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Measurement of the neutron flux parameters *f* and α at the Pavia TRIGA Mark II reactor

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Abstract

In this paper, evaluation of neutron flux parameters of TRIGA Mark II reactor in Pavia was carried out. For any of the three irradiation positions investigated, this work represented the first experimental evaluation of α. Moreover, in addition to α, values of other parameters such as *f*, $(_1)^{th}$ and $(_1)^{e}$ were also calculated and compared with the existent literature data from other TRIGA Mark II reactors and the Pavia's facility.
Results obtained in the present study represent a mandatory step ahead for future application of k_0-Neutron Activation Analysis method (k_0-NAA) at Pavia's facility.

Keywords

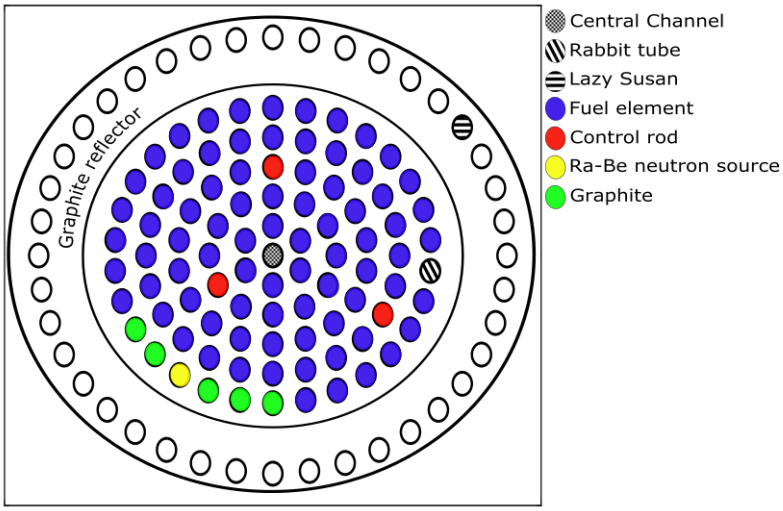
Neutron flux parameters, TRIGA Mark II, Cd-cover, k_0 method

Introduction

In the framework of the application of k_0 standardization method to Neutron Activation Analysis (k_0-NAA), reliable knowledge of two parameters of the neutron flux (f and α) is mandatory [1]. The first one indicates ratio between thermal and epithermal neutron flux while the second one provides a better representation of epithermal part of neutron spectrum shape. In this paper, evaluation of these two parameters was accomplished in three different irradiation channels of reactor TRIGA Mark II of Laboratorio di Energia Nucleare Applicata (LENA) situated in Pavia, with the aim to introduce the use of k_0-NAA in this facility on a permanent basis. In particular: the position 1 (closest to equator) of Central Channel (i), position number 27 of rotary specimen rack (codenamed Lazy Susan) (ii) and pneumatic transfer tube (codenamed Rabbit) (iii) (Fig. 1) channel's factors f and α were evaluated with methods based on irradiations with and without Cd cover.

The pool-type facility of Pavia is a 250 kW reactor working with 20% enriched ^{235}U fuel elements and moderated with demineralized light water [2].

Fig. 1 Cross section representation of TRIGA Mark II reactor channels in Pavia



Theory

Knowledge of α is of fundamental importance in order to quantify deviation from 1/E dependence of neutron spectrum in the epithermal range. The energy distribution of the neutron flux in the epithermal range, $\varphi_e(E)$, in an ideal spectrum varies as 1/E from conventional epithermal neutron flux evaluated at 1 eV (Φ_e). In real spectra, however, this dependence is not fulfilled due to core configuration and effect of moderator elements. In a real situation, the slope of function for epithermal neutrons deviates from 1/E shape, for this reason a dimensionless correction factor α has to be taken into account according to

$$\varphi_e(E) = \Phi_e \; 1/E^{1+\alpha} \quad . \tag{1}$$

To achieve experimental evaluation of α parameter, a series of flux monitors, nuclides with defined resonance energies (E_r) in the epithermal range, are used as neutron flux spots in the investigated region. In this case use of effective resonance energies ($\bar{E}_r$) is mandatory because they represent the energy of a fictitious single-resonance which provides the same activation rate of all epithermal resonances of the nuclide. In addition, they allow to consider the effect of α on resonance integrals according to

$$\bar{E}_r^{\alpha} I'_0(\alpha) = I'_0 \, , \tag{2}$$

where I'_0 and $I'_0(\alpha)$ are the reduced resonance integral in the case of ideal and real neutron spectrum, respectively [3].

