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Study of non-1/v reaction nuclides using k_0 - Neutron Activation Analysis at the Malaysian Nuclear Agency Research Reactor

(Kajian Tindak Balas Nuklid yang Tidak Mematuhi Hukum 1/v Menggunakan k_0- Analisis Pengaktifan Neutron di Reaktor Penyelidikan Agensi Nuklear Malaysia)

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ABSTRACT

The modified spectral index $r(\alpha)\sqrt{T_n / T_0}$; the Westcott $g_{Lu}(T_n)$ factor and absolute neutron temperature T_n were determined for the handling of non-1/v (n, γ) reaction based on the Westcott formalism using k_0-neutron activation analysis (k_0-NAA) method at the Malaysian Nuclear Agency (MNA) research reactor. The $r(\alpha)\sqrt{T_n / T_0}$ was determined by the bare bi-isotopic monitor method using measurement of radionuclides of ^{97}Zr and ^{95}Zr. The ^{176}Lu as non-1/v and ^{197}Au as 1/v monitors were utilized for determination of $g_{Lu}(T_n)$. The $r(\alpha)\sqrt{T_n / T_0}$ and $g_{Lu}(T_n)$ values ranged from 0.0715 to 0.1417 with a RSD of 15.24% and from 1.7832 to 2.0149 with a RSD of 3.58%, respectively. The accuracy of the method was evaluated based on the calculated absolute neutron temperature (T_n) value. The calculated average value of T_n was 40.56±9.32°C while the value reported by MNA was 40°C, which represents an acceptable level of consistency.

Keywords: Malaysian Nuclear Agency; neutron flux spectra; non-1/v (n, γ) reaction nuclides; TRIGA MARK II reactor; Westcott formalism

ABSTRAK

Nilai indeks spektrum diubahsuai $r(\alpha)\sqrt{T_n / T_0}$; faktor Westcott $g_{Lu}(T_n)$ dan suhu neutron mutlak T_n telah ditentukan menggunakan tindak balas yang tidak mematuhi hukum 1/v (n, γ) berasaskan rumus Westcott, menggunakan kaedah k_0-analisis pengaktifan neutron (k_0-APN) di reaktor penyelidikan Agensi Nuklear Malaysia (ANM). Nilai $r(\alpha)\sqrt{T_n / T_0}$ telah ditentukan dengan kaedah monitor dwi-isotop menggunakan pembilangan bagi radionuklid ^{97}Zr dan ^{95}Zr. Pemonitor ^{176}Lu yang tidak mematuhi hukum 1/v dan pemonitor ^{197}Au yang mematuhi hukum 1/v digunakan sebagai penentuan $g_{Lu}(T_n)$. Nilai $r(\alpha)\sqrt{T_n / T_0}$ dan $g_{Lu}(T_n)$ yang diperoleh masing-masing berjulat 0.0715 hingga 0.1417 dengan Sisihan Piawai Relatif (RSD) ialah 15.24% dan julat 1.7832 hingga 2.0149 dengan RSD ialah 3.58%. Kejituan kaedah yang digunakan telah dinilai berdasarkan pengiraan suhu neutron mutlak (T_n). Nilai purata T_n yang diperoleh dalam kajian ini ialah 40.56±9.32°C manakala nilai yang dilaporkan oleh ANM ialah 40°C, membuktikan aras kebolehterimaan yang konsisten.

Kata kunci: Agensi Nukleur Malaysia; formula Westcott; Reaktor TRIGA Mark II; Spektrum fluks neutron; Tindakbalas nuklid tidak mematuhi hukum 1/v

INTRODUCTION

The Malaysian Nuclear Agency (MNA) TRIGA Mark II research reactor was designed for use by research institutions and universities for education purposes, non-destructive testing and isotope production. Its base consists of uranium enriched to 20% in ^{235}U and zirconium hydride (U-ZrH). For neutron activation analysis, forty rotary rack (RR) irradiation channels are located around the top portion of the core and inside the reflector (Masood et al. 2008).

The k_0-NAA method is formulated based on the Høgdahl convention and Westcott-formalism (De Corte et al. 1993; De Corte 2001; Lin et al. 1997). The Høgdahl convention is regulated to (n, γ) cross sections that follow 1/v law in the thermal neutron energy region. The parameters such as thermal to epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α) are determined by the Høgdahl convention (Høgdahl 1962).

