# = ТЕХНІКА ТА МЕТОДИ ЕКСПЕРИМЕНТУ=

УДК 539.125.5

# M. Adib<sup>1</sup>, N. Habib<sup>1</sup>, I. I. Bashter<sup>2</sup>, M. Fathallah<sup>3</sup>, M. S. El-Mesiry<sup>1</sup>, A. Saleh<sup>2</sup>

<sup>1</sup> Reactor Physics Department, Nuclear Research Center, Atomic Energy Authority, Cairo, Egypt <sup>2</sup> Faculty of Science, Zagazig University, Egypt <sup>3</sup> Faculty of Science, Jazan University, Kingdom of Saudi Arabia

## 2 keV FILTERS OF QUASI-MONO-ENERGETIC NEUTRONS

A simulation study for the production of 2 keV filters of quasi-mono-energetic neutrons based on the deep interference minima in the <sup>45</sup>Sc total cross-section was carried out. A computer code QMENF-II was adapted to calculate the optimum amounts of the <sup>45</sup>Sc as a main filter element and additional component ones to obtain sufficient intensity at high resolution and purity of the filtered quasi-mono-energetic neutrons. The emitted neutron spectrum from nuclear reactor and from the reaction of 2.6 MeV protons on a lithium fluoride target at the accelerator beam port, are used for simulation.

*Keywords:* 2 keV quasi-mono-energetic neutron beams, <sup>45</sup>Sc neutron filters.

#### Introduction

Filtered neutron mono-energetic beams in keV region now-a-days are strongly needed for high precision measurements of the total and partial cross-sections [1] & [2], determination of calibration curves of various neutron detectors for applications in neutron dosimeters and spectrometry [3]. As well as the measurements of average total and radioactive neutron capture cross section in the keV region are important in the calculation and design of nuclear reactors [4]. Moreover it is used in Boron-Neutron Capture Therapy (BNCT) [5] & [6] and more recently in Gadolinium Neutron Capture Therapy (GdNCT) [7]. Recently the 2 keV beam is applied in neutron radiography, tomography, radiobiological studies and cancer treatments [8].

The main idea of neutron filter technique is the use of large quantities of a certain material which have the deep interference minima in its total neutron cross-section. By transmitting neutrons through bulk layer of such material, one can obtain the quasi-mono-energetic neutron lines instead of white spectrum. To select only one line (neutron filter) with high purity, a composition filter of some selected additional materials is usually used.

Recently Gritzay et al. [1] and Morcos & Adib [9] reported the components of the selected materials for forming filtered reactor- neutron beam at 2 keV. As reported by Gritzay et al. [1] the main filter material for 2 keV was <sup>45</sup>Sc and the additional materials are <sup>60</sup>Ni, <sup>54</sup>Fe, <sup>59</sup>Co, <sup>10</sup>B, S and <sup>27</sup>Al. While Morcos & Adib [9] used the same amount of <sup>45</sup>Sc along with natural V or Ti material as additional materials to simulate two new filters at 2 keV. The results reported by Gritzay et al. [1] showed that the filtered beam at 2 keV is strongly disturbed at 1.385 keV and at 2.253 keV. These may be due to the selected additional elements by Gritzay et al. [1].

While filters reported by Morcos & Adib [9] have higher neutron beam intensity at much lower purity than the later. In these works the authors did not give the basis for selecting the amount of <sup>45</sup>Sc as a main filter material or the additional ones. The use of the filtered reactor beam for BNCT in some cases is limited due to the low neutron intensity at the exit of the reactor channel. Morcos & Naguib [10] simulated a 2 keV neutron filters at the beam port of accelerator using the reaction <sup>7</sup>Li (p, n)<sup>7</sup>Be with protons of energy 2.6 MeV. The authors used high amounts of enriched isotopes. The cost of such filters may limit their applications.

Therefore in the present work, the basis of selecting the optimum amount of <sup>45</sup>Sc as a main element and the additional ones for filtered beam at 2 keV are given at both the exit of nuclear reactor channel and accelerator port. Moreover a suggestion of different additional materials to obtain higher intensity and resolution of the 2 keV filtered beam at low background are also given.

