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"Total neutron cross-section and transmission simulation for poly- & mono- Magnesium and Zirconium crystals"

Muhammad S. Mansy

PhD, Nuclear and Radiation Physics

Egyptian Atomic Energy Authority

Outlines:

- Types of neutrons and their production
- Neutron interaction with matter.
- Thermal neutrons monochromators and filters
- Transmission and cross-section measurements for thermal neutrons.
- HEXA-Filters code.
- Transmission and cross-section measurements for Poly and mono-crystalline Zr and Al crystals.

Neutrons and their production

- Neutron scattering is one of the most essential techniques for probing condensed matter. The zero net charge of the neutron means that is interacts very weakly with matter and penetrates deeply into a sample (Shirane *et al.*, 2002). The neutrons used in a scattering experiment produced in both nuclear reactors and spallation sources. The produced neutron beams have polychromatic wavelengths, consequently, to choose a narrow monochromatic band the use of neutron monochromator is encouraged. Bragg's law governs the wavelength accepted by a mono crystal.

Perfect and mosaic crystals

- A perfect monochromator crystal is very expensive; furthermore, the angular width of the diffracted beam is smaller than the beam divergence suitable for neutron spectrometers. As a result, imperfect (mosaic) mono-crystals utilization is recommended for obtaining the desired resolution. Unfortunately, the diffracted neutron beams contaminated by reflection from higher orders. Therefore, thermal neutron filters are required for effective removal of unavoidable contaminations and reducing both epithermal neutrons and γ-rays from fission reaction in the reactor. Monochromator and filter crystals are combined in certain setup to form a neutron spectrometer used in neutron diffraction experiments

Thermal neutrons and filters

• Favourable materials for neutron filters should have a low absorption coefficient for thermal neutrons, a large Debye temperature, and a small incoherent scattering cross-section. Filters may be poly or mono-crystals, in case of powder filters only neutrons with $\lambda > 2d_{max}$ will be transmitted, where (d_{max}) is the largest interplanar distance of Bragg planes. Several materials approved for the use as thermal neutron monochromator and filters, for example beryllium, beryllium oxide poly- and mono-crystals (Adib et al., 2015a; Adib et al., 2015b). Recently, zinc and zinc oxide (Adib et al., 2015a; Adib et al., 2015b), quartz poly- and mono-crystal (Adib et al., 2015b) and MgO (Adib et al., 2014) are also investigated.

- Freund (1983) proposed an empirical formula to calculate the total neutron cross-section of a crystalline solid as function of the neutron wavelength or energy with two adjustable fitting parameters (C_1 and C_2). The validity of this formula checked for several materials. Vogel (2000) calculated the total neutron cross-section of polycrystalline Mg using Granda (1984) model by "BETMAn" program; once more, Boin (2011) repeated the calculations using the same model by "nxs" program library. Cook et al., (1992) and Shull (1995) reported the utilization of Mg as a thermal neutron monochromator but in their works, they did not give the optimal parameters of the used mono crystal.

Theoretical treatment

Attenuation of neutrons by crystalline material can be determined by calculating the total neutron cross-section as:

$$\sigma_{total}(E,T) = \sigma_{abs}(E) + \sigma_{inel.}(E,T) + \sigma_{el.}(E,T) \qquad (1)$$

The first term $\sigma_{abs}(E)$ is the absorption cross-section and varies as ($E^{-1/2}$). The second term $\sigma_{inel.}(E,T)$ corresponds to inelastic scattering cross-section due to single- and multi-phonons in the solid material; it is energy and temperature dependent. The last term $\sigma_{el.}(E,T)$ is the elastic (Bragg scattering) cross-section.

Inelastic scattering cross-section $\sigma_{inel.}(E,T)$is proportional to neutron energy; with multi-phonon scattering dominant at high incident neutron energies ($E \geq k_B T$), where k_B is Boltzmann's constant and T is the crystal temperature.

The two fitting parameters $C_1$and $C_1$are material dependent and obtained from the following equations:

$$C_1 = \sigma_{abs}(E) \times \sqrt{E} \ , C_2 = 4.27 \exp[A_{av.}/61] \qquad (2)$$

Here, ($A_{av.}$) is the average molecular weight. Neutron energy related is to its wavelength as ($\lambda = 0.0286/\sqrt{E}$) where the energy ($E$) is expressed in electron volts and (λ) in nanometres.

Bacon (1975) expressed the total coherent scattering cross-section for a poly-crystalline material as:

$$\sigma_{el.}(\lambda, T) = \frac{N_c \lambda^2}{2} \sum_{d_{hkl}=0}^{2d_{hkl}<\lambda} |F_{hkl}|^2 d_{hkl} e^{-2w} \qquad (3)$$

Here, F_{hkl} is the structure factor and N_c is the atom number density, the summation runs for all sets of hkl with lattice spacing d_{hkl} smaller than half of the selected neutron wavelength (λ), such that Bragg edges appear at $\lambda = 2d_{hkl}$.

Naguib & Adib (1998) gave the contribution of elastic scattering $\sigma_{el.}(\lambda, T)$from a mono-crystal as:

$$\sigma_{el.}(\lambda, T) = \frac{1}{N_c t_o} \ln\left(1 \Big/ \prod_{hkl} (1 - P_{hkl}^{\theta})\right) \qquad (4)$$

Here, $t_o$is the thickness of the crystal in the beam direction, P_{hkl}^{θ} is the reflecting power of the (hkl) plane inclined by the angle θ_{hkl} to the incident beam direction and given by Bacon (1975) as:

For reflection method

$$P_{hkl}^{\theta} d\theta = \frac{a d\theta}{1 + a + (1 + 2a)^{\frac{1}{2}} \coth[A(1 + 2a)^{\frac{1}{2}}]} \qquad (5)$$

