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SINGLE CRYSTAL FILTERS FOR NEUTRON SPECTROMETRY

N. Habib

Reactor Physics Department, NRC, EAEA, 13759 Cairo, Egypt.
Email: nadiahabib15@yahoo.com

Abstract

A study of neutron transmission properties trough a large single crystals specimens of Si, Ge, Pb, Bi and sapphire at 300 K and 80 K have been made for a wide range of neutron energies.

The effectiveness of such filters is given by the ratio of the total cross-section of unwanted epithermal neutrons to that the desired thermal neutron beam and by the optimum choice of the crystal orientation, its mosaic spread, thickness and temperature.Our study indicates that sapphire is significantly more effective than the others for a wide range of neutron energies.

1. INTRODUCTION

The gamma radiation, fast and the thermal neutrons are all associated with fission reactions. However, to improve the effect-to-noise ratio for neutron scattering experiments, the development of thermal-neutron filters are required (Crawford et al., 1995).

As shown by several authors (Korneev et al., 1991; Ibel, 1994; Kim et al., 1993; Munter et al., 1996; Funer, 1996), curved guide tubes transport thermal neutrons by total internal reflection from the mirror surfaces coated with ^{58}Ni. Therefore, such neutron guide tubes are now-a-days used as thermal neutron filters. However, they are expensive to construct, therefore their use is limited

At high neutron energies, greater than about 1 eV, the total neutron cross-section σ_t of crystalline thermal neutron filter material is of the range of a few barns (1 bn =10^{-28} m^2), but at lower thermal energies, less than 0.1 eV, where much of the coherent "Bragg" scattering is disallowed, the effective cross-section for the single crystal specimens is much reduced. This effective cross-section is the sum of several contributions: (a) absorption, σ_a , normally proportional to neutron wavelength, (b) incoherent, σ_{inc} , (c) coherent inelastic, σ_{inel} and (d) any residual Bragg scattering, σ_{el}. Item (d) is highly dependent on neutron wavelength, crystal orientation and crystal perfection and item (c) depends upon crystal temperature (phonon population) and may be substantially reduced by cooling the filter. In most cases there is little advantage in cooling the above materials below liquid nitrogen temperature. The efficiency of a filter is thus determined by the ratio

$$R = \left(\sigma_a + \sigma_{inc} + \sigma_{inel}\right)/\sigma_t \tag{1}$$

The lower the value of R, the better the filter; a low value for the numerator implies good transmission of neutrons in the desired thermal energy range, while a large denominator ensures strong scattering of the unwanted epithermal and fast neutrons. The filter also acts to reduce the intensity of any γ -ray beam which accompanies the neutron beam, for example, from a conventional fission reactor.

In Table (1) we list the relevant cross-sections from which estimates of the values of R may be obtained. The values of σ_a refer to 1.8 $\overset{o}{\text{A}}$ (0.025 eV) neutrons: σ_{inc} values are taken to be independent of neutron wavelength; each σ_t is an average value for energies between 1 and 10^4 eV. All these cross-sections are reasonably well-known quantities, and are independent of the orientation, perfection and temperature of the filter crystal. The relationships between the well-known bound coherent cross-section, σ_{coh} , in Table (1), and the effective inelastic and elastic cross-sections, σ_{inel} and σ_{el} , in Eq. (1), are much more complex, however, and may not be reliably calculated. Measurements of the filter transmission are therefore required for particular single crystal specimens of each material, to determine realistic estimates of filter efficiencies. The best possible efficiencies will be obtained if the contributions σ_{el} and σ_{inel} are substantially reduced, the former by suitable orientation of highly perfect crystals and the latter by cooling to low temperatures to minimize the population of phonons, particularly those of low frequency. The absolute minimum value of R, assuming all coherent scattering processes may be eliminated, is given by the ratio

$$R_{\min} = \left(\sigma_a + \sigma_{inc}\right)/\sigma_t \tag{2}$$

Table (1) Relevant cross-sections of several elements suitable for thermal neutron filters. All cross-sections, except where otherwise noted, are taken from "Neutron Cross-Sections" Brookhaven National Laboratory report. BNL-325, third edditio (eds. Mughabghab and Garber), 1973. The absorption cross-sections σ_a refer to 0.025 eV neutrons, whereas the total cross-sections σ_t are average values for neutrons in the energy range 1 to 10^{-4} eV.

