

# Evaluation of thermal and epithermal neutron flux using the $k_0$ -NAA method with Gold and Zirconium monitors

## Abstract

Neutron flux analysis plays a crucial role in various nuclear research studies and sample analysis in research reactors. Accurate knowledge of the neutron flux spectrum and the ratio of thermal to epithermal neutrons is essential in these applications. In recent years, the newly  $k_0$  method has been utilized to determine thermal and epithermal neutron flux. This method involves the irradiation of two monitors, typically gold and zirconium, followed by the counting of emitted gamma rays using suitable detectors like HpGe. By analyzing the photonic peak area of the collected gamma-ray spectrum from the monitors, the thermal and epithermal neutron flux at the irradiation site can be calculated. In this study, the  $k_0$  method was employed to determine the thermal and epithermal neutron flux of Miniature Neutron Source Reactors (MNSRs). The method involved the irradiation of gold and zirconium monitors, followed by the measurement of emitted gamma rays. The photonic peak area of the gamma-ray spectrum obtained from the monitors was utilized to calculate the thermal and epithermal neutron flux. The research achieved a calculated error of 4 percent in determining the neutron flux. The  $k_0$  method, using gold and zirconium monitors, proved to be an effective approach for determining the thermal and epithermal neutron flux in the Isfahan's MNSR. The use of zirconium, with its suitable neutron absorption cross-sections, along with cadmium calculations, contributed to the accuracy of the flux determination. It is recommended to apply the  $k_0$  method in neutron and boron therapies and perform simulations using popular computational codes alongside the dual metal (gold and zirconium) measurement. This would enhance the accuracy of dosimetry calculations for absorbed doses resulting from neutron therapy.

**Keywords:**  $K_0$ , Thermal neutron, Epithermal neutron, Neutron flux, Nuclear reactor

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Mohammad Ali Shafaei, Shiva Torabi

Department of Physics, Yazd University, Iran.

**Correspondence:** Mohammad Ali Shafaei, Department of Physics, Yazd University, Yazd, Iran, Email mashafaei@yazd.ac.ir

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## Introduction

Measuring the thermal flux in a miniature reactor is not only important for educational purposes but also essential due to the decrease in neutron flux over time as fuel is consumed. To compensate for this decrease in excess reactivity, beryllium sheets are often added as reflectors. Accurate determination of the neutron flux becomes increasingly crucial during all stages of adding beryllium, both before and after. Epithermal neutron therapy, used for cancer treatment in many advanced countries, relies on simulation methods to determine the epithermal neutron flux. However, the method employed in this research offers a more precise determination of the epithermal neutron flux at any desired point, surpassing the simulation method. The flux parameters obtained through the  $k_0$  method, based on the Hogdahl convention, serve as reactor constants and enable the calculation of element concentrations with high accuracy.<sup>1,2</sup>

Accurate knowledge of the neutron flux spectrum and the ratio of thermal to epithermal neutrons is necessary for various nuclear investigations and neutron sample analyses in research reactors. Recent efforts have been focused on introducing a comprehensive method that overcomes the drawbacks of previous neutron flux determination methods. The  $k_0$  method, which divides the neutron flux spectrum into thermal and epithermal sections, has emerged as a promising approach. This method can be applied using either the Hogdahl convention or the Westcott formula. Although the Westcott formula has been preferred due to its simpler calculations, the need for accurate determination of thermal and epithermal neutron flux has led to a renewed interest in the Hogdahl convention. Currently,

most flux and flux parameter calculations for reactors are performed using computer codes and simulation programs due to the complexity of calculations based on the Hogdahl convention. However, blind reliance on these programs without a proper understanding of the underlying physical processes can lead to erroneous results. In this work, we aimed to manually perform all calculations associated with the latest method, which were typically conducted using computer codes. This approach allowed for a better understanding of the method and achieved more precise results. Specifically, we obtained the thermal and epithermal neutron flux of the Isfahan miniature reactor using the  $k_0$  method based on the Hogdahl convention, without relying on any simulation programs.

It is worth noting that the use of computational codes yielded unacceptable results, such as calculating a few milligrams of gold weight in tons. One possible reason for these incorrect results might be the failure to obtain the code from reputable international centers, which could have led to inaccuracies.<sup>3,4</sup>

## Methods and methodology

The  $k_0$  method, developed by Westcott and Hogdahl, is formulated into two relations known as the Hogdahl and Westcott relations.<sup>5-7</sup> These relations are used to determine parameters such as the ratio of thermal neutron flux to epithermal flux ( $f$ ), the epithermal flux factor ( $\alpha$ ), the thermal flux ( $G_{th}$ ), and the epithermal flux ( $G_{epi}$ ) using the Hogdahl method. Additionally, parameters characterizing the corrected spectrum, such as the Westcott factor ( $g_{tu}(T_n)$ ), and the absolute neutron temperature  $T_n$ , are determined using the Westcott

formula.<sup>8,9</sup> The  $k_0$  method, which was initially introduced to accurately determine the concentration of substances through neutron activation analysis ( $k_0$ -NAA), has found wider applications today.<sup>10,11</sup> From the beginning, gold and zirconium played a fundamental role in the  $k_0$  method due to their sensitivity to epithermal neutrons. Zirconium, due to having two isotopes ( $Zr^{94}$ ,  $Zr^{96}$ ) where  $Zr^{94}$  is sensitive to thermal neutrons and  $Zr^{96}$  is highly sensitive to epithermal neutrons, is a suitable choice alongside gold as a monitor and eliminates the need for cadmium to separate thermal from epithermal neutrons, and overall is a very suitable element for determining neutron flux parameters.

