

Determination of Epithermal Neutron Fluxes in the Cadmium-Lined Irradiation Channels of a Nuclear Reactor for Mineral Exploration



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ABSTRACT

Mineral exploration involves geochemical analysis which identifies potential mineral deposits in soil or rock samples using method such as neutron activation analysis (NAA). Neutron flux is an important requirement for this method. Epithermal neutrons induced nuclear reactions more efficiently than with thermal neutrons when analyzing elements with high resonance integrals, it is also useful in analyzing element such as Uranium, Thorium and certain transition metals. In this study, epithermal neutron fluxes in channels of Nigeria Research Reactor-1 (NIRR-1) installed with Cadmium (Cd) were determined for use in NAA procedures. The measurements were performed after Low Enriched Uranium (LEU) fuel substituted the High enriched Uranium (HEU) fuel in the reactor. The epithermal neutron fluxes were experimentally determined using Al-0.1%Au monitor foil via $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction. The epithermal neutron fluxes determined in the inner and outer Cd-lined irradiation channels A1 and A2 were found to be 2.539×10^{10} n/cm²s and 4.459×10^9 n/cm²s respectively at half operating power (15 kW) which corresponds to a thermal neutron flux of 5×10^{11} n/cm²s on the control console of NIRR-1. The values obtained before fuel conversion by previous authors in the A1-inner irradiation channel and in the A2-Outer Cd-lined irradiation channel were 2.72×10^{10} n/cm²s and 4.459×10^9 n/cm²s respectively. These values were both in agreement with the values of this study. This shows that the Cd installation and the fuel conversion from the (HEU LEU) does not affect the epithermal neutron fluxes. It also implies that the stability of the neutrons in NIRR-1 were not altered. Thus, the facility is good for Epithermal NAA. The use of additional flux monitors, implementation of ENAA in geochemical analysis and evaluation of epithermal neutron shaping factor (α) were recommended.

Keywords:

Reactor,
Neutron flux,
Highly-enriched Uranium,
Low-enriched Uranium,
Epithermal.

INTRODUCTION

Despite the availability of numerous resources in Nigeria, for decades the country depends solely on petroleum resources neglecting gold, coal, lead, limestone, quartz, iron ore, tin, silver, kaolin, zinc and tantalite which are found across the states. These resources have the potential to contribute significantly to the economic growth and development of Nigeria. However, in recent times efforts are being made to revitalize and harness the mineral resources for job creation, economic growth, infrastructure development, improved living standards

and increased government revenue. Mineral exploration involves range of the following activities: the analysis of soils and rocks (Geochemical surveys), geological mapping and sampling, geophysical surveys with the goal of identifying potential mineral deposits in the soil or rocks. There are numerous spectroscopic technique to identify minerals in samples, these includes: atomic absorption spectroscopy, x-ray fluorescence and neutron activation analysis (NAA) (Glaser B., & Lehr C (2018). Neutron Activation Analysis (NAA) is a nuclear technique for the qualitative and quantitative

determination of minerals in samples based on the measurement of characteristic radiation emitted when the samples are bombarded with stream of neutrons (IAEA, 2023). This nuclear technique requires a source of neutron which could be a: nuclear research reactors, isotopic neutron source, particle accelerators or generators (Jonah et al 2006). Nuclear reactor is considered the most important source of neutrons for NAA applications (Greenberg *et al* 2011). However, the highly enriched uranium (HEU) fuel used in some research reactors is associated with proliferation risk. This has made the regulating agencies to devise a means of reducing the fuel to the Low enriched uranium (LEU). A research coordinated by International Atomic Energy Agency (IAEA) highlights the feasibility of converting

HEU fuel in the Nigeria research reactor-1 (NIRR-1) to the LEU fuel with 12.5% fissile Uranium fuel ^{235}U as opposed to the HEU fuel enriched with 90 % of ^{235}U (Jonah *et al* 2009). NIRR-1 was successfully installed with LEU and the HEU core was returned to China in 2018 after the installation (Anas *et al* 2023). This fuel conversion should not compromise the utilization of the reactor especially with respect to the stability of its neutron fluxes (a requirement for NAA applications). The irradiation channels of NIRR-1 contains the thermal neutron flux with energy range 0.025 eV- 0.4 eV, the epithermal neutron flux with energy range 0.5 eV – 100 keV and the fast neutron flux with energy range 100 keV – 25 MeV. The distribution of neutron energy of this type of reactor is shown in Figure 1.

