Neutron Flux Variation at the Inner Irradiation Channel of the Nigeria Research Reactor-1 (NIRR-1)

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Abstract: In order to ascertain the level of flux variation in one of the inner irradiation channels of the Nigeria Research Reactor-1 (NIRR-1), the irradiation container used for routine activation analysis was employed with copper wires as flux monitors. Measurements were carried out with these wires arranged in axial direction to determine the thermal neutron flux at selected positions using absolute foil activation method. Our results show that there exists a slight flux variation from one position to another ranging from (4.57 ± 0.24) x 10^{11} to (5.20 ± 0.20) x 10^{11} $cm^{-2}s^{-1}$. Individual foil shows slight flux variation from one position to another in the same irradiation container but they all pointed toward a level of stability in spite of the recent installation of the cadmium lined irradiation channel. The values obtained in this work are in good agreement with the previously measured value of (5.14 ± 0.24) x 10^{11} $cm^{-2}s^{-1}$ after commissioning of NIRR-1. This shows that the cadmium lined installation does not affect the flux stability. In order to improve the accuracy of neutron activation analysis (NAA) using NIRR-1 facility, there is need for flux corrections to be made by miniature neutron source reactor (MNSR) users during NAA particularly long irradiation, where more than six samples are irradiated simultaneously in the same container.

Keywords: Nuclear Flux, Irradiation Channel, NAA. NIRR-1.

INTRODUCTION

The only Research Reactor in Nigeria is the Nigeria Research Reactor-1 (NIRR-1), which is a Miniature Neutron Source Reactor (MNSR) installed at the Centre for Energy Research and Training, Ahmadu Bello University, Zaria. The NIRR-1 was acquired extensively for soil fertility and geochemical mapping project in Nigeria and has also been used over the years for neutron activation analysis [1], with an increasing interests in the determination of cross section and trace elements in many fields of science and technology [2, 3]. In this perspective, NAA using reactor neutrons plays a vital role due to its high sensitivity and detection limits for many elements in a variety of matrices [4], but these could not be achieved without a proper knowledge of the neutron flux. For proper utilization of this reactor, parameters such as neutron flux, φ, thermal to epithermal flux ratio, f and neutron spectrum shaping factor, α, has to be monitored regularly.

The Nigeria Research Reactor-1 (NIRR-1) is a tool for neutron activation analysis and thermal neutrons caused the majority reactions of interest in the activation analysis, with the reaction rate being proportional to the neutron flux [5]. Any quantitative activation measurements will rely on the neutron flux of the reactor [6]. The distribution of the neutron flux however varies from one sample position to the other and from one irradiation channel to the next depending on the reactor type, core configuration, axial and radial distribution of the neutrons. This distribution does not remain constant even over relatively short distances [7] i.e. the neutron flux falls off with distance in both vertical and horizontal directions away from the reactor core. However, the neutron flux in a particular irradiation channel will depend on its distance from the reactor core. Therefore, determination of accurate information on the thermal neutron flux values to which the sample is exposed was necessary to obtain the accurate elemental concentrations.

Neutron flux measurement can be done either by relative or absolute method, but the former does not provide an absolute value of neutron flux at various points. However, the latter provides the absolute neutron flux in the irradiation channel or at some positions in the capsule [8]. As reported earlier [9], this drawback of the relative method has prompted an investigation into information about neutron flux in the irradiation position, and the nuclear data concerning the target and product nuclide that would be required to calculate element masses from the gamma-ray spectrum. But recent developments have shown that uncertainties in nuclear data are the major source of systematic errors in activation analysis using absolute method [10]. However, developments in nuclear data and in measurements of neutron flux parameters allow uncertainties to be reduced to below 10% [9].

An experimental method was designed in this work to obtain the thermal flux in the B2-inner irradiation

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channel with similar foil arrangement to the recent work done in the Ghana research reactor-1 (GHARR-1) [11]. For quantitative calibration in neutron activation analysis, several approaches exist; such as relative method, comparator method and absolute method. In this work, an absolute foil activation method is employed to determine the axial neutron flux distributions in this channel. When sample is irradiated with neutrons, the activation rates depend on the geometry effect due to irradiation position within the capsule, the variation and the differences within the irradiation site [3]. Neutron flux variation within the irradiation container could introduce an uncertainty in NAA if appropriate measures are not taken; hence, successful application of neutron activation analysis can only be guaranteed if flux monitoring is performed in a regular basis for analytical quality control. The neutron flux at B2 inner irradiation channel was reported to be (5.14 ± 0.24) x 10^{11} $cm^{-2}s^{-1}$ [12], but after more than 8 years of successful operation of the NIRR-1 facilities and recently the installation of the epi-cadmium lined channel in the large outer irradiation channel (Figure **1**) for epithermal activation analysis, we envisage the need to re-measure the flux in this channel to guarantee its application in neutron activation analysis with the aim of investigating its variation from one sample position to another in the axial direction.