### Basis of the Høgdahl convention in the MNSR:

This section provides an explanation of the Høgdahl convention and the necessary equations for calculating neutron flux parameters ( $f$ ,  $\alpha$ ) as well as the thermal and epithermal neutron fluxes in thermal research reactors using the  $k_0$  method. Additionally, the flux parameters and thermal and epithermal fluxes in the Isfahan Miniature Reactor will be computed and the methodology will be thoroughly described based on experimental data and the relationships discussed in this section. The Høgdahl convention, in conjunction with parameters  $Q_0(\alpha)$ , serves as the foundation of the method [insert method] for detectors  $1/v$ . According to this convention, the range of the neutron reaction rate integral with matter, in terms of the energy cut-off cadmium ( $E_{cd} = 0.55$ ), is divided into two parts:

$$R = \int_0^{E_{cd}} \sigma(E)\phi(E)dE + \int_{E_{cd}}^{\infty} \sigma(E)\phi(E)dE \quad (1)$$

The choice of cadmium is based on its significant absorption cross-section for neutrons with energy below 0.55 eV (thermal neutrons) and its low absorption cross-section for neutrons with energy above 0.55 eV (epithermal neutrons). This energy threshold of 0.55 eV is known as the cadmium cut-off energy and serves as the lower limit for the integral threshold energy in cadmium. Consequently, the  $I_0$  total cross-sectional area can be defined as follows:

$$I_0 = \int_{E_{cd}}^{\infty} \frac{\sigma(E_n)dE_n}{E_n} \quad (2)$$

In an ideal scenario, the energy distribution of epithermal neutrons can be represented by  $1/E$ . However, in practical situations, this fraction is often modified and deviates from the ideal distribution. Consequently, the relationship for  $1/E^{1+\alpha}$  can be expressed as follows:

$$I_0(\alpha) = (1 \text{ eV})^\alpha \int_{E_{cd}}^{\infty} \frac{\sigma(E_n)dE_n}{E_n^{(1+\alpha)}} \quad (3)$$

Finally, the reaction rate equation in accordance with the Høgdahl convention can be expressed as follows:

$$R = \phi_{th} \sigma_0 + \phi_{epi} I_0(\alpha) \quad (4)$$

Since neutron flux reduction due to foil thickness is applicable for both thermal and epithermal neutrons, the self-shielding correction factor for the foil can be divided into thermal and epithermal components. Thus, considering the thermal self-shielding correction factor ( $G_{th}$ ) and the epithermal self-shielding correction factor ( $G_{epi}$ ), the final reaction rate relation is as follows:

$$R = G_{th} \phi_{th} \sigma_0 + G_{epi} \phi_{epi} I_0(\alpha) \quad (5)$$

In the case where the samples are chosen to be extremely thin, meaning their thickness does not exceed 100 micrometers, the values

of the thermal self-shielding correction factor ( $G_{th}$ ) and the epithermal self-shielding correction factor ( $G_{epi}$ ), that can be assumed to be 1. However, if the sample thickness exceeds this limit, these correction factors need to be calculated based on the actual sample thickness.<sup>12</sup> The flux parameter ( $f$ ), which is the objective of calculation using the  $k_0$  method, is defined as the ratio of thermal neutron flux to epithermal neutron flux.

$$f = \frac{\phi_{th}}{\phi_{epi}} \quad (6)$$

The ratio of the integral of the resonance integral (i.e., the sum of cross-sections over the resonance energy range) to the thermal neutron activation cross-section is defined as the epithermal neutron  $f$ -factor ( $Q_0$ ). In the case of considering the flux parameter ( $\alpha$ ) as a correction factor for the deviation from the ideal state of epithermal neutrons, a modified definition of the  $f$ -factor can be given as follows:

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_0} \quad (7)$$

### The fundamental equation of Høgdahl's Convention based on the $k_0$ method

As mentioned, the detection of energy spectrum and neutron flux using neutron capture in a foil is based on the generation of a gamma-emitting radioisotope, followed by the measurement of emitted gamma radiation using a High-Purity Germanium (HpGe) detector. To analyze the spectrum, it is necessary to determine the energy of each peak and the corresponding count, which represents the area under the peak. The energy allocation to the peaks is achieved through the calibration of the detector using a gamma-emitting source with known energy and intensity. The relationship between the reaction rate and the net count under the peaks can be expressed as follows:<sup>7-9</sup>

$$R = \frac{N_p/t_c}{N_A \theta \epsilon_p \gamma / M} \quad (8)$$

$N_A$ : ( $6.023 \times 10^{23} \text{ mol}^{-1} \approx$ ) Avogadro's number

$N_p$ : Net count under the peaks

$T_c$ : Counting time:

$S$ : Saturation correction factor:  $1 - e^{-\lambda t_i}$  ( $t_i$  Irradiation time,

Decay constant  $\lambda = \frac{\ln 2}{T_{1/2}}$ ,  $T_{1/2}$  Half-life)

$D$ : Counting delay correction factor  $e^{-\lambda t_d}$  ( $\lambda$  Decay factor): ( $t_d$  Decay time)

$C$ :  $\frac{(1 - e^{-\lambda t_c})}{\lambda t_c}$  = Counting correction factor

$W$ : Target mass (in grams):

$\theta$ : Relative isotopic abundance of the target (constant value)

$\epsilon_p$ : Absolute full-energy peak efficiency of the detector at the energy of interest:

$\gamma$ : Probability that in each decay of the isotope, a photon with the energy of interest is emitted (gamma emission probability)

$M$ : Atomic weight (in grams per mole):

By combining equations 5 to 8, the theoretical specific activity equation is derived as follows:

$$A_{sp} = \frac{N_A \theta \gamma}{M} [G_{th} \phi_{th} \sigma_0 + G_{epi} \phi_{epi} I_0(\alpha)] \epsilon_p \quad (9)$$

Specific activity ( $s^{-1}g^{-1}$ ) is defined by the following relation:

$$A_{sp} = \frac{N_p / t_c}{SDCW} \quad (10)$$

By combining equations 9 and 10, the concentration of an element in a sample is calculated:

$$\rho = \frac{\left( \frac{N_p / t_c}{SDCW} \right)_a \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{G_{th,m} \cdot f + G_{epi,m} \cdot Q_{0,m}(\alpha)}{G_{th,a} \cdot f + G_{epi,a} \cdot Q_{0,a}(\alpha)} \cdot \frac{\epsilon_{p,m}}{\epsilon_{p,a}} \quad (11)$$

$\rho$ : Concentration

a: Analyte (Zirconium)

m: Monitor or comparator element (Gold)

The factor  $k_0$ , as defined, is an empirical value that essentially contains the constant nuclear values needed in activation analysis calculations, such as cross-section and isotopic abundance. The factors  $k_0$  for many nuclides have been determined by equations 12 and 13.

$$k_{0,m}(a) = \frac{M_m \gamma_a \theta_a \sigma_{0,a}}{M_a \gamma_m \theta_m \sigma_{0,m}} \quad (12)$$

$$Q_0 = \frac{I_0}{\sigma_0} \rightarrow Q_0(\alpha) = \frac{Q_0 - 0.429}{\bar{E}_r^\alpha} + \frac{0.429}{E_{Cd}^\alpha (2\alpha + 1)} (1eV)^\alpha \quad (13)$$

$\bar{E}_r$ : Effective resonance energy (eV)

### Methods for determining the Factor $\alpha$

The factor  $\alpha$ , deviation of the epithermal neutron flux from the ideal state  $1/E$ , is  $1/E^{1+\alpha}$ . (When the relations  $I_0(\alpha)$  and  $Q_0(\alpha)$  are used, it means that the distribution of the epithermal neutron flux is not in an ideal state.)

$$\phi_{epi}(E) = \phi_{epi} \cdot (1eV)^\alpha / E^{1+\alpha} \quad (14)$$

$\alpha$  is independent of the neutron energy and  $1eV$  indicates the energy reference. Also,  $\alpha$  depends on the physical properties of the reactor system and its radiation facilities. In this article, only the approach of using three monitors without cadmium coating (Bare-triple monitor) based on standard  $k_0$  is explained, which is due to the use of zirconium isotopes and having a suitable absorption cross-section in thermal and epithermal energies.

Parameter determination  $Q_0(\alpha)$

$Q_0(\alpha)$  as introduced in the previous sections, It is defined as

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_0} \cdot \sigma_0, \text{ The cross section of thermal neutrons and,}$$

$I_0(\alpha)$  the resonance integral for thermal neutrons with  $1/E^{1+\alpha}$  distribution is defined as follows:

$$I_0(\alpha) = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)dE}{E^{1+\alpha}} (1eV)^\alpha \quad (15)$$

Based on the definition of the effective resonance energy  $\bar{E}_r$ , the following relationship is established:

$$I_0(\alpha) = \frac{I_0 - 0.429\sigma_0}{\bar{E}_r^\alpha} + \frac{0.429\sigma_0}{E_{Cd}^\alpha (2\alpha + 1)} (1eV)^\alpha \quad (16)$$

As a result, we will have:

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{\bar{E}_r^\alpha} + \frac{0.429}{E_{Cd}^\alpha (2\alpha + 1)} (1eV)^\alpha \quad (17)$$

As a result,  $\alpha$  and  $Q_0(\alpha)$  can be obtained by numerical calculations.

