

## Slant tube characterization for the implementation of $k_0$ - standardization in Nigerian Research Reactor-1

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### Abstract

The result for large sample irradiation are continuously piping into the Center for Energy Research and Training (CERT), Ahmadu Bello University, Zaria. Therefore, characterization of the irradiation channels that can accommodate large sample becomes a challenge. The first series of trial for large samples irradiation through slant tube of the Nigeria Research Reactor-1 (NIRR-1) was necessary, in which the results were compared with standard values reported by the arena of researchers including nuclear regulatory bodies. The thermal to epithermal flux ratio ( $f$ ), epithermal neutron flux distribution parameters ( $\alpha$ ) were determined by using two slant tube channels of NIRR-1. The adopted methodology here is bare and cadmium covered monitor foils method. The vertical dipstick higher purity germanium detector (HPGe) was calibrated and used for the calculation. The  $f$  and  $\alpha$  values were calculated as 111.8 and -0.04977 respectively. The good agreement between the certified and experimental values of the shaping factors was achieved.

**Keywords:** NIRR-1, neutron spectrum, flux ratio, shaping factor

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### INTRODUCTION

The commissioning of NIRR-1 under custody of Ahmadu Bello University, Zaria on February 3<sup>rd</sup>, 2004, was seriously derived the pleasure and interest of many Nigerian scientists to study physics and engineering and perhaps it is highly contributing to the development of science, medicine, technology and nuclear engineering and the Nigeria as whole. Even though, NIRR-1 is a low power miniature source reactor (MNSR) with 31 kW thermal power, 3.77 mk of excess reactivity, with operational full power time of 4.5 hours and thermal neutron flux of  $1 \times 10^{12}$  ncm<sup>-2</sup> in its irradiation channels, but so many different analysis from different sectors like hospitals and health centers, can-food companies, meal industries, research institutes and university are sending on daily bases and to extent that some samples are bulky enough to be irradiated through existing reactor's irradiation channels. The only solution to this problem is to characterize the slant tubes, so that the large samples can be irradiated through it, and the guide tube would be able to accommodate large sample for Large Sample Neutron Activation Analysis (LSNAA) [1, 2, 3, 9]. To achieve this goal, High Purity Germanium (HPGe) detector has to be calibrated to ensure the efficiencies of the standard gamma-ray sources are quite within the range at all source-to-detector geometries for calculating the full energy efficiencies. The full efficiency,  $\epsilon_f$  for the three different regions, 50 – 90 keV, 90 – 200 keV and above 200 keV using equations 2, 3 and 4, respectively. Some selected standard gamma-ray sources for efficiency calibration used in this work, are Sodium-22, Manganese-54, Cobalt-57, Cobalt-60, Cesium-137, Europium-152 and Radium-226. The close geometry used was 1 cm and the far geometries are considered to be 5 and 10 cm, and the time of measurements for standard gamma-ray sources are 300, 600 and 900 seconds, respectively. The purified neutron monitor foils and alloys used for the slant tube site

characterization are Zinc-64, Zinc-69, Gold (Au)-197 and Gold (Au)-198, that is for determination of neutron spectrum parameters of the slant tube irradiation site. The foils were chosen to justify the previous results of the inner and outer irradiation channel characterization of the same reactor. The slant guide tube is located between the control rod guide tube and small inner irradiation channels. Bare multi monitor is the suitable method for characterizing reactor irradiation sites with unstable neutron flux and spectrum parameters [1,2,10]. However, in this study the Cadmium covered multi-monitor and bare multi-monitor methods were adopted in finding thermal to epithermal flux ratio  $f$  and epithermal shaping factor,  $\alpha$ .