THEORETICAL CONSIDERATION

The reaction rate in a thermal neutron flux of known density n can be calculated from the knowledge of the cross section σ_0 at a particular velocity v_0, provided that σ is proportional to $1/v$. The thermal flux distribution $\Phi_{th}(E)$ as a function of energy after the neutrons reach thermal equilibrium with the moderator atoms is Maxwellian distribution given by:

$$\Phi_{th}(E) = \frac{E}{E_m^2}\exp\left(-\frac{E}{E_m}\right) \quad (1)$$

where $\Phi_{th}(E)$ is the neutron flux at energy E, normalized to unit integral flux. $E_m = kT_m$ represents the neutron energy, corresponding to the most probable velocity v_0, and k is the Boltzmann's constant.

In absolute method, the thermal neutron flux is given as [13];

$$\varphi_{th} = \frac{N_p M}{wN_a\gamma\theta\varepsilon_p}\frac{1}{(1-e^{-\lambda t_i})e^{-\lambda t_d}}\frac{\lambda}{(1-e^{-\lambda t_m})c\sigma_{eff}} \quad (2)$$

N_p the net number of counts under the full-energy peak during counting time, t_m, w is the weight of irradiated element, $S = 1-e^{-\lambda t_{irr}}$ the saturation factor, $D = e^{-\lambda t_d}$ the decay factor with t_d being the decay time, $C = (1-e^{-\lambda t_m})/\lambda t_m$ the measurement factor correcting for decay during the measurement time, t_m, M the atomic weight, λ is the decay constant, θ the isotopic abundance, N_a the Avogadro's number, γ the absolute gamma-ray emission probability, ε_p the full energy peak detection efficiency, c is the concentration of the analyte and σ_{eff} is the effective neutron cross section in cm^2 as defined below [13];

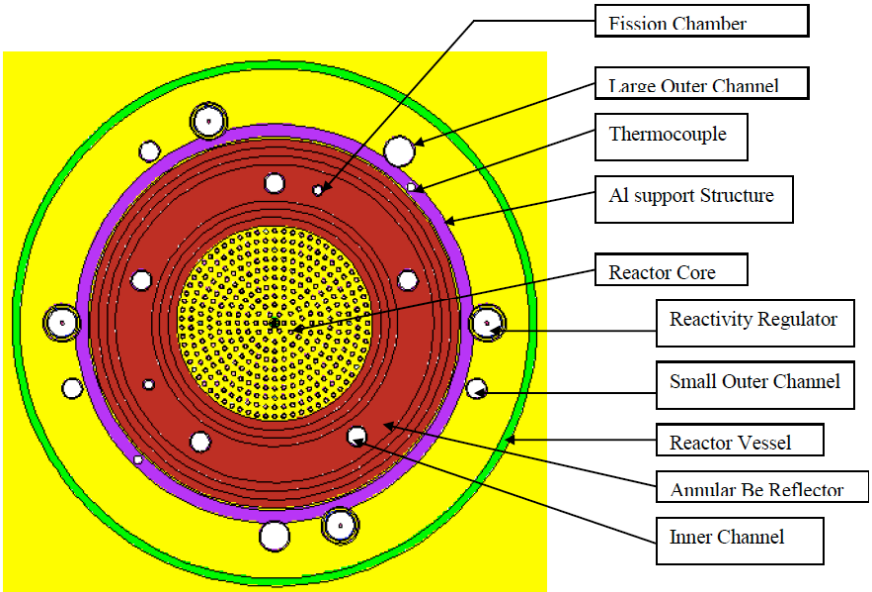

Figure 1: A geometric diagram of NIRR-1 in the x-y plane from MCNP.

$$\sigma_{eff} = \sigma_0\left(1+\frac{Q_0(\alpha)}{f}\right) \quad 3$$

Where $Q_0 = I_0/\sigma_0$ is the ratio of resonance integral to thermal cross-section, α is a measure of the non-ideal epithermal neutron flux distribution, and f is the thermal to epithermal neutron flux ratio.

