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Investigation of the Specific Activity of Al-0.1%Au Wire for Epithermal Neutron Activation Analysis for Nirr-1 after Conversion from Heu to Leu

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ABSTRACTS

The significance that Epithermal Neutron Activation Analysis (ENAA) play in the field of science research, technology and innovations and how humans interact with elemental components in food chain and environment, has led to the development of ENAA protocol in research reactors. Epithermal neutrons, which have energy between 0.5 eV and 0.5 MeV, are increasingly needed by reactor clients. In this study, we have examined the activity of the flux monitor in the irradiation site of the inner channel (A1) of Nigeria Research Reactor-1 (NIRR-1) after its conversion to low enriched uranium fuel. The flux monitor (Al-0.1%Au thin wires) was washed with acetone, dried for 2 minutes, weighed to be 0.0122 g, and thereafter placed in a 15.0 mm internal diameter polyethylene vials that has 1 mm wall thickness for irradiation. The activity of flux monitor Al-0.1%Au wire of 0.0122 g was calculated to be 5 x 10^8 count/s.g using A1 inner channels of the Cd line. This value obtained in this our studies found to be consistent with previous result and has shown that the epithermal channel A1 of the NIRR-1 remain suitable for ENAA after the reactor's fuel conversion from HEU to LEU.

Keywords: NIRR-1, NAA, ENAA, Specific Activity

INTRODUCTION

Samples containing specific rare earth elements were extremely radioactive after being exposed to a neutron source as shown in Figure 1, which led to the discovery of Neutron Activation Analysis (NAA) in 1936 (Ali 1999). This brought about the possibility of using nuclear processes on samples followed by measurements of the generated radioactivity to make it easier to identify the major, minor, and trace elements contained samples both qualitatively and quantitatively (Joseph *et al.,* 2011). The idea of NAA has gone through different development stages from theory to methodology and its sensitivities have been reported (Joseph *et al.,* 2019). In experimental particle physics, detectors are devices used for detecting, measuring and analyzing high-energy particles, such as those produced by nuclear decay, cosmic radiation or reactions in a particle accelerator (Joseph *et al.,* 2017). Detectors like High Purity Germanium (HPGe) and Sodium Iodide (NaI) uses ionization and excitations processes in identifying the radiant energy through the help of electric signals. Scintillation detectors are very sensitive radiation instruments and are used in both portable and stationary system. The properties of scintillation material required for good detectors are transparency,

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availability in large size, and large light output proportional to gamma ray energy (Joseph and Nasiru, 2013).