Cross-sections (barn; 1 bn = 10^{-24} cm^2)				
Element	**Absorption** σ_a	**Incoherent** σ_{inc}	**Total** σ_t	σ_{coh}
Al	**0.230 ± 0.003**	**0.10 ± 0.03**	**1.43 ± 0.04**	**1.496 ± 0.008**
Be	**0.0092 ± 0.001**	**0.04 ± 0.01**	**6.15 ± 0.02**	**7.53 ± 0.07**
Bi	**0.033 ± 0.004**	**0.02 ± 0.02**	**9.28 ± 0.04**	**9.35 ± 0.04**
C	**0.0034 ± 0.0002**	**0.005 ± 0.01**	**4.75 ± 0.02**	**5.569 ± 0.005**
Mg	**0.063 ± 0.003**	**0.12 ± 0.01**	**3.42 ± 0.04**	**3.57 ± 0.28**
O	**0.00027**	**0.01 ± 0.01**	**3.76 ± 0.02**	**4.23 ± 0.01**
Si	**0.16 ± 0.03**	**0.02 ± 0.02**	**2.25 ± 0.06**	**2.163 ± 0.001**

Table (2) lists $R_{\min}$ for several possible filter materials. All the very best candidates, such as diamond, graphite, Bi, Be, and simple compounds such as BeO, suffer from a major problem: the availability of large, highly perfect single crystals at a reasonable cost. In addition, there is in bismuth a high density of low frequency phonon states, leading to undesirable high values of σ_{inel} . Magnesium oxide crystals of several cm^3 sizes are available, but we have so far been unable to obtain large enough samples for use as filters. (The filter described by

(Holmryd and Connor, 1969) was a composite of many small pieces.) Nevertheless, MgO is indeed a very promising candidate, should large crystals become available.

Table (2) Minimum values of the parameter R for several possible filter materials, assuming that all coherent scattering processes may be eliminated.

Material	$R_{\min} = (\sigma_a + \sigma_{inc})/\sigma_t$
C	**0.0018**
Bi	**0.0057**
BeO	**0.0060**
Be	**0.0080**
SiO_2	**0.0205**
MgO	**0.0269**
Al_2O_3	**0.0488**
Si	**0.080**

The considerations thus lead us to consider in more detail the remaining three materials, quartz, sapphire and silicon. From Table (2), quartz would seem to be the most promising, with silicon the least. Experimentally, however, these two materials are quite comparable; at 80 K, their R values are the same to within 20% over much of the thermal energy range. We have made neutron transmission measurements of several specimens of the above materials. The most important results, for 2 specimens of sapphire, indicate that this material can be significantly more effective than Si or SiO_2 (at least twice as good at 300 K). It is also readily available in large sizes, up to 300 mm diameter. However, our results also show that it is quite difficult to eliminate unwanted Bragg scattering processes (σ_{el}) for all neutron energies simultaneously.

2. MONOCRYSTALS AS THERMAL NEUTRON FILTERS

The use of large, perfect single-crystals of various materials as filters for thermal neutron beams has been long known (Iyengar, 1965) Several materials such as quartz (SiO_2) (Harvey, et al.,1982), bismuth (Adib, et al.,2003), silicon (Brugger, 1976), germanium (Vertibni, et al., 1974), lead (Adib, et al. 2002) and sapphire (Al_2O_3) (Nieman, et al.,1980),

