PYROLYTIC GRAPHITE AS A TUNABLE SECOND ORDER NEUTRON FILTER

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A study has been carried out on the neutron transmission through pyrolytic graphite (PG) crystals in order to check its applicability as an efficient tunable second order neutron filter. The neutron transmission have been calculated as a function of neutron wavelengths in the range from 0.01 nm up to 0.7 nm at various PG mosaic spread, thickness and orientation of its c-axis with respect to the beam direction. The computer package GRAPHITE has been used to provide the required calculation.

It was shown that highly aligned (1°FWM on mosaic spread) PG crystal ~2 cm thick, may be tuned for optimum scattering of 2nd order neutrons within some favorable wavelength intervals in the range between 0.112 and 0.425 nm by adjusting the crystal in an appropriate orientation. However, a less quality and thinner PG was found to almost eliminate 2nd order neutrons at only fixed values of wavelength corresponding to the poison of the triple intersection points of the curves (hkl)^2 and (00l).

Keywords: Thermal neutron filters, Pyrolytic graphite crystals

INTRODUCTION

In this respect, the use of polycrystalline materials and pyrolytic graphite (PG) has led to a considerable improvement in neutron diffraction techniques.

It is well known that, the coherent elastic scattering by a crystalline material cannot occur for neutrons with wavelengths which exceed some maximum value of \( \lambda_{\text{max}} \), where \( \lambda_{\text{max}} \) is given by: \( \lambda_{\text{max}} = 2d_{\text{max}} \), and \( d_{\text{max}} \) is the largest \( d_{\text{max}} \) spacing of planes in the crystal. The variation of the scattering cross-section in the vicinity of \( \lambda_{\text{max}} \) for a number of polycrystalline filters, of which Be, BeO, and graphite are perhaps the most commonly used [1]. However, their use are limited specially when the selected neutrons having wavelengths less than 0.4 nm. Moreover, to increase their filtering efficiency, they usually cooled to liquid
nitrogen temperature. Such condition complicates and some time becomes inconvenient to carry out the experimental work.

PG has been in use for about 30 years as a filter. Since in PG, crystallites are preferentially oriented along the hexagonal c-axis. The transmission of neutrons thru PG with c-axis parallel to the beam versus neutron wavelength, exhibits "absorption" lines due to Bragg scattering. By applying PG as second-order filter in neutron powder diffractionmetry, Loopstra [2] and Shapiro [3] demonstrated its high efficiency for first-order neutrons with \( \lambda = 0.26 \text{ nm} \). Recently Adib et al [4] showed that five centimeters thick PG (mosaic 8°) cooled to liquid nitrogen temperature is a high efficiency for transmitting first-order of (4-7 meV) and (10-15 meV) neutrons incident along its c-axis.

However, Frikkee [5] reported an investigation that has been carried out on the neutron transmission through a PG filter as a function of the filter orientation with respect to the beam. It is shown that highly aligned PG may be tuned for optimum scattering of second-order neutrons in the wavelength range between 0.112 nm and 0.425 nm, by adjusting the filter in an appropriate orientation.

The measurement of neutron transmission through highly oriented (0.4° FWHM on mosaic spread) 1.85 mm thick crystal set at different angles to the incident beam, reported by Mildner et.al. [6], was found to justify the existence of the tuned intervals reported by Frikkee [5].

All the mentioned works do not study the effect of crystal mosaic spread value upon the width of the second order wavelength intervals nor the filtering factors within these intervals. Furthermore, the optimum thickness of PG crystal to be used as high efficient second order filter was not also estimated. Therefore the recent calculations of the neutron transmission through PG crystals in terms of mosaic spread, optimum filtering thickness and its orientation with respect to the beam, carried out by Adib et.al. [7,8] are reviewed and discussed in the present work. Since highly oriented PG crystal having few centimeters thick is required to obtain suitable filtering efficiency within the favorable selected intervals. Therefore, in the present work, a feasibility study is also carried on using less oriented and thinner PG crystals to almost eliminate 2nd order neutrons at only fixed values of wavelength corresponding to the poison of the triple intersection points of the curves (hkl)° and (00l)