$$\epsilon_p = \frac{N_p/t_m}{A_t I_a} \quad (1)$$

$$\epsilon_E = \epsilon_0 \exp - (\mu_E A_t) A L. \exp(\mu_{Ge} A_t) G_e \quad (2)$$

$$\ln \epsilon_E = A_1 + A_3 (4.816 \ln E + (\ln E)^2) \quad (3)$$

$$\ln \epsilon_E = \sum_{j=1}^6 a_j (\ln E)^{j-1} \quad (4)$$

### EXPERIMENTAL ANALYSIS

#### Site characterization

The site characterization directly referred to the neutron spectrum parameters in determination of the irradiation sites. Though in this work, slant guide tube (slant tubes) was characterized with prepared intention to stand as irradiation site for bulk samples for the implementation of Kayzero-Neutron Activation Analysis ( $k_0$ -NAA) [4]. Two sets of monitor foil oxides of <sup>64</sup>Zn, <sup>69</sup>Zn, <sup>197</sup>Au and <sup>198</sup>Au

were prepared with the following weights 0.0083 kg, 0.0448 kg 0.0139 kg and 0.0139 kg, respectively. One of the set was inserted in a stack of 1 mm thick and then into a vial with no cadmium cover for bare analysis, while the other set was arranged in the same manner but covered with a cadmium and then sent into reactor via slant tube. The irradiation and counting regimes for both bare and cadmium covered monitors are presented in Table 2. Gamma ray peaks spectral acquisition was done using vertical deep stick HPGe detector, after which a multi-purpose gamma-ray analysis software named WinSPAN-2004 was used for identifying and evaluating the peaks of the nuclide energies. Our concerned here focused on gold and zinc where their nuclear data and slant tube coating regimes are shown in Table 1 and 2, respectively [1, 9, 10]. The Figure 1 below illustrates the NIRR-1 core configuration with all its irradiation sites [10].

**Thermal to epithermal flux ratio *f***

The Cd-ratio for multi-monitor method was used for the determination of thermal to epithermal neutron flux ratio, *f*. The determined parameter is summarized by the following relations.

$$f = \phi_{0i}(\alpha) \cdot (F_{cdr}R_{cdr} - 1) \frac{G_{ei}}{G_{thi}} \tag{5}$$

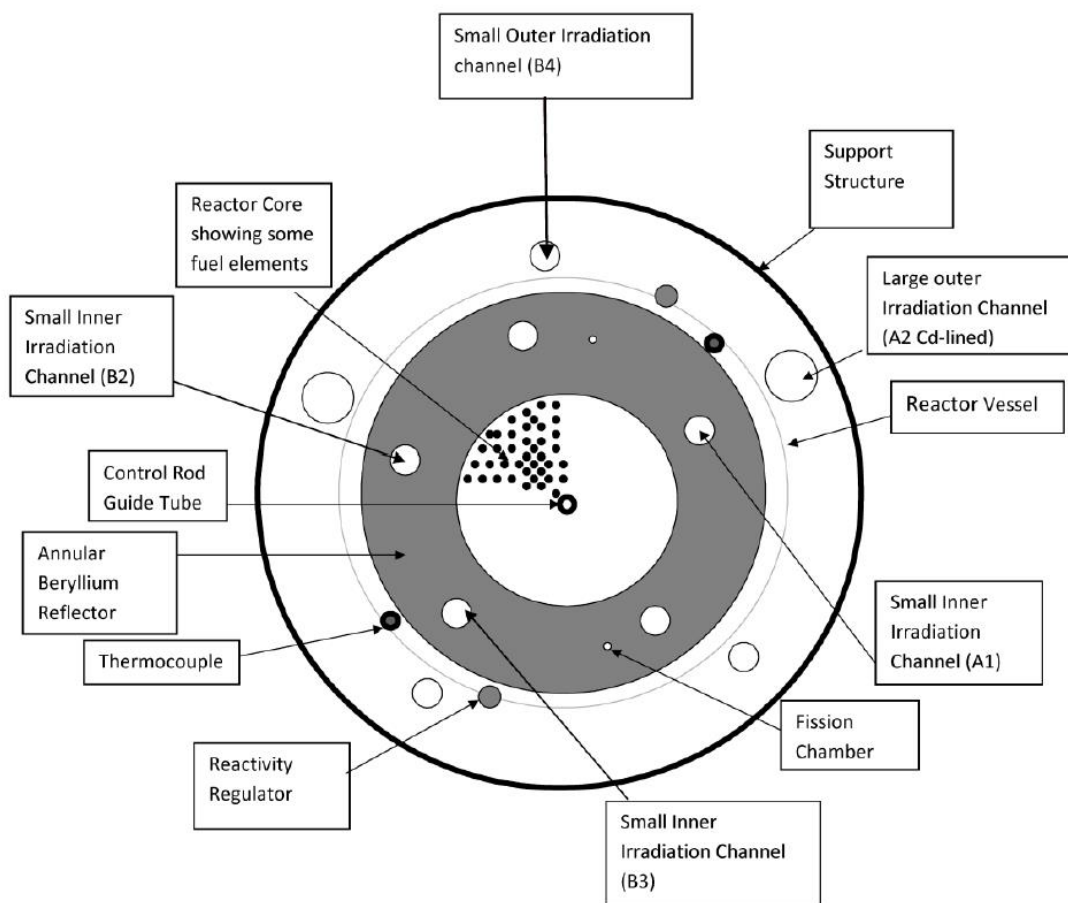
$$R_{cdr} = \frac{A_{sp}(bare)}{A_{sp}(cd-covered)} \tag{6}$$