For non-ideal situation, the Q_0 needs to be modified with an α-dependent term because the Q_0 -values are only valid for ideal spectra, and is not true for deviating spectra [14].

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_{th}} = \frac{Q_0 - 0.429}{E_r^{\alpha}} + \frac{0.429}{(2\alpha+1)E_{cd}^{\alpha}} \quad 4$$

For non-ideal conditions, the resonance integral I_0, must also be modified with an α-dependent term, hence $I_0(\alpha)$ values should be used instead of I_0 [15].

$$I(\alpha) = \left[\frac{I_0 - 0.426\sigma_0}{(\bar{E}_r)^{\alpha}} + \frac{0.426\sigma_0}{(2\alpha+1)(E_{Cd})^{\alpha}}\right] \quad 5$$

$$E_a^{\alpha} = \int_{E_{Cd}}^{\infty} \alpha(E)\frac{E_a^{\alpha}}{E^{1+\alpha}}dE$$

Where $(E_r)^{\alpha}$ = effective resonance energy, $E_a^{\alpha} = 1\,eV -$ arbitrary energy, $E_{Cd}^{\alpha} = 0.55\,eV -$ effective cadmium cut-off energy, $\sigma_0 = 2200\ m/s\ (n,\gamma)$ cross-section and α = an experimentally determinable characteristics of the reactor channel.

In gamma measurements of solid samples using HPGe detectors, it is of paramount importance to obtain the full energy peak efficiency in the calibrating geometry as a function of γ-emission energy E_γ.

$$\varepsilon_p(E_\gamma) = \frac{N_\gamma}{t_r P A_0 \exp(-\frac{\Delta t}{\tau})} \quad 6$$

Where N_γ is the number of counts in the photopeak, t_r is the real time taken for the run data, P is the branching ratio corresponding to the energy, A_0 is the activity of the source on the reference date, Δt is the time elapse since calibration up to measurement.

τ is the mean life time ($\tau = \frac{1}{\lambda}$ where λ is the decay constant)

MATERIALS AND METHOD

The activation material used was selected based on the purpose and conditions of the experiment, which required that the detecting material should be easy to acquire, have stable chemical properties and appropriate activation cross section and a proper half life [8]. Since cross-section spectra are similar in shape for a given type of neutron interaction, it is recommended not to choose specific interactions based on foils. Factors such as energy response, environmental conditions, exposure time and counting method were also taken into consideration.

Copper wire (99.99% purity) was used for this work due to its availability amongst others, high cross-sections for thermal neutrons compared to materials like Mo-99 and Zn-65, and the required nuclear data parameters are obtainable from nuclear data table as shown in Table **1** [16, 17]. The thermal to epithermal flux ratio, f and epithermal neutron flux shaping factor, α values for B2-irradiation channel were obtained from earlier work [18], and reported as 19.2 ± 0.5 and -0.052 ± 0.002 respectively. Values for $I_0(\alpha)$ and $Q_0(\alpha)$ were also determined from equations (4) and (5) respectively, while I_0 and Q_0 are obtainable from nuclear data [16].

The copper wires were arranged in axial directions in a typical irradiation container normally used for routine activation analysis. The wires were placed vertically as shown in Figure **2**, labelled X, Y and Z. After irradiation of 1 min in B2-inner channel, each wire was sectioned into three pieces to account for the axial flux profiles. A GEM- 30195 HPGe coaxial detector (ORTEC) consisting of MAESTRO Multi-channel analyzer (MCA) emulation software was calibrated and used for counting the induced radioactivity. Each of the sectioned copper wire was counted for 10 min at 15 cm from the detector's head, to obtain the net peak areas. The efficiency of the detector was determined from equation 6 using standard gamma-ray sources (^{241}Am, ^{152}Eu, ^{137}Cs and ^{60}Co). From the efficiency curve, the photo peak efficiency was obtained. The 1345.77 keV

Table 1: Nuclear Data for Activation Material Used [17, 16]

Nuclide	Nuclear Reaction	θ (%)	σ_{th} (barns)	R.I (barns)	Product Half-life	Eγ (keV)	γ (%)	Q_0	E_r (eV)
Cu	^{63}Cu(n,γ)^{64}Cu	69.17	4.28	4.88	12.7 h	1345.77	0.48	1.1	1040

