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NEUTRON TRANSMISSION OF GERMANIUM POLY- AND MONO-CRYSTALS

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The measured total neutron cross-sections of germanium poly-and mono-crystals were analyzed using an additive formula. The formula takes into account the germanium crystalline structure and its physical parameters. Computer programs have developed in order to provide the required analyzes.The calculated values of the total cross-section of polycrystalline germanium in the neutron wavelength range from 0.001 up to 0.7 nm were fitted to the measured ones at ET-RR-1. From the fitting the main constants of the additive formula were determined. The experimental data measured at ET-RR-1 of the total cross-section of high quality Ge single crystal at 440^o K, room, and liquid nitrogen temperatures, in the wavelength range between 0.028 nm and 0.64 nm, were also compared with the calculated values using the formula having the same constants. An overall agreement is noticed between the formula fits and experimental data.

A feasibility study for the use of germanium in poly-crystalline form, as cold neutron filter, and in mono-crystalline one as an efficient filter for thermal neutrons is undertaken. The filtering efficiency of Ge single crystal is detailed in terms of its isotopic abundance, crystal thickness, mosaic spread, and temperature. It can be concluded that the 7.5 cm thick ^{76}Ge single crystal (0.1^o FWHM mosaic spread) cooled at liquid nitrogen temperature is an efficient thermal neutron filter.

Keywords: *Thermal neutron filter, Neutron transmission of germanium isotopes.*

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 [1].

As shown by several authors [2-4], 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. Poly- and mono crystals have also been used as thermal-neutron filters [5, 6]. In these contexts, the dependence of the total cross-section on material parameters must be known, it being acknowledged that the thermal diffuse scattering (TDS) and neutron capture cross-sections represents dominant contributions to the total cross-sections at thermal-neutron energies. Freund [7], has reported a semi-empirical formula, which permits the calculation of TDS as a function of material constants, temperature and neutron energy, neglecting the contribution of Bragg scattering to neutron transmission through a large imperfect single crystal. Recently Naguib and Adib [8,9], have reported a general formula which allows calculation of the total thermal-neutron cross-section for an imperfect single crystal, as a function of crystal mosaic spread and orientation of the plane at which the crystal was cut.

Adib et al [10] adapted this formula carrying out a study for use of poly- and mono-crystalline silicon as cold and thermal-neutron filters respectively. The low-coherent scattering cross-section of silicon and the difficulty in producing 30 cm thick single crystal with well-defined mosaic spread were found to limit its use as a thermal-neutron filter.

The present work concerns the analysis of the measured, at ET-RR-1 reactor, total cross-sections of germanium poly- and mono-crystals using the additive formula, along with the developed computer programs. A feasibility study for use of Ge at its stable isotopes as a thermal-neutron filter is also given. Paralleling efforts that are being made by Vertibni et al. [11] to use ^{76}Ge with small thickness is also considered to be a better choice than silicon, its coherent scattering cross-section and Z number being higher than the latter.

THEORETICAL TREATMENT

The total cross-section determining the attenuation of neutrons by a solid crystalline, could be given as

$$\sigma = \sigma_{abs} + \sigma_{TDS} + \sigma_{Bragg} \tag{1}$$

Where the neutron capture cross-section (i.e absorption) σ_{abs} for most elements obeys the $1/\sqrt{E}$ law, and can be written as

$$\sigma_{abs} = C_1 E^{-1/2}$$

With E the neutron energy and C_1 is a constant which calculated from the values provided by Sears [12].

Following Freund [7] the TDS cross-section σ_{TDS} is given by

$$\sigma_{TDS} = \left[A/A+1\right]^2 \sigma_{bat}\left[1-e^{-(B_0+B_T)C_2 E}\right] + \frac{\theta_D^{1/2}\sigma_{bat}}{36A\sqrt{E}} \begin{cases} R \rightarrow X \leq 6 \\ 3.3X^{-7} \rightarrow X \geq 6 \end{cases} \tag{2}$$

where $X = \theta_D/T$ (T being the sample temperature), $\sigma_{bat} = S + s$ (the sum of coherent and incoherent scattering cross-sections, respectively, of the bound atom), A is the atomic mass number, C_2 is a constant which is independent of the scattering material, h is Plank’s constant and K_B is Boltzman’s coefficient, with

$$B_0 = 3h^2/2K_B\theta_D A.$$

In addition, for lower temperature, the series R is given by

$$R = \sum_{n=0}^{\infty} B_n X^{n-1} \Big/ \left[n!\left(n+\frac{5}{2}\right)\right], \tag{3}$$

With B_n being the Bernoulli numbers.