For transmission method

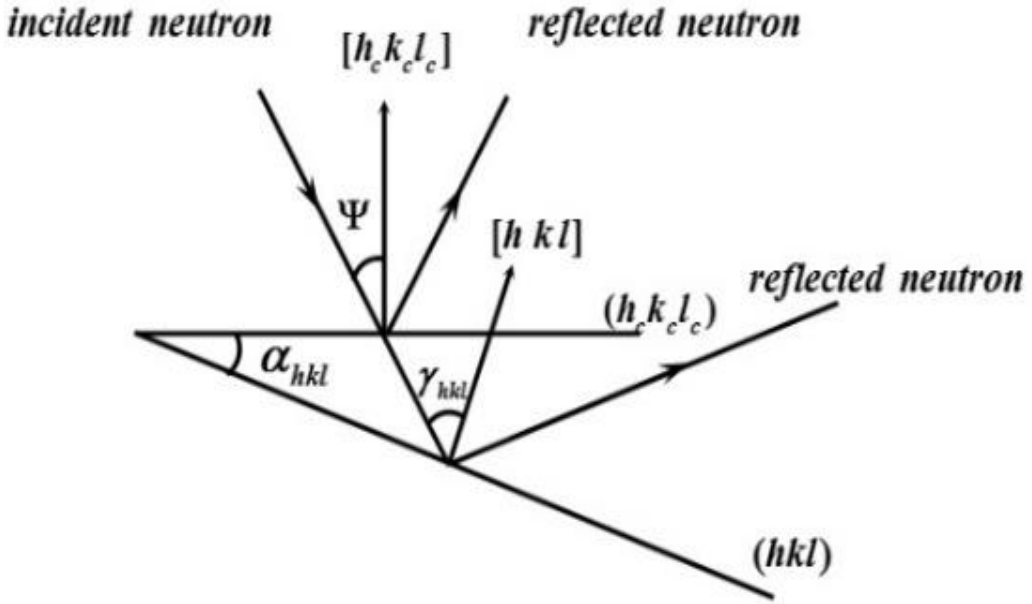
$$P_{hkl}^{\theta} d\theta = \sinh(Aa) e^{-A(1+a)} d\theta \qquad (6)$$

Here, $A = \mu t_o / \gamma_o$, $a = \frac{Q_{hkl}}{\mu} W(\Delta)$ and $d\theta = d\lambda / 2d_{hkl} \cos\theta$ in which μ is the linear absorption coefficient.

A mosaic crystal is composed of small perfect crystals with small relative mis-orientaions. These mis-orientaions assumed to have a Gaussian distribution with standard deviation η on mosaic blocks of the mono crystals given as

$$W(\Delta) = \frac{1}{\sqrt{2\pi}\eta} (e^{-\Delta^2} / 2\eta^2) \qquad (7)$$

A schematic diagram of the Bragg scattering from mono crystal cut along $(h_c k_c l_c)$ plane and the reflected neutrons from any (*hkl*) plane shown in Fig. 2



For Hexagonal crystal structure with lattice constant a_o, the equation describing the cutting plane $(h_c k_c l_c)$ which is parallel to the crystal surface given as:

$$\sqrt{h_c^2 + h_c^2 + h_c^2 Z} = a_o \qquad (10)$$

The relation gives the inter-planar distance d_{hkl} is

$$d_{hkl} = \frac{a_o}{\sqrt{h^2 + k^2 + l^2}} \qquad (11)$$

If the angle between the neutron beam direction and the direction, $[h_c k_c l_c]$ is ψ, then $\gamma_o = cos\,\psi$. While, the direction cosine of the diffracted beam γ_{hkl} from any (hkl) plane expressed as:

$$\gamma_{hkl} = \frac{(hh_c + kk_c + ll_c)\cos\psi + l_c\left[\frac{hh_c + kk_c}{\sqrt{h_c^2 + k_c^2}} - \frac{l\sqrt{h_c^2 + k_c^2}}{l_c}\right]\sin\psi}{\sqrt{h_c^2 + k_c^2 + l_c^2}\sqrt{h^2 + k^2 + l^2}} \qquad (12)$$

While, its inclination angle α_{hkl} to the crystal surface given by:

$$\cos\alpha_{hkl} = \frac{(hh_c + kk_c + ll_c)}{\sqrt{h_c^2 + k_c^2 + l_c^2}\sqrt{h^2 + k^2 + l^2}} \qquad (13)$$

If the cutting plane is $(00l_c)$, then γ_{hkl} becomes

$$\gamma_{hkl} = \frac{(l\cos\psi + k\sin\psi)}{\sqrt{h^2 + k^2 + l^2}} \text{ and } \cos\alpha_{hkl} = \frac{l}{\sqrt{h^2 + k^2 + l^2}} \qquad (14)$$