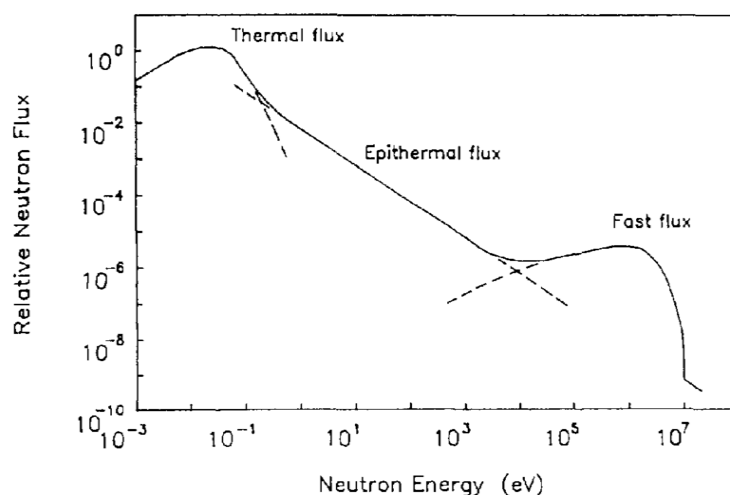


Figure 1: Neutron Energy Distribution (Greenberg et al., 2011)

Thermal neutrons being the majority of all the components of neutrons in a reactor tends to hinder the determination of some minerals in a material samples via NAA technique. Therefore, Cd was permanently installed in one of the inner irradiation channel (A_1) and one of the outer irradiation channel (A_2) of NIRR-1, in order to absorb or filter the thermal neutrons and enhance the measurement of minerals via epithermal neutron activation analysis.

The values of epithermal neutron flux in an irradiation positions has been reported by different authors using activation method: The epithermal neutron fluxes in the inner channel 2 and outer channel 7 of Ghana Research Reactor-1 (GHARR-1) was reported by Sobgadjí *et al* (2011) and the values were found to be $3.762 \times 10^{10} \text{ n/cm}^2 \text{ s}$ and $1.376 \times 10^{10} \text{ n/cm}^2 \text{ s}$ respectively. Neutron fluxes in channel 4 of Dalat Nuclear Research Reactor (DNRR) were measured by (Son *et al.*, 2016) using a set of pure metals thin foils as detectors. The values determined are 4.80×10^9 , 1.98×10^7 and $5.06 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ for the thermal, epithermal and fast neutron fluxes respectively.

Jonah *et al* (2005) determined the epithermal neutron fluxes in the inner irradiation channel (B_1) and the outer irradiation channel (B_4) of NIRR-1. The values are reported as $2.60 \times 10^{10} \text{ n/cm}^2 \text{ s}$ and $5.18 \times 10^9 \text{ n/cm}^2 \text{ s}$ respectively. Sadiq *et al* (2005) also determined the epithermal neutron fluxes in the inner irradiation channel (A_1) and the outer irradiation channel (A_2) of NIRR-1. The values are reported as $2.72 \times 10^{10} \text{ n/cm}^2 \text{ s}$ and $4.48 \times 10^9 \text{ n/cm}^2 \text{ s}$ respectively. The fast neutron flux value determined by Jonah *et al* (2008; 2014) in the inner irradiation channel (B_2) and the outer Cd-lined irradiation channel is $1.04 \times 10^{11} \text{ n/cm}^2 \text{ s}$ and $1.65 \times 10^{10} \text{ n/cm}^2 \text{ s}$ respectively. Distribution of radial and axial neutron flux in the inner irradiation channel (B_2) of NIRR-1 was investigated by Musa *et al.*, (2012) using foil activation technique and cadmium covered multimonitor method using the monitor set Al- 0.1% Au, Zr and Zn thin foils. The values of epithermal neutron flux was found to be $(4.51 \pm 0.01) \times 10^9 \text{ n/cm}^2 \text{ s}$. Njinga *et al* (2012) also determined epithermal and fast neutron fluxes using the same activation material as Musa *et al* (2012) via $\text{Au} - 197(n, \gamma)\text{Au} - 198$ and $\text{Al} - 27(n, p)\text{Mg} - 27$

reactions and the values are $3.96 \times 10^9 n/cm^2 s$ and $1 \times 10^{10} n/cm^2 s$ respectively, epithermal neutron flux in the inner channel was reported as $2.55 \times 10^{10} n/cm^2 s$. Epithermal neutron flux was also calculated in A_2 cadmium-lined channel of NIRR-1 by Sheyin *et al* (2015) using Al-0.1%Au foil and the value was reported as $4.51 \times 10^9 cm^{-2} s^{-1}$.