The basic technique that is intended in this work to determine the parameter  $f$  is the use of the bare bi-isotopic monitor method using two zirconium isotopes with the reactions  $^{94}Zr(n, \gamma)^{95}Zr$  and  $Zr^{96}(n, \gamma)Zr^{97} / Nb^{97m}$ .

$$f = \frac{G_{epi,1} \frac{k_{O,Au}(1)}{k_{O,Au}(2)} \times \frac{\epsilon_{p,1}}{\epsilon_{p,2}} \times Q_{O,1}(\alpha) - G_{epi,2} \frac{A_{sp,1}}{A_{sp,2}} \times Q_{O,2}(\alpha)}{G_{th,2} \frac{A_{sp,1}}{A_{sp,2}} - G_{th,1} \frac{k_{O,Au}(1)}{k_{O,Au}(2)} \times \frac{\epsilon_{p,1}}{\epsilon_{p,2}}} \quad (18)$$

This method is very suitable for determining  $f$  because it also takes into account the contribution of epithermal neutrons, but if the neutrons are well heated in the reactor irradiation systems and the amount of epithermal neutrons in the neutron flux is very small, the cadmium ratio method is more suitable.

### Determining the flux of thermal and epithermal neutrons without cadmium coating by the $k_0$ method

According to Wascott and Hogdel, the relationship that exists to determine the heat transfer ( $\Phi_{th}$ ) in terms of  $n.cm^{-2}.s^{-1}$  is as follows:<sup>5,6</sup>

$$\Phi_{th} = \frac{f \cdot A_{sp,Au}}{(f + Q_{0,Au}(\alpha)) \epsilon_{p,Au}} \quad (19)$$

In 2007, Khoo<sup>8</sup> introduced another relation for heat flux with the same parameters, which has a similar answer to relation 19:<sup>7,8,12</sup>

$$\Phi_{th} = \frac{AM}{wN_{AV} \theta \sigma_{eff}} \frac{1}{(1 - e^{-\lambda t_i})(e^{-\lambda t_d})} \frac{\lambda}{(1 - e^{-\lambda t_m}) \gamma \epsilon} \quad (20)$$

Of course, this relationship was introduced for the gold monitor that year, but now this relationship can be used for any other monitor, for example, according to the set of monitors used, any of the zirconium isotopes can also be used in this relationship. Using the definition of parameter  $f$  (Equation 6) and using any of the above relationships to calculate the flux of heat neutrons, the flux of epithermal neutrons can be calculated.<sup>7-12</sup> In the following, the relationships required to calculate neutron flux parameters ( $f$ ,  $\alpha$ ) and thermal and epithermal neutron flux in thermal research reactors, using the method without cadmium  $k_0$  coating, are presented. Finally, by using the measured experimental data and the relationships mentioned in this section, the flux parameters and thermal and epithermal fluxes in the Isfahan miniature reactor will be calculated. Because of its two special

isotopes, zirconium plays a significant role in the  $k_0$ -NAA method, because the reaction of  $Zr^{94}(n, \gamma)Zr^{95}$  to thermal neutrons and the reaction of  $Zr^{96}(n, \gamma)Zr^{97}$  to epithermal neutrons is sensitive. In fact, the ratio of integral intensity to the thermal cross-section,  $Q_0$ , for the first reaction is about 5, while for the second it is about 250, encompassing a wide range of  $Q_0$  values. Considering the  $Q_0$  value for different elements, the level of activity of that element with epithermal neutrons can be calculated. For example, for the reaction  $Zr^{94}(n, \gamma)Zr^{95}$  with  $Q_0 = 5.31$ , the radioactivity (activity) with epithermal neutrons is negligible and about 2%, for the reaction  $Au^{197}(n, \gamma)Au^{198}$  with  $Q_0 = 15.7$ , it is about 6%, and for the reaction  $Zr^{96}(n, \gamma)Zr^{97}$  with  $Q_0 = 251.6$ , it is about 86%. Therefore, the use of zirconium eliminates the need for cadmium to separate the spectrum of thermal and epithermal neutrons.

### Actual irradiation and calculations

After preparing the target materials, the next step is to select an appropriate irradiation time. Gold, due to its high cross-section for thermal neutrons, activates in a very short time, on the order of minutes. For this reason, the gold sample was exposed to neutron radiation in the reactor for only 5 minutes. Zirconium has a lower cross-section for absorbing thermal neutrons, hence the irradiation time for zirconium, with the same selected flux for gold, was set to about 5 hours. After starting the reactor and reaching the flux of  $5(10) \text{ n.cm}^{-2} \cdot \text{s}^{-1}$ ,<sup>13,14</sup> we first placed the polyethylene capsule containing the gold sample inside the sample transfer system and, noting the time, sent it to the desired internal irradiation center. After the irradiation time ( $t_i = 5$  minutes) was over, the gold sample was removed from the reactor and placed inside a lead shield. Then the capsule containing zirconium was placed inside the same sample transfer system, noting the time, it was sent to the same internal irradiation center. At the end of the irradiation time ( $t_i = 302$  minutes), the zirconium sample was also removed from the reactor and placed inside a lead shield. Considering the half-lives of the gold element and zirconium isotopes, the samples were counted the day after irradiation, three days after that, and one week after irradiation to use the best number for calculations. To count the samples, we first removed them from the lead shields, cut their polyethylene coatings using a cutter, and the samples were removed from the capsules using tweezers and then transferred to clean polyethylene capsules. On the other hand, the rabbit room in the Isfahan miniature reactor section is equipped with a system for pneumatic transfer of samples to the High Purity Germanium (HPGe) detector. We first set the distance from the end of the transfer tube to the top of the detector to 13 centimeters (to reduce the error to 5%) and calibrated the detector with a known energy source (Cobalt 60). After that, we sent the gold and zirconium capsules to the top of the detector via the transfer tubes in turn, and spectrometry began. The spectrum analysis device and the radioactivity (activity) of the Isfahan miniature reactor are located in a room next to the rabbit room and consist of a high purity germanium detector, electronics (such as, preamplifier and amplifier), and a multi-channel analyzer (MCA). The specific activity (radioactivity) for a single gold isotope and two zirconium isotopes was calculated. The measured parameters as well as the nuclear constants used to calculate specific radioactivity are presented in Table 1.