However, (Mildner and Crawford, 1993), (Mildner and Lamaze, 1998), (Mook and Hamilton, 2001), have been suggested sapphire single crystals as most successful filter materials. At high neutron energies, greater than about 1eV, the total neutron cross section σ_t of sapphire is in the range of a few barns, but at lower thermal energies, less than 0.1 eV, where much of the coherent Bragg scattering is disallowed the effective cross-section for single-crystal specimens 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, 1983) reported a semi empirical formula with two fitting parameters, which permits calculation of (TDS) as a function of material constants, temperature and neutron energy neglecting the contribution of Bragg scattering to the neutron transmission through a large imperfect single crystal. While (Mildner and Crawford 1993), reported a formula with four parameters to fit the experimental data assuming that the crystal is perfect. (Mildner and Crawford 1993), also reported that the inclusion of an unharmonic term significantly improve the fit around the minimum in the attenuation.

Since perfect single crystals do not exist, therefore the contribution of Bragg scattering from different (hkl) planes to the neutron transmission through a large imperfect single crystal may exceeds that of the scattering cross-section of the bound atom. Consequently, high attenuation factor may limit the use of such materials as a neutron filter.

Therefore, the present paper concerns the recent efforts to select the optimum parameters of single crystals to be used as efficient thermal neutron filters.

3. OPTIMUM PARAMETERS OF SINGLE CRYSTAL FILTERS

Recently, (Naguib, and Adib, 1996), reported a formula which allowed to calculate the total cross-section of an imperfect Cu single crystal of (FCC) structure as a function of crystal constants, temperature and neutron energy. The formula also includes the contribution of Bragg scattering to the total cross-section as a function of crystals mosaic spread and orientation of the plane at which the crystal was cut along.

(Naguib, and Adib, 1996) developed a computer code ISCANF to carry out the required calculations. The agreement obtained between the calculated and experimental cross-section values through copper single crystals justify the applicability of their computer code. Table (3) lists the physical parameters of the most promising single crystals.
Adib and Habib (2003), (2007), (2001), (2003), provided a detailed study of the attenuation of thermal neutron beams through a large Si, Ge, Pb, Bi and sapphire single-crystals. They gave a generalized formula which, for neutron energies in the range $10^{-4} < E < 10$ eV, permits calculation of their transmission properties. The optimum orientation, mosaic spread, temperature and the effect of tuning for the efficient transmission of thermal reactor neutrons are also given**.** Since the crystal structure type of each monocrystal is different as well as the number of atoms per unit cell is also not the same, therefore Adib and Habib (2003), (2007), (2001), (2003), developed a computer code for each l type of crystal structure to provide the required calculation. The applicability of each code was approved after the obtained agreement between the latest experimental results and calculated ones.
The selected optimum parameters of three of them which are of major interest and in use at the operating high flux reactors in several laboratories, are given below.

3.1 Silicon Single Crystal

To find the optimum thickness of silicon (111) crystal, the neutron transmission was calculated by Adib, (2003) assuming that the mosaic spread has the value of 0.5 mRad and 1mRad where the crystal was cooled at liquid nitrogen temperature. Fig. (1)a & b display the result of calculation for silicon single having mosaic 0.5 mRad and 1 mRad respectively. It seems that a 30 cm thick crystal with mosaic spread of 0.5 mRad and cooled at liquid nitrogen temperature is free from parasitic Bragg reflection.