THEORETICAL TREATMENT

The graphite absorption cross-section due to nuclear capture is very small (\( \approx 3 \text{ mb at } E_n = 0.025 \text{ eV} \)). Therefore the total cross-section determining the attenuation of neutrons is given by the sum:

\[
\sigma = \sigma_{\text{tds}} + \sigma_{\text{Bragg}}
\]  

(1)

Where, \( \sigma_{\text{tds}} \) is the thermal diffuse scattering and \( \sigma_{\text{Bragg}} \) correspond to Bragg scattering cross-section due to reflection from (hkl) planes.

As shown by Freund [9] \( \sigma_{\text{tds}} \) can be split into \( \sigma_{\text{mph}} \) (multiple phonon) and \( \sigma_{\text{sph}} \) (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 $\theta_D$ is the Debye temperature characteristic of the graphite. The second part of TDS is predominant in the range $E \geq K_B T$ where down scattering and multiphonon processes occur. As shown by Freund [9], the predicted empirical equation for $\sigma_{mph}$ fits the experimental results rather well except for graphite.

However, M. Adib [7] showed that best fit of the multi-phonon scattering cross-section term given by Freund [9] in the range $E \gg K_B \theta$ can be replaced by: the static incoherent approximation reported by Cassels [10].

Following Frikkee [5], 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 $\theta_{hkl}$ determined by:

$$\sin \theta_{hkl} = \frac{1}{c} d_{hkl}.$$  \hspace{1cm} (2)

Where $d_{hkl}$ is interplanar distance.

As shown by Frikkee [5] that, 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 $\psi$, and if the mosaic spread is negligible in comparison with $\psi$, the lattice planes $(hkl)$ will scatter neutrons in the following wavelength intervals:

$$2d_{hkl} \sin(\theta_{hkl} - \psi) \leq \lambda \leq 2d_{hkl} \sin(\theta_{hkl} + \psi) \quad \text{for} \theta_{hkl} \geq \psi$$

$$0 \leq \lambda \leq 2d_{hkl} \sin(\theta_{hkl} + \psi) \quad \text{for} \theta_{hkl} \leq \psi$$  \hspace{1cm} (3)

The planes $(00l)$, on the other hand, scatter neutrons with a discrete wavelength $\lambda = 2d_{00l} \cos \psi = 2d_{00l} \sin \theta$ Where $\theta$ is the glancing angle.

The Bragg scattering cross-section due to reflection from $(00l)$ planes of PG is given by k.Naguib & M. Adib [11] as:

$$\sigma_{Bragg}(00l) = -\frac{1}{Nt_o} \ln(1 - P_{00l})$$  \hspace{1cm} (4)

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.

However, it was shown by Frikkee [5] that the scattering cross-section due to non-$00l$ planes reaches pronounced maximum at the boundaries in the $(\lambda; \psi)$ plane given by:

$$\lambda^\pm = 2d_{hkl} \sin \theta_{hkl} \pm \psi$$  \hspace{1cm} (5)

Following, Adib et.al. [7] The Bragg scattering cross-section due to reflection from non-$00l$ planes of a PG crystal with standard deviation $\eta$ on mosaic blocks, and set at angle $\psi$, at wavelength $\lambda$ in the interval between $\lambda^-$ and $\lambda^+$, can be given as

$$\sigma_{Bragg}^{non-00l} = \frac{N_o \lambda^3 F_{hkl}^2 d_{hkl} e^{-2w}}{\sqrt{4d_{hkl} \sin \psi \cos \theta_{hkl} |\lambda - \lambda^\pm_{hkl}|}}$$  \hspace{1cm} (6)

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While at boundaries the Bragg scattering cross-section is decreased due to mosaic spread and can be expressed as

\[ \sigma_{\text{Bragg}}^{\text{non-00l}} = N_0 \frac{\lambda^2 F_{hkl}^2 d_{hkl} e^{-2\eta W(\Delta)}}{\sqrt{4d_{hkl} \sin \psi \cos \theta_{hkl} (\delta \lambda)}} \]  

(7)

where \( W(\Delta) \) is the Gaussian distribution having standard deviation \( \eta \) on graphite mosaic blocks, and \( \delta \lambda \) is the wavelength spread.

