CHARACTERIZATION OF IRRADIATION CHANNELS OF THE GHARR-1 MINATURE NEUTRON SOURCE REACTOR

BY

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DECLARATION

I, Yusuf Aminu Ahmed, hereby declare that this thesis titled **"Characterization of Irradiation Channels of the GHARR-1 Miniature Neutron Source Reactor"** is my own work and has not been submitted in any other form for another degree or diploma to any other University or Institution. Information derived from published or unpublished work of other workers has been fully acknowledged in the text.

_____ _____

Yusuf Aminu Ahmed Date

CERTIFICATION

This thesis titled **"Characterization of Irradiation Channels of the GHARR-1 Miniature Neutron Source Reactor "** by **Yusuf Aminu Ahmed** (MSc/Sci/23473/2000-2001) meets the regulations governing the award of the degree of Master of Science (Physics) of Ahmadu Bello University, Zaria and is approved for its contribution to knowledge and literary presentation.

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DEDICATION

To

Ummahani Aminu Ahmed

And

Zainab Ahmed Tijjani

(For their love, understanding and care)

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ABSTRACT

Determination of reactor-based essential parameters in activation analysis (neutron spectra shape-factor α and the thermal to epithermal flux ratio f) for inner channel (No. 5) and outer channels (No. 7) of the GHAna Research Reactor-1 (GHARR-1) a Miniature Neutron Source Reactor (MNSR) was undertaken in an effort to characterize the channels for the application of k_0-standardization in Neutron Activation Analysis with the reactor

Measurements were carried out according to the Hogdahl flux convention using cadmium ratio method. Specific activities of Gold and Zirconium monitors irradiated together with and without cadmium cover under the same neutron flux were used to obtain cadmium ratios. The thermal to epithermal flux ratio f of the reactor were measured and found to be 17.28 for the inner channel (No. 5) and 40.38 for the outer channel (No.7) while the epithermal flux-shaping factors α for these channels were obtained as -0.107 ± 0.0001 and -0.173 ± 0.0001 respectively. The α-values were obtained by iteration method using in-house computer programs (Q-Basic and C++) developed for this work. The results obtained compared favorably well with the previously determined ones by Akaho and Nyarko 2002 for inner channel No. 1 (f = 18.8 and α = - 0.104) and outer channel No. 6 (f = 49.0 and α = - 0.0261) of the same reactor.

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CHAPTER ONE

INTRODUCTION

1.1 Introduction

In an ideal thermal neutron reactor, the thermal and epithermal neutron flux per unit energy interval has been shown to be inversely proportional to the neutron energy at that interval. The variation of the flux with energy [$\Phi_{epi}(E) \approx 1/E$] is based on the assumption that the moderation process in an ideal reactor takes place in a homogeneous infinite medium and that absorption, inelastic and elastic scattering during moderation do not occur (Mukhin 1970). However, for epithermal flux distribution in a 'real' reactor spectrum, this is an over assumption of the moderation situation. Infact, moderation process differs from one reactor to another or even within the same reactor (Ryves, 1969); meaning that it deviates from ideal $1/E$ law and is better approximated by $1/E^{1+\alpha}$ where α is an experimentally determinable characteristics (De Corte *et. al*., 1981) of the reactor irradiation channel and also a measure of epithermal flux deviation from ideal distribution. It may have positive or negative values depending on the nuclides of interest, core configuration, geometry of irradiation channels e.t.c.

It has been shown by Jovanović *et. al*., 1989 that the epithermal neutrons contribute substantively to the (n,γ) reaction when using k_0-standardization in *Instrumental Neutron Activation Analysis* (INAA). Therefore, the effect of non-ideality of the epithermal spectrum should not be underestimated when calculating induced activity on a target nuclide. In general, the thermal and epithermal neutrons in a reactor are the

most widely used for Neutron Activation Analysis (NAA) because their fluxes are greatest and the cross section for *(n, γ)* reaction is the largest hence they offer the greatest analytical sensitivity available (De Corte *et. al*., 2000) for NAA. For this reason, knowledge of α-values in an irradiation channel is necessary for the conversion of resonance integral I_o and other parameters such as Q_o, valid only in ideal situation to α–dependent terms to take care of both thermal and epithermal contributions that are obtained in real reactor situations. The *(n, γ)* reactions that involve those parameters are hereby discussed.

The total reaction rate per target atom for *(n, γ)* reactions induced by both thermal and epithermal neutrons in a reactor is given by (IAEA; 1990):

$$R = R_{th} + R_{epi} = \Phi_{th}\,\sigma_{th} \;+\; \Phi_{epi}\,I_o \qquad (1.1)$$

Where Φ_{th} = thermal neutron flux

Φ_{epi} = epithermal neutron flux

σ_{th} = average thermal neutron cross-section

I_o = effective resonance integral

For an 'ideal' (*1/E*) epithermal distribution, I_o is defined by (Simonits *et. al*., 1984) as:

$$\mathrm{I_o} = \int_{E_{cd}}^{\infty} \sigma(E)\frac{dE}{E} \qquad (1.2)$$

E_{cd}=0.55eV (effective Cd cut-off energy)

$\sigma(E)$ =*(n, γ)* cross-section

However, for the 'non-ideal' (real) situations, the resonance integral I_0 needs to be modified with an α - dependent term because literature data for I_o, which are valid only

for ideal spectra, are not valid for deviating spectra (Moens *et. al.*, 1979). It is for these conditions that I_0 (α) values ought to be used. The conversion of I_o to an α - dependent term takes the form (Jovanović *et. al.*, 1987):

$$I_o(\alpha) = \left(\frac{I_o - 0.429\sigma_o}{\overline{E}_r^{\alpha}} + \frac{0.429\sigma_o}{(2\alpha+1)\overline{E}_{cd}^{\alpha}} \right) E_a^{\alpha} = \int_{E_{cd}}^{\infty} \sigma(E) \frac{E_a^{\alpha}}{E^{1+\alpha}} dE \qquad (1.3)$$

where $\overline{E}_r^{\alpha}$ = effective resonance energy

E_a^{α} = 1eV – arbitrary energy

$\overline{E}_{cd}^{\alpha}$ = = 0.55 eV - effective cadmium cut-off energy

σ_o =2200 m s^{-1} (n,γ) Cross section.

α = an experimentally determinable characteristics of the reactor channel.

Although the epithermal neutrons represent only a small fraction of the total reactor neutrons, they are sometimes useful in NAA for several elements (e.g. Br, Rb, Sr, Mo, Ba, Ta and U) that have higher relative reaction rates for epithermal neutrons than for thermal neutrons. The technique of taking advantage of those *(n, γ)* reactions with high resonance integrals through the irradiation of samples under a cadmium cover to shield out the thermal neutrons is commonly known as Epithermal Neutron Activation Analysis (ENAA).

The large number of resonance peaks for most nuclides makes calculation of effective cross sections for the epithermal neutrons slightly difficult. In order to avoid these problems, a gold standard is frequently used because the reaction ^{197}Au (n, γ) ^{198}Au has a single resonance (411 KeV) peak and it has been well investigated (Nisle, 1963; De Corte *et. al.;* 1987; Kennedy *et. al.*, 2000) and found to be 1550 barns. The activity ratio for an infinitely thin gold foil or alloy irradiated with and without cadmium

covers is a frequently used method (De Corte *et. al*., 1981; Jovanović *et. al*., 1989; Akaho and Nyarko, 2002) for measuring epithermal flux. The cadmium ratio method (sometimes called activity ratio) for a ^{197}Au standard solution irradiated with and without cadmium covers is used for the measurement of both thermal and epithermal flux. This method is also used as a calibration standard to measure the resonance integrals for *(n, γ)* reactions (Moens *et. al*., 1979). The equation describing this cadmium ratio method is:

$$R_{cd} = \frac{A_{bare}}{F_{CD} A_{CD}} = \frac{(\Phi_{th}\sigma_{th} + \Phi_{epi} I_0)}{\Phi_{epi} I_0} \tag{1.4}$$

Equation 1.5 can also be written as:

$$R_{cd} = \frac{R}{R_{epi}} = \frac{A_{bare}}{F_{CD} A_{CD}} \tag{1.5}$$

F_{cd} = cadmium transmission factor

A_{bare} = activity of bare monitor

A_{cd} = activity of cadmium covered monitor

Using equation (1.1) in (1.5) we obtain

$$(R_{cd} - 1) = \left(\frac{\Phi_{th}}{\Phi_{epi}}\right)\left(\frac{\sigma_{th}}{I_0}\right) = \left(\frac{f}{Q_0}\right) \tag{1.6}$$

The ratio of thermal to epithermal flux is defined as:

$$f = \frac{\Phi_{th}}{\Phi_{epi}}$$

where Φ_{th}*:* = thermal neutron flux

Φ_{epi} = epithermal neutron flux

And the ratio of resonance integral to thermal cross-section is given as:

$$Q_0 = \frac{\mathrm{I}_0}{\sigma_{th}}$$

Thus:

$$f = (R_{cd} - 1)Q_0 \tag{1.7}$$

R_{cd} = Cadmium ratio

I_0 = resonance integral

σ_{th} = thermal cross section at a neutron velocity of 2200 ms^{-1}

If the single comparator techniques is adopted for routine NAA, the effect of the non-ideality of the epithermal spectrum should not be neglected. Thus to be accurate in all relevant expressions, Q_0 should be replaced by $Q_0(\alpha)$ [where $Q_o(\alpha)$ is the α-corrected Q_o to take care of non-ideality of epithermal spectrum] as defined by De Corte, 1987:

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_{th}} = \frac{Q_0 - 0.429}{E_r^{\alpha}} + \frac{0.429}{(2\alpha+1)E_{cd}^{\alpha}} \tag{1.8}$$

$I_0(\alpha)$ = modified resonance integral.

