NEUTRON CHARACTERISTICS OF IRON OXIDE SINGLE-CRYSTAL

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Rec. 30/12/2013       Accept. 26/1/2014

A formula is given that permits the calculation of both neutron reflectivity and attenuation of Iron Oxide (FeO) single-crystals as a function of wavelength at both room and LN temperatures. A computer program FeO written in FORTRAN-77, has been developed to carry out the required calculations. The monochromatic and filtering characteristics of FeO single-crystals are detailed in terms of crystal cutting plane, mosaic spread, thickness and reactor moderating temperature within the wavelength band from 0.001nm up to 1.4nm. Calculation shows that, 10mm thick FeO single-crystal cut along its (200) plane having 0.4° FWHM on mosaic spread are the optimum parameters when, it used as a monochromator from a thermal reactor flux at neutron wavelengths shorter than 0.15nm. While, from a cold reactor flux, it is also appreciated as a neutron monochromator, free from higher order contaminations at wavelengths from 0.3 nm up to 0.42 nm. Calculation also shows that 50 mm thick FeO-single crystal cut along its (200) plane, with 0.01° FWHM on mosaic spread, is a good thermal neutron filter, with high effect-to-noise ratio.

Keywords: Iron oxide single–crystal, Thermal neutron monochromator and filter.

INTRODUCTION

A monochromator is a single-crystal that selects the neutron wavelength according to Bragg's law \( n\lambda = 2d\sin\theta \). The range of wavelengths accepted depends on the crystal structure and its mosaicity. A larger mosaicity increases the number of monochromatic neutrons that will make it to the sample, while reduces its wavelength resolution [1].

Common materials used as monochromator crystals are pyrolytic Graphite [2], Silicon, Copper, Beryllium and Iron, crystals. The choice of monochromator depends on the range of incident neutron wavelength required for the experiment and the
desired wavelength resolution. However, a beam of monochromatic neutrons, selected from the spectrum of a nuclear reactor by means of diffraction by a single crystal, will in general contaminate with higher-order components. Consequently the use of a filter is indispensable to eliminate higher order ones [3]. Therefore, silicon (111) reflection comes with the added advantage that second order (n=2) neutrons are forbidden [4]. However, Iron oxide seems to be preferable as a neutron monochromator, since its coherent scattering length is longer than for silicon [5].

The use of long-wavelength neutrons at reactor and pulsed sources often requires a filter for the removal of unwanted epithermal neutrons. Single–crystal filters are often used to transmit the thermal neutron beam with little epithermal and fast neutron contaminations. The filter also acts to reduce the intensity of γ-ray beam which accompanies the neutron beam, for example, from a conventional fission reactor. The use of large, perfect single-crystals of various materials as filters for thermal neutron beams has long been known [6]. Several materials such as quartz (SiO₂) [7], bismuth [8], silicon [9], lead [10], sapphire (Al₂O₃) [11, 12, 13], Iron [14], magnesium oxide (MgO) [15, 16], magnesium fluoride (MgF₂) [17] have been suggested as most successful filter materials. At high neutron energies, greater than about 1eV, the total neutron cross section σ 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. In this respect, MgF₂ cooled to liquid –nitrogen temperature to reduce the multi-phonon scattering seems to be an excellent choice. Iron would seem to be the worst, since it has the biggest absorption cross-section while it has the highest value of σₜₐₜ. However it is, in some cases, the most promising than the others especially when thermal reactor neutrons are accompanying with high intensity of γ-rays. Moreover Iron oxide more preferred than iron, since its TDS is lower and chemically more stable than the later.

Therefore, the present work concerns a detailed study for the use of FeO single-crystals as a thermal neutron monochromator and/or filter in terms of crystal orientation, mosaic spread, cutting plane, thickness and temperature.