Table (3): Physical parameters of the most promising single crystals

Element	Silicon	Germanium	Lead	Bismuth	Sapphier
Atomic weight	**28.08**	**72.61**	**207.21**	**209**	**20.4**
Crystal structure	**Diamond**	**Diamond**	**F.C.C.**	**Rhomb**	**Rhombohedral (n=2) or trigonal (n=6)**
Lattice parameters	**a_0=0.54307 nm**	**a_0=5431 nm**	**a_0=0.495 nm**	**a_0=b=c=0.4546nm $\alpha=\beta=\gamma=57.23^0$**	**Rhomohedral: a_0 =b=c=0.5128 and $\alpha=\beta=\gamma=55.28$**
No. of unit cells/m^3	**0.6243E+28**	**0.5519E+28**	**0.8245E+22**	**2.123E+28**	**4.71E+27**
θ_D	**420 K**	**363 K**	**200 K**	**119 K**	**1040 K**
Coherent scattering length fm	**4.2**	**8.185**	**9.3**	**8.532**	**b_{Al} = 3.449 and b_O = 5.805**
σ_{abc} at 0.025 V (barns)	**0.161**	**2.2**	**0.017**	**0.0338**	**0.231**
(σbat) (barns)	**2.180**	**8.42**	**11.106**	**9.156**	**15.7**

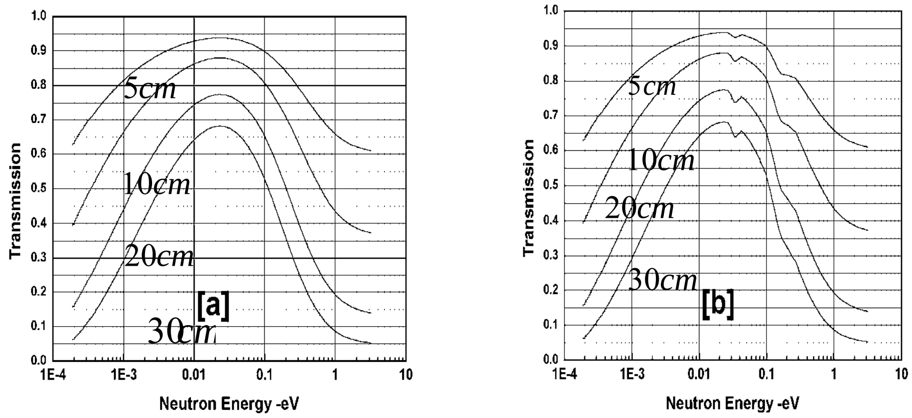

Fig. (1) Neutron transmission through silicon crystal for different thickness

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.(2), 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.

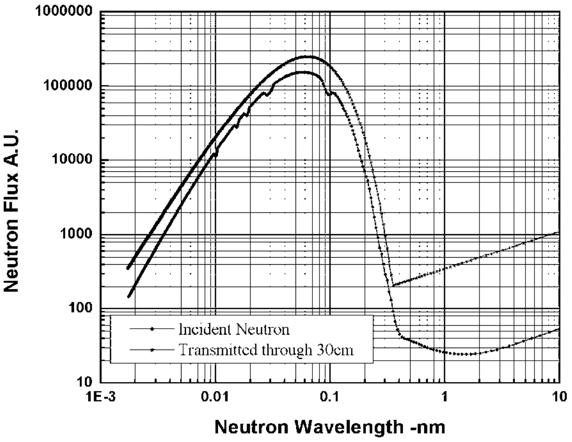

Fig. (2) Transmitted neutron thermal flux through silicon single crystal.

Its seems that 30 cm thick single crystal cooled at liquid nitrogen it sufficient of almost removing epi-thermal neutron and transmits less than 1% of fast ones with energies ~1 MeV and less than 5% of γ-rays with average energy Eγ =2MeV, while providing reasonable intensity of thermal neutrons(70%).

3.2 Germanium Single Crystal

Now-a-days enriched 76 Ge single crystals become one of the most promising thermal neutron filter especially after the availability of highly enriched Ge isotopes in reasonable price Therefore Habib, (2007) carried out a feasibility study of using Ge isotopes as efficient thermal neutron filter.

The calculated transmissions by Habib, (2007) for neutron energies from 1 meV to 10 eV for different Ge isotopes are displayed in Fig. (3). It is appear ante that, when used as a thermal-neutron filter, ^{76}Ge and ^{74}Ge crystals cut along the (111) plane are preferable than other isotopes, since their effects-to-noise ratio are higher.