E_r = effective resonance energy (eV)

E_{cd} = 0.55eV effective cadmium cut-off energy

Experimental determination of α, using the cadmium ratio method involves irradiating two or more monitors (e.g Au, Zr, Co) with and without cadmium alternately at the same channel (De Corte *et. al.*, 1981). When monitors such as ^{197}Au(1) and ^{94}Zr(2) are irradiated under uniform neutron flux, equation (1.7) transforms to:

$$(R_{cd,1} - 1)Q_0(\alpha)_1 = (R_{cd,2} - 1)Q_0(\alpha)_2 \tag{1.9}$$

Substituting (1.8) into (1.9) and rearranging we have:

$$(R_{cd,1}-1)/(R_{cd,2}-1)=\{[\frac{Q_o-0.429}{\overline{E}_r{}^{\alpha}}+\frac{0.429}{(2\alpha+1)\overline{E}_{cd}{}^{\alpha}}]_2/[\frac{Q_o-0.429}{\overline{E}_r{}^{\alpha}}+\frac{0.429}{(2\alpha+1)\overline{E}_{cd}{}^{\alpha}}]_1\} \tag{1.10}$$

$$\Rightarrow \quad \frac{(R_{cd}-1)_1}{(R_{cd}-1)_2}=\frac{Q_0(\alpha)_2}{Q_0(\alpha)_1}=f(\alpha) \tag{1.11}$$

Equation 1.11 is the iteration function used in this work to determine the epithermal flux-shaping factor (α).

1.2 Previous Work

The effect of the epithermal neutron flux distribution on the accuracy of absolute and single comparator standardization method in (n, γ) activation analysis has previously been studied by De Corte *et. al*., 1981; 1982; 1987; who reported that an error of 4 - 11% would be introduced in the calculation of epithermal contribution to the neutron flux by neglecting the effect of the neutron shape factor, (α). This error range is too large and unacceptable for any NAA result to be meaningful and accurate. Thus, to preserve the accuracy, it is essential to perform appropriate corrections for α in all irradiation channels of a reactor.

The possibility of adapting k_o –method for Miniature Neutron Source Reactors (MNSR) based on Hogdahl convention (Hogdahl, 1962) was investigated by Kennedy *et. al*., 2000. The neutron flux parameters using the Hogdahl-convention and Westcott-formalism (Westcott, 1960) were also used by Acharya *et. al*., 2001 to characterize two

irradiation channels of the Dalhousie University SLOWPOKE-2 Reactor (DUSR) which is a similar reactor to the Nigerian and Ghanaian MNSR.

Previously, Akaho and co-workers (Akaho, *et. al*. 2001, 2002) determined the thermal to epithermal flux ratio *f*, and the epithermal flux shaping-factor α, associated with inner channel 1 and outer channel 6 of the Ghana MNS reactor, out of the five inner channels and two outer channels installed by the Chinese vendors.

Earlier, Qazi *et. al*., 1996 performed neutronic calculations for the Ghanaian MNSR and reported that the cold-clean-core excess reactivity at 20^{o}C was 3.88 mk, which compared favourably with the experimental value of 3.99mk reported by Akaho *et. al*., 2000. In order to compensate for a drop in the reactor's excess reactivity (due to fuel burn-off) from 3.88mk measured by Qazi *et. al*., 1996 to 2.8mk measured recently, the technical group of GHARR-1 had to add 3mm thick beryllium shim.

The addition of the beryllium shim informed the need to re-characterize the irradiation channels of the reactor to validate the sub-to-epicadmium flux ratio *f* and the flux shaping factor α in order to be sure that the excess reactivity of 3.88-3.99 mk of the reactor is regained and that it is good enough to perform experiments.

This project is aimed at determining necessary nuclear parameters of GHARR-1 irradiation channels for application of k_o-methodology (see Appendix A) and also to extend the knowledge and experience gained during the work to NIRR-1 after commissioning.

1.3 Justification

One of the applications of a research reactor like the Ghanaian and Nigerian MNSR is in the field of Neutron Activation Analysis (NAA) of Geological, Biological, Environmental samples etc. for the determination of trace elements in matrices (Volkovic, 1975; Shu-De, 1984). Between the various forms of NAA that will be exploited with research reactors, the single comparator method has become the most popular and relatively faster analytical method (De Corte, 1987). Irradiation channels used for NAA require characterization in order to determine their sub-cadmium to epi-cadmium flux ratio f as well as the spectrum shape factor α that will be used in the relevant equations (Appendix A) for calculating concentrations of elements from an radiated sample. Use will be made of either inner or outer irradiation channels for NAA depending on the nature of the sample to be analysed and flux intensity required.

Determination of the neutron spectrum shape factor α and flux ratio f for each irradiation channel is necessary if the single comparator method is to be applied for routine NAA. In fact it is the first experimental assignment to be carried out by the Nigerian scientist on commissioning NIRR-1. This is the reason for undergoing this research work. The Ghanaian reactor was used with the hope that the knowledge gained will be applied to NIRR-1 on commissioning since both reactors are similar, supplied and installed by the same vendor (China). In addition, a research work like this will promote exchange of scientific data, standardization, quality assurance, and experience, which will greatly enhance scientific development of nuclear technology within the West African sub-region.

In view of the fact that NAA has wide application which include Agriculture (tracer technique, radio isotope production, seed mutation and soil analysis), Medicine (radio-immunoassay, radio therapy e.t.c.), Industry (materials testing and analysis) and Environment (trace element analysis), this type of research work will open a new frontier in scientific development of Nigeria and a long term benefit to the entire African community.

1.4 Objective

It has been shown earlier (Moens *et. al*., 1979; De Corte *et. al*., 1982; Simonits *et. al*., 1984) that the actual epithermal neutron flux distribution can deviate significantly from the ideal $\frac{1}{E}$ law, and can therefore be approximated by $\frac{1}{E^{1+\alpha}}$ function with α as the deviation parameter. Therefore, when using either absolute, relative or single comparator method in reactor neutron activation analysis, the deviation of epithermal neutron flux should not be neglected (see Appendix A).

Through elastic collisions with moderator nuclei, fission neutrons (with energies ranging between and up to 15 MeV) rapidly become thermalized creating a broad energy distribution consisting of three principal components namely *thermal* with mean energy of 0.025 eV, *epithermal* with energy between 0.5 eV and 1 MeV and *fast* from 1 to 15 MeV (Knoll, 1979). The thermal and epithermal neutrons are the most widely used for irradiating samples (Connolly *et. al*., 1963; Moens *et. al*., 1979; 1981; De Corte *et. al*., 1979; 1982; Simonits *et. al*., 1975; 1984) because their cross sections for *(n, γ)* reactions are the largest thus offering the greatest analytical sensitivity available for NAA. All

relevant equations in NAA calculations therefore have to be modified to take into account the deviation of the neutron flux.

Since the GHARR-1 is similar to NIRR-1 and for the fact that the latter is yet to be commissioned –it is the objective of this work to measure the neutron flux ratio *f* and the epithermal neutron deviation parameter α which is to be used in correcting Q_0 to $Q_0(\alpha)$ via the resonance integral for both inner and outer irradiation channels of GHARR-1 for the fact that both channels are often used for NAA. The corrections are necessary in preserving accuracy of all NAA results.

Another reason for undertaking this research work is that effective resonance energy $\overline{E_r}$ has earlier been calculated and tabulated for 96 and recently 128 isotopes (Moens *et. al*., 1979; Simonits *et. al*., 1984) using THETIS reactor, Gent. And as a check for these calculated values using Ghanaian and Nigerian MNSR, or for isotopes where no, or only questionable, resonance parameters are available (De Corte *et. al*., 1982), an experimental $\overline{E_r}$ determination has been developed by Simonits *et. al*., 1984 and recently proposed for determination using Nigerian Research Reactor-1 by Ahmed *et. al*., 2002. The significance of this energy $\overline{E_r}$ as concluded by Ryves, 1969 is that it represents a single average energy, which replaces all the resonance energies. And in the experimental determination of $\overline{E_r}$ the only determinable variables are the flux ratio *f* and neutron spectra shape factor α. By obtaining *f* and α in a number of irradiation channels, cadmium ratios of the respective channels can be deduced. The cadmium ratios are the ones used in evaluating effective resonance energy $\overline{E_r}$. It is therefore the objective of

this work to determine f and α as a basis for future experimental determination of effective resonance energy with MNSR reactors.

CHAPTER TWO

NEUTRON SOURCES

2.1 Neutron Sources

Neutron sources can be classified in several different ways; however, the most appropriate classification seems to be according to production types.

2.1.1 Neutron Generators

Nowadays so many types of neutron generators exist. Their basic operating principle is based on the fact that charged particles are accelerated to an adequate energy and then directed to strike a target. The most prominent reactions suitable for neutron production are the *(p, n)* and the (*d, n*) reactions.

A reaction which is frequently used for production of neutrons in the energy range of 2.5 MeV to about 7.0 MeV is ^{2}H (d, n) ^{3}He with Q = 3.27 MeV. Neutron Generators like the one available in CERT, ABU Zaria uses the reaction ^{2}H(d, n) ^{4}He with Q = 17.58 MeV to produce monoenergetic neutrons of energy 14 MeV.

