Determination of Thermal Epithermal Neutron Fluz Ratio (*f*), and Epithermal, Neutron Flux Shape Factor (α) in the Large Sample INAA Instrumental Neutron Activation Analysis Irradiation Channel at the Gharr-1 Facility

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ABSTRACT---- ***The thermal-to-epithermal neutron ratio (f) and epithermal neutron shaping factor (α) are essential parameters when calculating the concentration of samples using the k_0-standardization method in the Large sample neutron activation analysis (LSNAA). The work was performed in a newly installed Large Sample Neutron Activation Analysis facility in the Ghana Research Reactor-1 (GHARR-1) facility using Au/Zr monitor couple. The thermal to epithermal neutron ratio was found to be 8.25 ± 0.14 and epithermal neutron shape factor (α) was also found to be 0.32998 ± 0.012. These results aided us in the determination of elemental concentration of "large samples" using the k_0-standardization method at the GHARR-1 facility.***

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1. INTRODUCTION

Determination of neutron flux in a research reactor is important in providing accurate activation analysis, present interest focusing on the use of instrumental neutron activation analysis (INAA). Neutron flux can be precisely calculated by NAA methods such as the comparator and absolute methods. For example, one of the comparator methods is by using the k_0- standardization concept [1]. This k_0-INAA method, also termed as the single comparator method and due to the characteristics of the nuclides, is set relative to the characteristics of a single nuclide (^{197}Au). Basically in the conventional bare triple monitor method, Au-Zr (reactions ^{94}Zr (n,γ) ^{95}Zr, ^{96}Zr (n,γ) ^{97}Zr and ^{197}Au (n,γ) ^{198}Au [2] monitor set is commonly used.

Neutron Activation Analysis (NAA) at the Ghana Research Reactor–1 Center currently uses a maximum mass of 0.2g (200mg) for biological sample and 0.1g (100mg) for geological samples in determining elemental concentration. To employ the use of Large Sample NAA technique, where sample masses are in kilogram ranges, a new irradiation channel has been designed, fabricated and installed at the GHARR-1 facility. A ''large sample'' in NAA is defined as a test portion in which neutron and γ-ray self-attenuation cannot be neglected in view of the required degree of accuracy.

In this work, the epithermal neutron flux shaping factor, α, and the thermal to epithermal neutron flux ratio, *f*, in the newly designed and fabricated LSNAA irradiation channel in the GHARR-1 facility is determined using the "bare triple monitor" method and the "bi-isotopic monitor" method using a combination of ^{197}Au-^{94}Zr- ^{97}Zr monitors.

2. THEORY

To determine the thermal to epithermal neutron flux ratio (*f*), epithermal neutron flux shape factor (α) for selected irradiation channels in reactor, the zirconium bare triple monitor method using k_0 factors had been used. The use of Zr as

a multi-isotopic flux ratio monitor has some distinct advantages, as compared with other flux monitors. Lately, by combining Zr with Au, the epithermal neutron flux shape factor (α) could be determined.

The combination of ^{197}Au-^{94}Zr-^{97}Zr monitors has been applied due to no true coincidence corrections were needed even if small distances between source and detector were used. Neutron flux parameter is based on measuring the induced radioactivity of the irradiated Zr foil [3].

The bare bi-isotopic using Zr method

When using the bare bi-isotopic (Zr) method, the Zr monitors with reaction ^{94}Zr (n,γ) ^{95Zr} and ^{96}Zr (n,γ) ^{97}Zr/^{97m}Nb are used to calculate *f*:

$$f = \frac{G_{ep,^{95}Zr}\dfrac{k_{0,Au}\left(^{95}Zr\right)\varepsilon_{p,^{95}Zr}}{k_{0,Au}\left(^{97}Zr\right)\varepsilon_{p,^{97}Zr}}Q_{0,^{95}Zr}(\alpha) - G_{ep,^{97}Zr}\dfrac{A_{sp,^{95}Zr}}{A_{sp,^{97}Zr}}Q_{0,^{97}Zr}(\alpha)}{G_{th,^{97}Zr}\dfrac{A_{sp,^{95}Zr}}{A_{sp,^{97}Zr}} - G_{th,^{95}Zr}\dfrac{k_{0,Au}\left(^{95}Zr\right)\varepsilon_{p,^{95}Zr}}{k_{0,Au}\left(^{97}Zr\right)\varepsilon_{p,^{97}Zr}}} \quad (1)$$

where G_{ep} and G_{th} are the correction factors for epithermal and thermal neutron self-shielding, and A_{sp} = (peak area)/($SDCwt_c$). *S*, *D*, *C*, *w* and t_c are saturation (build-up) factor, decay factor, counting factor, mass of target element and counting time, respectively. $Q_0 = I_0/\sigma_0$, with I_0 the resonance integral and σ_0 the thermal (2200 m.s–1) cross section.

The "bare triple monitor" method

When using the bare triple monitor method, the a-calculation is based on the reactions presented in Table 1 using the following equation:

$$(a-b)Q_{0,^{95}Zr}(\alpha)\frac{G_{ep,^{95}Zr}}{G_{th,^{95}Zr}} - aQ_{0,^{97}Zr}(\alpha)\frac{G_{ep,^{97}Zr}}{G_{th,^{97}Zr}} + bQ_{0,^{198}Au}(\alpha)\frac{G_{ep,^{198}Au}}{G_{th,^{198}Au}} = 0 \quad (2)$$