THEORETICAL TREATMENT

The total cross-section determining the attenuation of neutrons by single-crystal is given by:

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

(1)
where $\sigma_{abs}$ is the absorption cross-section due to nuclear capture processes, $\sigma_{tds}$ is the thermal diffuse scattering (TDS) or inelastic scattering cross-section and $\sigma_{Bragg}$ corresponds to elastic or Bragg scattering. The contribution of Bragg scattering $\sigma_{Bragg}$ 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 Naguib and Adib [18] as:

$$\sigma_{Bragg} = \frac{1}{N t_o} \ln \left( \frac{1}{\prod_{hkl} (1 - P_{hkl}^\theta)} \right)$$

(2)

where $N$ is the atom number density, $t_o$ is the thickness of the crystal in the beam direction, and $P_{hkl}^\theta$ is the reflecting power of the $(hkl)$ plane inclined by an angle $\theta_{hkl}$ to the incident beam direction.

In the case of imperfect crystal of finite absorption the reflecting power $P_{hkl}^\theta$ of the $(hkl)$ plane inclined by an angle $\theta_{hkl}$ to the incident beam direction is given by Bacon [19] as:

$$P_{hkl}^\theta d\theta = \frac{a d\theta}{1 + a + (1 + 2a)^{1/2}} \coth[A(1 + 2a)^{1/2}]$$

(3)

where

$$a = \frac{Q_{hkl}}{\mu} W(\Delta)$$

(4)

In which $\mu$ is the linear absorption coefficient, $t_o$ is the crystal thickness and $\gamma_{hkl}$ the direction cosine of the diffracted beam. $Q_{hkl}$ is the well-known crystal-graphic quantity defined by Bacon [19]:

$$Q_{hkl} = \lambda^2 N_c^2 F_{hkl}^2 / \sin 2\theta_{hkl}$$

(5)

where, $F_{hkl}$ is the amplitude of the diffracted neutron beam for the $(hkl)$ reflection, $N_c$ is the number of unit cells per unit volume. $W(\Delta)$ has a Gaussian form with standard deviation $\eta$ of mosaic blocks of single crystal, $\Delta = \theta - \theta_{hkl}$ where $\theta$ is the glancing angle between the incident beam and the planes whose reflectivity is being considered and $\theta_{hkl}$ is the Bragg angle for these planes, and is written as:

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

(6)
As shown by Bacon [19], the integrated reflectively $R^\theta = \int_{-\infty}^{+\infty} P_{\theta}^d d\theta$, from imperfect crystal of finite absorption researches saturation for bulk crystal thickness of $t_o$. Such behavior is due to the multiple internal reflections inside the filter. Therefore, when single crystal is used as a neutron filter in transmission geometry, to take into account the effect of multi-internal reflections within the crystal thickness $t_o$, Eq. (3) is multiplied by a factor $\chi = t_o/t_s$ and $t_o$ in Eq (4) is replaced by $t_s$.

As shown by Naguib and Adib (20), the reflecting power $P_{\theta}^d$ for an imperfect single crystal depends upon the direction cosine of the diffracted beam $\gamma_{hkl}$.

$$\gamma_{hkl} = \frac{(h\beta \cdot k\alpha + l\gamma_{hkl}) \cos \psi + l\gamma_{hkl} - \frac{l\gamma_{hkl}}{h\beta + k\alpha}}{\sqrt{h^2 + k^2 + l^2}} \sin \psi$$

The reflected intensity $I_{Ref}$ from single-crystal when a neutron beam having Maxwellain distribution $\Phi(\lambda)$ is given by:

$$I_{Ref} = \Phi(\lambda) * P_{\theta}^d,$$

where $\Phi(\lambda)$ having neutron gas temperature $T$ is given by Gurevich, & Tarasov [21]:

$$\phi(\lambda) = \frac{\text{constant}}{\lambda^5} \exp(-h^2 / 2mkT\lambda^2)$$

While, for epithermal and higher neutron energies than 1eV, the neutron flux $\Phi(\lambda)$ is given by

$$\phi(\lambda) = constd\lambda / \lambda.$$

The software computer code MgO [16] has been used to calculate the neutron total cross-section and transmission of FeO single-crystals in energy range from 0.1meV to 10 eV (i.e. neutron wavelengths from 1.4 nm to 0.001 nm).