This study uses absolute foil activation method to determine the epithermal neutron fluxes in the two Cd-lined irradiation channels of NIRR-1 (A_1 and A_2). This is important in order to find the probable effects of the fuel conversion and the Cd installation. Also, to guarantee the application of ENAA using this reactor especially during analysis of the solid minerals.

Theory of Epithermal Neutron Flux

In order to determine the epithermal neutron flux, radionuclides in the samples are transformed by irradiating the sample with neutron flux for some time t_i , the nuclide in the sample decayed after some time t_d , and counted for some time t_c .

The rate of nuclear transformation is given by equation 1.

$$\frac{dN}{dt} = RN_o - \lambda N \quad (1)$$

Where R is the rate of the reaction and is given by equation 2

$$R = \varphi_{epi} \sigma \quad (2)$$

$$\text{Where } \sigma = \int_{E_{Cd}}^{E_{max}} \frac{\sigma(E_n) dE_n}{E_n} \quad (3)$$

The epithermal neutron cross section (σ) is proportional to $\frac{1}{E_n}$. This requirement is fulfilled by most of the (n, γ) reactions and it shows the dependence of the epithermal neutron cross section to the cadmium cut off energy ($E_n = 0.55 eV$). Where E_n is the neutron energy.

The disintegration rate of the product radionuclide at the end of the irradiation time t_i follows from:

$$D(t_i) = N(t_i)\lambda = N_o R (1 - e^{-\lambda t_i}) \quad (4)$$

The number of irradiated nuclei $N(t_i, t_d)$ present at the start of the measurement and the number of nuclei ΔN disintegrating during the measurement is given by equation 5.

$$\Delta N(t_i, t_d, t_m) = \frac{RN_o}{\lambda} (1 - e^{-\lambda t_i}) e^{-\lambda t_d} (1 - e^{-\lambda t_m}) \quad (5)$$

Replacing the number of target nuclei N_o by $(N_A M)/w$ and using equation (2) for the reaction rate (R), the resulting net counts N_p in a peak in the spectrum

corresponding with a given photon energy is approximated by equation 6.

$$N_p = \Delta N \gamma \varepsilon = \varphi_{epi} \sigma \frac{N_A \theta M}{w} ((1 - e^{-\lambda t_i}) e^{-\lambda t_d} \frac{(1 - e^{-\lambda t_m})}{\lambda}) I_\gamma \epsilon \quad (6)$$

Equation 6 can be rewritten in terms of the epithermal neutron flux

$$\varphi_{epithermal} = \frac{N_p \lambda M}{(1 - e^{-\lambda t_i}) e^{-\lambda t_d} (1 - e^{-\lambda t_m}) N_A w \theta \epsilon I_\gamma \sigma_{n,x}} \quad (7)$$

Where: ε is the corresponding detection efficiency, N_p is the net count in the gamma-ray peak of E_γ , M is the atomic mass of target element, N_A is the Avogadro's number, θ is the isotopic abundance of the target isotope, is the mass of target element in sample, I_γ is the gamma ray abundance, $\sigma_{n,x}$ is the spectrum average cross section.

The various and different number of epithermal neutron cross section available suggest the usage of a gold foil for monitoring of the fluxes because the $^{197}\text{Au} (n, \gamma) ^{198}\text{Au}$ reaction has a single epithermal neutron cross section and has been reported to be 1550 mb assuming an ideal situation ($\frac{1}{E}$ law).