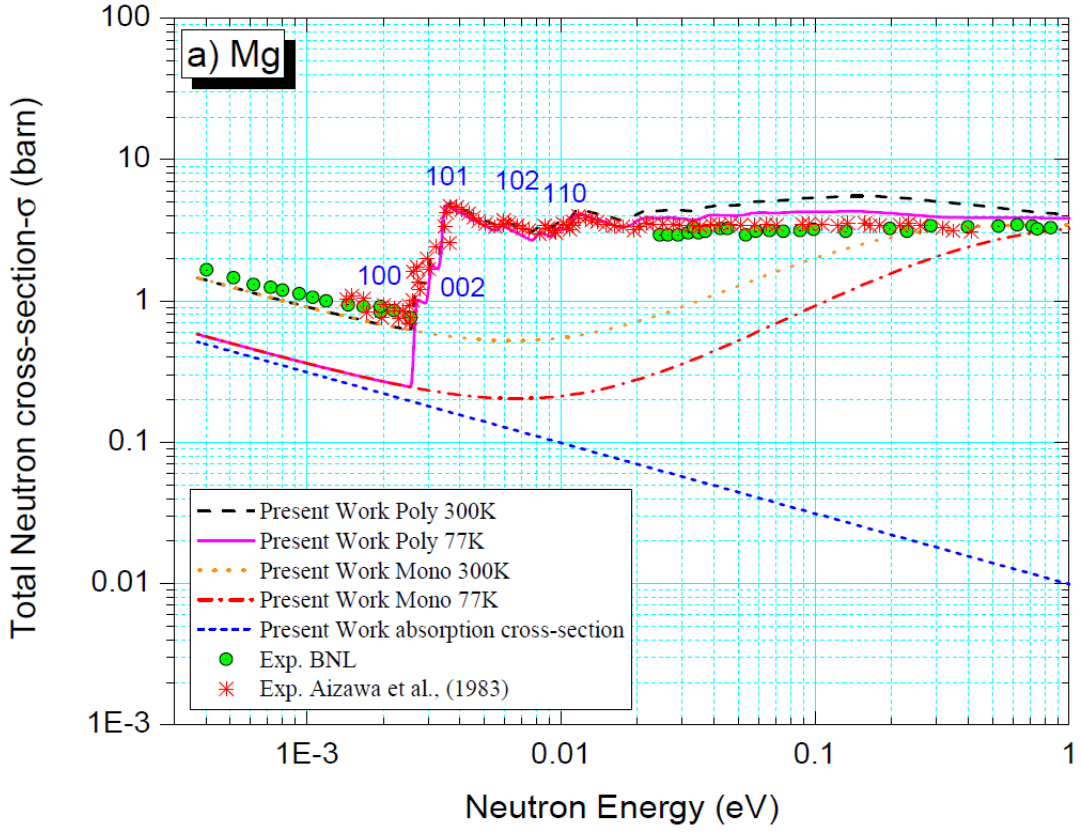
Results and discussions

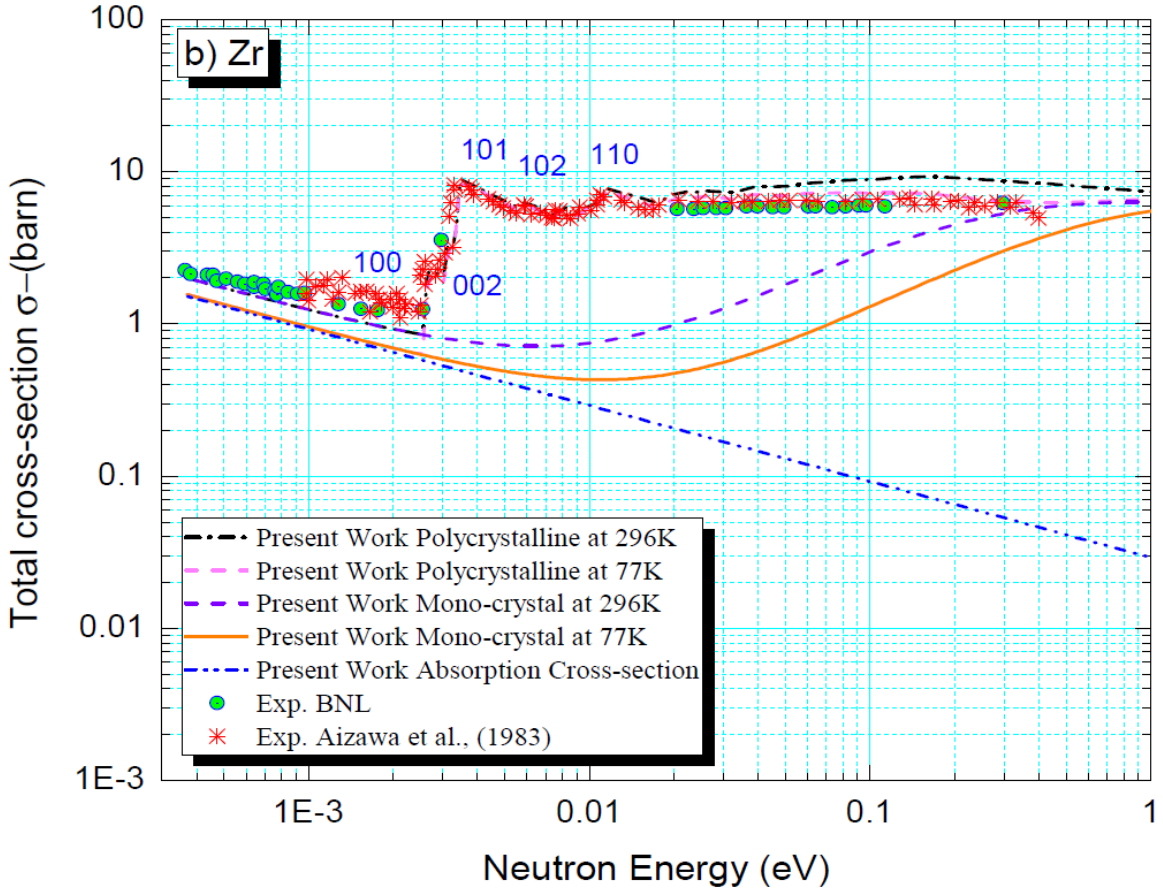
Erk (2011) reported that β-Zr crystallizes in the Hexagonal (fluorite) type structure (space group $Fm\,3m$, no =225, a_o= 0.5939 nm, Z=4). The Pb^{+2} ions occupy Wyckoff 4(a) sites at (0, 0, 0) (1/2, 0, 1/2) (0, 1/2, 1/2) (1/2, 1/2, 0). Whereas F^{-1} ions are in Wyckoff 8(c) positions F_1 at (1/4,1/4,1/4) (1/4,3/4,3/4) (3/4,1/4,3/4) (3/4,3/4,1/4) and F_2 at (1/4,1/4,3/4) (1/4,3/4,1/4) (3/4,1/4,1/4) (3/4,3/4,3/4). While Mg crystallizes in the Hexagonal (Rock-salt) type structure (space group $Fm\bar{3}m$, no =225, a_o= 0.5630 nm, Z=4). The Na^{+1} ion occupy Wyckoff 4(a) sites at (0,0,0) (1/2 ,0, 1/2) (0,1/2,1/2) (1/2,1/2,0), whereas Cl^{-1} ion is in Wyckoff 4(b) positions at (1/2,0,0) (0,0,1/2) (0,1/2,0) (1/2,1/2,1/2). At room temperature Zr and Mg have effective Debye temperature Θ_D = 237K and 321K respectively. Debye temperature is very important for the calculation of inelastic scattering cross-section, which is temperature dependent.