From eq. (1), appearance of the epithermal production rate per target nucleus (R_e), defined as $R_e = F_{th} G_e \Phi_e I_0(\alpha)$, can be deduced. R_e indicates the amount of radionuclide produced per target nucleus per second in the nuclear reactor that is expected when a cut-off is applied to the thermal component of neutron flux, as happens in an irradiation performed under Cd cover; R_e is straightly connected with specific activity ($A_{sp,Cd}$), where the subscript Cd denotes the presence of Cd-cover. In particular, $R_e = \frac{M A_{sp,Cd}}{N_A \theta \varepsilon_p}$ if the specific activity of irradiated monitor under Cd is evaluated from detection of delayed γ-rays with gamma spectrometry; where M is the molar mass, N_A is

the Avogadro constant, θ is the isotopic abundance of target nuclide, Γ is the γ yield for 100 disintegrations, ε_p is the detector efficiency at full-energy peak.

Accordingly, $\varphi_e(\bar{E}_r)$ can be defined as follows

$$\varphi_e\left(\bar{E}_r\right) = \frac{\bar{E}_r^{-\alpha} M A_{sp,Cd}}{N_A \theta F_{Cd} G_e (\varepsilon_p \bar{E}_r I_0(\alpha)}, \quad (3)$$

where $A_{sp,Cd} = \frac{N_p / tl}{SDCw}$; N_p is the net count of full energy peak corrected for coincidence losses; $S = 1 - e^{-(ti}$, is the saturation factor; $D = e^{-(td}$, is the decay factor; $C = (1 - e^{\dagger}(-^{-}(" tc))/(tc$, is the counting factor; *tl*, *tc*, *ti*, *td* are the live time of spectrum collection, real time of spectrum collection, irradiation time and time interval between irradiation and counting respectively; λ is the decay constant of the produced radionuclide; *w* is the mass of monitor element. $I_0(\alpha)$ is the resonance integral taking into account variation of epithermal neutron flux from ideal shape 1/*E*; F_{Cd} is a correction factor taking into account the shielding of epithermal flux due to Cd cover; G_e is epithermal self-shielding effect due to the sample.

Moreover, k_0 factors can be successfully deducted. $k_1(0,\mathrm{Au})\,(i) = (M_1\mathrm{Au}\,\theta_1 i\,\sigma_1(0,i)\,(_1 i)/(M_1 i\,\theta_1\mathrm{Au}\,\sigma_1(0,\mathrm{Au})\,(_1\mathrm{Au})$, from definition of k_0 comparator, where σ_0 is the (n,γ) cross section at 2200 m s^{-1} neutron speed and subscript *i* denotes parameters referred to analyte of interest while subscript Au refers to gold, the ultimate comparator. With inclusion of k_0 factor $\varphi_e(\bar{E}_r)$ can be represented as follows:

$$\varphi_e\left(\bar{E}_r\right)\bar{E}_r = \frac{\bar{E}_r^{-\alpha} A_{sp,Cd}\, nk}{k_{0,Au}(i) F_{Cd}\, G_e\, \varepsilon_p\, Q_0(\alpha)} \quad (4)$$

Where *nk* represents a factor composed by N_A and nuclear parameters,

$nk = M_{\mathrm{I}}\mathrm{Au}/(\theta_1\mathrm{Au}\,\sigma_1(0,\mathrm{Au})\,(_1\mathrm{Au}\,N_1\mathrm{A})$, and $Q_0(\alpha)$ is the ratio of $I_0(\alpha)$ to σ_0.