The contribution of Bragg scattering σ_{Bragg} to the total cross section taking into account the resulting reflection from different (*hkl*) planes, which are able of giving the Bragg reflection for the neutron wavelength λ was calculated. In case of poly- crystalline material the reflections are from all planes having spacing $d_{hkl} \geq \lambda/2$, while in case of mono crystal, reflections are from the (*hkl*) planes satisfying the Bragg equation:

$n\lambda = 2\, d_{hkl} \sin \theta_{hkl}$

where n is the order of reflection, θ_{hkl} is the glancing angle to the (*hkl*) plane.

It was shown by Bacon [13] that for a poly-crystalline material with grain size less than 10^{-4} mm, the total coherent Bragg scattering cross-section can be given as:

$$\sigma_{Bragg} = \frac{N_c \lambda^2}{2} \sum_{d_{hkl} \geq \lambda/2} F_{hkl}^2 d_{hkl} . e^{-2w} \tag{4}$$

where N_c is the number of unit cells per cubic centimetre , F_{hkl} is the structure factor of the unit cell and e^{-2w} is the Debye-Waller factor.

Following Naguib and Adib [8], the Bragg scattering cross-section by a single crystal is given by:

$$\sigma_{Bragg} = \frac{1}{N t_o} \ln\left(\frac{1}{T_{Bragg}}\right) \tag{5}$$

where N is the number of atoms per cubic centimetre and t_o is the effective thickness of the crystal in cm. T_{Bragg} is the resulting neutron transmission from different (*hkl*) planes given by:

$$T_{Bragg} = \prod_{hkl} \left(1 - P_{hkl}^{\theta}\right)$$

where P_{hkl}^{θ} is the reflecting power of the (*hkl*) plane inclined by an angle θ_{hkl} to the incident beam direction and is given by Bacon [13].

Two computer programs **PGe** (**P**oly-crystalline **Ge**rmanium), and **SGe** (**S**ingle crystal **Ge**rmanium) have been developed in order to calculate the total cross-section and the transmission of neutrons of energy ranging from 10^{-4} to 10 eV incidents on poly-crystalline Ge and on imperfect mono-crystal respectively.. SGe code is an adapted version of DSIC code developed to calculate the total cross-section and transmission through crystalline silicon for neutron energies below 10 eV. The adapted version can provide additional the calculations of the total cross-sections of Ge single crystal and its isotopes.

EXPERIMENTAL DETAILS

As reported by M.Adib [14] two polycrystalline samples with thicknesses of 2.2×10^{26} and 1.6×10^{26} atoms per m^2 were prepared from fine spec pure Ge powder ,while the mono-crystalline one was of cylindrical shape 0.03 m in diameter and 0.025 m thick and cut along the (111) axis. The total neutron cross-section measurements of poly- and mono-crystalline samples were performed using two time-of-flight spectrometers installed in front of two of the ET-RR-1 reactor horizontal channels. The spectrometer's resolution at different intervals of the whole energy range (2meV-1eV) could be varied from 20 to 3μs/m [14].

RESULTS AND DISCUSSIONS

For comparison of experimental neutron cross-section data with calculated values, the program takes into consideration the effects of both neutron wavelength resolution and incident neutron beam divergence as given by Habib [15].

Poly-crystalline Ge

Using the PGe program, the total neutron cross-section of Ge was calculated for neutrons in the wavelength range 0.001 nm up to 0.8 nm. The main physical parameters required in these calculations are listed in Table.1.

Table 1: The main physical parameters of Ge.

Atomic weight	72.61
Crystal structure	Diamond structure
Lattice parameters $a_o=b_o=c_o$	0.5431 nm
Atomic position	0, 0, 0; 1/2, 0, 1/2; 0, 1/2, 1/2; 1/2, 1/2, 0; 1/4,1/4,1/4;3/4,3/4,1/4;3/4,1/4,3/4; 1/4,3/4,3/4
Number of unit cell/m^3	0.5519 E+28 / m^3
Coherent scattering length b_c	0.8185 fm
Absorption cross-section for thermal Neutrons σ_a (E= 0.025 eV)	2.2 b
Total scattering cross-section (σ_{bat})	8.42 b