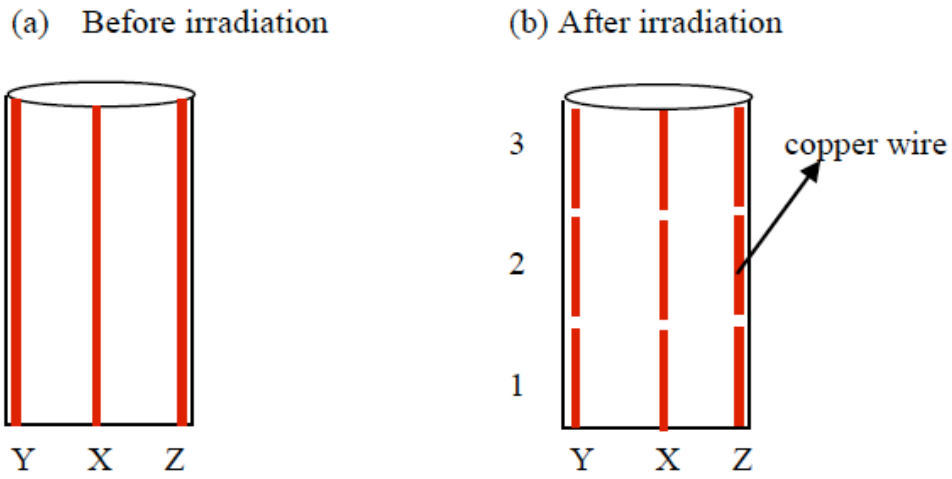

Figure 2: Schematic diagram showing the vertical wires before and after irradiation.

full energy peak from $^{63}Cu(n,\gamma)^{64}Cu$ reaction was employed because of its longer half-life (12.7 hrs) compared to (5.14 min) resulting from $^{65}Cu(n,\gamma)^{66}Cu$ reaction.

RESULTS AND DISCUSSIONS

The result for the efficiency calibration of the High Purity Germanium detector (HPGe) using standard gamma-ray sources is shown in Figure **3**. The photopeak efficiency of 1345.77 keV peak was found from the efficiency curve and later used for calculations of the neutron fluxes. However, 1345.77 keV peak was chosen and found to have less spectral interference when compared to 511 keV and as a consequence little error propagation. This is because 511 keV could be annihilation peak or from other radioisotopes such as ^{24}Na and ^{49}Ca-49 which may possibly interfere with that of Cu-64 [5, 17].

Table **2** shows the axial neutron flux distribution and indicate a variation of 2% - 4% along the vertical direction within the irradiation container. Figure **4** shows the overall axial neutron flux variation at positions X, Y and Z respectively. The axial neutron flux range from 4.57 x 10^{11} to 5.20 x 10^{11} $cm^{-2}s^{-1}$ and the mean value in the container was found to be 4.90 x 10^{11} $cm^{-2}s^{-1}$. 0.1%Au-Al foil was also irradiated at the bottom of the irradiation container and the measured flux was (4.53 ± 0.02) x 10^{11} $cm^{-2}s^{-1}$. We infer that the observed variation in flux with position may be due to the relative position of the foil to the core, its vertical distance from the core and the decay time. Meanwhile, the measured values of both Au and Cu-wire are in