2.1.2 Isotopic Neutron Sources

This is another neutron source that is also available in CERT, ABU Zaria. This type of neutron source comprises an alpha-radioactive material closely packed with neutron emitter like beryllium. The reaction producing the neutron is ^{9}Be (α, n) ^{12}C with Q = 5.7 MeV. Typical α-emitters are ^{226}Ra *(E_{α}= 4.78 MeV),* ^{239}Pu *(E_{α}= 5.16 MeV) and*

^{210}Po *(E_α= 5.30 MeV)*; the mixtures are known as Ra – Be, Pu – Be and Po – Be neutron sources. The α-particles experience considerable energy loss before interacting with beryllium, and as a result the energy spectra of the neutrons produced is continuous.

2.1.3. Nuclear Reactors

Among the numerous neutron sources available, nuclear reactors with high fluxes of neutrons from the fission of uranium –235 give the most intensive irradiations better than Neutron generators and Isotopic sources and hence, has the highest available sensitivities for Neutron Activation Analysis (NAA).

The neutron energy distributions of reactors vary considerably in relation to the type of reactor and between irradiation channels within a reactor. Fission produced neutrons have energies ranging up to 15 MeV with an average energy of about 2 MeV (IAEA; 1990). Through elastic collisions with moderator nuclei, the fission neutrons rapidly become thermalized creating a broad energy distribution consisting of three principal components-thermal, epithermal and fast.

The GHAna Research Reactor-1 (GHARR-1) is the source of neutrons for this work. The salient aspects of the reactor are discussed below.

a) Description of GHARR-1 Reactor

The Ghana Research Reactor-1 (GHARR-1) is a Chinese Miniature Neutron Source Reactor (MNSR) type with a thermal power rating of 30 kW. This reactor is similar to the installed NIgerian Research Reactor-1 (NIRR-1). GHARR-1 is a tank-in-pool reactor that uses 90.2% enriched Uranium fuel cladded with Aluminium. The

reactor is cooled and moderated with light water, while beryllium acts as a reflector (Yang, 1992).

There are 344 fuel pins concentrically arranged in 10 rings with six dummy rods uniformly spaced on the outer most ring (Table 2.1). Four tie rods located in the eight rings, hold the lower and upper grid plates in position. The fuel cage is located on a 50 mm thick metallic beryllium annulus. The water above the core acts as a reflector.

For long term reactivity compensation due to fuel burn-up and Samarium poisoning, beryllium shims of varied thickness are to be placed (as the demand arises) in an aluminium tray located on top of the reactor core. The addition of the shims is another way of elongating the life span of the core as will be discussed later.

Table 2.1 Technical specification of GHARR-1

Parameters	Description
Reactor Type	Tank-in-pool
Rated Thermal Power	30kW
Fuel	UAl_4Dispersed in Al base material
U-234 enrichment	90.2%
Core Shape	Cylinder
Core diameter	23.1 *cm*
Core height	23 *cm*
No. of fuel elements	344
Weight of U-235	998.116*g*
No. of irradiation Channels	10
Inner irradiation channels	5
Flux (inner channel)	1.0×10^{12} $ncm^{-2}s^{-1}$
Flux (outer channel)	5.0×10^{11} $ncm^{-2}s^{-1}$
Reactor cooling mode	Natural convection
Height of inlet orifice	6mm
Height of outlet orifice	7.5 mm
Diameter of fuel meat	4.3 mm
Diameter of fuel element	5.5 mm
Excess reactivity	3.99 mk
Length of Cd control rode	230 mm

The GHARR-1 core is a small compact system consisting of a central control rod guide tube, the fuel cage and fuel rods arranged in ten concentric lattice rings about the control guide tube. The average radial pitch between any two adjacent fuel rings is 10.95 mm. Table 2.2 shows the dimensions of the fuel lattice rings of the core.

Table 2.2: Geometrical dimension of GHARR-1 core lattice

Fuel Ring No.	No. of fuel rods	Diameter (mm)	Radial Pitch (mm)
1	6	21.90	11.47
2	12	43.80	11.47
3	19	65.70	10.86
4	26	87.70	10.98
5	32	109.50	10.78
6	39	131.40	10.58
7	45	153.50	10.70
8	52	175.20	10.58
9	58	197.10	10.68
10	65	219.00	10.58

Due to its small size, the core is heavily reflected on both sides and on the bottom by beryllium alloy to reduce neutron leakage as well as the criticality condition. The designed cold clean core excess reactivity is about 4 mk at 20^0C (Yang, 1992). The reactor has a pin-type fuel element with two end-plugs consisting of 90.2% highly

enriched Uranium with Aluminium as cladding material. The reactor has only one control rod made of cadmium and cladded with stainless steel. It has reactivity worth of about 7 mk (Yang, 1992; Anim-Sapong *et. al*., 2000). The design parameters of GHARR-1 fuel are shown in Table 2.3.

Table 2.3 Design Parameters of GHARR-1 Fuel

Description	Rate
Reactor thermal power	30 kW
Max. heat flux at fuel element surface	3.56×10^3 kW/m^2
Inlet/outlet coolant temp.	$30^oC/54^oC$
Coolant Velocity	15.30 mm/s
U^{234} content/fuel element	2.88g
Enrichment factor	90.2%
Uranium Weight fraction/fuel	27.5%
Fuel density	3.36203 g/cm
Volume porosity	3%
H/U Ratio	197

The use of a single control rod in the operation of MNSR is due to its inherent safety features and simplified drive mechanism. The rod performs shim, safety and regulatory control functions. This makes the reactor safe and compact enabling it to be located in densely populated area without harm to the public. Fig. 2.1 shows the schematic diagram of the reactor.

Fig. 2.1 Schematic representation of GHARR-1 core showing irradiation channels

(Top View)

Fig. 2.2 Schematic representation of GHARR-1 core showing irradiation channels

(Side View)

b) Safety features of GHARR-1

Ghana research reactor-1 is designed to incorporate some inherent safety characteristics in order to prevent radiation leakage. The objective of its design was to achieve a small compact core structure with under moderation achieved by choosing highly enriched uranium as fuel material with water serving as both moderator and coolant (Yang, 1992). The core structure is simplified by arranging the irradiation channels in the beryllium reflector and this reduces the effect on flux distribution on the irradiation experiments due to the movement of the central control rod (Yang, 1992; Akaho *et. al*., 2000) and therefore enhances the performance of the reactor from both mechanical and computational physics point of view.

Based on the computational reactor physics and thermal hydraulic calculations two major provisions were considered (Yang, 1992) to increase safety margin and to enhance the inherent safety of the reactor.

The ratio of Hydrogen to Uranium ($H/^{235}U$) atoms is reported to be 197 (Yang, 1992) in order to enlarge the degree of under-moderation of the core and thereby increase the negative moderation temperature coefficient and enhance the temperature feed back. The second consideration is that the ratio of the core height to diameter, which was optimized to 1.0 by the manufacturers, has a beneficial effect on the axial neutron leakage fraction and the worth of the top beryllium reflectors (shim plates) used for the extension of the lifetime of the core.

In addition, the optimization of the core height to unity increases the coupling of outlet and inlet flow mixing of the water coolant and enables part of the warm coolant to directly enter the core, thus shortening the time of the temperature feed back. Therefore, an appropriate choice of inlet flow orifice is necessary for the core thermal hydraulics design to yield the desired rise in temperature for ensuring an inherent safety of the reactor. The design is such that the temperature should not be too high to permit the occurrence of boiling in the core and cause a large temperature effect that will substantially reduce the excess reactivity for the compensation of Xenon-poisoning.

As a result of the above discussed features on inherent safety, stability of flux and moderate cost, the MNSR has recently, found enormous application in various fields of science particularly NAA (Kennedy, *et. al*., 2000; Akaho and Nyarko, 2002).

CHAPTER THREE

METHODOLOGY

3.1 Determination of thermal to epithermal flux ratio

During the operation of a reactor the flux is expected to be stable. In view of this, the thermal to epithermal flux ratio has to be determined for a new channel (uncharacterised) and monitored frequently for an old channel (characterised) of a reactor to establish the flux stability. Usually, the flux for an uncharacterised channel is experimentally determined (Simonits *et al*., 1976; De Corte *et. al*., 1981; 1982; 1987) by the following methods:

(i) Cadmium-ratio method

(ii) Bare-monitor method.

3.1.1 Cadmium- ratio method

The ratio of thermal to epithermal flux f of a reactor is defined in equation 1.6. It can be observed that the ratio is a factor of only two parameters Q_0 and R_{cd}, which can both, be determined experimentally. However, when studying the effect of non-ideal epithermal distribution, Q_0 has to be corrected with the epithermal shaping-factor α (equation 1.8) to take into account deviations from *1/E*- distribution associated with the epithermal flux.

Cadmium ratio method involves irradiating bare and cadmium covered monitors and determining their induced activity. The flux ratio is thereafter obtained from the experimentally determined cadmium ratio using the induced activity. The major advantage of the method is that it avoids the introduction of k_0-factors, which sometimes has to be determined experimentally to ensure accuracy. However, due to flux depression caused by insertion of a neutron absorber such as cadmium, the method may not be suitable for application at reactors that attain a considerable fuel burn-off such that their excess reactivity is much less than that recommended. To avoid situations like this, the second method is an alternative.