The results of calculations are displayed in Figure1. as solid lines. For comparison, the experimental data presented in Adib [14] after its correction for small angle scattering are also displayed in Fig.1 as dots. The calculated results are in reasonable agreement with experimental values for the fitted parameters C_2=1402 nm^{-2}eV^{-1} and Θ_D=363 K. The values of C_2 was found to be equal to that deduced from the semi-empirical form, while Θ_D is higher than the value of Θ_D=290 K, reported by Freund [7]. The discrepancies can be explained by the validity limits of Equations.(2) which come from the assumptions implied by Freund, namely, that there is no variation of Θ_D with temperature, and that simple superposition of single phonon and multi-phonon events can be assumed at neutron energies when both types of event are of the same order of magnitude.

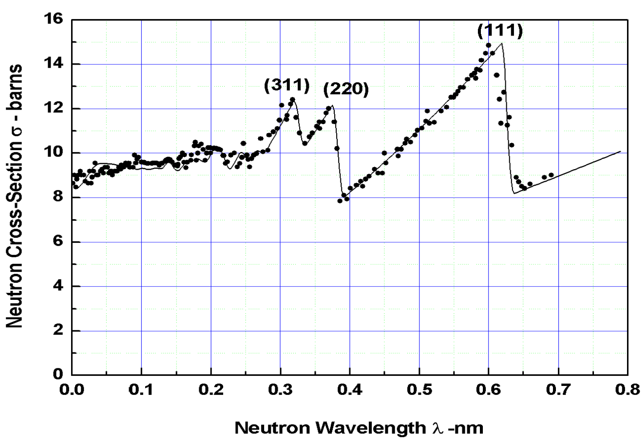


Figure 1: Total cross-section of poly-crystalline Ge.

To show the effect of both thickness and temperature of the poly-crystalline Ge on its filtering features, calculations was performed at room and liquid nitrogen (LN_2) temperatures, for neutron wavelengths in the range 0.001 – 0.8 nm. The results of those calculations are displayed in Figure 2, for room and (LN_2) temperatures.

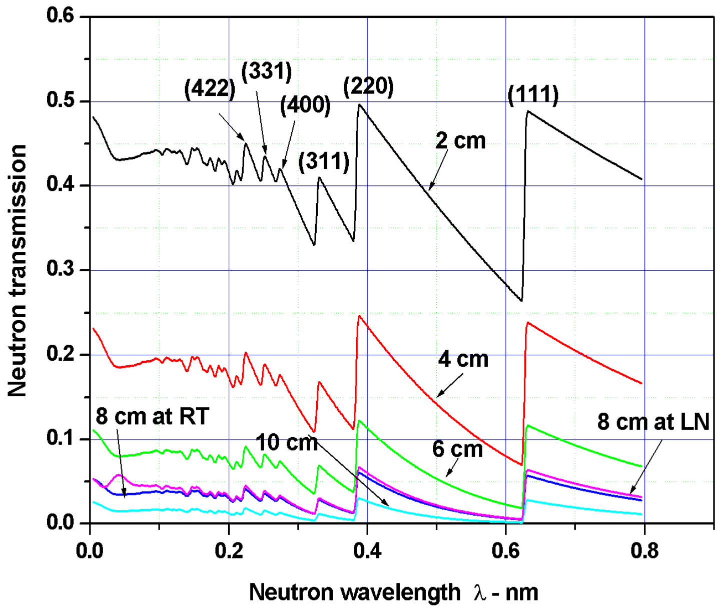


Figure 2: Neutron transmission through different thickness of poly-crystalline Ge.

The indication is that the reflections from (hkl) planes of poly-crystalline Ge are strongly disturbing the neutron transmission at any thickness. Such transmission behavior limits the application of poly-crystalline Ge when it used as a cold neutron filter.

Mono-crystalline Ge

The experimental data of mono-crystalline germanium as measured at 440 K, room, and liquid nitrogen temperatures, in the wavelength band from 0.64 nm to 0.028 nm, (energy range from 2 meV to 1 eV) presented in Adib et al.[14] are displayed in Figure. 3 as open triangles, dots and open squares respectively. Adib [14] reported that the total cross-section of a Ge mono-crystal as measured along the main axis (111) and normal to it at two different resolution values are not disturbed by Bragg reflections. They reported that such behavior may be due to the highly oriented Ge single crystals used during measurements.