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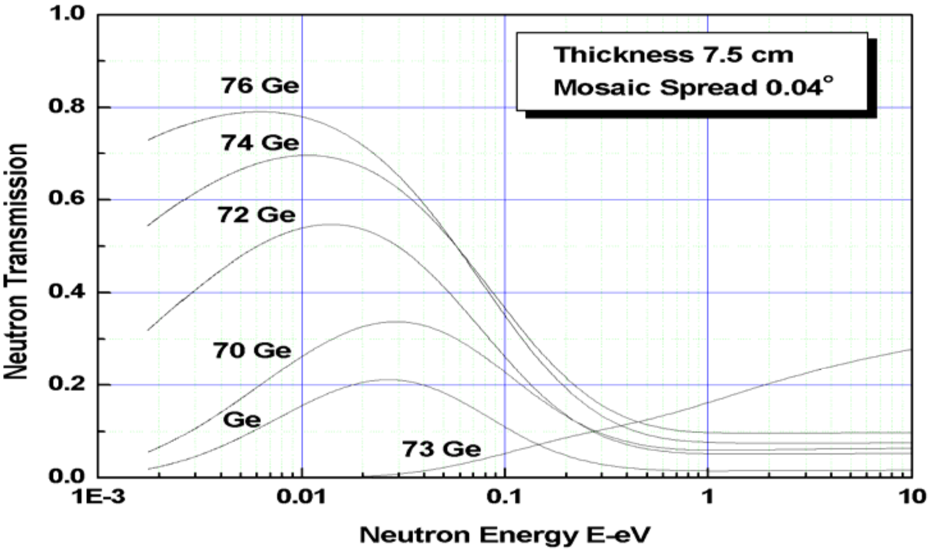
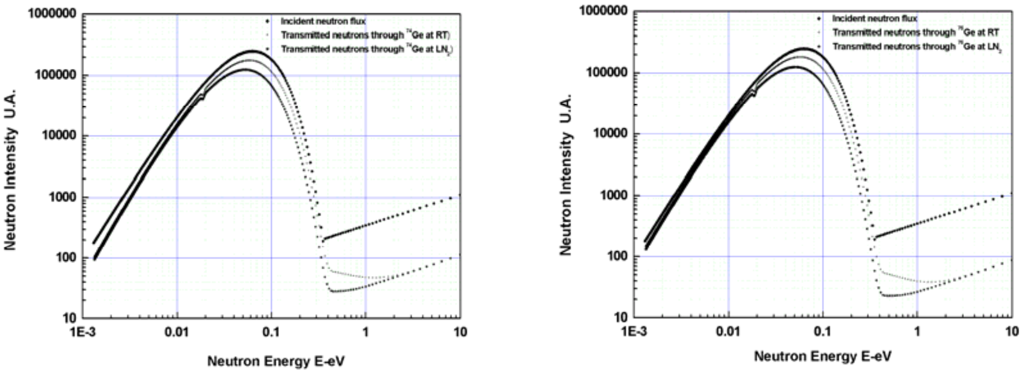


Figure (3) Neutron transmission through different Ge isotopes.

Calculation carried out by Habib, (2007) (Fig. 4) showed that 7.5 cm thick ^{76}Ge (111) crystals 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 energies E > 1 eV. Fig. (4) also shows that, there is an increase ≈ 20% at neutron energies < 0.05 eV in he neutron transmission through the cooled ^{76}Ge crystal at LN$_2$ temperature. Such improvement for many applications may be insufficient to warrant the expense and inconvenience of cryogenics.



(a) (b)

Figure (4) Thermal-neutron flux transmitted through 7.5 cm Ge (111) crystal.

The transmitted neutron spectrum is almost free from disturbing Bragg reflections. The filtering characteristics are less good for ^{74}Ge crystal. The final choice depends upon the experimental conditions required and the price of such crystal.