Results and discussions

Calculation of the total neutron cross-section of poly- and mono crystals

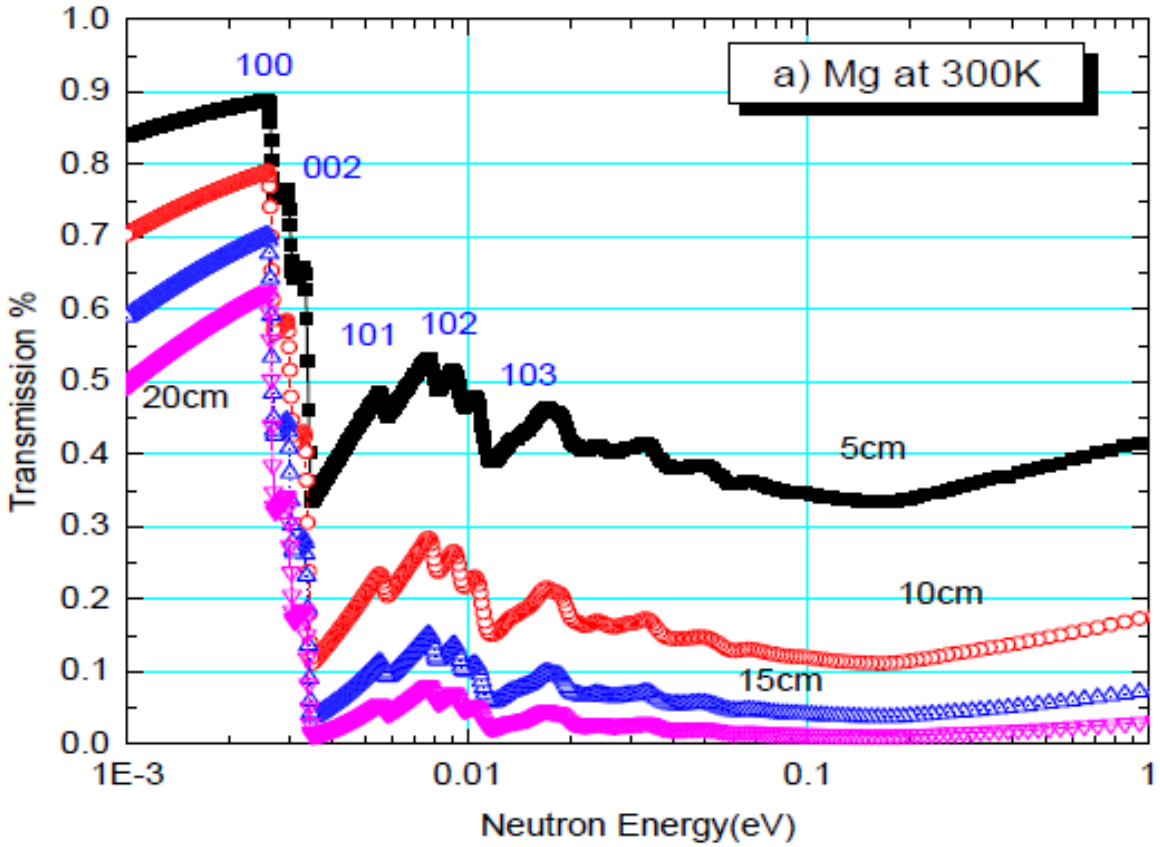
In order to check the validity of the designed code "HEXAGONAL-MOFI", the total neutron cross-section of polycrystalline Mg was calculated in the energy range of (8×10^{-4} to 1) eV and compared with that reported by Boin (2011). The result of calculations plotted in Fig. 3.