The formula given by Equation 1, was fitted to experimental results for mono-crystalline germanium measured at room temperature assuming 3 cm thick perfect mono-Ge crystal cut along (111) plane. . The calculated values in the neutron wavelength range from 0.001 nm to 0.7 nm with steps of 0.001 nm and wavelength spread $\Delta\lambda = 0.01$ nm using **SGe** code, where the values of the fitting parameters C_2 and θ_D are taken the same as those for poly-crystalline case. The result of calculations is also displayed in Figure 3. as solid line. Similar calculations of total cross-section values of perfect Ge crystal at 440 K and LN_2 were also carried out using the same Ge physical parameters and values of C_2 and θ_D as those for room temperature case.

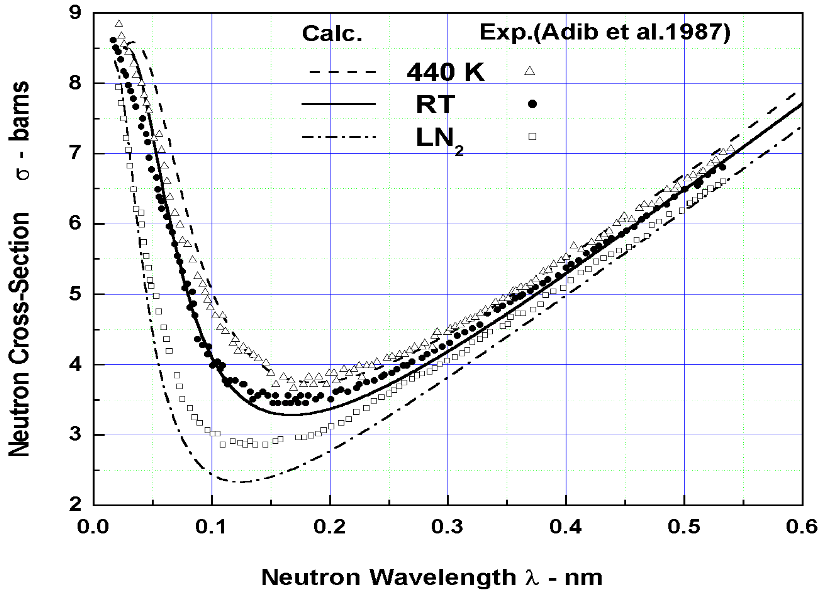


Figure 3: Total neutron cross-section of mono-crystalline germanium.

The results of calculation at 440 K and at LN_2 temperatures are also displayed in Figure 3. as dashed and dashed point lines respectively. From the figure one can notice that the agreement between the calculated values and measured ones at 440 K is reasonable. However, the calculated values at LN_2 temperature around 0.15 nm are slightly less than the measured ones. The discrepancy may be due to the fact that the sample temperature during measurement was higher than the LN_2 one. Such remark was also mentioned by Adib et al. [14].

The overall agreement obtained between the calculated Ge cross-section values at different temperatures supports the application of the DGEC code and permits calculations of the total cross-section of mono-Ge crystals with diamond structure, deduce within accuracy sufficient for determining the neutron filtering characteristics.

Since the aim of the present work is to select a Ge-single crystal filter with low enough mosaic spread and at reasonable price. Therefore, the Bragg reflections from such crystal must not be so pronounced. To decrease Bragg reflections occurring at thermal energies, an optimum choice of the crystal mosaic spread is essential. Neutron transmissions through 7.5 cm Ge (111) crystal at room temperature, for different values of mosaic spread was calculated and are displayed in Fig.(4). As may be observed, for standard deviation of mosaic spread > 0.8 mrad (0.1^o FWHM), parasitic Bragg reflections could limit the use of Ge as a thermal neutron filter.

