Association, 11 - 15 Oct., 2003) / Ed. by Prof. A. Comsan. - Cairo, 2004. - P. 257 - 269.
6. *Frikkee E.* // Nucl. Instr. and Meth. - 1975. - Vol. 125. - P. 307 - 312.
7. *Mildner D.F.R., Arif M., Werner S.A.* // J. Applied Crystallography. - 2001. - Vol. 34. - P. 255 - 262.
8. *Freund A.K.* // Nucl. Inst. Meth. - 1983. - Vol. 213. - P. 1553.
9. *Cassels J.M.* // Prog. Nucl. Phys. - 1950. - Vol. 1. - P. 185.
10. *Bacon G.E.* Neutron Diffraction. / 3rd edition. - Oxford: Claredon Oxford, 1975. - P. 65 - 93.
11. *Naguib K., Adib M.* // J. Physics D: Appl. Phys. - 1996. - 1996. - Vol. 29. - P. 1441 - 1445.

ПІРОЛІТИЧНИЙ ГРАФІТ ЯК СЕЛЕКТИВНИЙ НЕЙТРОННИЙ ФІЛЬТР

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Використовуючи адитивну формулу, було розраховано пропускання нейтронів через кристали піролітичного графіту (ПГ), установлені під різними кутами відносно падаючого пучка. Щоб забезпечити необхідні розрахунки, було створено комп’ютерну програму HOPG. Було отримано повне узгодження між розрахованими пропусканнями нейтронів через пластину ПГ кристала товщиною 1,85 мм та розходженням с-осей 0,4°, установленим під різними кутами до падаючого пучка, та експериментальними даними в діапазоні довжин хвиль від 0,02 до 1,4 нм. Було детально проаналізовано прийнятність використання ПГ кристала як ефективного нейтронного фільтра другого порядку від товщини кристала, розходження с-осей та його орієнтації відносно нейтронного пучка. Показано, що ПГ кристал с розходженням с-осей 0,8° прийнятний для оптимального нейтронного розсіювання другого порядку в діапазоні довжин хвиль (0,384 - 0,183) нм при виборі відповідної орієнтації фільтра.

ПИРОЛИТИЧЕСКИЙ ГРАФИТ КАК СЕЛЕКТИВНЫЙ НЕЙТРОННЫЙ ФИЛЬТР

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Используя аддитивную формулу, было рассчитано пропускание нейтронов через кристаллы пиролитического графита (ПГ), установленные под различными углами относительно падающего пучка. Чтобы обеспечить необходимые расчеты, была создана компьютерная программа HOPG. Было получено полное согласие между расчетными пропусканиями нейтронов через пластину ПГ кристалла толщиной 1,85 мм и расходимостью с-осей 0,4°, установленной под различными углами к падающему пучку, и экспериментальными данными в диапазоне длины волны от (0,02 до 1,4) нм. Была детально проанализирована пригодность использования ПГ кристалла как эффективного нейтронного фильтра второго порядка от толщины кристалла, расходимости с-осей и его ориентации относительно нейтронного пучка. Показано, что ПГ кристалла с расходимостью с-осей 0,8° приемлем для оптимального нейтронного рассеивания второго порядка в диапазоне длин волн (0,384 - 0,183) нм при выборе соответствующей ориентации фильтра.

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