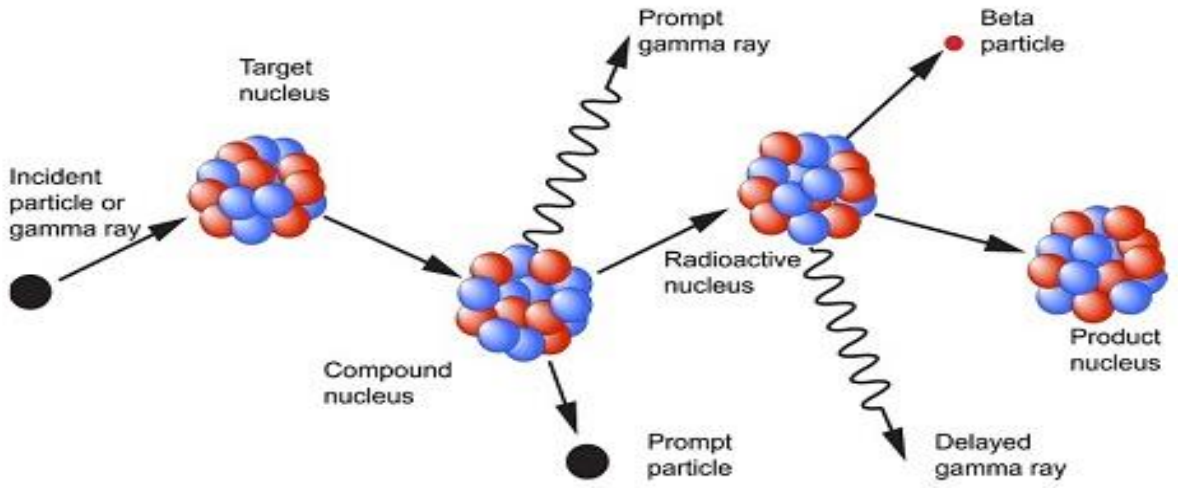

Figure 1: Process of Neutron Capture by a Target Nucleus (Jonah *et al.*, 2006).

Epithermal and thermal neutrons cause (n, γ) reactions on target nuclei, according to Jovanovi *et al.* (2010) Epithermal Neutron Activation Analysis is an NAA method that only uses epithermal neutrons to irradiate samples inside cadmium or boron shields against thermal neutrons causing (n, γ) reactions. ENAA is a valuable analytical method for carrying out multi-element analysis in samples, both qualitative and quantitative. Because NAA is used so often throughout the world, it is anticipated that each year, a few 10,000 samples are analyzed in practically every imaginable sector of science or technology (Ali, 1999). ENAA plays a vital role in exploring the new and old mining sites through identifying essential trace element in the geological, environmental and biological samples surrounding the mining areas.

The Nigeria Research Reactor-1 (NIRR-1) is a Miniature Neutron Source Reactor (MNSR). It uses light water as a moderator and coolant, more also, it uses highly enriched uranium as a fuel material in the core of the reactor (90.2%) which was converted to low enriched uranium core of 12.5% (Zhu, 1990; Balogun, 2003). The facility was judged to be working harmlessly for NAA from the time it reached its standard reactivity of operation on February 3, 2004 (Jonah *et al.*, 2005; 2006). According to Ahmed *et al.* (2006) and Joseph *et al*. (2011), the NIRR-1 is primarily modeled for neutron activation analysis (NAA), which is a tool for scientific and technological study. Additionally, instructors that teach nuclear engineering and radiation safety courses use it for their students (Azande *et al*., 2010; Ahmed *et al*., 2010). Previous researches (Ahmed *et al*., 2002; Ahmed *et al.*, 2008) published more information on the reactor's characteristics and its operational parameters.

The international nonproliferation community in its attempts to reduce the amount of highly enriched uranium (HEU) fuel in the reactor cores worldwide, this leads to the idea of converting reactors to low enriched uranium (LEU) cores, where the high enriched uranium tends to be used for nuclear weapons around the globe (Wilson *et al.*, 2020). The nuclear reactor is still recognized best origin of irradiation for the purpose of sample analysis with a variety of nature, despite the fact that many various irradiations have been introduced in the scientific field over the years (Joseph *et al*., 2011). In order to conduct element activation analyses, thermal and epithermal neutrons are increasingly being used in reactors all over the world.

However, there are several situations when using thermal neutrons has a number of disadvantages, in many

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matrices, the major elements are substantially activated by thermal neutrons (for example, Na and CI in biological samples and Na, Al, and Mn in rock samples), and their Compton interference prevents the measurement of some trace elements' gamma rays (Fedlix, 2015). Discussion of impacts on environmental soil samples have been made earlier that shows the need to improve Epithermal Neutron Activation Analysis (Joseph *et al.,* 2016). In some instances, it is impossible to identify some of the elements either because the created radionuclide has a too long half-life or because it does not emit gamma rays. As a result, these challenges can be overcome by the employment of epithermal neutron. Despite the comparatively moderate activity caused by the epithermal neutrons, large sensitivity can be achieved. Toxic elements pollution of the environment, even at low levels and their resulting long-term cumulative health effects are among the leading health concerns all over the world (Abubakar and Joseph, 2023). Discussions of specific issues and typical ENAA uses have taken place (Alfassi, 2001). The range of these elements' detection limits of ENAA are according to Xiaolin *et al*. (2016), better than those with standard NAA by a ratio of 1.5-7.