3.3 Sapphire Single Crystal

The neutron transmissions were calculated by Adib and Habib (2004) as a function of wavelength, for different thicknesses of 27, 54 and 81 mm of super optical quality sapphire single-crystals with [001] axis parallel to the incoming neutron beam at room temperature. The calculation was carried out within steps of λ=0.001 nm in the wavelength band from 0.001 nm up to 1.0 nm. The wavelength spread $\Delta\lambda$ was selected to be 0.004 nm, which is equal to that reported by Mildner et al (1993) .The result of these calculations are displayed in Fig. (5) along with the experimental data reported by Mildner et al. (1993). The agreement obtained between the calculations and the experimental data supports the application of additive cross-section formulae.

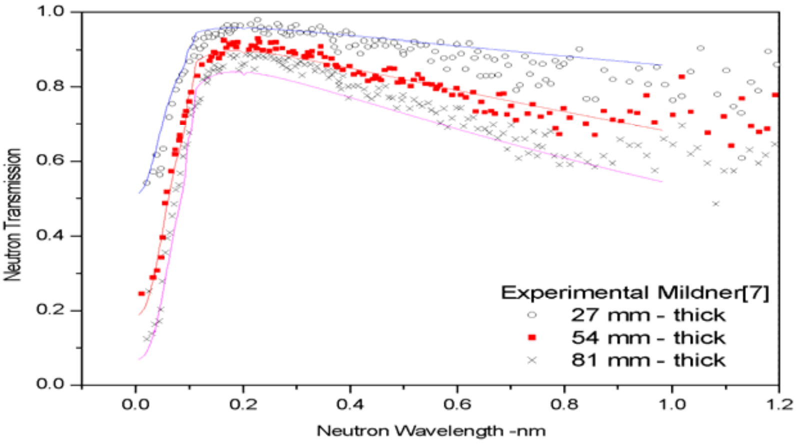
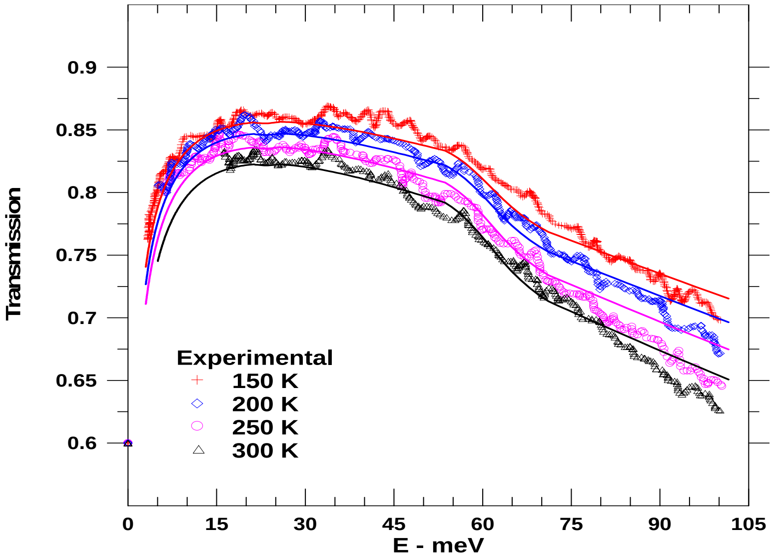


Fig. (5) Neutron transmission at different – sapphire crystal thickness.