**RESULTS AND DISCUSSION**

1. **Monochromatic features of FeO single-crystals:**

The main FeO physical parameters used for calculations are listed in Table 1. The main parameters determining the quality of FeO crystal as a neutron monochromator are: reflecting power, wavelength spread of the reflected beam and the ratio of high order contaminations to the first one.
Table 1. FeO Physical parameters.

<table>
<thead>
<tr>
<th>Physical property</th>
<th>FeO (wüstite)</th>
</tr>
</thead>
<tbody>
<tr>
<td>System and space group</td>
<td>(F.C.C.) Fm3m</td>
</tr>
<tr>
<td>Molecular weight</td>
<td>40.300</td>
</tr>
<tr>
<td>Density (gm/cm$^3$)</td>
<td>5.745</td>
</tr>
<tr>
<td>Lattice parameters (nm)</td>
<td>$a_0 = 0.4302$</td>
</tr>
<tr>
<td>No. of molecules / unit cell</td>
<td>4</td>
</tr>
<tr>
<td>No. of unit cells $N_c/m^3$</td>
<td>1.20435E+28</td>
</tr>
<tr>
<td>Atomic positions</td>
<td>Fe: (0,0,0);(0,0.5,0.5);(0.5,0,0.5);(0.5,0.5,0)</td>
</tr>
<tr>
<td></td>
<td>O: (0.5,0.5,0.5);(0,0,0.5);(0,0.5,0);(0.5,0,0)</td>
</tr>
<tr>
<td>Coherent scattering lengths b (m)</td>
<td>Fe = 0.945E-14</td>
</tr>
<tr>
<td></td>
<td>O = 0.5803E-14</td>
</tr>
<tr>
<td>Total scattering cross section</td>
<td>15.852</td>
</tr>
<tr>
<td>Debye temperature $\Theta_D (K)$</td>
<td>430</td>
</tr>
<tr>
<td>Absorption cross section at 0.025eV (barns)</td>
<td>0.407</td>
</tr>
</tbody>
</table>

From the Eq.(3), the highest reflectivity $P_{hkl}^{\theta}$ and the widest band of monochromatic wavelengths is expected from the surface of a single crystal cut along $(h,k,l)$ at glancing angle $\theta_{hkl}$, whose inter-planer distance $d_{hkl}$ is the longest and its structure factor $F^2_{hkl}$ is the highest. Therefore, the most promising cutting planes for FeO single crystals as a neutron monochromator are (111), (200), (220) and (311). The distribution of the reflected neutrons $P_{hkl}^{\theta}$ at $\lambda_c = 0.114\mu m$ from FeO single-crystal as a function of mosaic spread were calculated assuming the following input parameters listed in Table 2.

Table 2. Input parameters for reflecting power $P_{hkl}^{\theta}$ for constant $d\theta$.

<table>
<thead>
<tr>
<th>Cutting plane $(h,k,l)$</th>
<th>$d_{hkl}$ (nm)</th>
<th>$F_{hkl}$</th>
<th>Glancing angle $\theta$°</th>
<th>Thickness $t_s$ (nm)</th>
<th>$\lambda_{\min}$ (nm)</th>
<th>$\lambda_{\max}$ (nm)</th>
<th>$\Delta\lambda$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>111</td>
<td>0.24866</td>
<td>0.2127E-27</td>
<td>13.27</td>
<td>2.00</td>
<td>0.1E-2</td>
<td>0.3E-2</td>
<td>0.73E-3</td>
</tr>
<tr>
<td>200</td>
<td>0.21535</td>
<td>0.3726E-26</td>
<td>15.73</td>
<td>2.31</td>
<td>0.1E-2</td>
<td>0.3E-2</td>
<td>0.63E-3</td>
</tr>
<tr>
<td>220</td>
<td>0.15227</td>
<td>0.3726E-26</td>
<td>22.00</td>
<td>3.27</td>
<td>0.1E-2</td>
<td>0.3E-2</td>
<td>0.44E-3</td>
</tr>
<tr>
<td>311</td>
<td>0.12986</td>
<td>0.2127E-27</td>
<td>26.07</td>
<td>3.83</td>
<td>0.1E-2</td>
<td>0.3E-2</td>
<td>0.35E-3</td>
</tr>
</tbody>
</table>