Application of logarithm to eq. (4) shows the linear dependence between $\log\varphi_e\left(\bar{E}_r\right)\bar{E}_r$ and $\log\bar{E}_r$, with the first representing the dependent variable and the latter the independent variable:

$$\log[(\varphi]_e\left(\bar{E}_r\right)\bar{E}_r\right) = -\alpha\log\bar{E}_r + \log\frac{A_{sp,Cd}\, nk}{k_{0,Au}(i) F_{Cd}\, G_e\, \varepsilon_p\, Q_0(\alpha)}. \quad (5)$$

The Cd-cover method is suitable to determine α via eq. (5) by irradiating a set of monitors under a Cd cover and following counting of gamma emitted, through gamma spectrometry.

A monitor set with a wide and well distributed energy range is recommended to be chosen; for each monitor element, after irradiation and counting, eq. (5) can be applied using an initial guess value for α (α=0). Results can be plotted on a log($\varphi_e(\bar{E}_r)\bar{E}_r$) to log($\bar{E}_r$) graph and the slope of the straight line resulting from an iterative least-square regression fit leads to the determination of α [4].

For what concerns the measurement of *f*, a comparison between obtained activity from a similar monitor set deriving from the covered irradiation and a bare one can be performed. The cadmium ratio $R_{Cd}=\frac{A_{sp,b}}{A_{sp,Cd}}$, where $A_{sp,b}$ is defined as specific activity after a bare irradiation, corresponds to

$$R_iCd = (G_ith\,(_ith\,\sigma_i0 + G_ie\,(_ie\,I_i0\,(\alpha))/(G_ie\,(_ie\,I_i0\,(\alpha)F_iCd) \quad (6)$$

where Φ_{th} is the conventional thermal neutron flux and G_{th} is the self-shielding factor of the sample for thermal neutrons.

Consequently, *f* can be obtained from R_{Cd} for each monitor [5] according to

$$f = \frac{G_e}{G_{th}} Q_0(\alpha)(R_{Cd}F_{Cd} - 1) \quad (7)$$

Moreover, $(_ie$ can be obtained from eq. (4). In fact, according to eq. (1), $(_ie = \varphi_{\downarrow}e\,(E)\,E^{\uparrow}(1+\alpha)$, that leads to

$$(_ie = (A_{\downarrow}(sp,Cd)\,nk)/(\,[k_i(0,Au)\,(i)\,F_iCd\;\;G]\;_{\downarrow}e\;\;\varepsilon_ip\;\;Q_i0\;(\alpha)) \quad (8)$$

It means that the value of $(_ie$ can be determined by calculating eq. (8) on each monitor element. Thus, from the knowledge of *f*, also $(_ith$ is derived from

$$(_ith = (_ie\,f \quad (9)$$

Experimental

A similar monitor mixture composed by Au, Co, Zr and Rb was used. The amount of each monitor element was adjusted in order to obtain adequate activity with respect to the irradiation position. This monitor set allowed investigation on epithermal part of flux ranging from 5.65 to 6260 eV. (Table 1) provides information about relevant parameters of selected monitor elements.

Table 1 List of elements combined in a monitor set, all data were taken from [6] but F_{Cd} [7]. The F_{Cd} value for Rb was assumed as 1 because its $\bar{E}_r$ is distant enough from E_{Cd} to not perceive perturbation in epithermal part of flux at resonance energy. The standard uncertainties in parentheses apply to the last digits.