The applicability of the Høgdahl convention is restricted to (n, γ) reactions for which Westcott's g-factor is equal to unity, i.e. for which the cross section varies as $\sigma(v) \sim \frac{1}{v}$ where v is neutron velocity. In the thermal neutron energy region, this convention excludes handling of non-1/v (n, γ) reactions of nuclides (e.g. ^{176}Lu, ^{151}Eu etc.) where Westcott's g≠1. For the k_0-NAA to be generally

applicable for all nuclides, the Westcott formalism is adopted and parameters such as the modified spectral index $r(\alpha)\sqrt{T_n/T_0}$, the Westcott $g_{Lu}(T_n)$ factor and the absolute neutron temperature T_n are determined. For calculation of $r(\alpha)\sqrt{T_n/T_0}$, and $g_{Lu}(T_n)$, the values of f and α are required (Acharya & Chatt 2003; Akaho & Nyarko 2002; Alghem & Ramdhane 2008; De Corte et al. 1993; Dung & Hien 2003; Lin et al. 1997).

In the present work f and α were determined by the bare bi-isotopic monitor and bare triple monitor methods, respectively (De Corte et al. 1969, 1980, 1981, 1986). Calculation of f proceeds as:

$$f=\frac{\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}\cdot Q_{0,1}(\alpha)-\frac{A_{sp,1}}{A_{sp,2}}\cdot Q_{0,2}(\alpha)}{\frac{A_{sp,1}}{A_{sp,2}}-\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}}, \quad (1)$$

where 1 = ^{97}Zr/^{97m}Nb (743.3 keV) and 2 = ^{95}Zr (724.2 + 756.7 keV). The ε_p is the full energy peak efficiency; $k_{0,Au}(a)$ is a nuclear constant known as the k_0-factor of analysis versus the gold (Au) monitor; the A_{sp} is the specific count rate calculated as $A_{sp}=\frac{N_p/t_c}{SDCW}$, where N_p is measures gamma net peak area (counts); S is saturation factor, calculated as $S = 1 - e^{-\lambda t_i}$, where t_i is irradiation time (s); D is the decay factor, calculated as $D = e^{-\lambda t_d}$, with t_d representing decay time (from end of irradiation to start of counting); C is the counting factor calculated as $C = [1 - e^{-\lambda t_c}]/\lambda t_c$, correcting for decay during counting, with t_c counting time; W is the mass of the irradiated element (g). The $Q_0(\alpha)$ is calculated as (De Corte et al. 1969, 1980, 1981, 1986):

$$Q_0(\alpha)=\frac{Q_0-0.429}{\bar{E}_r^{a}}+\frac{0.429}{(2\alpha+1).(0.55)^{\alpha}}, \quad (2)$$

where $\bar{E}_r$ is effective resonance energy in eV; $Q_0 = I_0/\sigma_0$ such that I_0 is the resonance integral for the (n, γ) reaction and σ_0 is the thermal neutron cross section. $\bar{E}_r$ and Q_0 are nuclear constants for each nuclide. The α is calculated by solving the following equation:

$$(a - b)\,Q_{0,1}(\alpha) - aQ_{0,2}(\alpha) + bQ_{0,3}(\alpha) = 0, \quad (3)$$

where

$$a=\left(1-\frac{A_{sp,2}}{A_{sp,1}}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}\right)^{-1}$$

and

$$b=\left(1-\frac{A_{sp,3}}{A_{sp,1}}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(3)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,3}}\right)^{-1}, \quad (4)$$

where 1= ^{97}Zr/^{97m}Nb (743.3 keV), 2= ^{95}Zr (724.2+756.7 keV) and 3= ^{198}Au (411.8 keV) (De Corte et al. 1969, 1980, 1981, 1986).

CHARACTERIZATION OF NON-1/v (*n*, γ) REACTION

The Westcott formalism based on k_0-NAA was regulated for non-1/v (n, γ) reaction nuclides. To use the Westcott formalism in the calculation of elemental concentration, the modified spectral index $r(\alpha)\sqrt{T_n/T_0}$, the Westcott's factor $g_{Lu}(T_n)$ and $S_0(\alpha)$ factor must be determined.

The modified spectral index $r(\alpha)\sqrt{T_n/T_0}$ is a measure of the epithermal-to-total neutron density ratio. It must be considered as one single parameter, and as such it can be experimentally determined by the bare bi-isotopic monitor method using a Zr monitor:

$$r(\alpha)\sqrt{T_n/T_0}=\frac{G_{th,2}\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}g_1(T_n)-G_{th,1}\cdot\frac{A_{sp,1}}{A_{sp,2}}\cdot g_2(T_n)}{G_{r,2}\cdot\frac{A_{sp,1}}{A_{sp,2}}\cdot S_{0,2}(\alpha)-G_{r,1}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}\cdot S_{0,1}(\alpha)}, \quad (5)$$

where 1 = ^{97}Zr/^{97m}Nb (743.3 keV) and 2 = ^{95}Zr (724.2 + 756.7 keV). G_{th} is the correction factor for thermal neutron self-shielding; G_r is the correction factor for resonance self-shielding; $S_0(\alpha)=\frac{S_0}{\bar{E}_r^{a}}.(1eV)^{\alpha}$, where S_0 is the corresponding quantity for an ideal 1/E epithermal neutron flux distribution.