3.1.2 Bare-monitor method

This method is sometimes called "intensity ratio" method. It can be adopted in situations where the double irradiation technique discussed above cannot be used particularly for reactors in which the introduction of a cadmium box is not allowed.

For ideal reactor flux distribution, the ratio of thermal to epithermal flux f can be obtained (Simonits *et al.*, 1975) as:

$$f = \frac{\frac{k_{o,95}}{k_{o,97}} \bullet \frac{\varepsilon_{\gamma,97}}{\varepsilon_{\gamma,95}} Q_{o,97} - \frac{I_{sp,95}}{I_{sp,97}} Q_{o,95}}{\frac{I_{sp,95}}{I_{sp,97}} - \frac{k_{o,95}}{k_{o,97}} \bullet \frac{\varepsilon_{\gamma,97}}{\varepsilon_{\gamma,95}}} \qquad (3.1)$$

The subscript '95' refers to ^{94}Zr (n,γ) ^{95}Zr and '97' to ^{96}Zr (n,γ) ^{97}Zr reactions respectively; and their efficiencies (ε) and constants (k_0) for each of the Zr isotopes written in terms of the subscripts '95' and '97'.

When considering deviations from *1/E* distribution, Q_0 in equation 3.1 above has to be corrected with α. The Q_o values could either be determined experimentally or obtained as nuclear data from the literature compilations of Moens *et. al*. 1984; Jovanović *et. al*., 1987 and De Corte *et. al*., 1989 (Table 3.1).

Table 3.1 Nuclear data of the monitor isotopes used

MONITOR	$T_{½}$	E (k*e*V)	Q_O	$k_{O\ AU}$	E_r (eV)	F_{cd}	σ
$^{94}Zr(n, \gamma)^{95}Zr$	64.03 d	724.2	5.06	1.972E-4	6260.0	1.0	0.05
$^{197}Au(n,\gamma)^{198}Au$	2.696 d	411.8	15.71	1.0	5.65	0.99	98.8

The advantage of the method is that it is very favourable for situations in which the introduction of cadmium into the reactor is not acceptable. The method equally has low precision for some nuclides particularly for those that require short-lived irradiations. Also the following isotopic combinations ^{97} Ru – ^{103} Ru and ^{198} Au – ^{97} Zr (Table 3.2) could be used for flux ratio measurements in situation where Zirconium foils are not available in determining the ^{94} Zr (n,γ) ^{95} Zr reaction.

Table 3.2 List of suitable α-monitor pairs.

^{197}Au-^{94}Zr	^{197}Au-^{100}Mo	^{109}Ag-^{94}Zr
^{181}Ta-^{94}Zr	^{197}Au-^{68}Zn	

3.2 Determination of epithermal flux-shaping factor (α)

Apart from the flux ratio, another important reactor characteristic to be determined for each irradiation channel is the epithermal flux-shaping factor. It has been shown in earlier publications (Ryves, 1969; De Corte *et. al*., 1979; Moens *et. al*., 1979 and Jovanović *et. al*., 1987) that the factor α is essential for the correction of resonance integral, i.e. for the conversion of I_o-values (only valid in an ideal 1/E-epithermal reactor neutron spectrums) to $I_o\ (\alpha)$-values, for use in actual $1/E^{1+\alpha}$ spectra.

In practice, α-determination is required mainly in the calibration of an irradiation channel and in performing reactor-neutron activation without standards (e.g. using the k_o-standardization method). The former case requires the most accurate α-determination possible. De Corte *et. al*., 1981 recommended either the cadmium-covered method and the cadmium-ratio method stating the merits and demerits of each method.

In most analytical cases a method that uses a triple monitor set like ^{197}Au-^{96}Zr-^{94}Zr provides accepted precision and accuracy as tested and verified extensively by De Corte *et. al*., 1987 in the *THETIS* reactor and the *WWR-M* reactor and other authors in NAA (Table 3.3). Some α-determination methods were found to be extremely sensitive to the accuracy of the nuclear data used (De Corte *et. al*., 1981). In order to avoid this, the use of k_o-factor and carefully selected nuclear parameters- Q_o, and E_r was suggested (De Corte, 1987; Jovanović *et al.*, 1989; Kennedy *et. al*., 2000; Acharya *et. al*., 2001; Akaho and Nyarko, 2002).

Table 3.3 Experimentally determined values of f and α for different research reactors

Research Reactors	Monitors	Reactor Power	Channels	f	α	Reference
TRIGA MARK II Ljubljana, Yugoslavia	^{197}Au-^{94}Zr	250 kW Light-Water Moderated	CC	≅ 20	-0.057±0.010	Jovanović *et al.*, 1989
			PT	≅ 20	-0.046± 0.011	
			CR25	≅ 20	-0.014 ±0.006	
WWR-M, Budapest, Hungary	^{197}Au-^{238}U-^{98}Mo-^{100}Mo-^{64}Zn	5,000 kW Light-Water Moderated	MILA	36.01± 0.82	-0.014 ±0.006	De Corte *et al.*, 1987
DR-3,Risø, Denmark	^{197}Au-^{238}U-^{98}Mo-^{100}Mo-^{64}Zn	10,000 kW Heavy Water Moderated	R4V4	337± 7	-0.158 ± 0.11	De Corte *et al.*, 1987
THETIS, Gent, Belgium	^{197}Au-^{238}U-^{98}Mo-^{100}Mo -^{64}Zn	250 kW Light-Water Moderated	7	110.2± 1.9	-0.0713±0.0088	De Corte *et al.*, 1987
			14	38.35± 0.70	-0.0298±0.0089	
			16	104.4± 1.8	0.0792±0.0089	
SLOWPOKE-2 Halifax, Canada	^{197}Au-^{94}Zr-^{96}Zr	20 kW Light-Water Moderated	5	18.9	-0.0427	Akaho and Nyarko, 2002
			1	56.5	-0.0098	
CIRUS, Bhabha Atomic Centre, India	^{197}Au-^{94}Zr-^{96}Zr	40,000 kW Heavy-Water Moderated	PT	80.0±5.3	-0.011±0.004	Acharya *et al.*, 2002

Apart from Au-Zr combination, which was used for this work, other monitor sets may as well be used (De Corte *et. al*., 1981) giving also results with acceptable precision and accuracy (Table 3.2). However, it should be emphasized that, even with otherwise suited monitors, extreme care should be taken about the choice of the accurate Q_o-values which is to be introduced in the expression for α-calculation. It is necessary to correct the values with epithermal shaping-factor α to take into account deviations from 1/E-distribution associated with the epithermal flux.

Procedures for experimental determination of α-values were intensively discussed by Ryves, 1969 and De Corte *et. al*., 1981. They all point to three distinct approaches:

1. Cadmium covered method
2. Bare monitor method.
3. Cadmium ratio method

Comparing the three methods in terms of simplicity and accuracy, the bare method seems to be the simplest (particularly for reactors where introduction of cadmium is not allowed) and suited for rapid α-determination. However, from the accuracy point of view, the cadmium ratio method yields the most accurate results (De Corte *et. al*., 1981; Akaho and Nyarko, 2002) and that is why this option was chosen for this project.

3.2.1 Cadmium-covered method

For monitors that have cross-section dependence [$\sigma(v) \approx 1/v$] up to 1-2 eV, α can be obtained by irradiating simultaneously N monitor-sets in one container, or by

performing N subsequent irradiations of analogous monitor sets. The α-factor can then be computed from the specific count rates of the irradiated monitors.

A number of isotopes do not have well–established nuclear data which can be obtained from the literature. Such nuclear data used for α-determination should be treated with caution. Hence the arbitrary selection of monitor isotope pairs should be viewed with good precision of the nuclear data used for the calculation of α.

The fixed accuracy and the overall uncertainty of the method will generally improve when replacing the nuclear data (with their associated large uncertainties on σ_o and γ) by the corresponding k_o-factor determined. Experimentally determined $k_{o,Au}$ factors with accuracy < 2% (usually 1%) are found in literature (Moens *et. al*., 1984). Also, De Corte. *et. al*., 1981 suggested a list of practically suited α-monitor pairs with known k_o-values and acceptable error propagation factors. Some of these monitors are listed in Table 3.4. This method was not used in this work for the reasons discussed in 3.2.2 and 3.2.3 below.

Table 3.4: Suggested triple monitors for α-determination

$^{197}Au - {}^{94}Zr - {}^{96}Zr$	$^{109}Ag - {}^{94}Zr - {}^{96}Zr$
$^{181}Ta - {}^{94}Zr - {}^{96}Zr$	$^{175}Lu - {}^{94}Zr - {}^{96}Zr$
$^{121}Sb - {}^{94}Zr - {}^{96}Zr$	$^{123}Sb - {}^{94}Zr - {}^{96}Zr$
$^{45}Sc - {}^{175}Lu - {}^{96}Zr$	$^{50}Cr - {}^{181}Ta - {}^{96}Zr$
$^{181}Ta - {}^{68}Zn - {}^{96}Zr$	$^{197}Au - {}^{112}Sn - {}^{94}Zr$

3.2.2 Bare monitor method

This method is the only technique enabling α-determination without cadmium-cover irradiations. The method is therefore important in (n, γ) activation analysis, using k_o-standardization method, and in all nuclear experiments (e.g. cross-section measurements e.t.c) where cadmium-covered irradiations is not allowed. Since no cadmium-cover is used in this method, the uncertainty on cadmium cut-off energy is not involved, thus allowing the use of a 1/v – monitor.