**Table 1** Shows the measured and nuclear parameters of the samples for calculating specific activity

Isotopes	E(keV)	$\gamma$	Np	w(gr)
Au <sup>197</sup>	411.8	0.955	299862	272(10 <sup>-5</sup> )
Zr <sup>94</sup>	724.2	0.441	2510	414(10 <sup>-4</sup> )
	756.7	0.545	2990	414(10 <sup>-4</sup> )
Zr <sup>96</sup>	743.3	0.979	20747	414(10 <sup>-4</sup> )

**Continues table 1**

Isotopes	ti(min)	td(min)	tc(min)	t1/2 (min)	$\lambda$ (min-1)
Au <sup>197</sup>	5	8745	21.667	3880.04	179(10 <sup>-6</sup> )
Zr <sup>94</sup>	302	1517.5	21.667	92188.8	7.25(10 <sup>-6</sup> )
	302	1517.5	21.667	92188.8	7.25(10 <sup>-6</sup> )
Zr <sup>96</sup>	302	1517.5	21.667	1014.6	683(10 <sup>-6</sup> )

Given that  $\gamma$  for the energy (keV) of the Zr<sup>94</sup> isotope is greater, we use this Zr<sup>94</sup> energy in all calculations. Using equation 8, the specific radioactivity values obtained are shown in Table 2.

**Table 2** Shows the values of specific activity

i	S <sub>i</sub>	D <sub>i</sub>	C <sub>i</sub>	A <sub>sp,i</sub>
Au <sup>198</sup>	893(10 <sup>-6</sup> )	209665(10 <sup>-6</sup> )	998067(10 <sup>-6</sup> )	272(10 <sup>8</sup> )
Zr <sup>95</sup>	2268(10 <sup>-6</sup> )	988655(10 <sup>-6</sup> )	999919(10 <sup>-6</sup> )	1.49(10 <sup>6</sup> )
Zr <sup>97</sup>	18612(10 <sup>-6</sup> )	354618(10 <sup>-6</sup> )	992635 (10 <sup>-6</sup> )	2.05(10 <sup>6</sup> )

After calculating the values related to the specific radioactivity of the used isotopes, we calculate the  $k_0$  coefficients for each one, using the following equation and the values from Table 3.

**Table 3** Shows the nuclear parameters of isotopes and their  $k_0$  values

a	M	$\gamma$	$\theta$	$\sigma_0$	$K_{0,m}(a)$
Au <sup>197</sup>	196.97	0.955	1	98.7	1
Zr <sup>94</sup>	91.22	0.545	0.1738	0.053	1.15(0 <sup>-4</sup> )
Zr <sup>96</sup>	91.22	0.979	0.028	0.0213	1.30(10 <sup>-5</sup> )

$$k_{0,m}(a) = \frac{M_m \gamma_a \theta_a \sigma_{0,a}}{M_a \gamma_m \theta_m \sigma_{0,m}} \quad (21)$$

$$\{m = Au^{197}\}, \{a = Zr^{97}, Zr^{96}, Au^{197}\} \quad (22)$$

Before we proceed with the calculations, it should be noted that the thickness of the used foils was more than 100 micrometers and it is necessary to first calculate the correction factors for the used isotopes. Because as mentioned, if we cannot ignore the thickness of the used foil, we must also consider the foil self-shielding correction factor so that the effect of the neutron flux drop inside the foil is applied to the radioactivity. The reason for applying such a correction is the possibility of absorption of thermal and epithermal neutrons in the foil that does not result in activation. Therefore, the average neutron flux inside the foil ( $\phi$ ) is always less than the average neutron flux on the surface of the thin foil under the same conditions. The foil self-shielding correction factor is defined as follows:<sup>7,8</sup>

$$G = \frac{\bar{\phi}}{\phi_0} \quad (23)$$

Since the reduction of neutron flux due to the foil thickness is possible both for thermal neutrons and epithermal neutrons, we can

generally divide the foil self-shielding correction factor into two thermal and epithermal parts. The thermal self-shielding factor ( $G_{th}$ ) is defined as follows:

$$G_{th} = \frac{\text{(average neutron thermal flux inside the foil)}}{\text{(average neutron thermal flux on the foil surface)}} = \frac{\bar{\Phi}_{th}}{\bar{\Phi}_{0,th}} \quad (24)$$