$$\phi_{0i}(\alpha) = \frac{\phi_{0i}-0.429}{E_{ri}^\alpha} + \frac{0.42}{(2\alpha-1)0.55^\alpha} \tag{7}$$

$$A_{sp} = \frac{N_p/t_m}{WSDC} \tag{8}$$

- where *i* =  $\phi_0$ -value monitor
- $G_{thi}$  = self shielding thermal neutron correction factor
- $G_{ei}$  = self epithermal neutron correction factor
- $F_{cdr}$  = cadmium transmission factor for epithermal neutron
- $R_{cdr}$  = cadmium ratio
- $A_{sp}(bare)$  = specific count states of bare target
- $A_{sp}(cd - c)$  = specific count states of Cd-covered target
- $\phi_{0i}(\alpha) = I_0/\sigma_0$
- $I_0$  = Isotopic resonance abundance of target nucleus
- $\sigma_0$  = Cross sectional at neutron velocity of 2200 m/s for monitor
- W = weight
- $S = (1 - \exp(-\lambda t_{ir}))$
- $D = \exp(-\lambda t_d)$
- $C = 1 - \exp(-\lambda t_m)$ , counting factor
- $t_m$  = time of measurement
- $\lambda$  = decay constant

The equations above are used iteratively on Excel spread sheet for the computation of thermal to epithermal flux ratio [1, 9, 10].



**Figure 1.** NIRR-1 core configuration showing irradiation sites.

**Table 1** Nuclear data for the used nuclides.

Monitor	$E_\gamma$ (eV)	$Q_0$	$F_{cd}$	$T_{1/2}$
Au-198	5.65	15.7	441.8	2.695 days
Zn-65	590	3.19	438.6	13.76 hrs

**Table 2** Irradiation, decay and counting regimes slant tube.

Irradiation site	Isotope	Sample weight (mg)	Irradiation time, $t_{irr}$ (sec)	Cooling time, $t_d$ (sec)	Measurement time, $t_m$ (sec)
Slant-tube	Au-197 (bare)	1.39E-02	2.70E+03	3.60E+03	1.80E+03
	Au-197 (Cd-covered)	1.39E-02	4.50E+03	1.50E+03	1.80E+03
	Zn-69 (bare)	5.00E-03	2.70E+03	2.58E+03	6.00E+02
	Zn-69 (Cd-covered)	4.90E-03	4.50E+03	3.80E+03	1.80E+03
	Au-198 (bare)	1.39E-02	2.70E+03	3.40E+05	3.00E+02
	Au-198 (Cd-covered)	1.39E-02	4.50E+03	3.44E+05	3.00E+02
	Zn-64 (bare)	5.10E-03	2.70E+03	6.09E+05	1.20E+03
	Zn-64 (Cd-covered)	4.90E-03	4.50E+03	6.09E+05	4.50E+03