#### Modeling calculation of neutron filtered spectra

A computer code QMENF-II (Quasi-<u>Monochromatic Epithermal Neutron Filter</u>) in the FORTRAN language reported by Morcos & Naguib [11] was adapted to calculate the neutron spectra formed by filters. The filter components and their amounts are optimized to get the highest possible intensity without disturbed lines of the main energy line and the lowest one of the parasitic energy lines in filtered neutron spectrum.

The input preparation contains the total crosssection data for different materials calculated by the PrePro-2010 code [12] using the JENDL-3.3 and ENDF/B-VII libraries. The output results are the transmitted neutron spectrum through the filter. The transmission T(E), of neutrons through the filter is given by

© M. Adib, N. Habib, I. I. Bashter, M. Fathallah, M. S. El-Mesiry, A. Saleh, 2014

$$T(E) = \prod_{i} T_{i}(E), \qquad (1)$$

where  $T_i(E)$  – the neutron transmission of the *i* filter component, given by

$$T_i(E) = \exp(-t_i \cdot N_0 \cdot \sigma_t(E)/A_i), \qquad (2)$$

 $t_i$  – the thickness of component filter *i*, gm/cm<sup>2</sup>;  $\sigma_t(E)$  – the total neutron cross-section, cm<sup>2</sup>/atom;  $N_0$  – Avogadro number;  $A_i$  – atomic weight, gm.

The transmitted neutron spectrum is given by

$$\phi_{\mathrm{T}}(\mathrm{E}) = \mathrm{T}(\mathrm{E}) \cdot \phi(\mathrm{E}), \qquad (3)$$

where  $\phi(E)$  – the incident neutron spectrum. The purity of the filter *P* is given by

$$p = \frac{\text{area under the main peak}}{\text{area under the whole energy range}} \times 100\%$$

Since the cross section data tables calculated by PrePro-2010 program are not at equal energy steps, so to calculate the area under curves we use the trapezoidal rule for the area under the curve calculation as

$$\sum_{a}^{b} f(x) dx = \sum_{n=0}^{n-1} \frac{f(x_{n+1}) + f(x_n)}{2} [x_{n+1} - x_n],$$
(5)

where  $x_0 = a$ ,  $x_n = b$  and N is the total number of tabulated points.

The optimum thickness t of the main filter element is determined at the peak of the rate of change in purity  $\xi(t)$ , where  $\xi(t)$  is given as

$$\xi(t) = \{P(t + \Delta t) - P(t)\} / \Delta t.$$
(6)

## **Results and discussion**

## 3.1. 2 keV filters at nuclear reactor

The incident reactor neutron spectrum  $\Phi(E)$  is taken as function composed of 3 parts: Maxwellian for thermal neutrons, 1/E – dependence for resonance one and fission spectrum. This spectrum was normalized to unit:  $\int \Phi(E) dE = 1$  in limits from  $10^{-5}$  eV to 20 MeV [1].

The filtered beam intensity of main peak (2 keV) of <sup>45</sup>Sc and its purity at different thicknesses are calculated assuming a normalized reactor spectrum [1]. The result of calculation is displayed in Fig. 1.



(4)

Fig. 1. Intensity of main <sup>45</sup>Sc peak and its purity versus thicknesses.

Table 1. Components of 2 keV filters at nuclear reactors

	Thickness, gm/cm <sup>2</sup>						
Element	Gritzay et al.	Morcos & Adib [9]	Morcos & Adib [9]	Present work	Present work		
	[1]	Filter I	Filter II	Filter (R1)	Filter (R2)		
<sup>45</sup> Sc	104.6	104.6	104.6	90	90		
<sup>54</sup> Fe	39.35	78.6	3.935	70	70		
<sup>60</sup> Ni	80.2	_	-	80	_		
<sup>59</sup> Co	26.7	_	_	20	_		
V (natural)	_	3.48	_	_	_		
Ti (natural)	_	_	22.7	_	_		
<sup>64</sup> Zn	_	_	_	_	80		
S (natural)	56.0	_	_	20	20		
<sup>27</sup> Al	0.54	8.1	4.05	10	10		
$^{10}\mathbf{B}$	0.2 (85 %)	0.2 (85 %)	0.2 (85 %)	0.2 (85 %)	0.2 (85 %)		

Fig. 1 shows that with increasing the thickness of <sup>45</sup>Sc the purity is increasing while; the transmitted neutron beam intensity is decreasing. At 104.6 gm/cm<sup>2</sup>

thickness the filtered beam intensity is only 0.0064 a.u. at purity 43.2 %.