This factor is calculated using the following formula:

$$G_{th} = \frac{1 - 2E_3(\tau)}{2\tau} \quad (25)$$

In which,  $\tau = \sum_a \sigma_a t$  is the product of the macroscopic absorption cross section and the foil thickness. (The total cross-section of all the nuclei present in a unit volume of matter is called the macroscopic cross section, and ( $\Sigma = N\sigma$ ) the unit is  $cm^{-1}$ . The third part of the exponential integral functions, which are defined as follows and their values exist in the tables related to the method constants:

$$E_n(x) = \int_1^{\infty} \frac{e^{-ux}}{u^n} du \quad (26)$$

Therefore,  $E_3(\tau)$  is:

$$E_3(\tau) = \int_1^{\infty} \frac{e^{-\tau u}}{u^3} du \quad (27)$$

With a simple approximation, the following equation has also been proposed to calculate the thermal self-shielding factor:

$$G_{th} = 1 - \frac{\tau}{2} (0.923 + \ln \frac{1}{\tau}) \quad (28)$$

To calculate  $G_{epi}$ , for zirconium isotopes, the following formulas are available [10]:

For  $Zr^{96}$ :

$$G_{epi} = 1 - 1.543 \times 10^{-4} \times d + 1.143 \times 10^{-7} \times d^2 \quad (29)$$

Where  $d$  is the thickness of zirconium foils in micrometers. Also for  $Zr^{94}$ :

$$G_{epi} = 1 - 1.543 \times 10^{-4} \times d + 1.143 \times 10^{-7} \times d^2 \quad (30)$$

So, we will have:

Considering the obtained results, now we can calculate the flux parameters. The final equation for calculating the parameter  $\alpha$  by the method using 3 isotopes, without cadmium covering, is:

$$(a - b)Q_{o,1}(\alpha) \frac{G_{epi,1}}{G_{th,1}} - aQ_{o,2}(\alpha) \frac{G_{epi,2}}{G_{th,2}} + bQ_{o,3}(\alpha) \frac{G_{epi,3}}{G_{th,3}} = 0 \quad (31)$$

$$(a - b)Q_{o,1}(\alpha) \frac{G_{epi,1}}{G_{th,1}} - aQ_{o,2}(\alpha) \frac{G_{epi,2}}{G_{th,2}} + bQ_{o,3}(\alpha) \frac{G_{epi,3}}{G_{th,3}} = 0 \quad (32)$$

In which, coefficients  $b$ ,  $a$ , and also  $Q_{o,i}(\alpha)$  can be calculated from the following formulas:<sup>7,8,12</sup>

$$a = \left\{ 1 - \frac{A_{sp,2}}{A_{sp,1}} \cdot \frac{k_{o,Au}(1)}{k_{o,Au}(2)} \cdot \frac{\epsilon_{p,1}}{\epsilon_{p,2}} \right\}^{-1}$$

$$b = \left\{ 1 - \frac{A_{sp,3}}{A_{sp,1}} \cdot \frac{k_{o,Au}(1)}{k_{o,Au}(3)} \cdot \frac{\epsilon_{p,1}}{\epsilon_{p,3}} \right\}^{-1}$$

$$Q_o(\alpha) = \frac{Q_o - 0.429}{E_r^\alpha} + \frac{0.429}{E_{Cd}^\alpha (2\alpha + 1)} (1.eV)^\alpha$$

Using Table (5), parameters and  $b$  are obtained:

**Table 4** Self-shielding correction factors of gold and zirconium foils

Isotopes	d (μm)	m (gr)	M	ρ	Gepi	Gth
Au <sup>197</sup>	18	272(10 <sup>-5</sup> )	196.97	19.282	1	1
Zr <sup>94</sup>	310	414(10 <sup>-4</sup> )	91.22	6.52	0.963151	0.99926
Zr <sup>96</sup>	310	414(10 <sup>-4</sup> )	91.22	6.52	0.942153	0.99994

**Table 5** Required parameters for calculating coefficients  $a$  and  $b$

i	A <sub>spi</sub>	k <sub>o,Au</sub> (i)	ε <sub>pi</sub>	I <sub>0</sub>	Q <sub>0</sub>	E <sub>r</sub>
Zr <sup>97</sup>	2.05(106)	1.30(10 <sup>-5</sup> )	0.080797	5.28	248	338
Zr <sup>95</sup>	1.49(106)	1.15(10 <sup>-4</sup> )	0.07988	0.268	5.06	6260
Au <sup>197</sup>	2.72(1010)	1	0.14347	1550	15.71	5.65

Now, for calculating  $\alpha$ , we have:

$$a = \left\{ 1 - \frac{1.49 \times 10^6}{2.05 \times 10^6} \cdot \frac{1.3 \times 10^{-5}}{1.15 \times 10^{-4}} \cdot \frac{0.080797}{0.07988} \right\}^{-1} = 1.09064$$

$$b = \left\{ 1 - \frac{2.72 \times 10^{10}}{2.05 \times 10^6} \cdot \frac{1.3 \times 10^{-5}}{1} \cdot \frac{0.080797}{0.14347} \right\}^{-1} = 1.10759$$