**Determination of epithermal neutron shaping factor ( $\alpha$ )**

Epithermal neutron shaping factor ( $\alpha$ ) is a function of physical properties of the reactor. The method adopted to determined the  $\alpha$ -value in this work was the iterative procedure which based on MS-Excel spreadsheet utilities. The equation (10) was used for the  $\alpha$ -value calculation. Although for comparative reason, it was also calculated using simple multi-monitor for bare and cadmium covered method, given in equation (9) by taking a slope of the logarithmic ratio of

effective resonance energy of the monitors  $\alpha$ -corrected to the flux ratio ( $A_i(\alpha)$ ) against logarithmic effective resonance energy (Log  $E_r$ ) [7, 10].

$$A_i(\alpha) = \log \frac{E_{y,i}^\alpha}{(F_{cdr}R_{cdr-1})\phi_{oi}(\alpha)\frac{G_{ei}}{G_{thi}}} \tag{9}$$

where  $E_{y,i}^\alpha$  is the effective resonance energy of the monitors. The epithermal neutron shaping factor ( $\alpha$ ) was summarized by the relation.

$$\alpha + \frac{\sum_{i=1}^N \left[ \left( \log \bar{E}_{y,i} - \frac{\sum_{i=1}^N \log \bar{E}_{y,i}}{N} \right) \left( \log \frac{\bar{E}_{y,i}^{-\alpha}}{(F_{cd,i}R_{cd,i-1})Q_0(\alpha)G_{e,i}/G_{th,i}} - \frac{\sum_{i=1}^N \log \frac{\bar{E}_{y,i}^{-\alpha}}{(F_{cd,i}R_{cd,i-1})Q_0(\alpha)G_{e,i}/G_{th,i}}}{N} \right) \right]}{\sum_{i=1}^N \left( \log \bar{E}_{y,i} - \frac{\sum_{i=1}^N \log \bar{E}_{y,i}}{N} \right)} = 0 \tag{10}$$

The two points of energies and their corresponding efficiencies were selected from the semi empirical method calculation for the three different geometries and used for determining the thermal to epithermal neutron flux ratio,  $f$ . Perhaps, our working thermal to epithermal neutron flux ratio,  $f$ , was obtained by taking the average of the calculated values of  $f$ , as presented in Table 4 [1]. Despite that for confirmatory the  $f$ -value had been determined as an intercept of the plotted graph shown in Figure 2 [3, 10]. The epithermal neutron shaping factor ( $\alpha$ ) was determined as a slope of the graph of  $A_i(\alpha)$  against Log  $E_r$  as shown in Figure 2 from the source data of Table 4 [1,8,10].

The slope and intercept were obtained from logarithmic value of the widely spaced energies and their corresponding efficiencies, that are 661 keV and 1332 keV, respectively, for the three geometries, as in Table 3 [1,10].

**Table 3** Slopes and intercept data used in semi empirical method.

Geometry (cm)	Energy (keV)	Efficiency ( $\epsilon_p$ )	Intercept	Slope ( $a_i$ )
1	pt – 661	0.002102	-6.1063	-0.61841
	pt – 1332	0.001363		
5	pt – 661	0.000692	-7.21	-0.7068
	pt – 1332	0.000422		
10	pt – 661	0.000264	-8.16	-0.86554
	pt – 1332	0.000144		

**Table 4** Determinations of  $f$  and  $\alpha$ .

Isotope	Log $E_\gamma$	$R_{cd}$	$A_i (\alpha)$	Flux-ratio ( $f$ )
Au-197	0.752048	9.487592	-2.12057	131.9986
Zn-64	3.40824	26.2987	-1.68368	48.26992
Zn-69	2.770852	39.6079	-2.09047	123.1592
Au-198	0.752048	10.2338	-2.1572	143.616

The average  $f$  value is 111.8 [1]