MATERIALS AND METHODS

NIRR-1

NIRR-1, the only nuclear reactor in Nigeria, is the number eight among the Chinese Miniature neutron Source Reactors (MNSR) in the world. It has a tank-in-pool structural configuration with a nominal thermal power rating of 34 kW similar to the Canadian slowpoke (Agbo *et al.* 2015). Low enriched Uranium (LEU) fuel substituted the Highly Enriched Uranium (HEU) fuel earlier used in this reactor. Out of the 10 irradiation channels of this reactor, six irradiation channels were connected to the pneumatic transfer systems types A and B, among the connected channels, cadmium was permanently installed in two channels: one inner (A_1) and one outer (A_2) for the purpose of extending the utilization of the reactor to ENAA and FNAA. A detailed description of the reactor and the irradiation facility has been described elsewhere (Ahmed *et al.*, 2002, Balogun, 2003, Jonah *et al* 2006, Sheyin *et al.*, 2015).. A schematic core configuration of NIRR-1 showing inner A_1 and outer A_2 irradiation channels is shown in figure 2.

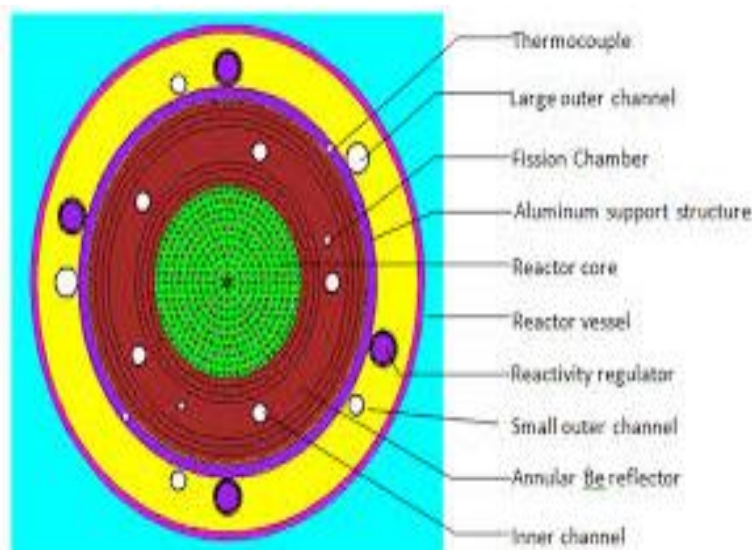


Figure 2: Core Configuration of NIRR-1 Showing the Irradiation Channels

NaI (TI) Scintillation Detector and Other Materials

A 2×2 NaI(Tl) cylindrical scintillation detector coupled to EC & G ORTEC multichannel buffer (MCB) was used for data acquisition and processing through connection-32 program MAESTRO-32. Other materials used in the analysis includes weighing balance, Al-0.1%Au disc, cadmium box, plastic vial, acetone solution, cotton wool and stop watch for taking the irradiation and cooling time.

Energy Calibration of 2×2 NaI Detector

Energy calibration was performed by matching the channel number of the detector with the spectrums of the standard gamma source. This was achieved by placing each of the gamma source on the surface of the detector (0 cm) and counted for 600 seconds. Since the MCA MAESTRO data acquisition software allowed the computer to search and mark the interested peak(s), the peak position of each of the gamma standard sources used was determined and the region of interest (ROI) was created on the peak through the knowledge of its profile. On this mode, the computer required for the peaks data which when supplied makes the calibration complete (Faanu *et al.*, 2018).

Efficiency Calibration of 2"×2" NaI Detector

Efficiency calibration of the NaI (TI) gamma energy spectrometer was performed using the gamma standard sources: ^{241}Am , ^{137}Cs and ^{60}Co . Each of the gamma standard sources was placed on the surface of the detector and counted for 600 seconds. The detector converted the gamma absorbed into electrical pulses (signals) and the strength of these pulses were increases and further amplified by preamplifier and the main amplifier. The analog signals obtained were converted to digital signals and sorted according to their energies by multichannel analyzer (MCA). Since the MCA MAESTRO data acquisition software allowed the computer to search and mark the interested peak(s), the peak position of each of the gamma standard sources used was determined and the region of interest (ROI) was created on the peak through the knowledge of its profile. On this mode, the count rates at each of the gamma photo peaks were determined and the data were subsequently substituted in efficiency equation. High voltage (900 v) was supplied to the detector to provide the necessary potential difference to bias the detector and create the needed depletion region. Lead was also used to shield all sides of the detector in order to reduce the unwanted background radiation. Figure 3 shows a block diagram for determination of NaI (TI) detector efficiency (Akar Tarim., 2018).