The components of the filter suggested by

Gritzay et al. [1] are listed in Table 1. Gritzay et al. [1] reported that the main peak at 2 keV with purity  $\approx 98$  % is strongly disturbed by dips at 1.385 and 2.253 keV. These dips are caused due to neutron resonances of <sup>59</sup>Co and <sup>60</sup>Ni respectively. While the two small disturbing dips at 1.06 and 2.737 keV are due to the neutron resonances of <sup>45</sup>Sc at these energies. To eliminate such disturbing dips in the main peak Morcos & Adib [9] replaced <sup>60</sup>Ni and <sup>59</sup>Co by natural vanadium (filter I) Their filter components are also listed in Table 1.The intensity of the peak at 2 KeV is two and half times more than that reported by Gritzay et al. [1]. However, its purity is only 80 %. Morcos & Adib [9] suggested another filter (Filter II) by replacing natural V by natural Ti and the amount of <sup>54</sup>Fe was taken ten times less than that given by Gritzay et al. [1]. However, the neutron intensity of their Filter II is increased while; its purity is only 38 %.

In these works they selected the same thickness of <sup>45</sup>Sc as a main filter material and did not give the basis of selecting such amount or the additional ones.

In the present work, to select the optimum  $^{45}Sc$  thickness the rate of change in purity  $\xi(t)$  at different thicknesses is calculated. The result of calculation is displayed in Fig. 2.



Fig. 2. Rate of change in purity at different thicknesses of <sup>45</sup>Sc.

Fig. 2 shows that the optimum thickness of <sup>45</sup>Sc is 90 gm/cm<sup>2</sup>. At such thickness the intensity is 0.008 a.u. The same producer was used to select the optimum amounts of the additional materials. Using the optimum thickness of <sup>45</sup>Sc, two filters (R1 & R2) are suggested. The selected components of these filters along with those reported by Gritzay et al. [1] and Morcos & Adib [9] are listed in Table 1.

The filter is usually installed at the exit of the horizontal reactor channel, consequently neutrons emerging from a steady state reactor with energies less than 1 eV obey the Maxwellian energy distribution and their intensities are higher than the 1/E spectrum. Such neutrons are parasitic for the filter at 2 keV. To remove their contribution <sup>10</sup>B (85 %) with thickness 0.2 gm/cm<sup>2</sup> was added to the filter material. Such addition decreases the intensity of the main peak by a factor 8 %.

The distribution of the filtered beam intensity (R1&R2) was calculated using the components listed in Table 1. The result is displayed in Fig. 3. For comparison the distribution of the filter suggested by Gritzay et al. [1] is also displayed in Fig. 3 as dashed line.



Fig. 3. Transmitted Neutrons through filters R1 and R2.

The transmitted filtered beam intensity (R1) was found about 1.6 times higher than that given by Gritzay et al. [1] at purity about 98.14 %. The intensity of such filtered beam is more preferable than the later for BNCT. However, the obtained intensity difference between filter (R1) and that given by Gritzay et al. [1] for almost the same filter components may be due to the fact that the total cross-section data for different materials were calculated, in the present work, by the PrePro-2010 code [12] using the JENDL-3.3 and ENDF/B-VII libraries while, Gritzay et al. [1] by the PrePro-2002 code [12] using the JENDL-3.3 and ENDF/B-VI libraries.

The FWHM of the filtered beam (RI) as well as the disturbing dips in the peak may limit its use as mono-energetic beam for high precision measurements of the total and partial cross-sections. These disturbing dips may in some cases smoothed due to multiple scattering of neutrons in a filter, having large thickness and also due to the neutron energy resolution of the experimental facility. To overcome such filter inconveniences, Zn-64 was suggested as additional element for <sup>45</sup>Sc filter (R2).

Fig. 3, *b* shows that the filtered beam is almost free from disturbing dips and the FWHM of the peak is only 0.55 keV at purity 97.9%. However, the addition of isotope  $^{64}$ Zn as a new component of the filter is not cheap, but the filter quality is much appreciated. The final choice depends upon the experimental conditions required and the price of such filter.