**RESULTS AND DISCUSSION**

**Characterization**

The characterization was purely depended on the determined characteristic features of the irradiation site and the site can either be a channel or start up tube (slant guide tube). Therefore the characteristic features here referred to as a neutron spectrum parameters. These parameters are thermal to epithermal flux ratio ( $f$ ) and epithermal neutron shaping factor,  $\alpha$ . Starting with thermal to epithermal flux ratio,  $f$  parameter as it was determined by the use of equation 5. The flux ratio,  $f$  obtained in this work has higher value more than the values obtained when characterizing irradiation channels of the same NIRR-1 reactor due to the fact that the slant tube site was located near the core of the reactor, while irradiation channels both type A and type B were far to the core on comparison, example for B<sub>2</sub> :  $\alpha = -0.052 \pm 0.002$  and  $f = 19.2 \pm 0.5$ , while for B<sub>4</sub> :  $\alpha = -0.029 \pm 0.003$  and  $f = 48.3 \pm 3.3$ . Secondly, the self epithermal neutron correction factor was found very high near the reactor core guide tube compared to the self shielding thermal neutron correction factor. Lastly, their ratio (that is the self epithermal neutron correction factor to the self shielding thermal neutron correction factor) was also found quite negligible compared to the product of the cadmium transmission factor for epithermal neutron and the cadmium ratio. These three reasons have made our result reliable and the thermal to epithermal flux ratio ( $f$ ) parameter value obtained was 111.8, according to [1]. While by adopted graphical method the determined thermal to epithermal flux ratio ( $f$ ) parameter value was 2.2564.

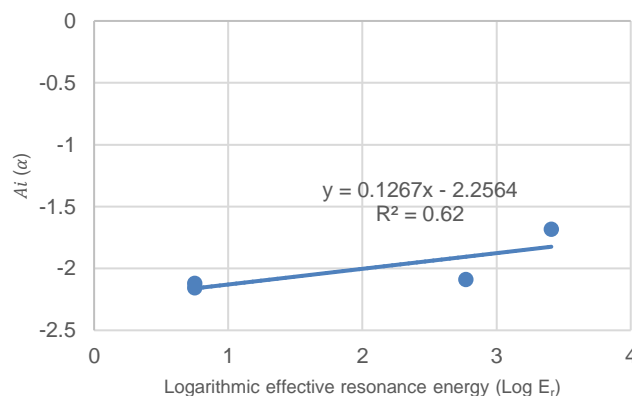
Therefore, the epithermal neutron shaping factor  $\alpha$ -value in this work was conveniently determined by iterative procedure which based on MS-Excel spreadsheet utilities. Eventhough it was shown in Figure 2 which was originated from equation (9) and table 4 [1,2,3,10]. Notwithstanding, the site characterizations of neutron parameters depend on the outcome of measured energies and efficiency calibration with HPGe detector [5]. The full energy efficiency calibration was performed in two different methods (conventional and semi empirical) and Table 5 shown one of six data obtained, three for each method and for the three different geometries (1 cm, 5 cm and 10 cm) with their corresponding energies range, as stated in the introduction of this work. Thus, the slopes and intercept for the irradiation energies and their corresponding full efficiencies of the selected point for the slant tube's decay and counting regimes were summarized in Table 3 [1]. Importantly noted that the efficiency calculation in the energy region above 250 KeV based on the respective polynomial expressions given in equation (11) below [1,3,10].

$$\log \varepsilon_p = a_0 + a_1 \log E + a_2 \log E \tag{11}$$

The efficiencies of their corresponding gamma energies were determined using the equation (1). Then, for the semi empirical efficiency calibration, the intercept and slopes from Table 3 are used as the constants  $a_0$ ,  $a_1$  and  $a_2$ , respectively of the equation (11) [8,10]. The determined neutron spectrum parameters of the slant guide tube is referred as the site characterization. However, the monitors used in this study does not obey the dependence  $\sigma(v) \sim 1/v$ . Therefore, the epithermal neutron shaping factor,  $\alpha$ , cannot be obtained from the slope of a straight line in Figure 2. Meanwhile, it was determined by iteration using equation 10.