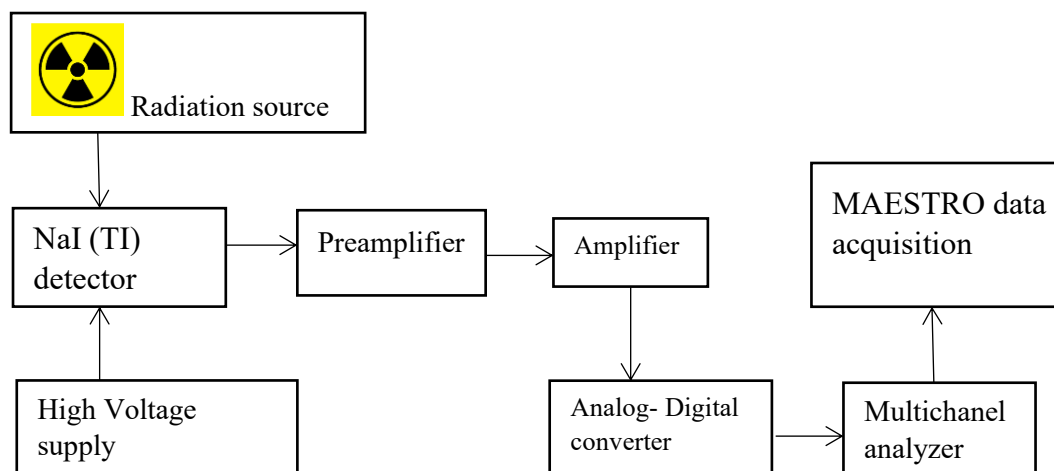


Figure 3: A Block Diagram for Efficiency Calibration of NaI(Tl) Detector

Absolute efficiency against energy was plotted in Microsoft Excel and a mathematical equation that described the absolute efficiency curve and an equation that described it was generated (Figure 4). The generated equation from the curve was used to get absolute efficiency at energy of interest. The determination of the absolute efficiency curve was carried out in the energy range of 59.5 to 1368.5 keV. However, due to the decay of standard gamma source of interest the efficiencies of the point source energy of ^{60}Co at 1332.5 keV and 1173.2 keV were used to interpolate the efficiencies of the ^{24}Na peak. This was done by plotting a graph of log of efficiencies against the log of their energies (Figure 5). From this graph, the efficiency of 1368.5 keV photo peak of ^{24}Na was determined since the log of this energy will be very close to the log 1332.5 keV photo peak of ^{60}Co on the graph. It also follows that the log of their efficiencies will also be close and the exponential of the value gives the absolute efficiency of 1368.5 keV photo peak of ^{24}Na . the value was shown in table 3.

Determination of Epithermal Neutron Flux

Two Al-0.1%Au disc were put in a polyethylene vials after cleaning with acetone and weighing with a digital

balance. They were subsequently filled with cotton to make it weighty, heat sealed and to stay at the bottom of the polyethylene vials. One Al-0.1%Au foil disc was activated in the inner cadmium-lined irradiation channels of (A_1) for 30 minutes and the other Al-0.1%Au foil disc was activated in the outer cadmium-lined irradiation channel (A_2) for 1 hour. After the activation, the induced radioactivity was counted on the calibrated NaI (TI) γ -ray counting system. At the end of the counting, the disc was allowed to decay totally and then placed in a cadmium box for storage. The irradiation time, cooling time and counting were all recorded. The activation of Al-0.1%Au foil in the cadmium lined irradiation channels allowed the monitoring of epithermal neutron flux via the ^{197}Au (n, γ) ^{198}Au reaction and the fast neutron flux via the ^{27}Al (n, p) ^{27}Mg and ^{27}Al (n, α) ^{24}Na reactions. Epithermal neutron fluxes was deduced relative to the spectrum averaged cross section data library shown in table 1. The nuclear data were obtained from experimental nuclear data library (ENDL) and Equation 7 was used to determine the epithermal neutron fluxes in the inner and outer cadmium-lined irradiation channel of NIRR-1.