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

\[ \sigma_{\text{Bragg}} = \sigma_{\text{Bragg}}^{00l} + \sum_{hkl} \sigma_{\text{Bragg}}^{\text{non-00l}} \]  

(8)

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

A Computer package GRAPHITE has been developed by Adib and Fathalla [12] in order to calculate the total cross section and transmission of neutrons of energy range from 0.1meV to 10eV through crystalline graphite.

**FEATURES OF PG CRYSTALS AS A 2nd ORDER NEUTRON FILTER**

The total effect of the various Bragg reflections, with exception of \((00l)\) reflections, is, that neutrons in the short-wave-length region bounded by the maximum value of \(2d_{hkl}\sin(\theta_{hkl}+\psi)\), will be removed to some extent from the beam. On the other hand, the filter should be transparent for first-order neutrons. This region was found [5] to cover the wave-length interval \(1.12\text{Å} < \lambda/2 < 4.25\text{Å}\).

The possibility to tune a PG 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 \((\lambda, \psi)\) plane given by equation (5).

Hence, one may expect to realize optimum scattering of neutrons by the \((hkl)\) planes at the boundary curves \((hkl)\) in \((\lambda, \psi)\) space defined by equation (5). Possible tuned positions of second-order neutrons are calculated and displayed in Fig.1.

On the basis of structure factors and multiplicities of Bragg reflections the best results may be expected at the curves \((1l)^2\) for even \(l\), \((002)\) and \((006)\). The favorable intervals for continuous tuning reported by Frikkee[5] are listed in Table 1. It may be noticed that selective filtering of second-order neutrons by the \((002)\) reflection fails for \(\lambda/2 < 0.2026\text{ nm}\), because the, first-order neutrons are scattered by the reflections \((100), (101)\).
Figure 1: Tuning diagram for a PG filter.

Table 1: Favorable interval for continuous tuning.

<table>
<thead>
<tr>
<th>Boundary</th>
<th>$\psi$ (deg)</th>
<th>2nd-order $\lambda$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(002)</td>
<td>72.40 - 54.79</td>
<td>0.2026 - 0.3863</td>
</tr>
<tr>
<td>(006)</td>
<td>40.00 - 0</td>
<td>0.1711 - 0.2234</td>
</tr>
<tr>
<td>(112)$^+$</td>
<td>14.59 - 69.88</td>
<td>0.1313 - 0.2305</td>
</tr>
<tr>
<td>(114)$^+$</td>
<td>0 - 53.77</td>
<td>0.1170 - 0.1980</td>
</tr>
<tr>
<td>(114)</td>
<td>1.92 - 0</td>
<td>0.1116 - 0.1170</td>
</tr>
<tr>
<td>(116)$^+$</td>
<td>0 - 32.79</td>
<td>0.1222 - 0.1629</td>
</tr>
<tr>
<td>(116)</td>
<td>4.12 - 0</td>
<td>0.1139 - 0.1222</td>
</tr>
</tbody>
</table>

Moreover a striking systematic feature of Fig (2) is the large number of triple intersection points of the curves $(hkl)^+ (00l)$ and $(00l)$. These intersections occur for the combinations: $(hkl)^{\pm} - (hkl)^{\pm} - (00l \pm l')$ and are independent of the $c/a$ ratio. The most efficient triple intersections as promising second order filter are listed in Table (2) along with those reported by Frikkee[5].
Table 2: Tuned positions at triple boundary crossings.