**Table 5** Corresponding gamma energies.

$A_i$	$t_m$	$N_p$	$E_\gamma$	$N_p/t_m$	$A_i^*E_\gamma$	$E_p$
36.286	900	35587	36	39.5411	1291306	0.00306
23.453	900	40229	99.9	44.6989	943505	0.00474
23.453	900	36480	99.9	40.5333	855579	0.00474
1.0621	900	7749	85.5	8.61	8230.31	0.10461
1.0621	900	944	10.7	1.04889	1002.63	0.10461
33.875	900	2E+05	84.6	243.62	7427390	0.00328
31.344	900	63932	28.2	71.0356	2003914	0.00354
31.344	900	12968	7.42	14.4089	406475	0.00354
31.344	900	635	0.42	0.70556	19903.7	0.00354
31.344	900	35945	26.4	39.9389	1126677	0.00354
31.344	900	2562	3.08	2.84667	80304.5	0.00354
31.344	900	9204	4.16	10.2267	288494	0.00354
31.344	900	8729	14.5	9.69889	273606	0.00354
31.344	900	6578	11.8	7.30889	206184	0.00354
31.344	900	7440	13.6	8.26667	233203	0.00354
31.344	900	839	1.74	0.93222	26298	0.00354
31.344	900	9965	20.7	11.0722	312348	0.00354
37.339	900	8424	3.41	9.36	314541	0.00298
37.339	900	60	0.43	0.06667	2240.32	0.00298
7.339	900	44745	44.3	49.7167	1670718	0.00298
37.339	900	1313	1.49	1.45889	49025.6	0.00298
37.339	900	2206	3.05	2.45111	82369.1	0.00298
37.339	900	9423	14.7	10.47	351842	0.00298
37.339	900	3322	5.71	3.69111	124039	0.00298
37.339	900	407	0.79	0.45222	15196.8	0.00298
37.339	900	674	13	0.74889	25166.2	0.00298
37.339	900	1060	2.08	1.17778	39579	0.00298
37.339	900	463	1.08	0.51444	17287.8	0.00298
37.339	900	1370	2.83	1.52222	51153.9	0.00298
37.339	900	6826	15.1	7.58444	254874	0.00298
37.339	900	376	1.18	0.41778	14039.3	0.00298
37.339	900	1694	4.96	1.88222	63251.7	0.00298
37.339	900	10	1.51	0.01111	373.386	0.00298



**Figure 2** Determination of  $f$  and  $\alpha$  by cadmium multi monitor method for NIRR-1 slant tube.

## CONCLUSION

From the obtained  $f$ -values of the four monitor foils ( $^{64}\text{Zn}$ ,  $^{69}\text{Zn}$ ,  $^{197}\text{Au}$  and  $^{198}\text{Au}$ ), the average was found to be 111.8, while the  $\alpha$ -value was -0.04977. The negative value of the shaping factor has to do with the nature of the spectrum and it implies that the spectrum is hardened. The values of  $f$  and  $\alpha$  obtained graphically are 2.2564 and -0.1267, respectively and proved that the monitor foils disobey  $\sigma(v) \sim 1/v$ . The  $f$ -value obtained from the intercept of the graph was scientifically not reliable because as the sample moved inward toward the reactor core the thermal to epithermal flux ratio,  $f$  was expected to be high. Secondly, the obtained slant tube average  $f$ -value have showed a comparable behaviour with  $f$ -values of irradiation channels of type A and B, even though different monitor foils were used. Generally, the determined flux parameters can serve as operational working documents for LSNAA and  $k_0$ -NAA implementation. Moreover, we suggest that the experiment needs to be repeated over time with different combination of monitor foils to ensure the reliability and efficiency of the reactor facility. Finally, this experiment has paved the way for further slant guide tube research on other neutron spectrum parameters like spectrum index,  $r(\alpha)[T_n/T_0]^{1/2}$ , Maxwellian neutron temperature  $T_n$  and thermal to fast neutron flux ratio  $f_T$ , in order to maximized the NIRR-1 utilization.

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