RESULTS AND DISCUSSION

Table 1: Nuclear Data Used in Determination of the Epithermal Neutron Fluxes at 15.5 Kw

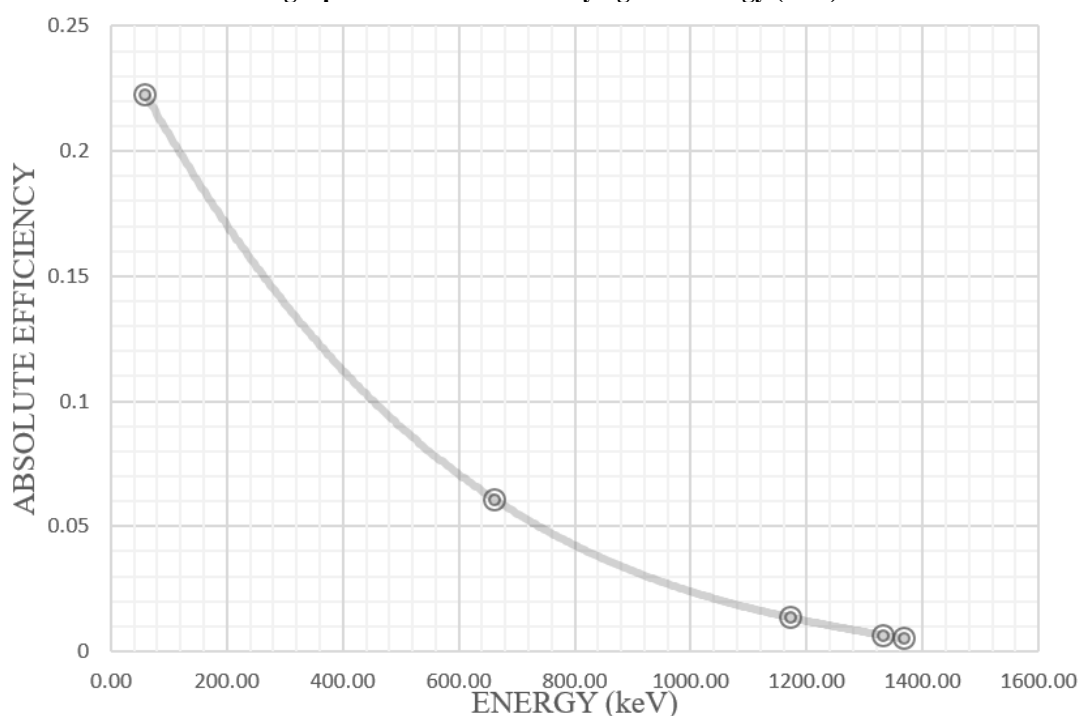
Reaction	$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$
Energy	411.8 KeV
Gamma abundance	96.52%
Half-life	2.333×10^5 s
Avogadro's number	6.023×10^{23} atoms/mol
Atomic mass of target element	196.967 Amu
Isotopic abundance	100%
Epithermal Cross section in $\frac{1}{E}$	1550 mb

Result of Efficiency Calibration**Table 2: Nuclear Parameters of the Gamma Sources Used and the Determined Absolute Efficiencies**

Nuclide	Energy (KeV)	Half-life (s)	I_γ (%)	A_t (Bq)	ϵ_{abs}
^{241}Am	59.54	1.36×10^{10}	35.9	3.55×10^4	0.222725
^{137}Cs	661.60	9.5×10^8	85.1	2.44×10^4	0.060788
^{60}Co	1173.20	1.66×10^8	99.9	3.59×10^4	0.013558
^{60}Co	1332.50	1.66×10^8	100	3.59×10^4	0.006173

Table 2 shows the specifications of the IAEA gamma standard sources, their activities and absolute efficiency measured on the surface of the detector (0 cm). The initial activities of the gamma sources were measured on 1st July, 2004 and the present activities were measured on the 12th July 2022. The decay constant was determined by taking the $\ln 2$ to half-life ratio of the radionuclides and the decay time was determined by subtracting the

time of initial measurement from the time of current measurement (12th July 2022- 1st July 2004). The net count was obtained on the detector under gamma energy of interest and the counting time is 600s. The absolute efficiency was determined by multiplying the net count by 100 and dividing the result by the product of present activities, gamma abundance and the counting time. All other data were gotten from literature.

A graph of absolute efficiency against energy (keV)**Figure 4: Absolute Efficiency-Energy Curve of NaI(Tl)**

$$y = -7\text{E-}11x^3 + 3\text{E-}07x^2 - 0.0004x + 0.2481 \quad (8)$$

$$R^2 = 1$$

Where y is the absolute efficiency, x energy (keV) and R is the regression.