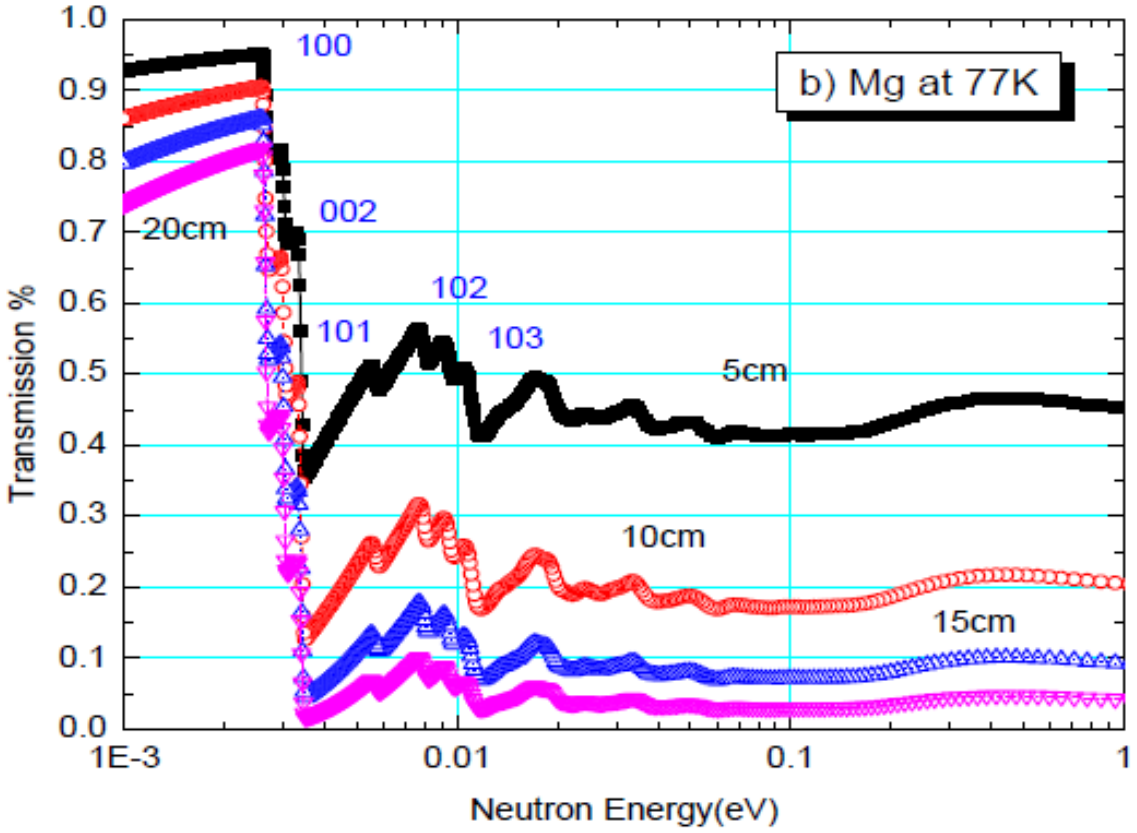




Results and discussions

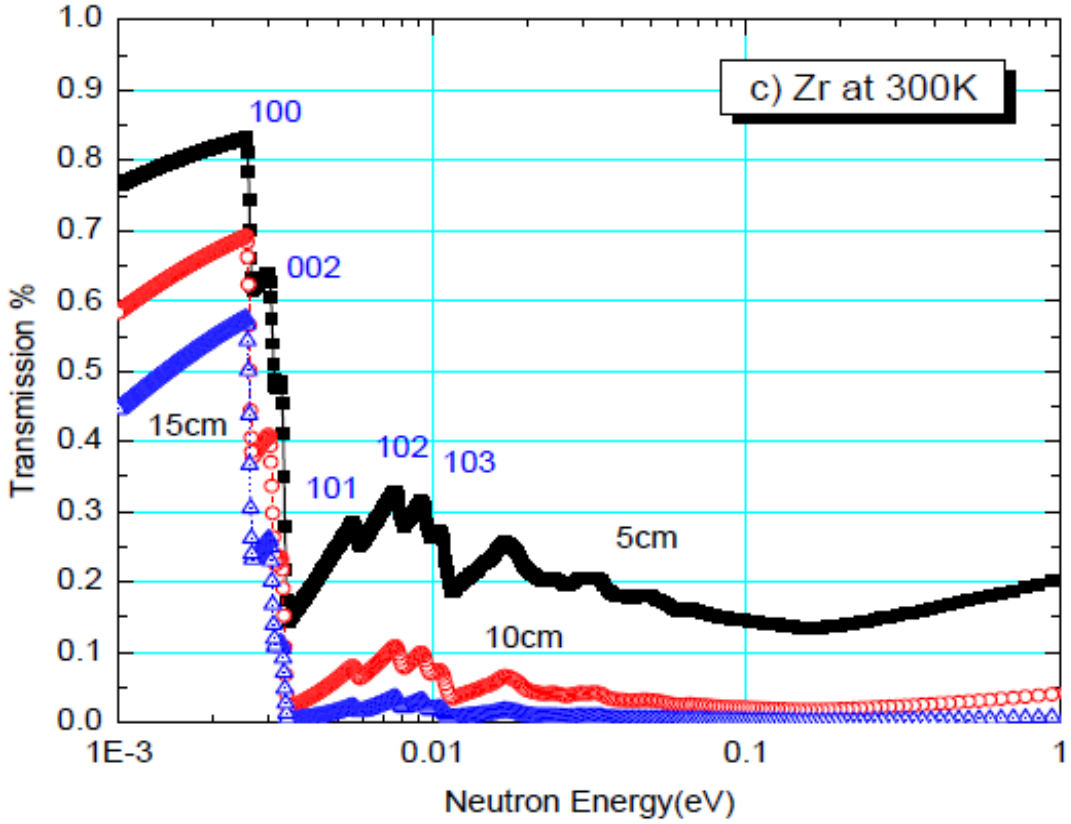
Calculation of the Neutron transmission for poly- and mono crystals