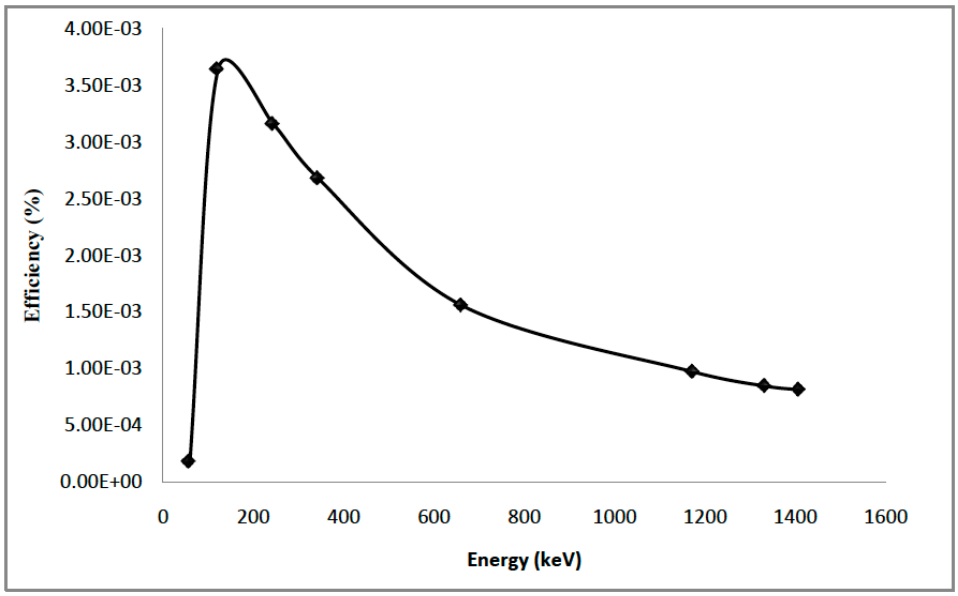

Figure 3: Graph of efficiency – energy function for the energy range 59.5 keV-1408 keV.

Table 2: Axial Flux Distribution in B2 Irradiation Channel

Axial Position	Section	Thermal Neutron Flux x 10^{11} ($cm^{-2}s^{-1}$)
Centre (X)	1	4.92 ± 0.18
	2	5.09 ± 0.19
	3	4.83 ± 0.18
Side (Y)	1	5.20 ± 0.20
	2	4.92 ± 0.20
	3	4.81 ± 0.18
Side (Z)	1	4.96 ± 0.19
	2	4.81 ± 0.19
	3	4.57 ± 0.21

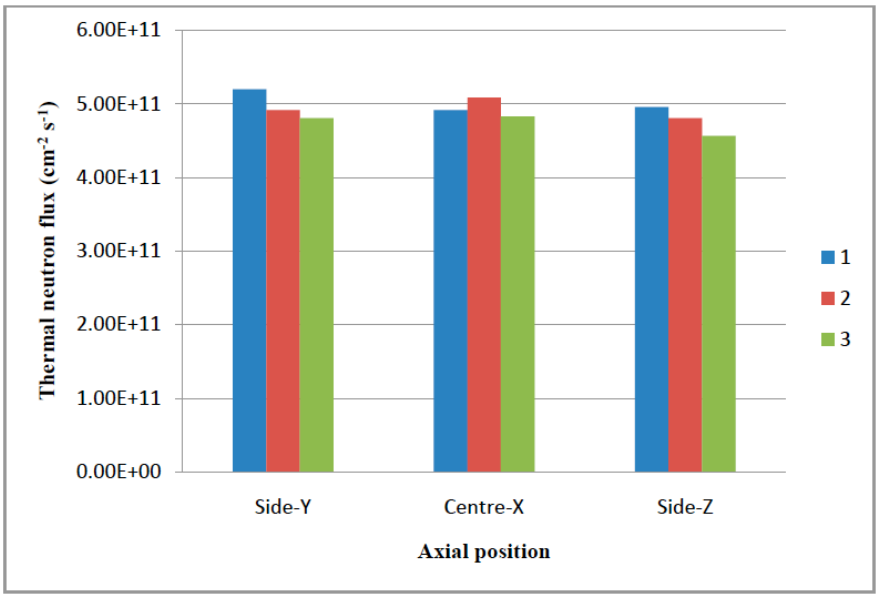

Figure 4: Axial neutron flux distribution within the irradiation container.

good agreement with the previously measured value [12].

CONCLUSION

Measurements of the axial flux distribution in the irradiation container at B2-irradiation channel have been carried out by foil activation method using $^{63}Cu(n,\gamma)^{64}Cu$ reaction, specifically by absolute method. Knowledge of the axial neutron flux in the core and the variation of the flux from one sample position to another are important in improving NAA technique. The axial neutron flux profiles indicate that variations occur from one position to another. The neutron flux for each of the positions measured can be used to predict the irradiation conditions of samples at those positions particularly during long irradiation in which many samples were jam-packed in one vial. This may facilitate flux corrections for the samples if need be. It can also be seen that even with the recent installation of the cadmium-liner in the outer channel of the reactor; the thermal neutron flux of the B2-channel has not deviated from 5.14 x 10^{11} $cm^{-2}s^{-1}$ measured earlier [12].

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