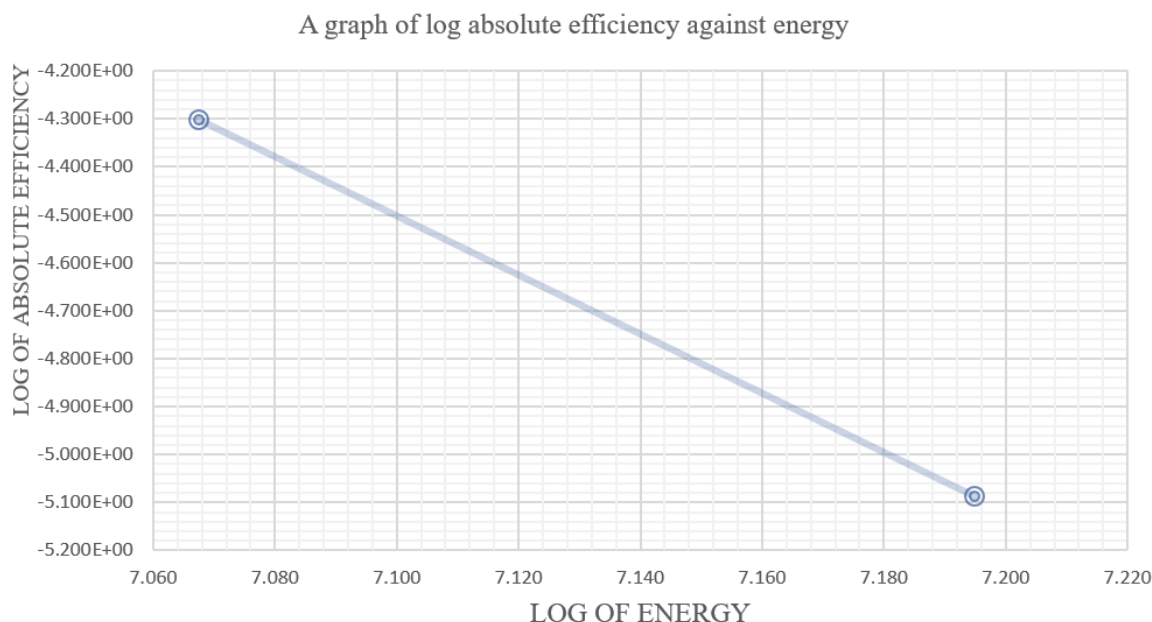


Figure 5: log of absolute efficiency against log of energy for the two gamma energy of ^{60}Co

$$y = -6.1802x + 39.378 \quad (9)$$

$$R^2 = 1$$

Where y is the log of absolute efficiency, x is log of the energy and R is the regression

The absolute efficiency of Na-24 with energy 1368.5 was determined through interpolation of the ^{60}Co gamma energy (1173.2 keV and 1332.5 keV). Since the energies are close, it implies that the log of their efficiencies was also close. Thus, equation 9 was used to determine the log of efficiency of 1368.5 keV and the exponential of this value gives the absolute efficiency at 1368.5 keV. The result was shown in table 2

Figure 4 shows the absolute efficiency against energy curve measured on the surface of NaI (TI) detector for the energy range 59.5 keV to 1368.5 keV. It can be observed that the efficiencies are high at low energy and decreases with increasing gamma energy. This may be due to decreased in the number of photoelectric events and increase in Compton scattering as the energy increases. The generated equation 8 can be used to determine the efficiencies at interested gamma energy within 59.5 to 1368.5 keV.

Table 3: Comparison of the Experimental and Calculated Absolute Efficiencies of Nai(Ti) Detector Measured on the Surface of the Detector

Radionuclide	Energy	Experimental absolute efficiency	Calculated absolute efficiency
Am-241	59.5	0.223	0.225
Cs-137	661.6	0.061	0.095
Co-60	1173.2	0.014	0.079
Co-60	1332.5	0.006	0.082
Na-24	1368.5		0.083

It can be observed from Table 3, the absolute efficiencies values obtained from the polynomial equation 8 is in good agreement with the experimental values obtained using IAEA gamma standard sources. Thus, by means of this polynomial equation, the efficiencies of gamma energy involved in the determination of the epithermal neutron fluxes were evaluated.