<table>
<thead>
<tr>
<th>Intersection</th>
<th>$\Psi$ (deg)</th>
<th>2nd order $\lambda$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(100)-(102)$^\gamma$-(002)</td>
<td>57.5</td>
<td>-</td>
</tr>
<tr>
<td>(103)$^\gamma$-(101)-(002)</td>
<td>63.92</td>
<td>64.02</td>
</tr>
<tr>
<td>(110)$^\gamma$-(112)$^\gamma$-(002)</td>
<td>69.88</td>
<td>69.88</td>
</tr>
<tr>
<td>(104)$^\gamma$-(102)-(002)</td>
<td>69.48</td>
<td>-</td>
</tr>
<tr>
<td>(102)$^\gamma$-(104)$^\gamma$-(006)</td>
<td>5.75</td>
<td>-</td>
</tr>
<tr>
<td>(101)$^\gamma$-(105)$^\gamma$-(006)</td>
<td>14.57</td>
<td>14.62</td>
</tr>
<tr>
<td>(100)-(106)$^\gamma$-(006)</td>
<td>27.68</td>
<td>-</td>
</tr>
<tr>
<td>(112)$^\gamma$-(114)$^\gamma$-(006)</td>
<td>33.65</td>
<td>33.65</td>
</tr>
<tr>
<td>(101)$^\gamma$-(109)$^\gamma$-(0010)</td>
<td>1.67</td>
<td>1.70</td>
</tr>
<tr>
<td>(114)$^\gamma$-(116)$^\gamma$-(0010)</td>
<td>5.97</td>
<td>6.07</td>
</tr>
<tr>
<td>(108)-(102)-(0010)</td>
<td>10.95</td>
<td>-</td>
</tr>
</tbody>
</table>

It may be noticed that the triple intersection points of the curves (004) and (008) reflection fail as second order neutron filter, because the, first-order ones are scattered by the reflections (002) and (004) respectively.

Filtering Efficiency of PG within the Favorable Intervals:

The filtering efficiency of PG with various mosaic spread values and thickness were calculated by Adib et al.[7] as a function of wavelength $\lambda$ at different setting angles $\psi$ at boundaries (002) and (006) using the computer package GRAPHITE. Adib [12] showed that, 1 cm thick PG crystal (0.4$^\circ$ mosaic) is an efficient second-order filter in the wavelength range (0.228 – 0.384) nm at boundary (002). While, they found that, 3 cm thick (0.8$^\circ$ mosaic) was also sufficient for removing more than 97% of second-order neutrons within wavelengths (0.183-0.228) nm at boundary (006), while transmitting more than 87% of the first –order one.

The tunable intervals determined by Adib et al. [7], were found to be narrower than that predicted by Frikkee[5] and listed in Table 1. Such difference may be due to the effect of PG mosaic spread on the broadening of reflecting peaks at boundaries. However, more calculations are needed to justify the applicability of the remaining five favorable intervals for continuous tuning of second –order neutrons, listed in Table 1, within which PG can be used as efficient filter especially for neutrons with wavelengths shorter than 0.1711 nm.

To show PG filtering efficiency of 2nd-order neutrons with wavelength shorter than 0.18 nm, the neutron transmission through 1 cm thick PG crystal (1$^\circ$ FWHM on mosaic spread) were calculated by Adib [8] as a function of $\lambda$ from 0.1 up to 0.9 nm with step of $\delta\lambda$=0.004 nm and at different setting angles $\psi$ as given in Table1. At each setting the neutron transmission at boundaries $(11l)^{\gamma}$ (at 2nd order i.e. $\lambda_{1/2}$) is deduced along with that transmission at double $(11l)^{\gamma}$ (i.e. at $\lambda$). The results of these calculations are displayed in Figs.2-4 as a function of setting angles $\psi$ at the boundaries from (112)$^{\gamma}$, (114)$^{\gamma}$ and (116)$^{\gamma}$.
reflections respectively. For comparison the range of setting angles reported by Frikkee [5] are also displayed in Figs.2-4 as dashed lines. From Fig.2 the indication is that, the range of setting angles and consequently, the neutron wavelength band within which the neutron filtering factor $T(\lambda)/T(\lambda')$ is high and constant due to reflection from $(112)^+$, and the band is narrower than that given by Frikkee [5].