As reported by Gritzay et al. [1], the neutron flux in the core of Kyiv research neutron is about  $10^{14}$  n/(cm<sup>2</sup> · s and the flux at the exit of the channel is about  $5 \cdot 10^9$  n/(cm<sup>2</sup> · s). For comparison, the filtering features of the 2 keV mono-energetic neutrons along with the purity for different filters are listed in Table 2.

Table 2. The filtering features of the 2 keV at nuclear reactor

Filter	Energy, keV	FWHM, keV	Neutron flux density $\times 10^{6} (n/cm^2 \cdot s)$	Purity, %	Gamma attenuation factor $E_\gamma \approx 2 \ MeV$
Gritzay et al. [1]	2.029	1.04	2.8	98.03	76000
Morcos & Adib [9] Filter I	2.029	1.05	7.6	81.28	490
Morcos & Adib [9] Filter II	1.87	1.11	4.9	38.86	38.0
Present work Filter R1	2.03	1.045	4.5	98.14	43800
Present work Filter R2	2.081	0.55	3.1	97.90	6800

Since the filter is usually installed at the exit of the horizontal reactor channel, consequently the accompanying  $\gamma$ -rays may limit its use for BNCT. The absorption coefficient of  $\gamma$ -rays with average energy 2 MeV was calculated for each filter. The calculated absorption coefficients are also listed in Table 2.

Table 2 shows that the suggested filters in the present work are more preferable than the others for applications. However, the low flux density and the

high accompanying gamma rays may limit their use for BNCT.

#### 3.2. 2 keV filters at accelerator beam port

In fact compared to nuclear reactors, acceleratorbased sources may allow producing epithermal neutron beams with high spectral purity and lower contamination of  $\gamma$ -rays. Moreover, the use of the lower energy accelerator may enable the operation of BNCT facilities in metropolitan areas [6].



Fig. 4. Energy spectrum simulated at the beam port [13].

The emitted neutron spectrum from the reaction of 2.6 MeV protons on a lithium fluoride target is used for simulation. The energy spectrum at the accelerator beam port reported by Minsky et al. [13] is displayed in Fig. 4.

Fig. 4 shows that the neutron flux intensity is higher than that from a nuclear reactor at lower contamination of y-rays. Moreover, the neutron spectrum with energies centered on 10 keV is considered the ideal spectrum for deep seated tumors. Such facility can treat tumors in less than 60 min.

Morcos & Naguib [10] suggested two filters with average energy 2.15 keV to be installed at accelerator beam port. The main element of the two filters was <sup>45</sup>Sc and the secondary elements were either <sup>60</sup>Ni & <sup>64</sup>Zn or <sup>76</sup>Ge respectively. The components and amounts of elements for both filters are listed in Table 3.

	Thickness, gm/cm <sup>2</sup>					
Element	Morcos & Naguib [10]	Morcos & Naguib [10]	Present work	Present work		
	Filter (2-a)	Filter (2-b)	Filter Acc1	Filter Acc2		
<sup>45</sup> Sc	2.989	110.593	90	10.0		
<sup>54</sup> Fe	_	_	70	-		
<sup>60</sup> Ni	62.314	_	50	50		
<sup>64</sup> Zn	356.65	_	150	250		
<sup>74</sup> Ge	—	61.2145	_	—		

Table 3. Components of 2 keV filters at accelerator beam port

The purity of filter (2-a) is 94 % while filter (2-b) is only 79.5 %. Moreover, Morcos & Naguib [10], used a large amounts of enriched isotopes especially <sup>64</sup>Zn as filter components. Consequently, the cost of such filters may limit their applications.

In the present work two filters Acc1 and Acc2 are suggested taking into consideration their high performance at reasonable price. Moreover the amount of isotope <sup>64</sup>Zn is selected much less than given by Morcos & Naguib [10].

less than 1 eV is too small, therefore  ${}^{10}B$  (85 %) was not added as a filter component. Moreover the contribution of neutrons with energies more than 100 keV is also too small. Consequently, natural sulfur is also omitted as a filter component.