Table (2) shows that the structure factor of the (odd, odd, odd) planes are 20 times less than that of (even, even, even) ones. Therefore FeO single crystal cutting along (200) and (220) planes satisfies these requirements among the other cutting ones. However at
long monochromatic neutron wavelengths, FeO (111) may be preferred than the others since the $d_{hkl}$ is the longest among the others.

The calculated reflecting power from cutting planes (111), (200) & (220) are displayed in Fig.1-a, b&c respectively. Figure 1 shows that the values of the reflective power from (200) and (220) are more than that of (111). Such result is in agreement with the fact that structure factor of the even-even-even plane ($0.3723E-26m^2$) is higher than that of the odd-odd-odd one ($0.2127E-27 m^2$). Figure 1 also shows that the broadening of the reflecting power distribution is decreasing with the decrease of $d_{hkl}$.

Such result is in agreement with the fact that at the same selected monochromatic wavelength the glancing angle $\theta_{hkl}$ is increasing with the decrease of $d_{hkl}$.

**Figure1.** The wavelength distribution of $P_{hkl}$ from FeO at various mosaics spread- $\eta$.

The wavelength resolution $\Delta \lambda/\lambda$ as a function of mosaic spread was determined as a ratio of the FWHM of the distribution $P_{hkl}$ to the wavelength $\lambda_{hkl}$. The values of peak reflecting power and $\Delta \lambda/\lambda$ versus $\eta$ are displayed in Figure 2. Figure 2 shows that $\Delta \lambda/\lambda$ is increasing with the increase of the mosaic spread $\eta$. This is in consistent of the
fact that low quality crystal (i.e., high value of $\eta$) the distribution of the reflected neutrons is broadened consequently the resolution becomes worst.

Figure 2 shows also that $P_{\theta hkl}^0$ reaches a maximum value at FWHM on mosaic spread $\eta = 0.1^\circ$ for cutting plane (111) (Fig.2a) and $\eta = 0.3^\circ$ for cutting planes (200) (Fig.2b) & (220) (Fig.2c). Such behavior may be due to the fact that the assumed incident neutron beam divergence (0.3o) is comparable with the mosaic spread value. However, FeO (200) with $\eta = 0.3^\circ$ (0.3o) FWHM on mosaic spread $\eta$ is in some cases may be more preferred than (111) cutting plane when it used as monochromator since the resolution of the reflected beam is better than the later.

The integrated reflecting power $R^\theta$ was calculated versus effective thickness $t_{eff} = t_o / \cos \theta$. The calculated $R^\theta(\eta)$ of 1st order reflection from single-crystal FeO ($\eta = 0.4^\circ$) for cutting plane (111) along with the higher order contaminations accompanying the 1st-one is displayed in Fig.3a while for cutting planes (200) & (220) crystal in Fig.3b&c, respectively.

![Figure 2](image)

**Figure 2.** Maximum $P_{\theta hkl}^0$ and $\Delta \lambda / \lambda, \%$ versus mosaic spread $\eta$. 
From Fig. 3 one can notice that FeO crystal with thickness from 7 to 10 mm is a suitable thickness for FeO crystal when it is used as a neutron monochromator. The choice depends upon the experimental requirements, its price and the ratio of the accompanied higher order reflections to the 1st one. Fig. 3 also shows that the integrated reflectively from FeO (111) is much less than from FeO (200) and (220). Moreover the second order contamination is about 50% of the first order one. Thus FeO(200) and FeO (220) single crystals are the better choice, however one must take into account the value of effect-to-background ratio when a thin FeO with small integrated reflections of 1st order is selected.