![Figure 2: Selective filtering of 2nd neutrons by (112)$^+$](image)

Moreover from Fig. 2 it is seen that selective filtering of 2nd-order by $(112)^+$ reflection fails at setting angle $\psi \approx 49^0$ (i.e. for $\lambda \approx 0.2247 \text{nm}$) because the 1st-order neutrons are scattered by reflection from (002) and also fails at setting angles $\psi \approx 23^0$ (for $\lambda \approx 0.1577 \text{nm}$), since 1st–order one are scattered from (004). Consequently, the setting angular interval at the boundary $(112)^+$ with constant filtering factor is divided to three sub intervals (SI).

![Figure 3: Filtering of 2nd neutrons by (114)$^+$](image)

![Figure 4: Filtering of 2nd neutrons by (116)$^+$](image)

Fig. 3 and Fig. 4 show that the widths of the wavelength intervals of continuous tuning due reflection from $(114)^+$ and $(116)^+$ respectively are also narrower than those given by Frikkee [5]. Moreover each interval is divided by two sub intervals.
To obtain wider wavelength interval, where filtering factor is constant, a highly oriented bulk PG crystals are needed. Such crystals, if available, are very expensive. On the other hand, low quality ones, the interference from different (hkl) planes may provide that filtering factor is not constant within the wavelength interval. Therefore an optimum choice of the crystal mosaic spread is essential to meet the experimental requirements.

The remaining two tuning intervals given in Table 1 at boundaries (114') and (116) are seems to be very narrow and their setting angles are close to $\psi = 0^\circ$. The filtering efficiency at boundaries (114') and at (116) for various setting angles were calculated as for (112)' with step of $\delta \lambda =0.002$ nm and for $0.5^\circ, 1.0^\circ$ and $2.0^\circ$ FWHM on PG mosaic spread. The results are displayed in Fig.5 & 6 for (114)' and (116) boundaries respectively.

One can notice that, the calculated angular setting intervals and consequently the wavelength bands for efficient removing 2nd -order neutrons, are also narrower than those reported by Frikkee [5]. Moreover, their band widths when using PG crystals with 2° mosaic spread are only 0.0035 nm and 0.0045 nm at the boundaries (114) and (116)' respectively. Therefore, the use of PG crystals at these boundaries are limited and restricted to highly oriented PG ones.

As a result, one can conclude that, in order to obtain high attenuation filtering factor of 2nd order neutrons within the selected neutron wavelength bands, one must have a highly oriented PG crystal (~1° FWHM) with thicknesses more than 1cm. Such crystals are expensive to manufacture. Moreover for some experimental facilities, the requirements of such wide tuning wavelength intervals are not needed. Therefore a feasibility study of using PG crystals as efficient 2nd order neutron filter at triple intersection points is worthwhile.

**Filtering Efficiency of PG at Triple Intersection Points**

To show PG filtering efficiency of 2nd -order neutrons at triple points given in Table 2, the neutron transmission through 1 cm thick PG crystal with various mosaic spread values, were calculated by Adib [13] as a function of $\lambda$ with step of $\delta \lambda =0.001$ nm and at different setting angles $\psi$ at both sides from the position of intersection. At each setting angle, the lowest neutron transmission value between the boundaries is deduced. Consequently, the
corresponding neutron wavelength ($\lambda_{\psi}$) at this minimum transmission is determined, along with that transmission at double wavelength (i.e. at $\lambda$). The results of these calculations for the most promising triple points as 2nd order filter virus $\psi$, are displayed in Figs. 7-9 with the boundaries of curves (002), (006) and (0010) respectively.

Fig.(7) shows that, the dip of the transmission curve as a function of setting angle $\psi$ around the boundary crossing position $(103)^{+}-(101)^{-}(002)$ is slightly broadened by increasing the PG mosaic spread value. However, the neutron transmission value at triple boundary crossing is about one order less than that at the boundary of only (002). The same effect is obtained for boundary crossing position $(110)^{+}-(112)^{-}(002)$. Therefore one can select a cheaper PG crystal (4FWHM) 0.5 cm thick to attenuate the 2nd order neutrons by 10 times at these triple crossing poissons, while the transmission of the first order ones is (~95%) higher than at boundary (002).