Results of the Epithermal Neutron Flux

The results of the epithermal neutron fluxes for inner cadmium-lined irradiation channel A_1 and outer cadmium-lined irradiation channel A_2 computed using

equation 7 is presented in table 4. The analysis of the result shows that the epithermal neutron flux in the inner channel is about six times the epithermal neutron flux in the outer channel A_2 for the same mass of target element and decay constant this can be associated with proximity of the inner channels to the core. Table 5 compared the epithermal neutron flux values in the present study with results in other irradiation channels of NIRR-1. It can be observed that the epithermal neutron flux determined in both the inner cadmium-lined irradiation channel (A_1) and the outer cadmium-lined irradiation channel (A_2) agreed well with the previously measured values

obtained by (Jonah *et al.*, 2005; Sadiq *et al.*, 2010; Njinga *et al.*, 2012; and Sheyin., 2015) before the cadmium installation and nuclear fuel conversion. This shows that the cadmium installation and the fuel conversion (HEU to LEU) does not affect the epithermal neutron flux

values. It also implies that the stability of neutrons in NIRR-1 were not altered. Thus, the facility is good for ENAA especially in mineral exploration survey: geochemical survey and other NAA analysis.

Table 4: Nuclear Data Generated and the Calculated Epithermal Neutron Fluxes Values

Irradiation channel	Inner cadmium-lined (A ₁)	Outer cadmium-lined (A ₂)
Mass of target element in sample	$1.31 \times 10^{-5} \text{ kg}$	$1.30 \times 10^{-5} \text{ kg}$
Decay constant	$2.971 \times 10^{-6} \text{ s}^{-1}$	$2.971 \times 10^{-6} \text{ s}^{-1}$
Irradiation time	1800 s	3600 s
Decay time	$7.56 \times 10^4 \text{ s}$	$9.36 \times 10^4 \text{ s}$
Counting time	7200 s	7200 s
Net count	5.973×10^6	1.969×10^6
Absolute efficiency	0.129365	0.129365
Saturation factor	5.333×10^{-3}	1.064×10^{-2}
Decay factor	7.988×10^{-1}	7.573×10^{-1}
Counting factor	2.116×10^{-2}	2.116×10^{-2}
Epithermal neutron fluxes	$2.539 \times 10^{10} \text{ n/cm}^2 \cdot \text{s}$	$4.459 \times 10^9 \text{ n/cm}^2 \cdot \text{s}$

Table 5: Comparison of Epithermal Neutron Flux Values in the Cadmium-Lined Irradiation Channel with Results in Other Irradiation Channels of Nirr-1

Author	Irradiation channel	Epithermal flux (n/cm ² .s)
Jonah et al (2005)	B1- inner	2.60×10^{10}
Sadiq et al (2010)	A1- inner	2.72×10^{10}
This study	A1- inner (cadmium lined)	2.539×10^{10}
Jonah et al (2005)	B4- outer	5.18×10^9
Sadiq et al (2010)	A2- outer	5.05×10^9
Njinga et al (2012)	A2- outer	4.48×10^9
Sheyin (2015)	A2- outer (cadmium-lined)	4.51×10^9
This study	A2- outer (cadmium-lined)	4.459×10^9

CONCLUSION

Absolute method was used to determine the epithermal neutron fluxes in both the inner and outer Cd-lined irradiation channels of NIRR-1 using $^{97}\text{Au}(n, \gamma)^{198}\text{Au}$ reaction. The measurement was performed using a calibrated NaI scintillation detector and the epithermal neutron flux values measured in the inner and outer Cd-lined irradiation channels were $2.539 \times 10^{10} \text{ n/cm}^2 \cdot \text{s}$ and $4.459 \times 10^9 \text{ n/cm}^2 \cdot \text{s}$ respectively. The result analysis agrees closely with the previous measurements before Cd installation and fuel conversion of NIRR-1. This shows that the cadmium installation and the fuel conversion on this reactor does not affect the epithermal neutron fluxes. It also implies that the stability of neutrons in NIRR-1 is not altered. Thus, the facility is good for ENAA especially mineral exploration survey. The use of additional flux monitors, implementation of ENAA in geochemical analysis and evaluation of epithermal neutron shaping factor (α) were recommended.

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