The composition of Acc1 filter is similar to R2 one, without addition of <sup>10</sup>B and Sulfur. <sup>60</sup>Ni is added to reject neutrons with energies higher than 3 keV. The distribution of the filtered beam intensity was calculated using the components listed in Table 3. The result is displayed in Fig. 5.

Since the contribution of neutrons with energies







Fig. 6. Transmitted neutrons through filter Acc2.

Fig. 5 shows that neutron flux density through filter Acc1 has the same value as reported by Morcos & Naguib [10] however; the purity is higher than the later. To increase the neutron flux density

and resolution, filter Acc2 is also suggested. The calculated distribution of filtered beam through filter Acc2 is displayed in Fig. 6.

Filter	Energy, keV	FWHM, keV	Main peak intensity $\times 10^{6} (n/cm^2 \cdot s)$	Neutron flux density $\times 10^9 (n/cm^2 \cdot s)$	Purity, %
Morcos & Naguib [10] Filter 2-a	2.12	0.22	20.60	4.98	94.5
Morcos & Naguib [10] Filter 2-b	2.18	1.17	5.46	5.81	79.5
Present work Acc1	2.103	0.339	6.2	4.68	99.8
Present work Acc2	2.10	0.282	20.5	6.56	95

*Table 4.* The filtering features of the 2 keV at accelerator beam port

For comparison the filtering features of the 2 keV mono-energetic neutrons at the accelerator beam port along with the purity of different filters are listed in Table 4.

Table 4 shows that the suggested filters are more preferable than that given by Morcos & Naguib [10] for applications. Their high flux density and purity are appreciated for their use for BNCT.

# Conclusion

The adapted computer code was found to be sufficient for calculating the optimum component amounts of a quasi-mono-energetic neutron beam at 2 keV.

The method used to select the optimum <sup>45</sup>Sc thickness as the maximum of the rate of change in purity is proved to be useful and economic for filtering composition. Based on <sup>45</sup>Sc and <sup>64</sup>Zn as an additional element the obtained quasi-mono-energetic beams were found free from the disturbing dips.

The high flux density and purity of the suggested filters at the accelerator beam port are appreciated for the use for BNCT.

## REFERENCES

- Gritzay O., Kolotyi V., Psheniehnyi V. et al. Neutron Filter technique and its use for Fundamental and applied investigations // 6<sup>th</sup> Conf. on Nuclear and Particle Physics. NUPPAC'07 (Luxor, Egypt, 2007).
- Moreh R., Block R.C., Danon Y. Generating a multiline neutron beam using an electron Linac and a U-filter // Nucl. Instrum. Methods Phys. Res. - 2006. -Vol. A562. - P. 401 - 406.
- Nascimento F., Ramos A.R., Fernandes A.C. et al. Optimization of filtered neutron beams for the calibration of superheated droplet detectors at the RPI // Nucl. Instrum. Methods Phys. Res. - 2001. -Vol. A580(1). - P. 282 - 285.
- Tan H.V., Hai C.N., Son N.P., Anh T.T. Neutron capture cross-section measurements of <sup>109</sup>Ag <sup>186</sup>W and <sup>185</sup>Gd on filtered neutron beams of 55 and 144 keV // IAEA, INDC (VN)-011. - 2004.
- Viaggi M., Dagresa M.A., Longhino J. et al. Boron neutron capture therapy for undifferentiated thyroid, carcinoma: Preliminary results with the combined use of BPA and BOPP // J. Appl. Rad. Isot. - 2004. -Vol. 61. - P. 905 - 909.
- Bisceglie E., Colangelo P., Colonna N. et al. On the optimal energy of epithermal neutron beams for BNCT // J. Phys. Med. Biol. - 2000. - Vol. 45. - P. 49 - 58.