From Fig.(8), one can notice almost the same behavior of the transmission curve at the triple crossing poissons $(101)^{+}-(105)^{-}(006)$ and $(112)^{+}-(114)^{-}(006)$. At these triple crossing boundaries even more thinner PG crystals may selected.

While Fig.(9) shows that at triple crossings boundaries $(101)^{+}-(109)^{+}(0010)$ and $(114)^{+}-(116)^{+}(0010)$, the selected PG crystals must have not less than 2° on mosaic spread. This is due to the fact that the setting angles at these triple crossing poissons are close to zero setting angles.

As a result, one can conclude that, a thinner and less oriented bulk PG crystals can be used as an efficient 2nd order neutron filter at the fixed wavelengths corresponding to the poissons of the triple intersection boundaries curves $(hkl)^{\pm}$ and (00l).

**CONCLUSIONS**

The computer package GRAPHITE has been successfully applied for the feasibility study on using PG crystals as efficient second order filter. The main optimum parameters and filtering features of PG crystals when used as selective neutron filter within the defined wavelength intervals for continuous tuning are summarized in Table 3.

**Table 3:** Optimum parameters of PG crystal (1°FWHM) as 2nd order filter.

<table>
<thead>
<tr>
<th>Boundary</th>
<th>$\psi$ degree</th>
<th>$\lambda$ 2nd order nm</th>
<th>Thickness cm</th>
<th>Transmission $\lambda$ of 1st order</th>
<th>Attenuation factor of 2nd order</th>
</tr>
</thead>
<tbody>
<tr>
<td>(002)</td>
<td>72.0 – 54.5</td>
<td>0.2070 – 0.3880</td>
<td>0.88</td>
<td>89.9%</td>
<td>10</td>
</tr>
<tr>
<td>(006)</td>
<td>35.0 – 10.0</td>
<td>0.1830 – 0.2200</td>
<td>1.91</td>
<td>91%</td>
<td>10</td>
</tr>
<tr>
<td>(112)$^+$</td>
<td>18.0 - 21.0</td>
<td>0.1424 - 0.1517</td>
<td>2.45</td>
<td>79.9%</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>23.0 - 48.0</td>
<td>0.1577 - 0.214</td>
<td>2.60</td>
<td>87.4%</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>52.0 - 68.0</td>
<td>0.2950 - 0.23</td>
<td>2.32</td>
<td>81.8%</td>
<td>10</td>
</tr>
<tr>
<td>(114)$^+$</td>
<td>1.0 - 16.0</td>
<td>0.1199 - 0.1566</td>
<td>3.07</td>
<td>75.1%</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>20.0 - 51.0</td>
<td>0.1647 - 0.1978</td>
<td>2.31</td>
<td>89%</td>
<td>10</td>
</tr>
<tr>
<td>(116)$^+$</td>
<td>1.0 - 20.0</td>
<td>0.1242 - 0.1529</td>
<td>2.61</td>
<td>76.7%</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>24.0 - 30.0</td>
<td>0.1569 - 0.1614</td>
<td>2.76</td>
<td>83.5%</td>
<td>10</td>
</tr>
<tr>
<td>(114)$^-$</td>
<td>1.50 - 0.0</td>
<td>0.1129 - 0.1171</td>
<td>1.24</td>
<td>80.2%</td>
<td>10</td>
</tr>
<tr>
<td>(116)$^-$</td>
<td>3.25 - 0.0</td>
<td>0.1157 - 0.1222</td>
<td>1.79</td>
<td>74%</td>
<td>10</td>
</tr>
</tbody>
</table>
While the main optimum parameters and filtering features of PG crystals when used as efficient 2\textsuperscript{nd} order at the fixed wavelength values corresponding to poisons of the triple crossing curves (hkl)\textsuperscript{2} and (00l) are listed in Table 4.

REFERENCES