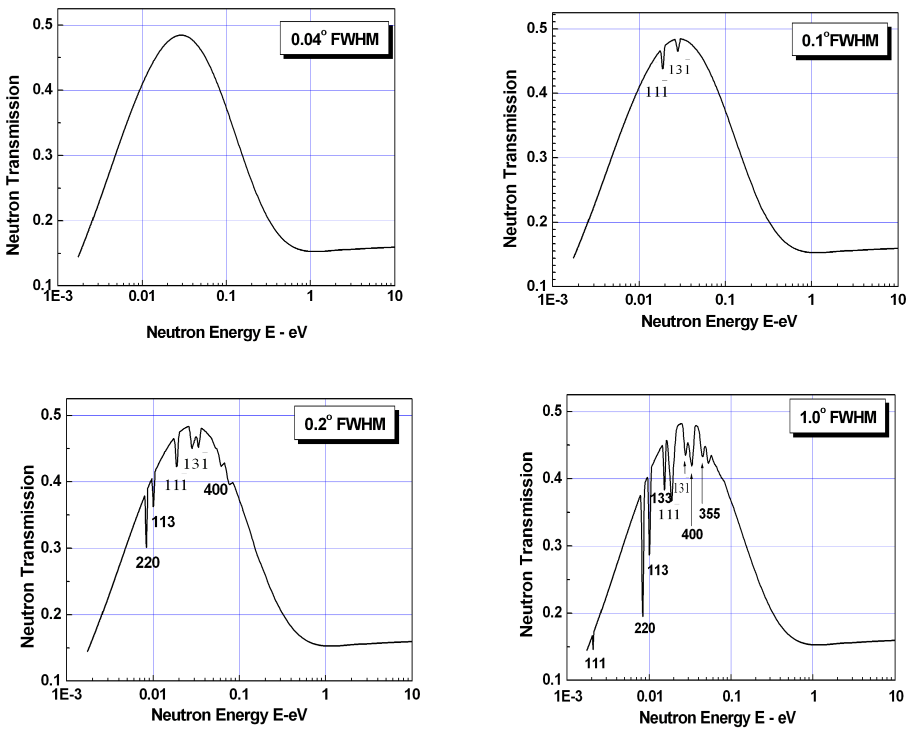


Figure 4: Neutron transmission through 7.5 cm Ge (111) crystal for various values of mosaic spreads.

From Fig.(4), one can notice that 7.5 cm Ge (0.04^o FWHM) transmits 48 % of incident thermal neutron, while at 1 eV, it transmits about 15 %. Therefore the effect-to-noise ratio of such filter is too low and consequently limits its use as a thermal neutron filter. To improve the filtering features, calculations were performed for neutron transmission through 7.5 cm Ge single (0.04^o FWHM) made from different enriched isotopes. The main parameters of different isotopes required in these calculations are listed in Table (2)[12].

Table 2: Neutron scattering length and cross-section of Ge isotopes.

Isotope	**Abundance %**	**Coherent scattering length fm**	σ_{bat} **barns**	σ_{abs} **(at 0.025 eV) barns**
^{70}Ge	**20.5**	**10.0(1)**	**12.6(3)**	**3.0(2)**
^{72}Ge	**27.4**	**8.51(10)**	**9.1(2)**	**0.8(2)**
^{73}Ge	**7.8**	**5.02(4)**	**3.17(5)**	**15.1(4)**
^{74}Ge	**36.5**	**7.58(10)**	**7.2(2)**	**0.4(2)**
^{76}Ge	**7.8**	**8.2(1.5)**	**8.0(3.)**	**0.16(2)**

The calculated transmissions for neutron energies from 1 meV to 10 eV for different Ge isotopes are displayed in Fig.(5). It is apparent 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.

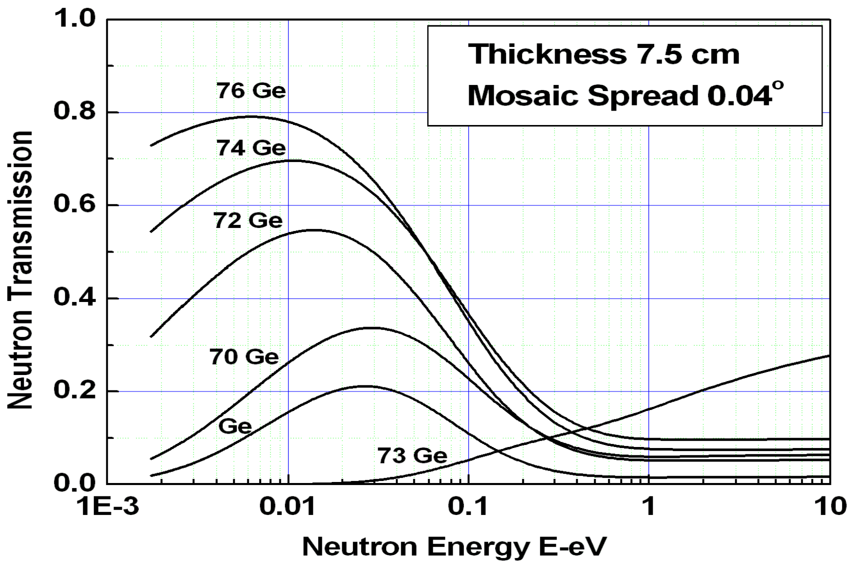


Figure 5: Neutron transmission through different Ge isotopes.