Theoretically, the best condition for this method would be:

1. Combination of one pure 1/v- monitor with two monitors showing a large spread on their E_r – values and with almost equal, high Q_0 – factors (since no cadmium-cover is involved).
2. Another, second best choice appears to be a set of three monitors with large spread in E_r and in Q_0- values, and if $Q_{0,1} < Q_{0,2} < Q_{0,3}$, then $E_{r,2} < E_{r,1} < E_{r,3}$ is preferred (De Corte *et. al*. 1979; 1981). If the above condition cannot be achieved, other sequence of E_r have to be accepted, where it should be absolutely avoided that the middle E_r-value corresponds to the middle Q_0-value.

The advantage of the ^{197}Au – ^{94}Zr – ^{96}Zr set is that none of the measured isotopes emits gamma rays in cascade, so that no true coincidence effect should be corrected for at a small source-detector distance (De Corte *et. al*., 1981; Gunnik, 1990). Thus, simple co-irradiation of a standardized Au and thin Zr foil enables the simultaneous determination of f and α.

Despite its numerous advantage, this method could not be adapted for this work because Q_0-values for so-called low 1/v monitors are generally known with a much worse

accuracy than for high ones. This type of sets turns out to give unacceptable error propagation factors as investigated by De Corte *et. al*.; 1981. Therefore, a combination of Au with Zr cannot be recommended for this method but very useful for other methods like cadmium ratio method discussed.

The disadvantage of the method is that it suffers from high error propagation factors, and thus considerable uncertainty on the determined α has to be expected. When it became inevitable that no any other method is suitable except this method (e.g. introduction of neutron flux absorber like cadmium or boron is not allowed into the reactor) then the following ways of reducing uncertainties (De Corte *et. al*., 1981) should be considered:

1. Judicious choice of monitors with accurately known k_o and Q_0 – values.
2. Utmost experimental care to minimize gross errors.
3. Improvement on counting statistics by irradiating several sets of monitors to obtain better standard deviations on the means of the A_{sp}-values.

Error analysis by De Corte *et. al.,* 1981 reveals that selection of a number of monitors with uniformly distributed E_r-values, ranging from low to high, will yield good results. Furthermore, the reference monitor should preferably have a relatively low Q_0-value, with all other monitors (within pairs or series) showing high Q_0-values. As observed by De Corte *et. al*., 1981 the accuracy and precision on α-values are markedly improved by increasing the number of monitors in the bare method. However, it should be emphasized that such an improvement can only be obtained after careful selection of the monitor isotopes, and especially of the reference monitor. Table 3.4 above gives some possible combination of triple monitor pairs as suggested by De Corte *et. al*., 1981.

IAEA, 1990 document shows that an isotope, like ^{197}Au, in combination with two Zirconium isotopes is a favourable one, since Zr additionally can serve as a flux ratio monitor. The advantages of using such combination are discussed in the method below.

3.2.3 Cadmium-ratio method

Using R_{cd} values determined experimentally for a set of monitor pair and their respective Q_o, E_{cd} and E_r values taken from literature, the α-value could be obtained by solving equation 1.11 through an iteration method with the aid of a computer program as will be discussed below in Section 3.3. This method has the advantage of avoiding the introduction of absolute nuclear data (M, θ, γ, σ) and could be achieved via simple measurements using a detector. Also the method is very flexible in that if similar monitor sets are irradiated with and without cadmium-cover, it has the important advantage of cancelling out gross errors due to γ-attenuation, different counting geometries for both monitors, true coincidence effects and inaccurate peak area determination.

The major disadvantage or setback of the method is the sensitivity of the method to time stability of reactor neutron spectrum, due to double irradiation required. However, this problem is taken care of by the type of reactor used for this work. The MNSR reactor like the Nigerian NIRR-1 and Ghanaian GHARR-1 has long-term stability of the neutron flux as reported by Yang, 1992; Qazi *et. al.*, 1996; Akaho and Nyarko, 2002. The core configuration of these reactors are not altered until a burn-up of about 1% is achieved after the core is operated for more than 10 years. This informed the choice of the method for this work.

Although this method is undoubtedly superior to the cadmium-covered method with respect to fixed accuracy, the overall uncertainty may become worse if care is not taken when selecting monitor pairs, which may contribute in error due to counting statistics. This holds especially for isotopes with high Q_o-values, which should not be used. The only possibility to reduce this contribution is trying to obtain good precision on the specific contribution rates, e.g. by performing a large number of experiments as was carried out in this project. Thereafter, the data obtained for R_{cd} , Q_o, E_{cd} ,E_r were used for a computer iteration in order to obtain the value for α.

3.3 Computer program for iteration

Computer programs and codes have recently found enormous applications in problem solving (Middleton *et. al.*, 1989; Maps, 1993). For so many years now, physicists use their research findings to develop iterative computer programs (Eckstein, 1993; Richard, *et. al*., 1996; Kozielska, 1996; Balogun, 2003).

In an effort to ease the somehow cumbersome and time-consuming manual iteration method for the determination of the epithermal flux-shaping factor, we developed a routine computer program (Q-Basic) based on equation 1.11. The program is developed in an Exec-format, and using relevant nuclear parameters and experimental values as input data (Table 3.5).

Table 3.5: Nuclear parameters used for evaluation of cadmium ratio

Reactor	Channel	Monitor	Q_o	E_r	E_{cd}	R_{cd} (Expt.)
GHARR-1	5 (inner)	Au-197	15.71	5.65	0.55	2.10
		Zr-96	251.6	338	0.55	
		Zr-94	5.06	6260	0.55	2.68
GHARR-1	7 (outer)	AU-197	15.71	5.65	0.55	3.57
		Zr-96	251.6	338	0.55	
		Zr-94	5.06	6260	0.55	3.52

To obtain a more rapid convergence to a solution of the function being iterated (equation 1.11) and to validate results obtained with the Q-Basic formalism, another program was written in C++ language based on local convergence theorem of a fixed-point iteration (Turner, 1994). Salient features of the two programs are hereby discussed.

3.3.1 Discussion on the Computer programs

The success of a program as a tool to problem solving may depend on control and data structures available within the program that will give confidence to results produced (Watts, 1987; Fenton, 1991; Hopkins, 1996). BASIC (Beginners All-Symbolic Instructive Code) and C++ programs are general-purpose computer languages with in-built structures that make them easily understandable while maintaining the precision necessary in computer language (Penfold, 1993). Both languages have advanced string-handling features that make them strong mathematical tools. The iteration of equation 1.11 could manually take days to complete due to large possibilities for trial-values of

several possible step lengths that exist, whereas this could be achieved within minutes using computer programs.

3.3.2 Development of the programs

The Q-Basic program was developed based on the linear method of iteration and it works by requesting nuclear parameters (R_{cd} Q_o, E_r and E_{cd}) of the monitor isotopes (Table 2) and 'trial' α-values. The initial trial α-value is used to generate a sequence of values by iterating the function $f(\alpha)$ (Equation 1.11) within a specified step-length as expressed in Equation 3.2 below. Choice of trial α-values for every step increment depends on the iteration results until a finite point of convergence is reached.

$$\alpha_{n+1} = f(\alpha) \qquad 3.2$$

For the iteration function $f(\alpha)$ of this work as obtained from Eq. 1.11 it was noticed that a trial value of -0.5 makes the function undefined (zero denominator). To avoid this type of non-convergence the program is designed to skip this oscillatory point (Fig. 3.1) by assigning a unity solution to it and send a warning signal to the user before accepting the next expected value. However, this method makes the function slow in converging and so a faster method had to be developed in C++ language.

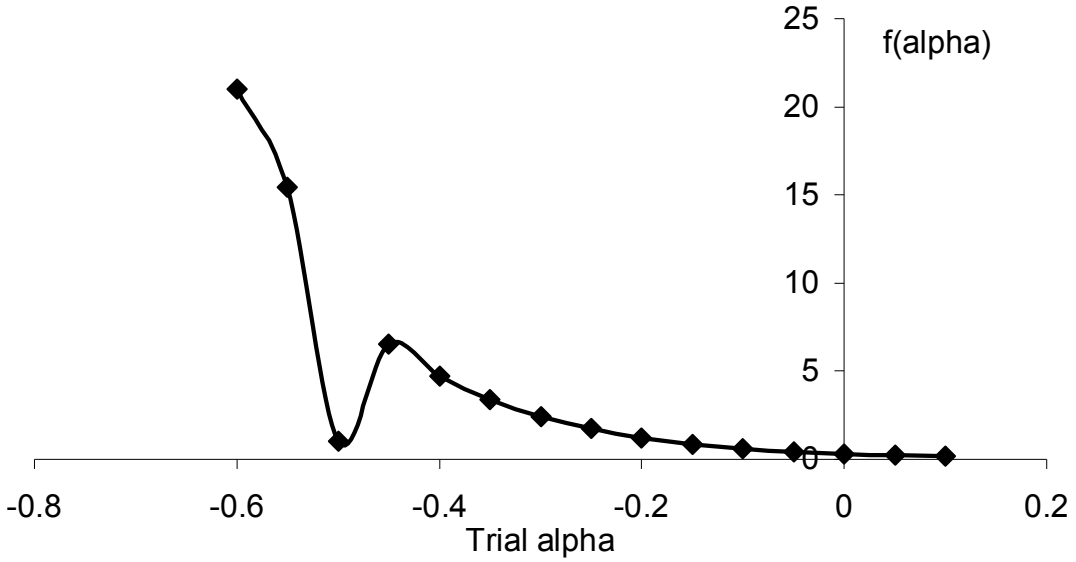


Fig.3.1 Oscillatory behavior of trial alpha with $f(\alpha)$ for a linear iteration