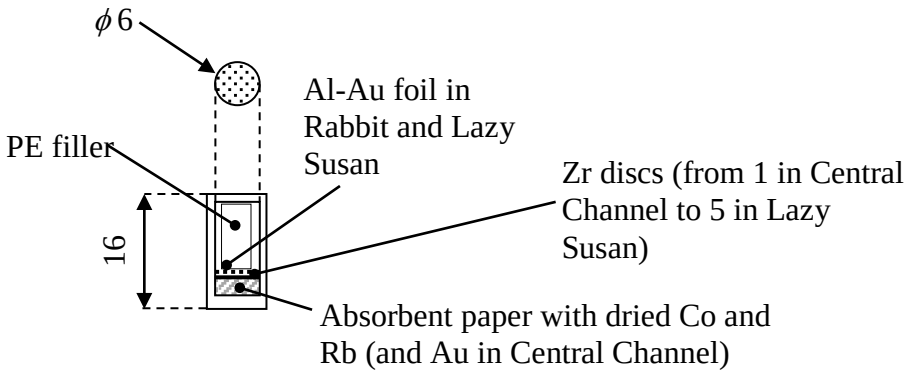
Monitor	$\bar{E}_r$ / eV	Q_0	$t_{1/2}$	E / keV	$k_{0,Au}(\alpha)$	F_{Cd}
^{197}Au(n, γ)^{198}Au	5.7(4)	15.7(3)	2.6950(2) days	411.8	1	0.991
^{59}Co(n, γ)^{60}Co	136(7)	1.99(6)	1925.3(3) days	1332.5	1.320(7)	1
^{85}Rb(n, γ)^{86}Rb	839(50)	14.8(4)	18.63(2) days	1077.0	$7.65(8) \times 10^{-4}$	1
^{94}Zr(n, γ)^{95}Zr	6260(250)	5.31(18)	64.02(6) days	756.7	$1.10(1) \times 10^{-4}$	1

A standard solution was used for Co (1000 µg mL^{-1}, VWR Chemicals) and Rb (1000 µg mL^{-1}, VWR Chemicals), while in the case of Zr, discs (0.992 g g^{-1} mass fraction, 6 mm diameter and 0.1 mm thickness, ZR000260 GoodFellow) were preferred. For Au, a 100 µg mL^{-1} solution (diluted from a 1000 µg mL^{-1} standard solution, VWR Chemicals) or an Al-Au(0.1%, IRMM-530RA) foil 0.1 mm thickness were used depending on irradiation channel. All the used standard materials were traceable to SI.

Flux monitors were irradiated and left always in the same vial also during the γ-counting with the result of a safer and simpler handling of sample in opposition with a longer data acquisition. The reason is due to the different activity between short-lived and long-lived radionuclides and the convenience to wait for quite complete decay of the firsts for a handy acquisition of the seconds. All monitors were put together in a cut 1 mL Polyethylene vial of 16 mm height (with cap) and 6 mm internal diameter; Co and Rb solutions (and Au in Central Channel irradiation) were pipetted in the vial on a stack of 12 layers of absorbent paper (width of about 2 mm) and dried with an IR lamp Zr discs

were piled above them ranging from 1 disc for Central Channel to 5 discs for Lazy Susan. Polyethylene filler, a scrap (between 4-5 mm large and 1 mm thick) cut from the wall of a 1 mL vial, was added to avoid misplacing of the setup within the vial in the following steps. (Fig. 2) shows the sample assembling scheme. Vials were sealed and put (bare or surrounded by Cd cover) at the bottom of the Polyethylene container used for irradiation. Io aggiungerei una frase dicendo che probabilmente sia il polietilene dell'irradiation container, sia del vial con cui hai preparato il campione hanno un effetto termalizzante. Anche il filler ha effetto termalizzante ma probabilmente con contributo minore date le dimensione. Al momento questi effetti non sono stati valutati ma, quando nel caso in cui questo è il setup di normale irraggiamento (e avviene nella maggior parte delle volte) il valore che stimiamo di f è quello giusto. Certo che se facessimo l'irraggiamento con il contenitore di irraggiamento in alluminio oppure senza I vial in PE (penso ad un silicio tal quale dentro al contenitore) i valori corretti di f potrebbero essere significativamente diversi. Sarebbe interessante fare misure di verifica rifacendo tutto usando l'alluminio e vedendo cosa cambia sul risultato di f.