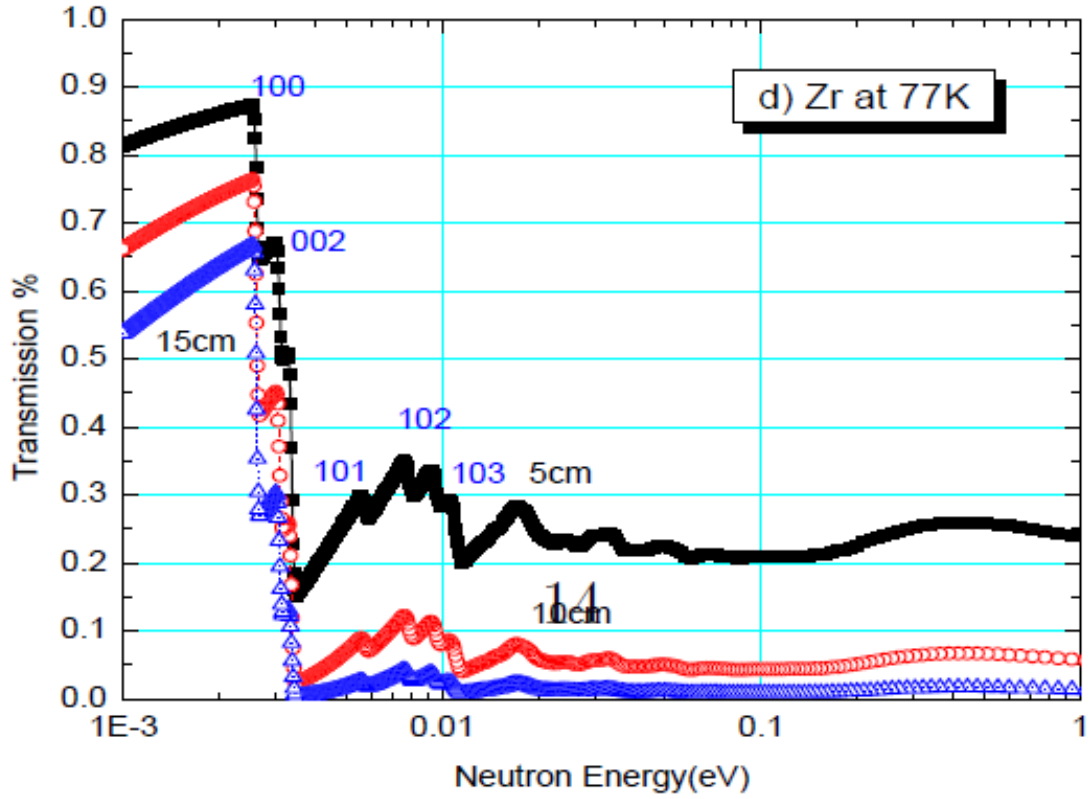




Results and discussions

Calculation of the Neutron transmission for poly- and mono crystals





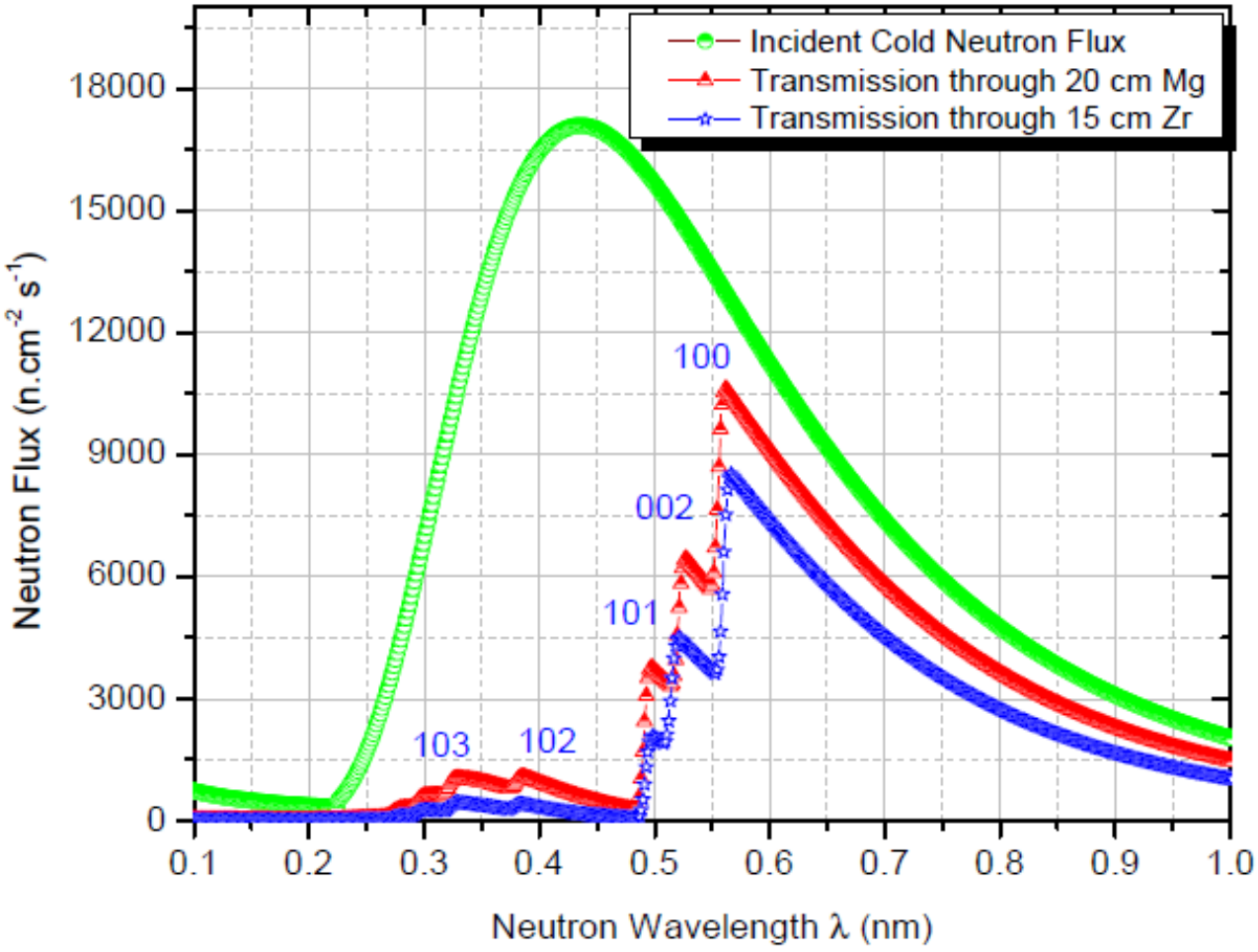
Neutron transmission of poly-crystalline Mg and Zr were calculated as a function of thickness at room (R.T. = 300 K) and liquid nitrogen (L.N. = 77 K) temperatures in the energy range from 1 meV up to 10 eV.

Results and discussions

Calculation of the Neutron transmission for poly- and mono crystals

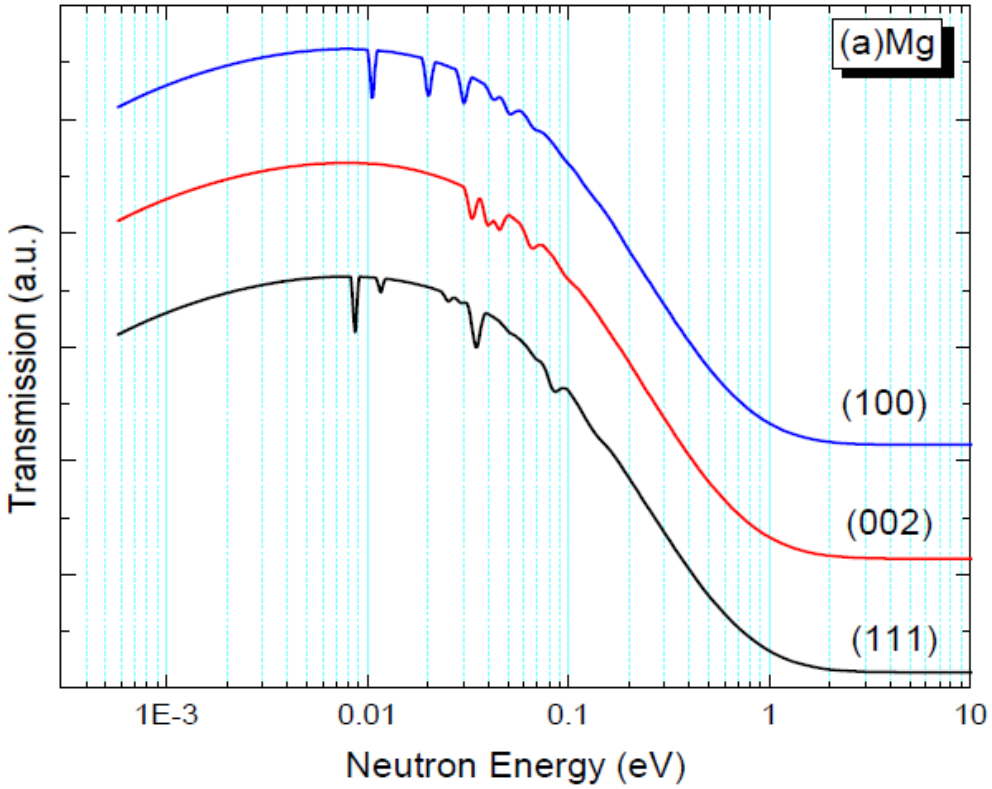
It seems that Mg polycrystal with 20 cm thick cooled with (L.N.) can transmit about 82% of the incident neutrons with high attenuation for neutrons with energies less than 33 meV.