The C++ is designed to estimate the number of iterations necessary to achieve convergence for a given function by relying on derivative of the function, which normally yields a linear first order convergence if $f'(\alpha)$ is not equal to zero or does not vanish at the solution - a situation that usually leads to divergence as given later (Eq. 3.6). This method is popularly called *Newton-Raphson* (Turner, 1994). The method implies that the function $f(\alpha)$ is differentiable and its graph has a definite slope at each point hence a unique tangent line (Fig. 3.2). By extending the tangent line to the curve of $f(\alpha)$ at an initial alpha value say α_0 to intersect the α-axis at α_1 and similarly α_n to α_{n+1}, we obtain an acceptable value of alpha that satisfies $f(\alpha)$. Analytically it means that:

$$\alpha_1 = \alpha_0 - \delta\alpha$$

.

$$\alpha_n = \alpha_{n-1} - \delta\alpha$$

$$\alpha_{n+1} = \alpha_n - \delta\alpha \tag{3.3}$$

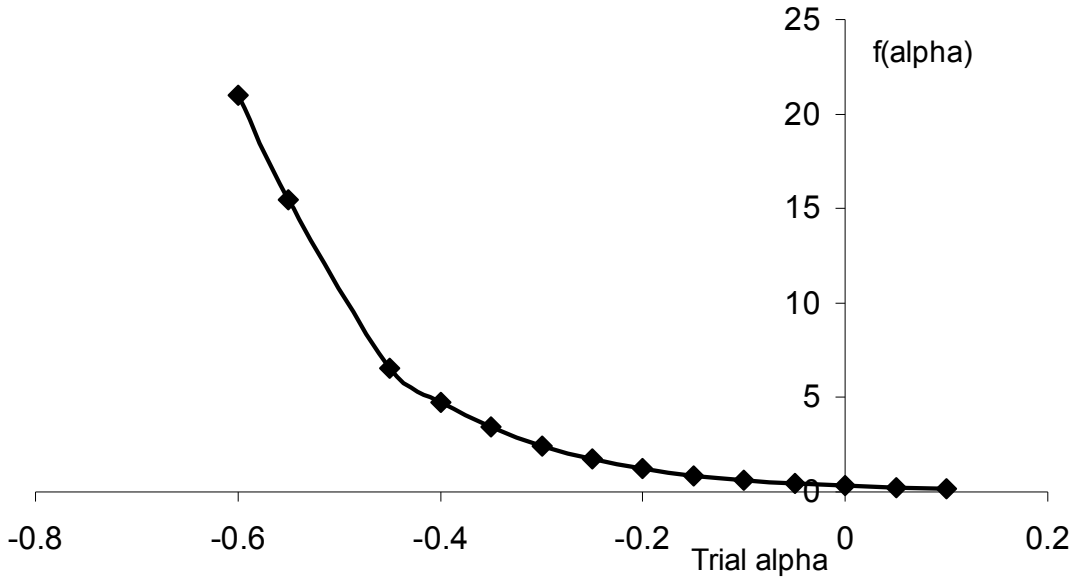


Fig. 3.2 Graph of trial alpha with $f(\alpha)$ for *Newton-Raphson* and *Secant* methods

The tangent to the curve at point $(\alpha_0 f(\alpha_0))$ on the graph (Fig. 3.2) is Thus:

$$f^1(\alpha_0) = \frac{f(\alpha_0)}{\delta\alpha} \Rightarrow \delta\alpha = \frac{f(\alpha_0)}{f^1(\alpha_0)} \tag{3.4}$$

So that substituting Equation 3.4 into 3.3 we have:

$$\alpha_1 = \alpha_0 - \frac{f(\alpha_0)}{f'(\alpha_0)} \tag{3.5}$$

In general Equation 3.5 could be written as:

$$\alpha_{n+1} = \alpha_n - \frac{f(\alpha_n)}{f'(\alpha_n)} \tag{3.6}$$

The *Newton-Raphson* method converges faster than the linear method used in the Q-Basic program, however, the major disadvantage of the method is failure to converge to the correct alpha value if $f'(\alpha_n)$ becomes very small or vanishes – a situation that permits the second term in the right hand side of Equation 3.6 to become large and consequently cause the iteration to diverge.

In situations where the derivative of the function in Equation 3.6 (*Newton-Raphson* method), vanishes, the C++ program is designed to use *divided difference approximation method* usually known as *Secant method* (Turner, 1994) to get solution of the iterate between any two selected points (Equation 3.9) say α_{n-1} and α_n. Instead of drawing a tangent to the graph of $f(\alpha)$ (Fig. 3.2) a *Secant* line that intersect the α-axis at a point say α_{n+1} is drawn and the troublesome $f'(\alpha_n)$ in Equation 3.6 above is replaced by the definition of $f'(\alpha)$. Thus:

$$f^1(\alpha) = \lim_{y \to \infty} \frac{f(\alpha + y) - f(\alpha)}{y} \approx \frac{f(\alpha + y) - f(\alpha)}{y} \tag{3.7}$$

If $\alpha = \alpha_n, y = \alpha_{n-1} - \alpha_n$ then Equation 3.7 transforms to:

$$f^1(\alpha_n) \approx \frac{f(\alpha_{n-1}) - f(\alpha_n)}{\alpha_{n-1} - \alpha_n} \tag{3.8}$$

So that on substituting Equation 3.8 into Equation 3.6 and rearranging we have:

$$\alpha_{n+1} = \alpha_n - \frac{f(\alpha_n)(\alpha_n - \alpha_{n-1})}{f(\alpha_n) - f(\alpha_{n-1})} = \frac{\alpha_{n-1} f(\alpha_n) - \alpha_n f(\alpha_{n-1})}{f(\alpha_n) - f(\alpha_{n-1})} \tag{3.9}$$

This was the method adopted for the development of the C++ Computer program used for the determination of the epithermal flux-shaping factor (α) in this work.

CHAPTER FOUR

EXPERIMENTAL

4.1 Sample Preparation and Standards

4.1.1 *Gold Standard*

A standard gold solution of certified purity > 99.999% and concentration 1000 ppm supplied by Seigniory Chemical Products ® (SCP), Canada limited was used. About 5 µL of the standard solution was pipetted using an *Eppendorf* ® pipette and spotted onto a small piece of Whatmann® 4.7 cm filter paper. The filters with the sample were then placed inside a small plastic polyethylene vial (manufactured by *Emerald plastics Inc.* ®) of 1.3 cm internal diameter and 2.3 cm high. Three additional replicate standards were prepared in a similar fashion and the capsules heat-sealed. Out of the four standards, two were wrapped with cylindrical cadmium box of 1 mm thickness for cadmium cover irradiations. The four samples prepared were then sent to the reactor through a pneumatic transfer system for one-hour irradiation.

4.1.2 *Zirconium Foils*

Four zirconium foils of thickness 0.125 mm with masses between 4.7 – 5.0 mg were prepared with two of the samples wrapped with cadmium, placed inside a plastic polyethylene capsule and heat-sealed. The heat-sealed capsules were further inserted into slightly bigger polyethylene vials of internal diameter 2.1 cm for inner channel

irradiations and 5.9 cm for outer channel irradiations. All the vials were then heat sealed and transferred to the reactor for one-hour irradiation through pneumatic system.

4.2 Irradiations

Among the four sets of Au and Zr (one set being cadmium-covered and the other bare) as prepared above, two sets of the samples were irradiated at the outer channel 7 and the remaining two sets irradiated at the inner channel 5 of the GHARR-1 research reactor for an hour. The samples were irradiated with the reactor operating at half its rated thermal power (15 kW), with a neutron flux of 5×10^{11} n cm^{-2} s^{-1} in one of the inner irradiation channels (Table 4.1). At the end of irradiations, the samples were allowed to decay for an average of 24 hours before counting.

4.3 Counting Schemes

Counting of the irradiated samples was carried out using a gamma-ray spectroscopic system consisting of an N-Type High Purity Germanium (HPGe) detector model GR2518 with relative efficiency of 25% and with an energy resolution of 1.8 keV (FWHM) at 1332.5 keV gamma ray of ^{60}Co. The system is connected to a high voltage supply model 3105 manufactured by Camberra® Industries Inc. The spectroscopy amplifier model is 7611/L and NIM power supply model is PS01-B manufactured by SILENA® Milano. It also has an 8k ACCUSPEC Multi Channel Analyzer (MCA) emulation software, a 486-microcomputer for spectral data acquisition/evaluation, and SPAN 5.0 which is a multi-purpose gamma analysis software developed by China Institute of Atomic Energy. The detector operates on a bias voltage of (negative) 3000V.