Fig.2 Monitor set up in an irradiation vial. 1 mL vials cut at the same height were used in all three irradiation channels. All dimensions are in mm



For samples diluted enough or for low values of cross sections, as in this case, G_{th} can be approximated to 1 with negligible uncertainty. The same holds also for Au in form of foil due to the high dispersion of gold (0.1%) in the thin Al matrix (0.1 mm) and for Zr in

form of disk; $F_{Cd} = 1$ for great part of nuclides when thickness of Cd cover is 1 mm and has a cylindrical shape with height to diameter ratio of 2 and sample placed at the center of the cylinder [5], also in this case uncertainty is considered negligible. Au differs because of interaction between $\bar{E}_r$ for Au and Cd main resonance around 0.55 eV. Thus, $F_{Cd} = 0.991$. G_e was calculated according to [5] for Zr ranging from 0.99 in case of 1 disc to 0.95 in case of 5 piled discs, while, for the other monitors $G_e = 1$ because of the dispersion of Co and Rb solutions within the absorbent paper and the high dilution of Au in solution and solid phase.

For the Cd cover, a pure Cd foil with 1 mm thickness was cut and bent in a cylindrical shape with 18 mm height and 11 mm external diameter; two discs of the same diameter and 1 mm of thickness were cut and used as upper and lower caps. Cd cylinder was surrounded with an aluminum foil to maintain correct position among caps and cylindrical body.

The 'Cd-cover' irradiation always followed the 'bare' one. Delay times between two subsequent irradiations in the same channel varied from a minimum of 5 min in Rabbit tube to a maximum of 90 min in Central Channel.

All γ-spectra were acquired on a CANBERRA HPGe detector with 35% relative efficiency and an ORTEC DSPEC 502 multi-channel analyzer. Samples were placed at 6 cm from end-cap of detector to minimize γ–coincidence effects and at the same time to obtain adequate count rate for every radionuclide; collection time of spectra was adjusted in order to reach satisfying (i.e. ≈ 0.5%) statistical uncertainty due to counting. The net area of the peaks was obtained from the γ-spectra using the fitting algorithm of the ORTEC GammaVision 7 software. Energy and efficiency calibration of the detection system was performed using a point-like multi-gamma source LEA 12ML01EGMA15 placed at the same counting distance.

(i) Central Channel

For what concerns samples preparation, Au solution was used instead of solid standard because of the extremely low quantities needed in case of use of foil and the subsequent issues due to possible lack of homogeneity.

Both bare and Cd-cover irradiations lasted 30 min. The samples were dropped in the channel when 20 min have passed after the reactor reached the 250 kW critical power.

A long and a short γ-acquisition were acquired for each sample. For the bare, short counting started 1728 min after irradiation end, lasted 100 min with 7% dead time, while the long counting started 10005 min after irradiation, lasted 1440 min with dead time below 1%. The short acquisition of Cd-covered sample started 1745 min after irradiation, lasted 250 min with 6% dead time while the long acquisition started 1995 min after irradiation, lasted 2500 min with dead time below 5%.

(ii) Lazy Susan

In this case Au foil was used. It was cut in the shape of a disc with 6 mm diameter. Both bare and Cd-cover irradiations lasted 30 minutes. Samples were dropped in the channel when reactor was already at critical power.

For the bare, short γ-counting started 1398 min after irradiation end, lasted 67 min with a 6% dead time, while the long counting started 18663 min after irradiation, lasted 3666 min with a dead time below 1%. The short acquisition of Cd-covered sample started 1394 min after irradiation, lasted 67 min with 5% dead time while the longest started 1483 min after irradiation, lasted 3167 min with dead time below 3%.

(iii)Rabbit

Au foil was cut in the shape of a disc with 6 mm diameter. Three Zr discs were used in this case.

Bare and Cd-cover irradiations lasted 5 minutes. Samples were pneumatically driven in the channel when reactor was already at critical power as the usual protocol for that kind of irradiations.

The short γ-counting of bare sample started 55 min after the end of irradiation, lasted 73 min with a dead time of 4%; the long acquisition started 130 min after irradiation, lasted 1500 min with similar dead time. Short acquisition for the Cd-covered started 1650 min after the irradiation, lasted 147 min while the long acquisition started 1798 min after the irradiation end and lasted 8333 min. In both cases dead time was below 2%.