Where

$$a = \left(1 - \frac{A_{sp,^{97}Zr}k_{0,Au}\left(^{95}Zr\right)\varepsilon_{p,^{95}Zr}}{A_{sp,^{95}Zr}k_{0,Au}\left(^{97}Zr\right)\varepsilon_{p,^{97}Zr}}\right)^{-1}$$

$$b = \left(1 - \frac{A_{sp,^{198}Au}k_{0,Au}\left(^{95}Zr\right)\varepsilon_{p,^{95}Zr}}{A_{sp,^{95}Zr}k_{0,Au}\left(^{198}Au\right)\varepsilon_{p,^{198}Au}}\right)^{-1}$$

$$Q_{0,i}(\alpha) = \frac{Q_{0,i} - 0.429}{\left(\overline{E}_{r,i}\right)^{\alpha}} + \frac{0.429}{(2\alpha+1)(0.55)^{\alpha}} \quad (3)$$

Where $\overline{E}_r$ is the effective resonance energy (eV) for the nuclide. [4,5,6.7,8,9]

The comparator factor (F_c) can be predicted by using equation below

$$F_{c,Au} = \frac{N_A\theta_{Au}\gamma_{Au}\sigma_{0,Au}}{M_{Au}}\phi_e 10^{-6} \approx \frac{\phi_e}{(3.47)(10^6)} \quad (4)$$

where N_A is the Avogadro's number, M is the molar mass, θ is the isotopic abundance, σ_0 is the (n,γ) cross section and γ is the absolute gamma-intensity.

The ϕ_{th} is the thermal neutron flux ($cm^{-2}s^{-1}$); the thermal neutron flux (ϕ_{th}) and epithermal neutron flux (ϕ_{epi}) are calculated respectively as follows [5];

$$\phi_{th} = \frac{f \cdot A_{sp,Au} \cdot 3.47}{\left(f + Q_{0,Au}(\alpha)\right) \cdot \varepsilon_{p,Au}} \quad (5)$$

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

Table I, Monitors and relevant nuclear data (Soliman et al., 2011, Dung and Sasajima, 2003)

Monitor	E_r, eV	Q_0	Half-Life	E, keV	$k_{0,Au}$
$^{197}Au(n,\gamma)^{198}Au$	5.65	51.71	2.695 d	411.8	1
$^{96}Zr(n,\gamma)^{97}Zr/^{97m}Nb$	338	248	16.9 h	743.3	1.30×10^{-05}
$^{94}Zr(n,\gamma)^{95}Zr$	6260	5	64.02 d	724.2	9.32×10^{-05}
				756.7	1.15×10^{-04}

3. EXPERIMENTAL

The thermal to epithermal neutron flux ratio (*f*) and epithermal neutron flux shape factor (α) used in k_0-NAA were determined using special Au/Zr technique. High purity (97.2%) Zr wire, diameter 0.5mm and reference material Al–0.1% Au alloy wire (IRMM-527a) diameter 1 mm, length 10 mm were taken into the polyethylene vials.

The samples were lowered manually into the newly designed, fabricated and installed large sample INAA irradiation site. The samples were irradiated for 4 hours after which the Zr monitor was counted immediately to determine the ^{97}Zr isotope (^{96}Zr (n,γ) ^{97Zr}) which has a half-life of 16.74 hours. The ^{197}Au monitor was counted after some few hours to determine the ^{198}.Au isotope and the Zr monitor was counted again after some few days to determine the ^{95}Zr isotope which has a half life of 64.02 days.

After appropriate cooling, the monitors were counted for gamma activity using an n-type high purity germanium (HPGe) detector Model GR 2518 (Canberra Industries Inc.) with a resolution of 1.8 keV (FWHM) for ^{60}Co gamma-ray energies of 1332 keV. The detector operated at a bias voltage of (-ve) 3000 V with relative efficiency of 25% to NaI detector. A Microsoft window based software MAESTRO (Maestro for windows model A 65-B 32, Version 6.05, Advanced Measurement Technology, Inc.) was used for the spectrum analysis. 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 10 minutes after one day decay time. The irradiated zirconium was counted for 10 hours after a week of decay time for the measurement of ^{95}Zr.

4. RESULTS

The irradiation site has been characterized to determine the epithermal shaping factor, α, the thermal-epithermal flux ratio, *f*, and the thermal flux in the irradiation site. The results are shown in the Table 2 below. The flux ratio, *f*, the epithermal neutron shaping factor, α, the thermal neutron flux, ϕ_{th}, and the epithermal neutron flux, ϕ_{epi} were calculated from experimental values using the relations in Equations (1), (2), (5) and (6) respectively

Table 2: Determination of f and α in the LS-NAA irradiation site in the GHARR-1 facility

	f	α	ϕ_{th} (10^9 n cm^{-2} s^{-1})	ϕ_{epi} (10^8 n cm^{-2} s^{-1})
LS-INAA channel	8.247 ± 0.14	0.32998 ± 0.012	6.23	7.56

5. CONCLUSION

The characterization of a newly installed irradiation channel for a large sample neutron activation analysis in the Ghana Research Reactor-1 (GHARR-1) facility is presented. For the determination of elemental concentration of Large samples using the k_0-standardization method, an accurate thermal flux value is very essential. Therefore having accurate f and α in the irradiation site is a necessity. In this work the combination of ^{197}Au-^{94}Zr- ^{97}Zr monitors was used in the determination of f and α, employing the "bare bi-isotopic using Zr method" and the "bare triple monitor" method. The average thermal neutron flux and the epithermal neutron flux in the irradiation site was measured to be 6.23 x 10^9 n $cm^{-2}s^{-1}$ and 7.56 x 10^8 n $cm^{-2}s^{-1}$ respectively. The thermal-to-epithermal neutron flux ration, f, and the epithermal neutron shaping factor, α, was measured to be 8.247±0.14 and 0.32998±0.012 respectively. The obtained results would be subsequently applied in future work using k_0 in Large Sample-NAA at the GHARR-1 facility.

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7. REFERENCES

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