The spectral intensities of the samples were accumulated for 30 (thirty) minutes by means of an MCA card. The samples were counted at a distance of 7.2 cm from the top of the detector surface. The efficiency parameters for NAA calculations were obtained by fitting the efficiency curve (Fig. 4.1) with a polynomial function using parameters earlier determined by Osae *et. al*., 1998

Table 4.1 Irradiation, decay and counting schemes for this work

Sample I.D.	Reactor Channel	Sample Weight	Time In	Time Out	Gamma Dose	Decay Time	A_{SP} (Bq/mg)
ZrCd-5	7(outer)	4.9 mg	11:07	12:07	20.7μsv/h	23.855 hr	1.945265
ZrB-6	5(inner)	4.9 mg	11:09	12:09	21.21μsv/h	26.654 hr	25.50205
ZrCd-7	5(inner)	4.7 mg	12:15	13:15	54.8μsv/h	21.992 hr	9.504894
ZrB-8	7(outer)	5.0 mg	12:17	13:17	457nsv/h	28.345 hr	6.84440
AuCd-6	5(inner)	5.0 mg	10:02	11:02	61.2μsv/h	22.853 hr	3003.893
AuB-7	7(outer)	4.5 mg	10:05	11:05	3.9μsv/h	27.257 hr	1635.62
AuCd-8	7(outer)	5.2 mg	13:22	14:22	22.7μsv/h	20.06 hr	457.87
AuB-9	5(inner)	5.3 mg	13:24	14:24	19.3μsv/h	24.988 hr	6307.19

Note: Reactor flux = 5×10^{11} n cm^{-1} s^{-1}

Irradiation date = 15th October 2002.

Irradiation time = 1 hour

Counting time = 30 minutes.

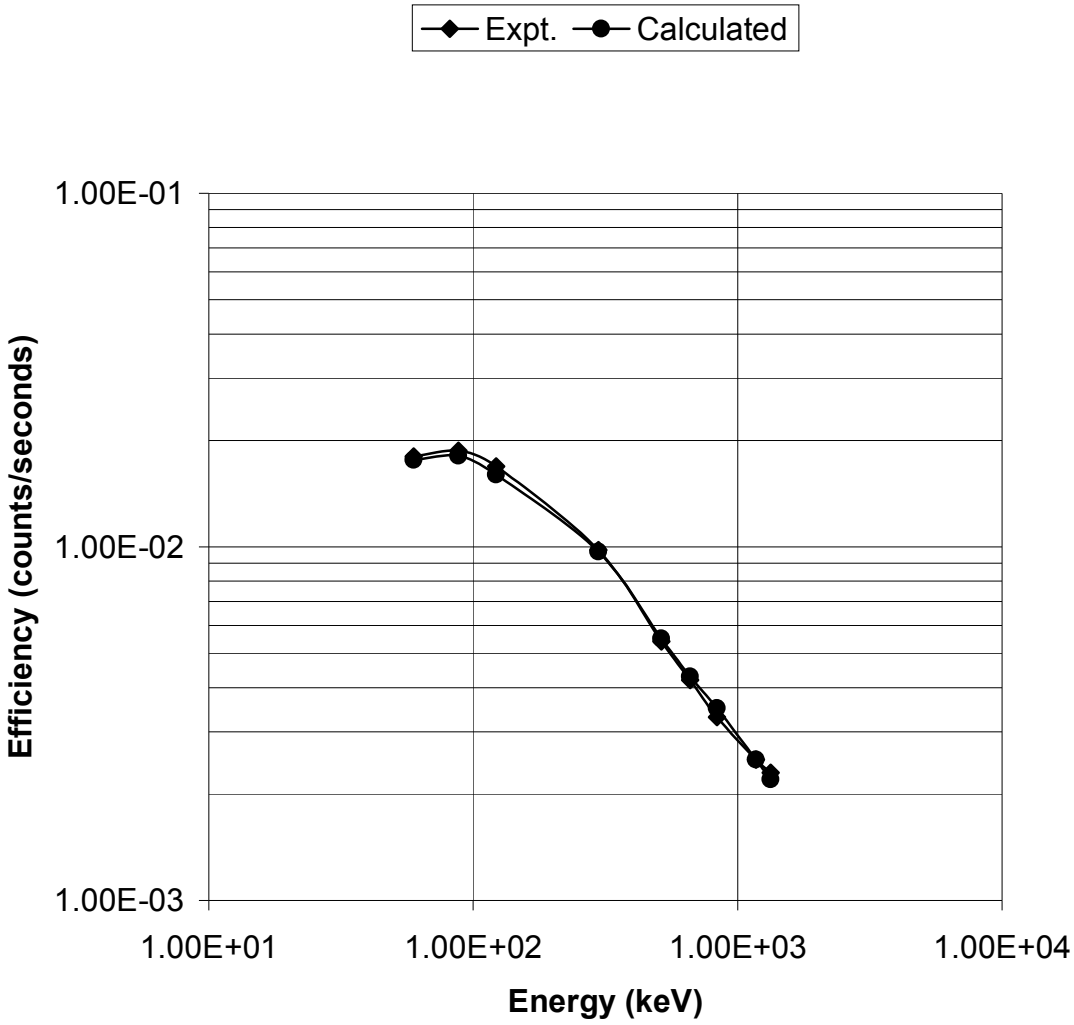


Fig. 4.1 FEPE curves at a geometry of 7.2 cm for an HPGe detector used.

4.4 Precaution

To permit measurement of epithermal flux, and also to ascertain that the filter used (cadmium foil of 1 mm thickness) does not itself interfere significantly with the resonance activation and does not also depress the reactor flux to an undesirable level, the choice of monitors used for this work (Au and Zr) was influenced by their activation

cross sections and energies of the main resonance contributing to activation integral. The nuclear data of the monitors involved were also carefully selected from literature.

Another aspect we considered during experimental process is contamination of samples. Care has been taken when handling samples to avoid contamination by hands or tools. Sample size as well as irradiation, decay and counting schemes are also chosen to give a fair representation of the samples irradiated.

CHAPTER FIVE

RESULTS

5.1 Results and Discussions

The measured activities of gold and zirconium foils for both bare and cadmium-covered irradiations are tabulated in Table 4.1 above. The activities were used in the determination of cadmium ratios (R_{cd}) for each of the samples irradiated via equation 1.7. The R_{cd} value obtained for gold is 2.1 for the inner channel (No. 5) and 3.57 for the outer channel (No. 7). For zirconium, the R_{cd} values obtained are 2.68 and 3.52 for inner and outer channels respectively as tabulated earlier in Table 3.5. Thereafter the thermal to epithermal neutron flux ratio f and epithermal neutron flux shaping-factor α were then evaluated for both the inner and outer channels using the R_{cd} values.

In the determination of α, Computer programs in Q-Basic and C++ languages were developed to ease the iteration method required for the evaluation of α from R_{cd} values. For completeness and comparison of their merits, each of the two programs developed (in Q-Basic and C++ languages) were used in this thesis (independently) to determine alpha for the inner channel (5) and outer channel (7) of the Ghana Research Reactor-1 (GHARR-1) with Au and Zr serving as monitor pairs.

The nuclear parameters for the gold and zirconium monitors used in this work were obtained from literature as reported and updated by Simonits *et al*., 1976; De Corte *et. al*., 1981; 1982; Jovanović *et. al*., 1989 (Table 3.1).. The results reported earlier by Akaho and Nyarko, 2002 for other channels of the same reactor were also re-evaluated with this two computer programs to validate the reliability of the programs (Table 5.1).

Although no data is available as to the type of computer program or method adopted by Akaho and Nyarko, 2002 for their iteration work, however, when their reported input nuclear data and R_{cd} values were computed with the C++ and Q-Basic programs, the results are in good agreement except for the inner channel 1which has a deviation of about 9.6% (Table 5.1). The deviation could be attributed to their $Q_0\ (\alpha)$ ratio (0.6387489), which was found to have a difference of 0.0963862 from R_{cd} ratios (0.7351351) of the two monitors. For an iteration program, which normally allows a convergence at a null difference between the $Q_0\ (\alpha)$ and R_{cd} ratios, a difference of the order 0.096 as obtained in the result of Akaho and Nyarko, 2002 is quite significant. To avoid this type of deviation in this work, the $f(\alpha)$ (equation 1.11) was floated to the finite convergence.

The results in Table 5.1 shows that the Q-Basic program yields a better precision compared to the C++. However, during the process of running the iteration, it was observed that the Q-Basic lacks the speed at which the C++ does its iteration. Infact, the Q-Basic program takes so many inputs of trial values and as such one is expected to carefully observe every output before reaching a point of convergence, which is pains-taking and mathematically unrealistic for small step-lengths.

Having independently arrived at the same results using both programs (Q-Basic and C++) for the two channels (5 and 7) investigated in this thesis and also successfully reproducing the results reported by Akaho and Nyarko, 2002 for other channels of the same reactor (Table 5.1), the two programs are self-assuring for the determination of the epithermal flux-shaping factor (alpha) in this work and could be adopted for future related works.

Table 5.1 Comparison of iteration results obtained from both C++ and Q-Basic programs

Program	Channel	R_{cd} Ratio	$Q_0(\alpha)$Ratio	Deviation	α-Value	Reference
C++	5 (inner)	0.6547619	0.6547619	Nil	-0.10778	This work
Q-Basic	5 (inner)	0.6547619	0.6547619	Nil	-0.10768	This work
C++	7 (outer)	1.019841	1.019841	Nil	-0.17318	This work
Q-Basic	7 (outer)	1.019841	1.019841	Nil	-0.17308	This work
Unreported program	1 (inner)	0.7351351	0.6387489	0.096	-0.104	Akaho and Nyarko, 2002
C++	1 (inner)	0.7351351	0.7351351	Nil	-0.1246	Reproduced with this program.
Q-Basic	1 (inner)	0.7351351	0.7351351	Nil	-0.1249	Reproduced with this program.
Unreported program	6 (outer)	0.3813946	0.3762887	0.0051	-0.0261	Akaho and Nyarko, 2002
C++	6 (outer)	0.3813946	0.3813946	Nil	-0.0241	Reproduced with this program.
Q-Basic	6 (outer)	0.3813946	0.3813946	Nil	-0.0240	Reproduced with this program.