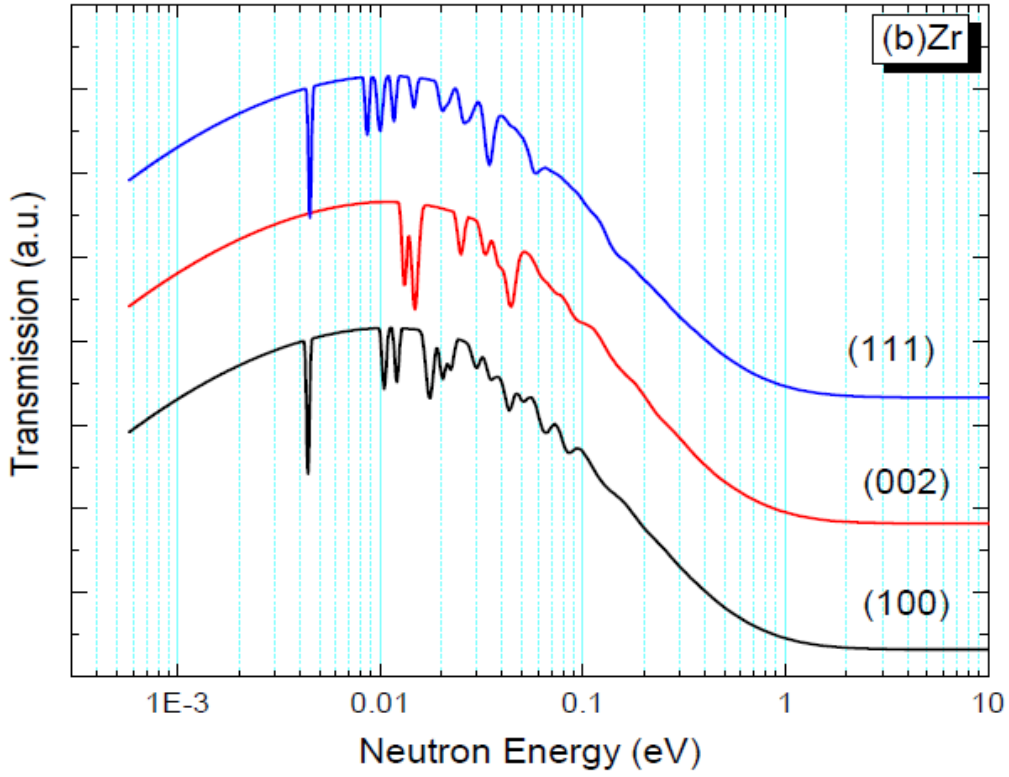
While a 15 cm thick for Zr can transmit only 68% for neutrons with energies less than 32 meV.



Results and discussions

Calculation of the Neutron transmission for poly- and mono crystals

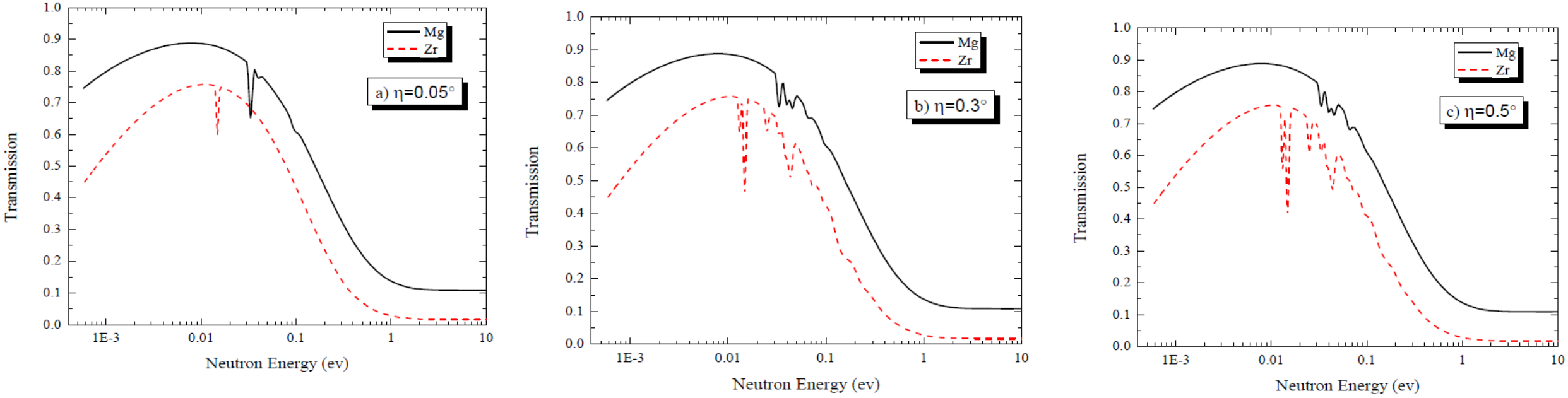




The results presented in Figs.5(a and b) is giving indication that the appropriate cutting-plane is the (0 0 2) plane for both Mg and Zr i.e., the crystal will be cut along its c-axis. This is because the small amount of parasitic Bragg-reflections comes form the other parallel planes to the plane (0 0 2), this leads to less disturbance of the transmitted neutrons which can cause any undesirable effect.