After calculation of $S_0(\alpha)$ and $r(\alpha)\sqrt{T_n/T_0}$, the $g_{Lu}(T_n)$ factor can be determined by bare irradiation of a non 1/v monitor such as Lu and a 1/v monitor such as Au and use of the following expression:

$$g_{Lu}(T_n)=\left[\frac{\left[\frac{A_{sp}}{k_{0,Au}.\varepsilon_p}\right]_{Lu}}{\left[\frac{A_{sp}}{k_{0,Au}.\varepsilon_p}\right]_{Au}}.\left(g_{Au}(T_n)+r(\alpha)\sqrt{T_n/T_0}.S_{0,Au}(\alpha)\right)\right]-r(\alpha)\sqrt{T_n/T_0}.S_{0,Lu}(\alpha). \quad (6)$$

The T_n values can be determined by calculating the values of $g_{Lu}(T_n)$ from equation (6) and using literature values of $g_{Lu}(T_n)$ vs. T_n (Holden 1999).

According to the Westcott formalism, the concentration ρ_a (in g/g) of a Westcott element is calculated as:

$$\rho_a=\frac{\left[\frac{N_p/t_c}{SDCW}\right]_a}{A_{sp,Au}}\cdot\frac{1}{k_{0,Au}(a)}\cdot\frac{g_{Au}(T_n)+r(\alpha)\sqrt{T_n/T_0}.S_{0,Au}(\alpha)}{g_a(T_n)+r(\alpha)\sqrt{T_n/T_0}.S_{0.a}(\alpha)}\cdot\frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}}. \quad (7)$$

For calculation of the concentrations of Westcott elements such as ^{176}Lu(n, γ)^{177}Lu, ^{151}Eu(n, γ)^{152}Eu, ^{151}Eu(n, γ)^{152m}Eu, ^{153}Eu(n, γ)^{154}Eu, ^{164}Dy(n, γ)^{165m}Dy, ^{164}Dy(n, γ)^{165}Dy, ^{168}Yb(n, γ)^{169} Yb and ^{175}Lu(n, γ)^{176m}Lu, equation (7) must be utilized. These elements are rare earth elements dealt with as non-1/v (n, γ) reactions (De Corte et al. 1993).

For utilizing entirely in k_0-NAA at MNA research reactor, handling of non-1/v (n, γ) reactions nuclides and determination of Westcott parameters are necessary. In

the present investigation, we aim to show the results of the calculations for $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n using the Westcott formalism based on k_0-NAA at MNA research reactor.

Experimental Details

Monitors of Au, Zr and Lu were irradiated simultaneously in twenty rotary rack (RR) irradiation channels of MNA research reactor for determination of $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$, T_n, f, and α. The monitors used were made of Al-0.1%Au alloy wire (IRMM-527a, diameter 1 mm, length 10 mm); Zr foils (IRMM, 99.9%, 125 μm thick) and Lu foil (Santoku America Inc, 99.9%, diameter 1 mm). The monitors were cut and carefully weighed so that the Au, Zr and Lu monitors ranged in weight from 13.6 to 18.9 mg, 15.7 to 26.9 mg and 13.2 to 33.1 mg, respectively. Monitors were heat-sealed inside polyethylene vials (1 cm diameter and 1 cm length) and packed in heat-resistant plastic so that each vial included 1 Au, 1 Zr and 1 Lu monitor. The monitors were prepared in twenty RR channels and irradiated for 3 hours. After appropriate cooling, monitors were counted for gamma activity using a HPGe detector coupled with a Canberra Accuspec multichannel analyzer (MCA), and the computer codes namely "Gamma Acquisition Analysis" were used for peak area evaluation. Full energy peak efficiency calibration of the detector was carried out using ^{241}Am, ^{109}Cd, ^{57}Co, ^{137}Cs and ^{60}Co point calibration sources placed at the reference position 15.8 cm from the detector, where true coincidence effects are negligible. The energy ranged from 58.91 keV to 1332.58 keV.