- Gelsomina De Stasio et al. Are gadolinium contrast agents suitable for gadolinium neutron capture therapy? // Neurological Research. - 2005. - Vol. 27. -P. 387 - 398.
- Suzuki M., Sakurai Y., Hgiwara S. et al. First attempt of boron neutron capture therapy (BNCT) for hepatocellular carcinoma // Japanese Journal of Clinical Oncology. - 2007. - Vol. 37(5). - P. 376 - 381.
- Morcos N.H., Adib M. 2 keV filters of quasimonochromatic epithermal neutrons // Journal of Nuclear and Radiation Physics. - 2011. - Vol. 6, No. 1 & 2. - P. 69 - 77.
- 10. *Morcos N.H., Naguib K.* Production of Optimal Epithermal Neutron Beams for BNCT. Sop transactions on applied physics. (In press. 2014).
- 11. *Morcos N.H., Naguib K.* QMENF- G: A computer package for quasi-mono-energetic neutron filters // Ann. Nucl. Energy. 2012. Vol. 40. P. 237 240.
- Cullen D.E. PREPRO-2010 (2010 ENDF/B Pre-Processing codes) // IAEA-NDS-39, Rev. 14, October 31, 2010.
- Minsky D., Kreiner A., Valda A. AB-BNCT beam shaping assembly based on <sup>7</sup>Li (p, n)<sup>7</sup>Be reaction optimization // J. Appl. Rad. Isot. - 2011. -Vol. 69(12). - P. 1668 - 1671.

# М. Адіб<sup>1</sup>, Н. Хабіб<sup>1</sup>, І. І. Баштер<sup>2</sup>, М. Фатхала<sup>3</sup>, М. С. Ель-Месірі<sup>1</sup>, А. Салех<sup>2</sup>

<sup>1</sup> Відділ фізики реакторів, Центр ядерних досліджень, Управління з атомної енергії, Каїр, Єгипет <sup>2</sup> Факультет природничих наук, Університет Заказік, Єгипет <sup>3</sup> Факультет природничих наук, Університету Джазан, Королівство Саудівська Аравія

# ФІЛЬТРИ КВАЗІМОНОЕНЕРГЕТИЧНИХ НЕЙТРОНІВ З ЕНЕРГІЄЮ 2 кеВ

Виконано дослідження фільтрів квазімоноенергетичних нейтронів з енергією 2 кеВ, яка відповідає глибокому інтерференційному мінімуму повних перерізів для ядра <sup>45</sup>Sc. Комп'ютерний код QMENF-II було пристосовано для розрахунку оптимальної кількості <sup>45</sup>Sc, як головного елемента фільтра, а також інших компонентів для одержання достатньої інтенсивності при високій роздільній здатності та чистоті пучка фільтрованих квазімоноенергетичних нейтронів. Для дослідження використовувалися нейтронні пучки з ядерного реактора та реакції прискорених протонів з енергією 2,6 МеВ на мішені фториду літію.

Ключові слова: квазімоноенергетичні пучки нейтронів з енергією 2 кеВ, нейтронні фільтри <sup>45</sup>Sc.

# М. Адиб<sup>1</sup>, Н. Хабиб<sup>1</sup>, И. И. Баштер<sup>2</sup>, М. Фатхала<sup>3</sup>, М. С. Эль-Месири<sup>1</sup>, А. Салех<sup>2</sup>

<sup>1</sup> Отдел физики реакторов, Центр ядерных исследований, Управление по атомной энергии, Каир, Египет <sup>2</sup> Факультет естественных наук, Университет Заказика, Египет <sup>3</sup> Факультет естественных наук, Университета Джазана, Королевство Саудовская Аравия

# ФИЛЬТРЫ КВАЗИМОНОЭНЕРГЕТИЧЕСКИХ НЕЙТРОНОВ С ЭНЕРГИЕЙ 2 кэВ

Выполнены исследования фильтров квазимоноэнергетических нейтронов с энергией 2 кэВ, соответствующей глубокому интерференционному минимуму полных сечений для ядра <sup>45</sup>Sc. Компьютерный код QMENF-II был приспособлен для расчета оптимального количества <sup>45</sup>Sc, как главного элемента фильтра, а также других компонентов для получения достаточной интенсивности при высоком разрешении и чистоте пучка фильтрованных квазимоноэнергетических нейтронов. Для исследования использовались нейтронные пучки с ядерного реактора и реакции ускоренных протонов с энергией 2,6 МэВ на мишени фторида лития.

*Ключевые слова*: квазимоноэнергетические пучки нейтронов с энергией 2 кэВ, нейтронные фильтры <sup>45</sup>Sc.

Надійшла 21.07.2014 Received 21.07.2014