The following measurements were obtained for each of the channels.

Outer Channel No. 7:

For this channel, the thermal to epithermal flux ratio was found to be 40.38 while the epithermal flux shaping factor α obtained by running the iteration programs is

– 0.173 ± 0.0001. The previous flux ratio measured for the outer channel No. 6 by Akaho and Nyarko, 2002 and this result (outer channel 7) are found to follow a normal trend of having higher values than that of the inner channels due to the presence of beryllium reflectors. By the MNSR design the inner channels are columns drilled-out within the beryllium annulus whereas the outer ones are channels positioned outside the annulus (Fig 2.2). The reflectors were designed by manufacturers to help in slowing down the fast neutrons released from the core to highly thermalized flux-values outside the reflectors (where incidentally the outer irradiation channels are located). Infact, the strong thermalization of flux by low power reactors is responsible for the negative alpha values associated with these type of reactors, as evident in Table 3.3 above and also as obtained in this work for the Ghana research reactor.

The slight deviation of my α-value from the one reported earlier by Akaho and Nyarko, 2002 for outer channel No. 6 of the same reactor (Table 5.2) could be attributed to the channel location with respect to the core (such as nearness of channel 7 to reactivity regulators; Fig. 2.1, 2.2), geometrical dependence (in which case channel no. 7 measured in this work is larger than channel No. 6; Fig. 2.1, 2.2), and effects of moderation (like ^{235}U burn-up with time and hydrogen to ^{235}U ratio; Qazi *et. al*., 1996), which could all lead to slight differences in spectral indexes between irradiation channels. Indeed, this is one of the reasons why it is advisable (De Corte *et. al.,* 1982) to measure the epithermal flux-shaping factor in each channel. The values in Table 3.3, shows the variation of α-values for different channels of the same reactor as published for many laboratories.

Table 5.2 Experimental parameters determined for GHARR-1 research reactor

Reactor	Channel	Monitors	R_{cd}	Q_o	$Q_o(\alpha)$	E_r	α	f	Reference
GHARR-1	5	^{197}Au	2.10	15.71	18.91788	5.65	- 0.107	17.28	This
(inner)		^{96}Zr		251.6	4711.0421	338			Work
		^{94}Zr	2.68	5.06	12.39631	6260			
	1	^{197}Au	2.36	15.71	18.793	5.65	- 0.104	18.8	Akaho
		^{96}Zr		251.6	446.826	338			and
		^{94}Zr	2.85	5.06	12.615	6260			Nyarko, (2002)
GHARR-1	7	^{197}Au	3.57	15.71	21.2039	5.65	- 0.173	40.38	This
(outer)		^{96}Zr		251.6	689.209	338			work
		^{94}Zr	3.52	5.06	21.64129	6260			
	6	^{197}Au	3.92	15.71	16.423	5.65	- 0.0261	49.0	Akaho
		^{96}Zr		251.6	290.599	338			and
		^{94}Zr	8.76	5.06	6.336	6260			Nyarko, (2002)

Inner Channel No. 5:

The thermal to epithermal flux determined is 17.28 and the epithermal flux shaping factor α as obtained from iteration is - 0.107±0.0001. These results can

generally be said to have compared well with the ones reported earlier by Akaho and Nyarko, 2002 for inner channel No. 1 of the same reactor and followed similar trend obtained for low power research reactors (Table 3.3). The experimental data obtained in this work is tabulated in Table5.2.

5.2 Conclusion

The thermal to epithermal flux ratio *f* of Ghana Research Reactor-1 (GHARR-1) were measured and found to be 17.28 for the inner channel No. 5 and 40.38 for the outer channel No.7 while the epithermal flux-shaping factor α for these channels were obtained as –0.107 ±0.0001 and –0.173±0.0001 respectively. The result shows good agreement with previous ones determined by Akaho and Nyarko 2002 for inner channels No. 1 and outer channel No. 6 of the same reactor. With these results, the reactor is said to have complete characteristics of flux behavior in four of its seven installed irradiation channels. The channels characterized were intentionally selected to give a good representation of the entire reactor core (Figure 2.1).

The two programs developed (Q-BASIC and C++) in this thesis independently gave the same results for the two channels (5 and 7) investigated and also successfully reproduced the results reported by Akaho and Nyarko, 2002 for other channels of the same reactor (Table 5.1). The programs are said to be self-assuring for the determination of the epithermal flux-shaping factor (alpha) and could be adopted for future related works.

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APPENDIX A: *k*-factor equations

Since detected peak intensities of an irradiated sample depend on the emitted induced activities; the specific intensity (I_{sp}) could be obtained from:

$$\mathrm{I}_{sp} = \left(\phi_{th}\sigma_{th} + \phi_e \mathrm{I}_o\right)\frac{N_{av}}{M} P_i f_\gamma \varepsilon_\gamma \qquad \text{A1}$$

where

$$\mathrm{I}_{sp} = \frac{N_p / t_m}{S \bullet D \bullet C \bullet m} \qquad \text{whence} \qquad \mathrm{I} = \frac{N_p / t_m}{S \bullet D \bullet C} \qquad \text{A2}$$

The specific intensity of a co-irradiated monitor (e.g. Gold) used as the monostandard or comparator (Gold symbols denoted by asterisks *) is given as:

$$\mathrm{I}^*_{sp} = \left(\phi_{th}\sigma_{th} + \phi_e \mathrm{I}_o\right)^* \frac{N_{av}}{M^*} P_i^* f_\gamma^* \varepsilon_\gamma^* \qquad \text{A3}$$

Girardi *et al.,* 1965; defined *k*-factors in terms of a constant *k* using equation A2 and A3, as:

$$k = \frac{\mathrm{I}_{sp}}{\mathrm{I}^*_{sp}} \qquad \text{A4}$$

where I^*_{sp} = Specific intensity of Gold

I_{sp} = Specific intensity for any given nuclide

Putting equations A2 and A3 into A4, we get:

$$k = \frac{M^* P_{ii} f_\gamma \sigma_{th} \varepsilon_\gamma \left(f + Q_o\right)}{M P_i^* f_\gamma^* \sigma_{th}^* \varepsilon_\gamma^* \left(f + Q\right)^*} \qquad \text{A5}$$

with $f = \frac{\phi_{th}}{\phi_e}$ and $Q_o = \frac{I_o}{\sigma_o}$ A6

The concentration of an element in the sample for a stable 'ideal' neutron flux could therefore be written in terms of k_0 as:

$$Conc.\ (ppm) = \frac{I_{sp}\varepsilon_\gamma (f + Q_o)}{I^*_{sp} k_0 \varepsilon^*_\gamma (f + Q)^*} \quad \text{A7}$$

where the k_0 factor containing the nuclear constants is given by:

$$k_o = \frac{M^* P_i \sigma_{th} f_\gamma}{M P_i^* \sigma^*_{th} f^*_\gamma} \quad \text{A8}$$

In an effort to optimize the application of the single comparator method, De Corte, 1987 introduced conversion of the *k*-values so that once these factors are determined in one 'reference' reactor channel with known *f* it can then be re-evaluated for any other "analysis' channel using the expression:

$$k_{anal} = k_{ref} \frac{\{f_{ref} + Q^*_o\}\{f_{anal} + Q_o\}}{\{f_{ref} + Q_o\}\{f_{anal} + Q^*_o\}} \quad \text{A8}$$

For application of Single comparator in an "ideal" reactor flux where both thermal and epithermal flux contribute significantly to the total flux, an accurate determination of α is essential for not only the correction of the resonance integral as it transforms from I_o to take the form of $I_o(\alpha)$ but also Q_0 as it transforms to $Q_0(\alpha)$ (Jovanović *et al.,* 1987) thus:

$$\mathrm{I}_o(\alpha)=\left[\frac{\mathrm{I}_o-0.429\sigma_o}{\overline{E}_r^{\alpha}}+\frac{0.429\sigma_o}{(2\alpha+1)E_{Cd}^{\alpha}}\right]E_a^{\alpha} \qquad \text{A9}$$

where $\sigma_o = 2200\ ms^{-1}$ is the (n, γ) reaction cross-section;

$\overline{E}_r$ = effective resonance energy in eV

E_{Cd} = cadmium cut-off energy in eV.

$E_a^{\alpha} = (1\ \mathrm{eV})^{\alpha} \equiv 1$ which could be omitted being a normalizing factor.

$$Q_0(\alpha)=\frac{I_0(\alpha)}{\sigma_{th}}=\frac{Q_0-0.429}{E_r^{\alpha}}+\frac{0.429}{(2\alpha+1)E_{cd}^{\alpha}} \qquad \text{A10}$$

Using the above transformations, the expression for non-ideal conditions that incorporates the flux-shaping factor α for the determination of the concentration of an element in the irradiated sample (equation A7) will now be of the form:

$$Conc\ (ppm) = \frac{\mathrm{I}_{sp}\ \varepsilon^{*}\left[f + Q_o(\alpha)\right]}{\mathrm{I}^{*}_{sp}\ \varepsilon\left[f + Q_o(\alpha)\right]k_o} \qquad \text{A11}$$

The f and α terms in equation A11 are reactor channel dependent parameters, which are to be determined for each irradiation channel that is dedicated for routine Neutron Activation Analysis (NAA) and this is what this work is out to achieve.