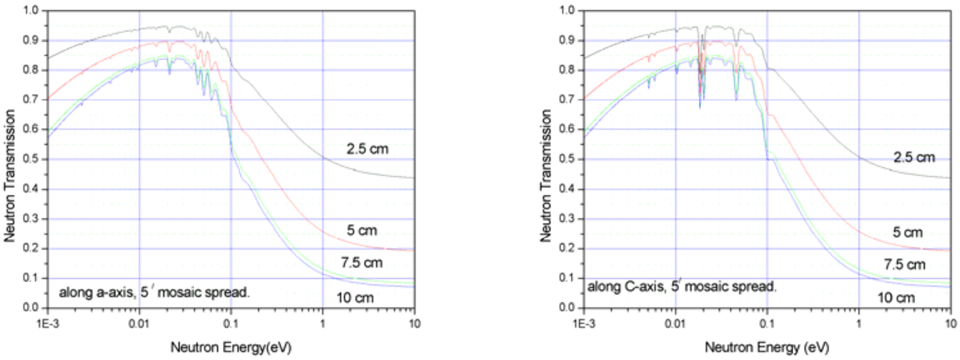
While, Figure (6) shows the neutron transmission reported by Mook and Hamilton, (2001) for a sapphire crystal for the a-axis direction over a large energy range. Also displayed as solid lines, are the calculated values assuming an 8 cm thick sapphire single crystal cut along (100) plane (i.e. along the a-axis) and with a FWHM mosaic spread of 1 min of arc . The agreement is reasonable within the range of fluctuations of the experimental values. However, a slight disagreement is obtained for, the c-axis neutron transmission of sapphire at different temperatures as a function of neutron energy.



The experimental data are taken from Mook & Hamilton (2001)

Figure (6) Neutron transmission of sapphire in the a-axis direction at different temperatures

To find the optimum sapphire thickness, the neutron transmission through different crystal thickness, were calculated. Fig. (7) a,b shows the result of calculation through a sapphire (100) crystal having mosaic spread of 5' while Fig.18b 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)**

Figure (7) Neutron transmission through sapphire, for different crystal thickness

Fig. (8) 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.

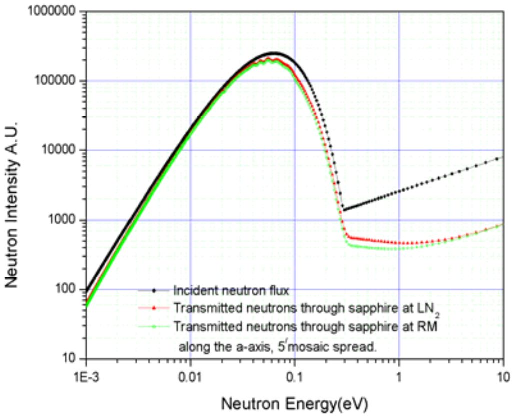


Figure (8) Thermal-neutron flux transmitted through 7 cm sapphire (100) crystal.

Figure (8) 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 LN_2 temperature. Such small improvement as reported by Mildner et al. (1993) & (1998) for many applications, this may be insufficient to warrant the expense and inconvenience of cryogenics.

4. CONCLUSION

Monocrystalline materials have been successfully used as thermal-neutron filters. The very best candidates are diamond, graphite, lead, Bi, Be, depleted uranium and sapphire. The major problem to choose the suitable monocrystalline is the availability of large and highly perfect single crystals at a reasonable cost. Our study indicates that sapphire is significantly more effective than the others for a wide range of neutron energies.The optimum filtering properties of the most promising monocrystals cooled to LN_2 are summarized in Table (4)

Table (4) Optimum Filtering properties of monocrystals.

Single crystal	**Mosaic spread FWHM-degree**	**Cutting plane** $h_c k_c l_c$	**Thickness t cm**	**Neutron transmission** $E_n < 0.025$ **eV**	**Neutron transmission** $E_n > 1$ **eV.**	**Attenuation factor** $E_\gamma \approx$ **2 MeV**
Silicon	**0.15**	**(111)**	**25.0**	**80 %**	**< 10 %**	**12**
^{76}Ge	**0.10**	**(111)**	**7.0**	**91 %**	**< 10 %**	**5.5**
Lead	**0.20**	**(111)**	**5.0**	**75 %**	**< 10 %**	**7.2**
Bismuth	**0.15**	**(111)**	**10.0**	**90 %**	**< 7 %**	**43.0**
Sapphire	**0.08**	**(100)**	**7.5**	**85 %**	**< 8.5 %**	**4.5**

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