These contaminations are true when the incident neutron beam distribution is constant. However the incident neutron beam distribution from steady state reactor obeys Maxiwellain one with neutron gas temperature T. Therefore the reflected neutron intensities from FeO cut along (200) and (220) were calculated at same selected reflected monochromatic neutrons of 0.114nm, 0.15 nm from thermal reactor flux and displayed in Fig.4a&b respectively. While, Fig.4c displays the reflected wavelength distribution of monochromatic neutrons from FeO cut along (200) at 0.35 nm from cold reactor flux.
Figure 4. The reflected neutron wavelength distribution from FeO single-crystal.

Since, from Fig. (4) the reflected neutron monochromatic intensities $I_{\text{Re}f}$ and its higher orders contaminations from FeO crystal was found to depend upon both the reactor moderating temperature and the value of the selected monochromatic wavelength, therefore the integrated intensity of monochromatic neutrons $\sum I_{\text{Re}f}(n)$ from 3 mm thick FeO crystal and the accompanying higher orders were calculated as a function of the selected neutron wavelength. The result of calculation is displayed in Fig.5a&b for cutting planes (200) and (220) respectively assuming that the neutron beam is incident from thermal reactor, while Fig.6a&b for cold reactor one.
Fig. 5. The integrated intensity of monochromatic neutrons and its higher orders from thermal reactor flux versus wavelength $\lambda$.

Fig. 6. The integrated intensity of monochromatic neutrons and its higher orders from cold reactor flux versus wavelength $\lambda$.

Fig. 5 shows that FeO (200) is the best choice as a neutron monochromator without need for a filter for selected monochromatic neutrons from a thermal reactor flux up to $\lambda \leq 0.15$. While, from Fig. 6 one can see, that if a cold reactor is available the use of the single crystal FeO (200) as a neutron monochromator free from higher order contaminations up to $\lambda = 0.42$ nm is also appreciated. While FeO (220) can be used as neutron monochromator only up to $\lambda = 0.3$ nm.

2. Filtering features of FeO single-crystals.

Since the required thickness of FeO when it used as a neutron filter, must be thick enough to remove the accompanying epithermal neutrons and gamma rays, therefore following Adib et.al [16], the filter thickness $t_\omega$ is assumed to consist of multi-layers of thickness $t_s = 50 \mu m$, where $t_s$ is much less than $t_\omega$. 
To show the contribution of Bragg reflections on neutron transmitted through a 20 mm FeO mono-crystal, cutting along different \((h,k,l)\) planes, calculations were performed using the FeO code, assuming room temperature and crystals which have the same FWHM on mosaic spread of 0.5°. Fig. (7) a,b,c &d displays the result of calculations for FeO crystals cut along the (111),(200), (220) and (311) planes at \(\psi = 0°\) respectively.

It is apparent that, when used as a thermal-neutron filter, FeO crystal cut along the (200) plane is preferable to other cuts since no Bragg reflections occur for neutron energies < 2meV and is a slight improvement on the (111) cut, due to the strong reflections from (220) and (311) planes in the latter.

Figure 7. Neutron transmission through FeO single-crystal, cut along different planes.
To decrease Bragg reflections occurring at higher energies an optimum choice of the crystal mosaic spread is essential. Neutron transmission through a 20mm FeO (200) crystal for different values of FWHM on mosaic spread were calculated and displayed in Fig.8. As may observed, for FWHM on mosaic spread > 0.5° parasitic Bragg reflections could limit the use of FeO as a thermal neutron filter.