Now to calculate  $\alpha$  we will have:

$$(a - b)Q_{o,1}(\alpha) \frac{G_{epi,1}}{G_{th,1}} - aQ_{o,2}(\alpha) \frac{G_{epi,2}}{G_{th,2}} + bQ_{o,3}(\alpha) \frac{G_{epi,3}}{G_{th,3}} = 0$$

$$(1.09064 - 1.10759) \left[ \frac{248 - 0.429}{(338)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] \frac{0.94215}{0.99999}$$

$$- 1.09064 \left[ \frac{5.06 - 0.429}{(6260)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] \frac{0.963151}{0.999926}$$

$$+ 1.10759 \left[ \frac{15.71 - 0.429}{(5.65)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] = 0$$

$$\Rightarrow (-0.01695) \left[ \frac{247.571}{(338)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] (0.9422)$$

$$- (1.09064) \left[ \frac{4.631}{(6260)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] (0.96322)$$

$$+ (1.10759) \left[ \frac{15.281}{(5.65)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] = 0$$

$$\Rightarrow (-0.01597) \left[ \frac{247.571}{(338)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right]$$

$$- (1.05053) \left[ \frac{4.631}{(6260)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right]$$

$$+ (1.10759) \left[ \frac{15.281}{(5.65)^\alpha} + \frac{0.429}{(0.55)^\alpha (2\alpha + 1)} \right] = 0$$

By solving the above relationship with the help of MATLAB software and choosing the best answer from the obtained answers, the final result for  $\alpha$  is as follows:

$$\alpha = -0.0174$$

After calculating  $\alpha$ , first the values of  $Q_0(\alpha)$  should be calculated for all three isotopes used:

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{\bar{E}_r^\alpha} + \frac{0.429}{E_{cd}^\alpha (2\alpha + 1)} (1eV)^\alpha$$

Zr<sup>97</sup>:

$$Q_0(\alpha) = \frac{248 - 0.429}{338^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 274$$

Zr<sup>95</sup>:

$$Q_0(\alpha) = \frac{5.06 - 0.429}{6260^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 5.83$$

Au<sup>197</sup>:

$$Q_0(\alpha) = \frac{15.71 - 0.429}{(5.65)^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 16.2$$

Now, using the above values, we can calculate the flux parameter  $f$  from equation 6:

$$\Rightarrow f = \frac{(0.9421) \frac{1.3 \times 10^{-5}}{1.15 \times 10^{-4}} \cdot \frac{0.080797}{0.07988} (274) - (0.9632) \frac{2.05 \times 10^6}{1.49 \times 10^6} (5.83)}{(0.999926) \cdot \frac{2.05 \times 10^6}{1.49 \times 10^6} - (0.999994) \cdot \frac{1.3 \times 10^{-5}}{1.15 \times 10^{-4}} \cdot \frac{0.080797}{0.07988}}$$

By solving the above equation using MATLAB software and selecting the best answer from the obtained solutions, the final result for  $\alpha$  is as follows:

$$\alpha = -0.0174$$

After calculating  $\alpha$ , first, the  $Q_0(\alpha)$  values for each of the three used isotopes should be calculated:

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{\bar{E}_r^\alpha} + \frac{0.429}{E_{cd}^\alpha (2\alpha + 1)} (1eV)^\alpha$$

Zr<sup>97</sup>:

$$Q_0(\alpha) = \frac{248 - 0.429}{338^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 274$$

Zr<sup>95</sup>:

$$Q_0(\alpha) = \frac{5.06 - 0.429}{6260^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 5.83$$

Au<sup>197</sup>:

$$Q_0(\alpha) = \frac{15.71 - 0.429}{(5.65)^{(-0.0174)}} + \frac{0.429}{(0.55)^{(-0.0174)} [2(-0.0174) + 1]} = 16.2$$

Now we can calculate the flux parameter  $f$  from equation 6 using the above values:

$$\Rightarrow f = \frac{(0.9421) \frac{1.3 \times 10^{-5}}{1.15 \times 10^{-4}} \cdot \frac{0.080797}{0.07988} (274) - (0.9632) \frac{2.05 \times 10^6}{1.49 \times 10^6} (5.83)}{(0.999926) \cdot \frac{2.05 \times 10^6}{1.49 \times 10^6} - (0.999994) \cdot \frac{1.3 \times 10^{-5}}{1.15 \times 10^{-4}} \cdot \frac{0.080797}{0.07988}}$$

By solving the above equation using Excel software, the final result for the flux parameter  $f$ , is as follows:

$$f = 20.136$$

In the last step, using the obtained flux parameters, and using the final equation for the thermal flux with the substitution of the nuclear parameters of gold and the calculated values for it, both relations 19 and 20 related to the thermal flux will have the same result as follows:

$$\varphi_{th} = 5.066 \times 10^{11} (\text{n.cm}^{-2}.\text{s}^{-1})$$

Using the definition of the flux parameter  $f$ , the epithermal flux is calculated:

$$\varphi_{epi} = \frac{\varphi_{th}}{f} = 3.13 \times 10^{10} (\text{n.cm}^{-2}.\text{s}^{-1})$$