To find the optimum Ge thicknesses, the neutron transmission through different crystal thickness, at room temperature were calculated. Fig.6(a) shows the result of calculation through a ^{74}Ge (111) crystal having mosaic spread of 0.4^{o}, while Fig.6(b) shows the situation for ^{76}Ge.

It would appear that 7.5 cm thick ^{76}Ge at room temperature is sufficient for removing neutrons with energies > 1eV (T_n < 7 %), while providing high transmission (T_n > 80 %) for neutrons with energies < 0.025 eV.

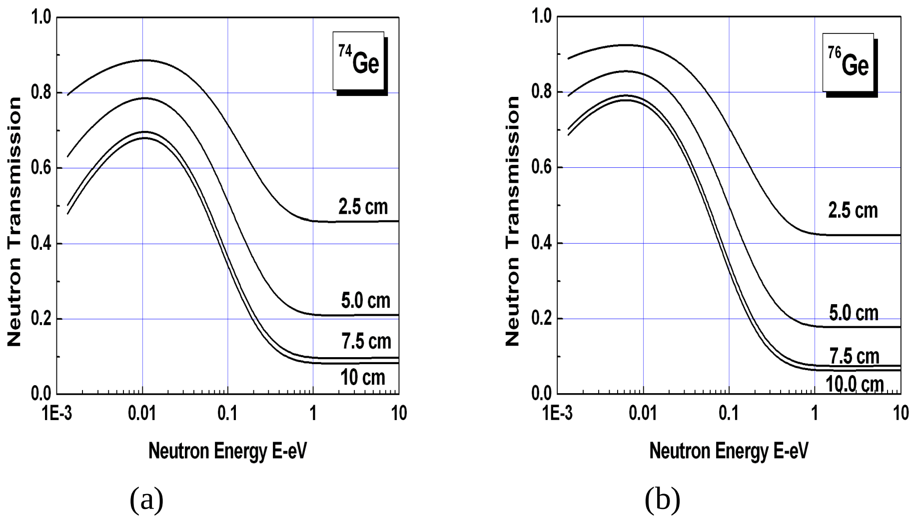
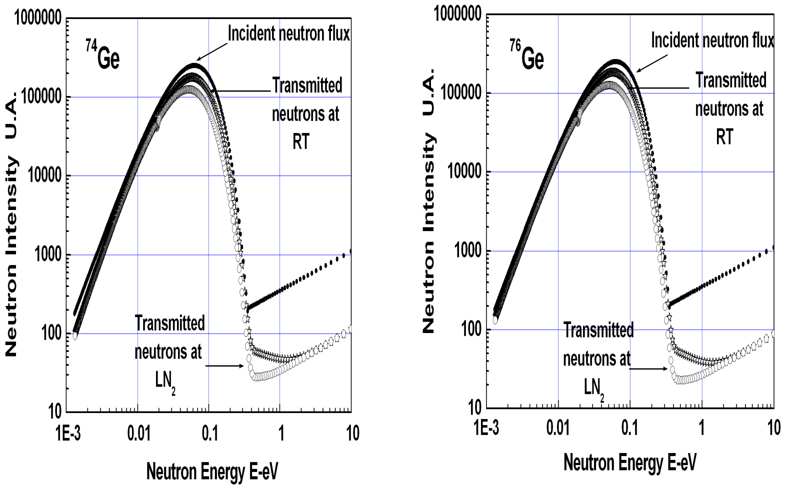


Figure 6: Neutron transmission through Ge(111)- for different crystal thickness.

Fig.(7) shows 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.(7) also shows that, there is an increase ≈ 20% at neutron energies < 0.05 eV in the 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 7: 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.

CONCLUSION

Use has been made of the simple additive formula determining the attenuation of neutrons by poly- and mono-crystalline solid, together with the **PGe** and **SGe** codes respectively, which have been developed and presented in this paper. Calculation shows that the application of poly-crystalline Ge is limited when it used as a cold neutron filter. While 7.5 cm thick ^{76}Ge single crystal cut along its (111) plane, with FWHM on mosaic spread < 0.1^{o} is a good thermal neutron filter, with high effect-to-noise ratio.

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