To find the optimum FeO thicknesses, the neutron transmission through different crystal thickness, were calculated. Fig.9 shows the result of calculation through FeO (200) crystal having FWHM on mosaic spread of 0.01° at 293 K Fig. 9a, while Fig.9b at 77K.
Figure 9. Neutron transmission through FeO - cut along (200) plane, for different crystal thickness.

It would appear that 50mm thick FeO cooled at liquid nitrogen is sufficient for removing neutrons with energies > 0.5 eV ($T_n<5\%$) while providing high transmission ($T_n>85\%$) for neutrons with energies < 0.02 eV. The transmitted neutron spectrum is almost free from disturbing Bragg reflections. The reflecting characteristics are less good for crystals having a mosaic spread of 0.01°. The final choice depends upon the experimental conditions required and the price of such crystals.

To show the filtering features of FeO, the incident neutron flux having Maxwellian distribution, with neutron gas temperature close to 300 K along with its transmission through FeO crystals at both 77 K and 293 K, were calculated with constant steps of $\Delta \lambda = 0.001 \text{ nm}$ using the computer program. The result of calculation is displayed in Fig. 10.

Figure 10. Thermal-neutron flux transmitted through 10cm FeO (200) crystal.

Fig.10 shows that FeO (200) crystals can be successfully used to transmit a thermal reactor flux having a Maxwellian distribution, with neutron gas temperature close to 293K, while significantly rejecting the accompanying slowing down flux ($dE/E$), with neutron energies $E>1$ eV. While, if it is necessary to install a cryogenic
FeO filter on a beam line in order to obtain lower attenuation at longer wavelengths it is worth considering and is readily available in large single-crystal blocks.

CONCLUSION

Use has been made of the simple additive formula determining the reflectivity and attenuation of neutrons by a single crystal, together with the FeO code, which have been developed and presented in this paper. This permits calculation of the reflecting power and the total neutron cross-section of mono-FeO when it used as a neutron monochromator and/or filter.

Calculation shows that, 7-10 mm thick of FeO (200) crystal having 0.5° FWHM on mosaic spread are the optimum parameters when, it used as a monochromator at selected neutron wavelengths shorter than 0.15nm. However, the integrated neutron intensity of 2nd and 3rd orders from a thermal reactor flux is even higher than that of the 1st order one at neutron wavelengths longer than 0.2 nm. While, from a cold reactor flux, the use of FeO (200) single- crystals as a neutron monochromator free from higher order contaminations up to λ= 0.42 nm are more appreciated. Calculation also shows that 50 mm thick FeO-single crystal cut along its (200) plane cooled at 77K, with 0.5° FWHM on mosaic spread, is a good thermal neutron filter, with high effect-to-noise ratio.

REFERENCE


**NEUTRON CHARACTERISTICS OF IRON OXIDE...**

[الخصائص النيوترونية لبللورة أكسيد الحديد الأحادية

مرفط المسيرى

قسم طبيعة المفاعلات - مركز البحوث النووية - هيئة الطاقة النووية

تم وضع معادلة لحساب انكساسة النيوترونات والتوهين من بللورة أكسيد الحديد الأحادية في علاقة مع الطول الموجي عند درجة حرارة الغرفة ودرجة النيتروجين السائل. تم تصميم برنامج مكتوب بلغة الفورتران- 77 لإجراء الحسابات. الحسابات أظهرت أن سمك 10 مليمتر من بللورة أكسيد الحديد الأحادية مقطوعة على المستوى (002) وتوثيق توزيع سيفيساني 0.4 درجة أمثل البارامترات للبللورة كموجود للنيوترونات الحرارية عند طول موجي أقل من 0.15 نانومتر بينما عند طول موجي من 0.3 إلى 0.42 للنيوترونات الباردة.

الحسابات أظهرت أيضا أن سمك 50 مليمتر من بللورة أكسيد الحديد الأحادية مقطوعة على المستوى (002) وتوثيق سيفيساني 0.01 درجة مرجع جيد للنيوترونات الحرارية للحصول على نيوترونات أحادية الطول الموجي بكفاءة عالية.**