Since the half-lives of radionuclides of ^{198}Au and ^{97}Zr/^{97m}Nb are short (2.695 day and 16.74 hours, respectively), both monitors were counted for about 5 minutes after one day decay time. The irradiated zirconium and lutetium monitors were counted after 3 days decay time for measurement of ^{95}Zr and ^{177}Lu over 15 minutes and 2 minutes counting time, respectively. The reactions and gamma-lines used for estimation of neutron flux parameters were: ^{94}Zr(n, γ)^{95}Zr (724.20 keV + 756.73 keV); ^{96}Zr(n, γ)^{97}Zr/^{97m}Nb (743.32 keV); ^{197}Au(n, γ)^{198}Au (411.80 keV), and ^{176}Lu(n, γ)^{177}Lu (208.4 keV). The bare bi-isotopic and bare-triple monitor methods were used (as shown in Equation (1) and (3)) to determine f and α, respectively, with the irradiation of Au and Zr monitors. The values of $r(\alpha)\sqrt{T_n/T_0}$ were calculated using the bi-isotopic method (Equation 5). The g_{Lu} (T_n) values were determined by irradiation of Au and Lu monitors and using Equation (6). The T_n values were the literature values of $g_{Lu}(T_n)$ vs. T_n (Holden 1999).

Results and Discussion

Table 1 presents the results of the calculations of $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$, T_n, f, and α at the twenty irradiation RR channels of the MNA research reactor. The $r(\alpha)\sqrt{T_n/T_0}$ and $g_{Lu}(T_n)$ values ranged from 0.0715 to 0.1417 with a relative standard deviation (RSD) of 15.24% and from 1.7832 to 2.0149 with RSD of 3.58%, respectively. The f ranged from 13.48 to 32.21 with a RSD of 20.87%, and α was found to range from -0.006 to 0.071. As shown in Table 1, the greatest deviation of α and f were observed in channels #12 and #18. These variations may have been affected by such factors as control rod height, different cores and modification in research reactor (Crow et al. 1995).

However, it is expected that the greatest variation in all of the results ($r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$, f, and α was due to the position of the monitors in RR irradiation channels. As shown in Figure 1, two positions irradiate the monitors in each RR container, so that the lower part receives more neutron flux than does the upper part because the upper part is placed at a level higher than the core (Khoo et al. 2008; Yavar et al. 2011). We note, however, that this current study involved 20 RR irradiation channels whose average values of $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$, T_n, f, and α, were calculated to obtain highly accurate results. The average values of f and α were 19.59±4.09 and 0.011±0.03, respectively, which are consistent with those found in a previous study (Wee et al. 2006) at the MNA research reactor, which found 17.2±0.9 and 0.016±0.005 for f and α, respectively.

The average values of $r(\alpha)\sqrt{T_n/T_0}$ and $g_{Lu}(T_n)$ were 0.1050±0.0160 and 1.9032±0.0681, respectively. The obtained value of $r(\alpha)\sqrt{T_n/T_0}$ was utilized for computation of $g_{Lu}(T_n)$, which in turn was used for calculation of T_n. Therefore, accuracy of the $r(\alpha)\sqrt{T_n/T_0}$ and $g_{Lu}(T_n)$ can be evaluated by the T_n results. The T_n values ranged from 24.13 to 55.86°C with a RSD of 22.99%. As shown in Figure 2, fluctuations of T_n were observed in the irradiation channels, suggesting that anisotropy flow of light water as a moderator in the core led to varied neutron temperatures in the RR channels. In the present study, the average value of T_n was 40.56±9.32°C, which was quite consistent with value of 40°C previously reported for the MNA research reactor. The T_n value found is practically equal to the temperature of the cooling water average temperature in the reactor neighborhood, as expected (De Corte et al. 1993).

Conclusion

The $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n parameters were determined at twenty RR irradiation channels of the MNA research reactor. The average values of $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n were 0.1050±0.0160; 1.9032±0.0681 and 40.56±9.32°C. Fluctuations observed in the calculated values of $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$, T_n, f, and α in the irradiation channels were mostly the effect of variations in the positioning of monitors within the RR containers, i.e. whether irradiation took place in the upper (above core level) or the lower (at core level) parts of the monitor, resulting in lower or higher neutron flux, respectively. The results of $r(\alpha)\sqrt{T_n/T_0}$ and $g_{Lu}(T_n)$ were evaluated by the value of T_n. The calculated value of T_n in present study was good conformity with the value reported for the MNA research reactor. The

parameters of $r(\alpha)\sqrt{T_n/T_0}$ and $g_{Lu}(T_n)$ were utilized for calculation of the concentration of elements which behave as a non-1/v (n, γ) reaction. The present work is the first study of the Westcott formalism based on k_0-NAA method at MNA research reactor and obtained results can be used for concentration calculation of Westcott elements in other studies which use this reactor.

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