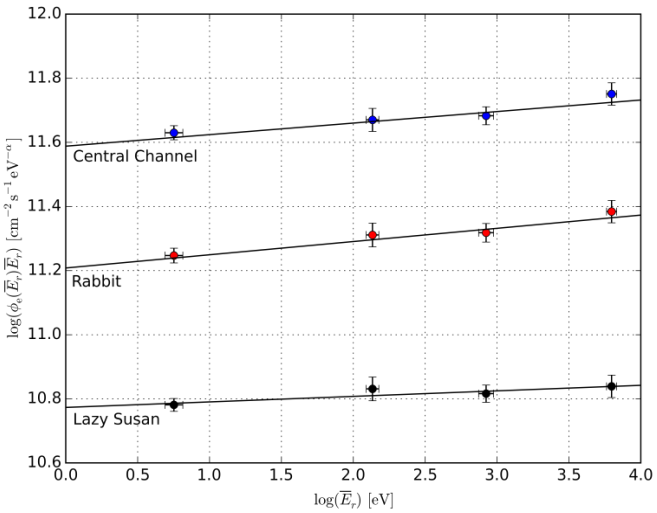
All uncertainties concerning irradiation, decay and counting times were negligible.

Results and discussion

For each set of monitor used in a defined irradiation position, the linear regression analysis of the data was performed until convergence to yield numerical values for α. The uncertainty was calculated as described by De Corte [7]. Here and hereafter, uncertainties in parentheses (k = 1) apply to the last digits.

A value of α equal to −0.036(6) was found in the Central Channel, −0.041(7) in the Rabbit and –0.016(6) in the Lazy Susan resulted. In (Fig. 3) the resulting linear regression fit on experimental values is showed for all three channels.

Fig. 3 Linear regression on data from monitor set in the different irradiation channels are showed, uncertainty bars correspond to k=2 confidence level



The presence of negative values for α indicates an imperfect thermalization of the epithermal part of the neutron flux. Specifically, the epithermal flux in the Central Channel and Rabbit tube (i.e. close to reactor core) shows a significant deviation from the ideal trend while far from reactor core, as in Lazy Susan channel, it deviates only marginally from ideality. This is in agreement with the results obtained in similar reactors, as showed in (Table 2).

Thermal to epithermal flux ratios were calculated with eq. (7) for the complete monitor set in any irradiation channel, and averaged. The f value deriving from Rb showed a clear

offset due to unidentified reasons. Thus, this monitor wasn't included in the average. The results of eq. (7) are reported in (Fig. 4) for each monitor element in the three channels. Obtained values of *f* were 15.6(3) in Central Channel, 17.4(4) in Lazy Susan and 15.7(4) in Rabbit pneumatic tube.

Fig. 4 Results of using Cd-ratio method from monitor set in different irradiation channels are showed, uncertainty bars correspond to *k*=2 confidence level

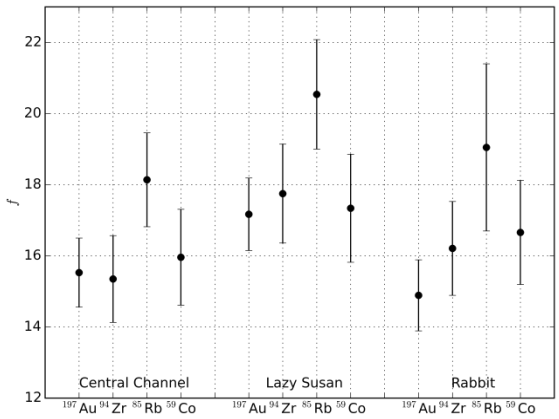


Table 2 Comparison among evaluation of *f* and α on others TRIGA facilities

	Central Channel		Rabbit		Lazy Susan	
	f	α	*f*	α	*f*	α
TRIGA Mark II Pavia (this work)	15.6(3)	−0.036(6)	15.7(4)	-0.041(7)	17.4(4)	-0.016(6)
TRIGA Mark II Ljubljana[1]	20.4(8)	−0.051(8)	19.4(7)	-0.048(5)	19.6(8)	-0.009(4)
TRIGA Mark II Morocco[8]	-	-	20.0(9)	-0.013(9)	38.3(13)	-0.017(9)