Results and discussions

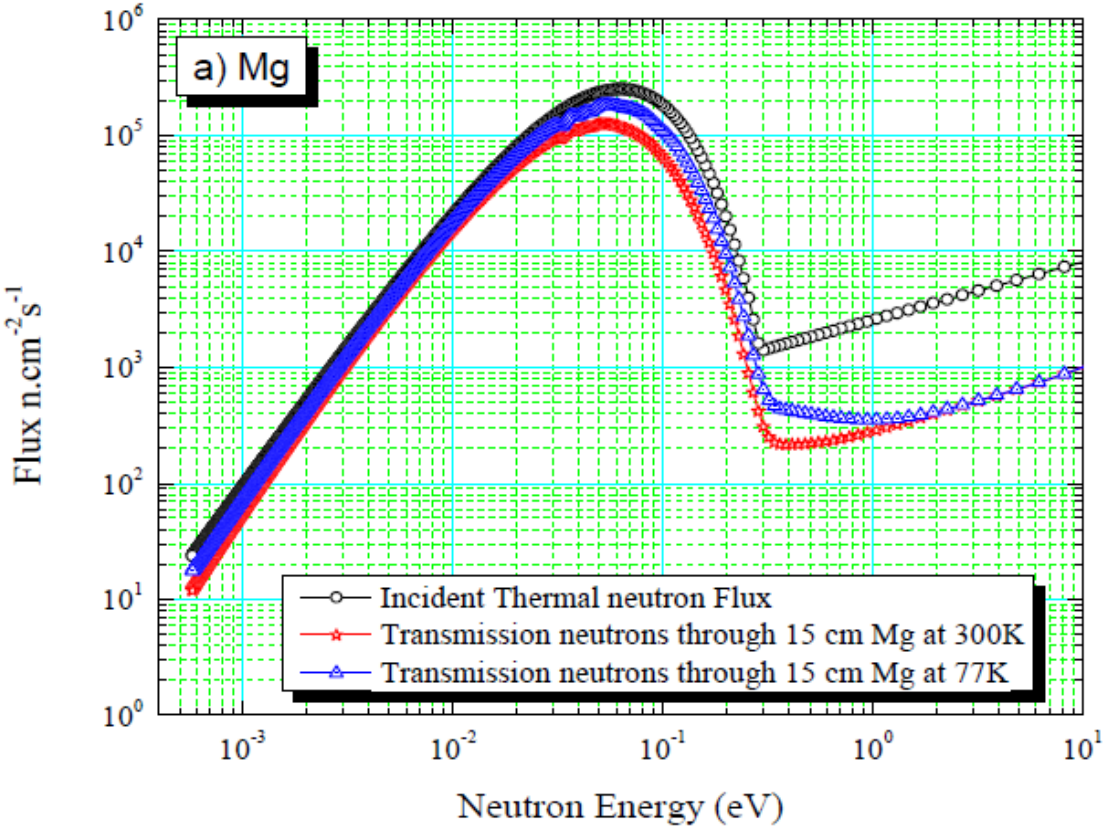
Calculation of the Neutron transmission for poly- and mono crystals

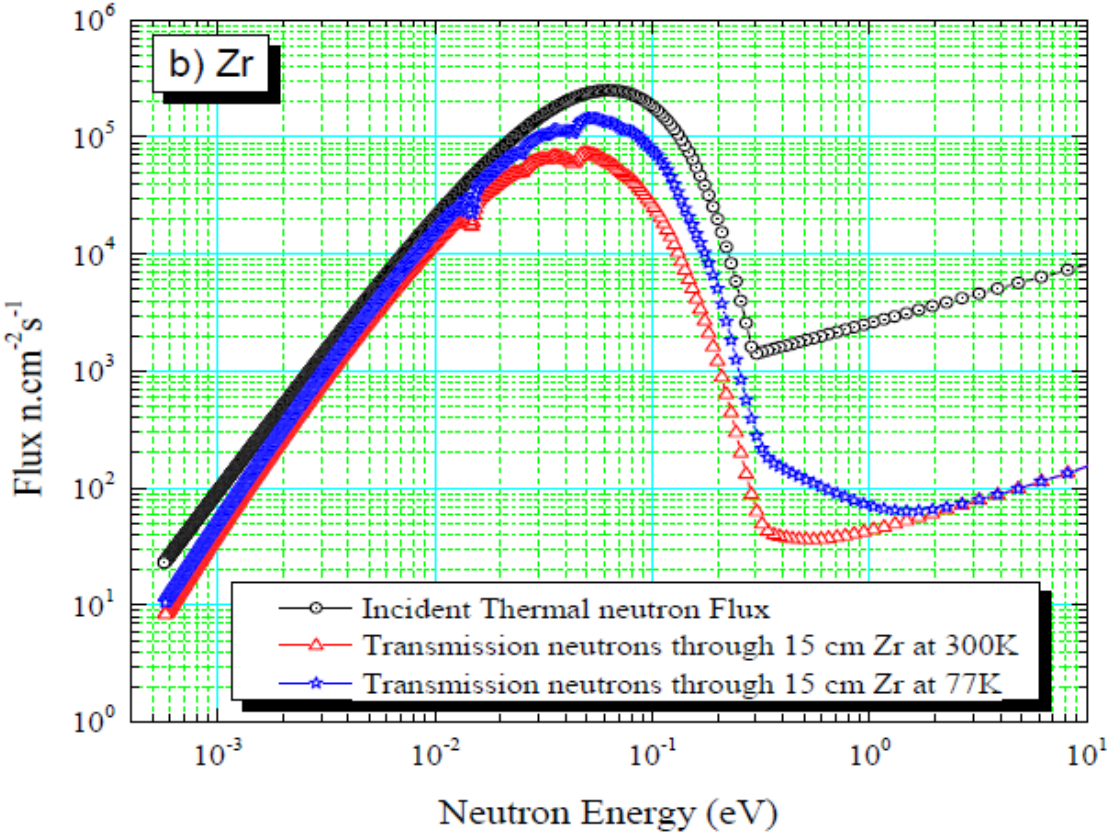


As seen from Figs.6 (a, b and c) the mosaic spread value of (η= 0.3°)can be used as the optimum value because the effect of the parasitic Bragg- reflections is low compared to other mosaic spread values. In addition, the cost of synthetic of such crystal will not be too expensive like perfect one.

Results and discussions

Calculation of the Neutron transmission for poly- and mono crystals





It is seen that 15 cm thick of both Mg and Zr crystal shows high neutron transmission in the desired neutron energies with high attenuation value for the unwanted epithermal neutrons.

Conclusion

- In the present study the validity of the developed HEXA-CODE is checked and shows high performance in the calculation of the total neutron cross-section and transmission for poly- and mono-crystals by comparison with the experimental data.
- The real situation of incident and transmitted thermal and cold neutron fluxes are also simulated. In addition, the optimal parameters controlling the efficiency of the neutron filters are also calculated.

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Thank you …