## Result and discussion

The results obtained for the Isfahan miniature reactor, including the errors made, are shown in Table 6. In the "Isfahan" miniature reactor, the ion fission chamber is used to read the thermal neutron flux. One of these two is connected to the computer and the other to the console, and therefore, at any moment, reading the two values should be as close as possible, and as stated in the documents related to the reactor, the difference between the two should not be more than 5%. Our obtained  $k_0$  method is less than 4%. Due to the limitation in the use of reactor equipment, there was only one chance to perform this test. As a result, in order to find out how much error we had compared to the real answer, the error calculation was used using a logarithmic method. As a result, the two target materials chosen for this work were gold and zirconium. It should be kept in mind that gold (Au) is the only comparator element in the " $k_0$ " method, but before that, it was widely used in other analysis methods and because of its unique characteristics, it was often used in very accurate flux measurements. Will be some of these features are:

**Table 6** The results obtained for neutron flux and flux parameters

$\alpha_{(10^{-4})}$	$f$	$\varphi_{th} = 10^{11} (\text{n.cm}^{-2}.\text{s}^{-1})$	$\varphi_{epi} = 10^{10} (\text{n.cm}^{-2}.\text{s}^{-1})$
$174 \pm 37$	$20.14 \pm 0.23$	$5.07 \pm 0.18$	$3.13 \pm 0.36$

Simple decay scheme, mono-isotopicity, precise determination of the cross-section and decay half-life, the possibility of making very small thicknesses in order to reduce self-protection effects and appropriate half-life, which make this element widely used.

## Suggestions

- A) The combination of gold + molybdenum + rubidium (Au + Mo + Rb) can be a suitable option for the  $k_0$  method.
- B) b) Used a monitor set, which can be counted and did spectroscopy 4 to 7 days after the radiation, in a maximum period of 30 minutes.
- C) c) A set whose elements are easily activated and have a wide range of  $E_r$  should be used in calculating the flux of thermal and epithermal neutrons.
- D) In addition to the mentioned cases, choosing the same masses for the elements of the monitor set can simplify the calculations.

## References

1. Hana Ch, Dasari KB, Myung L Ch, et al. Application of  $k_0$ -INAA Method in Preliminary Characterization of KRIS Urban Airborne Particulate Matter Certified Reference Material. *Applied Sciences*. 2020;10(19):6649.
2. Measurement, simulation and uncertainty quantification of the neutron flux at the McMaster Nuclear Reactor, L. Elizabeth, MacConnachie, R. David, Novog, *Annals of Nuclear Energy* 151,107879 McMaster University, 1280 Main St. W, Hamilton, ON, Canada, 2021.
3. Zs Ravay, Kenney G. Application of the  $k_0$  method in neutron activation analysis and in prompt gamma activation analysis, *Radiochim. Acta*. 2012;100:687–698.
4. Agostino GD, M Di Luzzio, M Oddone. The  $k_0$ -INRIM software: a tool to compile uncertainty budgets in neutron activation analysis based on  $k_0$ -standardisation. *Measurement Science and Technology*. 2020;31(1).
5. Ch. Westcott, Effective Cross Section values for well-moderated thermal reactor spectra, AECL-1101, 3<sup>rd</sup> edn. Chalk River Laboratory, Chalk River, 1960.
6. Ne Holden. Temperature dependence of the Westcott  $g$  factor for neutron reactions in activation analysis. *Pure Appl Chem*. 71:2309-2315.
7. X Lin, H Gerstenberg, Ch Lierse von Gostomski, et al. Determination of  $k_0$ -values for the reactions  $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$  and  $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}$ - $^{97}\text{mNb}$  by irradiation in highly thermalized neutron flux, *Applied Radiation and Isotopes*, 2009.
8. KS Khoo, SB Sarmani, IO Abugassa. Determination of thermal to epithermal neutron flux ratio ( $f$ ), epithermal neutron flux shape factor ( $\alpha$ ) and comparator factor ( $F_c$ ) in the Triga Mark II Reactor, Malaysia. *Journal of Radioanalytical and Nuclear Chemistry*. 2007;271(2):419–424.
9. SK Samanta, A Sengupta, R Acharya, et al. Standardization and validation of  $k_0$ -based Neutron Activation Analysis using Apsara-U reactor and its application to pure iron metal and coal sample for trace element determination, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*. 2021;1018:165856.
10. F De Corte, The  $k_0$ -standardization of NAA: germs, launch, breakthrough - on stage and in the wings. *Journal of Radioanalytical and Nuclear Chemistry*. 2018;318(3):1559–1563.
11. BJ Nyarko, Y Serfor-Armah, EH Akaho, et al. Reactor Neutron Activation Analysis of Lichens by  $k_0$ -NAA Standardization method. *International Journal of Pure and Applied Physics*. 2005;1(2)181–190.
12. K Khoo Siong, S Sarmani, A Abdul Majid, et al. Assessment of Neutron Flux Gradients in Irradiation Channels at the TRIGA Reactor Au-Cr-Mo Monitor Set Based on  $k_0$ -INAA, *Sains Malaysiana*. 2008;37(4):401–404.
13. M Rezvanifard. Guide to radioisotope production by Esfahan Miniature Neutron Source Reactor, MNSR Department, Esfahan Fuel Research and Production Center, Project Number: 430, Report: RPT4-430-MAN/NR-0015, Autumn 1377 (in Persian).