A trend in the values of *f* through the channels of Pavia reactor can be recognized starting from the less thermalized Central Channel and arriving to the most thermalized Lazy Susan. This particular behavior can be explained with the geometry of the reactor concerning the difference between inner irradiation channels (Central Channel and Rabbit are inserted in the reactor core) and outer irradiation channel (Lazy Susan is inserted in the graphite reflector and is likely to be more affected from the moderation capacity of graphite). Discrepancy with Ljubljana reactor *f* trend might derive from a different arrangement and title of fuel elements (homogeneous 20% enriched ^{235}U fuel elements in the Pavia facility against 20% and 70% enriched ^{235}U fuel elements interspersed in

Ljubljana [1]). However the uncertainties affecting the two series of *f* values don't allow to have a clearer magnitude of the trends. Moreover, from eq. (8) and eq. (9), values for α_e and α_{th} were obtained and listed in (Table 3). Also the *f*, α_e and α_{th} parameters, derived from integral flux data reported in [2] and converted according to definition of conventional fluxes described in [5], are showed in the same table.

Table 3 Thermal and epithermal conventional fluxes values obtained in this work and in previous investigations [2] after conversion of relative integral flux data; for the latter set of results, also the *f* value is showed.

		Central Channel	Rabbit	Lazy Susan
this work	Φ_{th} / cm^{-2} s^{-1}	6.11(16) × 10^{12}	2.54(7) × 10^{12}	1.02(3) × 10^{12}
	Φ_e / cm^{-2} s^{-1}	3.92(6) × 10^{11}	1.62(3) × 10^{11}	5.88(9) × 10^{10}
[2]	Φ_{th} / cm^{-2} s^{-1}	6.84 × 10^{12}	3.04 × 10^{12}	1.13 × 10^{12}
	Φ_e / cm^{-2} s^{-1}	4.26 × 10^{11}	1.90 × 10^{11}	7.93 × 10^{10}
	f	16.1	16.0	14.2

Manca commento a: After the results in Table 3, a very short discussion about the neutron flux anisotropy in each irradiation channel will be a plus. Declared uncertainties for *f* and α_e parameters were evaluated calculating the weighted uncertainty from single values obtained with propagation of variances in eq. (7) and eq. (8) respectively. For what concerns uncertainty of α_{th} , it was calculated by propagation of variances of *f* and α_e in eq. (9). Major contribution to uncertainty of *f*α_e α_{th} was found to be due to the $Q_0(\alpha)$ values of monitor elements: contribution of $Q_0(\alpha)$ on uncertainty of each singularly calculated *f* varied from about 45% for Au to about 85% for Zr and Co in all irradiation channels. $Q_0(\alpha)$ resulted the major uncertainty contribution also in α_e determination in which it was abount the 80% of the total for all monitors in every channel; however, in *f* determination, also contribution due to (R_{Cd}-1) factor is another important component, in particular for low thermalized reactors as the TRIGA Mark II is and especially for isotopes that prevalently absorb neutrons in the epithermal range (high Q_0). In fact, for *f* values calculated from Au, it contributed up to 60% to its uncertainty while for Zr and Co (low Q_0) the contribution was always below 20%.

Conclusions

With measurement of flux parameters (f and α) at the Pavia TRIGA Mark II reactor, use of k_0 standardization method can be actually exploited with samples irradiated in this facility.

The comparison of the α values here reported with those obtained in similar reactors showed good an agreement. Similarly, the f, G_e and G_{th} values derived from integral flux data reported in [2] and converted, were close to the values evaluated in this work. A relative difference of a few tens of percent was observed in the worst case.

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