According to Abugassa *et al.* (2004) said that the ENAA's superiority and significance were confirmed by the IAEA inter-comparison test, which rigorously analyzed epithermal NAA in comparison to alternative techniques including experimental neutron activation analysis based on the k_0-standardization approach. Recently, the ENAA technique has been significant in advances in biological and medical studies using reactors (Ahmed *et al.*, 2006; Kapsimalis *et al.*, 2009; Joseph *et al.*, 2016)). The most effective way to produce epithermal neutrons was to wrap samples in cadmium sheet, a technique that is still widely employed in laboratories today (Filby, 1995; Anderson and Cunningham, 2005; Gharib *et al.*, 2001). Based on simulations showing core excess reactivity and flux depression, the results show that adding cadmium-line in the outer channel has far less effect than adding it in the inner channel (Ahmed *et al*., 2013).

According to Joseph *et al*. (2013), to ensure accurate quantification as is the case for all analytical techniques, γ-ray spectrometry requires standard samples to establish an experimental efficiency calibration which is the most accurate method. Hossain *et al.* (2012), have demonstrated that errors of 4–18% can be introduced into ENAA for several isotopes at a typical channel of the 250 kW Thetis reactor if is disregarded, consequently, when using the ENAA's single comparator approach (k_0-ENAA) in the NIRR-1, the values of α in the irradiation sites should not be neglected. The significant negative value of in the cadmium-lined irradiation channel of NIRR-1 indicates a toughened (poorly thermalized) epithermal neutron spectrum (Fedlix, 2015).

Flux monitors are nuclides that have been transformed to radionuclide by neutron capture processes and have their emitted γ-rays monitored. Estimates of flux parameters are derived from knowledge of the quantities and nuclear properties of the selected monitors, as well as the results of γ-spectrometry measurements (Luzio, 2019). The epithermal neutron flux of flux monitors like Mo thin wire, Zr and Zn thin foils, and Al-0.1%Au can be found from the specific activities of the monitors after irradiation. From the previous report the epithermal neutron flux of NIRR-1 in A1 inner channel was determined to be $(4.51\pm0.01) \times 10^9$ n.cm^{-2}s using the activity 5.34 x 10^8 count/s.g of ^{198}Au in the thin Al-0.1% Au foil irradiated with the monitor set for determination (Fedlix, 2015).

Consequently, the significance of ENAA will play vital role in research and educational institutions in Nigeria, therefore this study has focused on investigating specific activity of epithermal neutron flux monitor (Al-0.1%Au wire) for ENAA after the conversion of the Nigeria Research Reactor 1 (NRR-1) from High Enriched Uranium to Low Enriched Uranium core.

MATERIALS AND METHODS

The Nigeria Research Reactor-1 (NIRR-1) was considered in this study, Table 1 list the primary

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characteristics of the NIRR-1 and depicted in Figure 2:

Table 1: NIRR-1 Technical Specifications

Parameters	**Description**
Reactor type	Tank-in-pool
Rated Thermal Power	30 kW
Fuel	U^{235} in Al cladding
^{235}U Enrichment	12.5%
Core shape	Cylinder
Core diameter	23.0 cm
Core height	23.0 cm
No. of fuel elements	347
Weight of ^{235}U	999.36g
Total number of irradiation sites	10
Inner and outer channels in use	6
Flux in inner channel	1×10^{12} n cm^{-2} s^{-1}
Flux in outer channel	5×10^{11} n cm^{-2} s^{-1}
Reactor cooling mode	Natural convection
Hight of inlet orifice	6 mm
Height of outlet orifice	7.5 mm
Diameter of fuel meat	4.3 mm
Diameter of fuel element	5.50 mm
Excess reactivity	3.77 mk
Length of Cd control rod	230 mm

(Jonah *et al.*, 2005; Ahmed *et al.*, 2006; Sadiq *et al.*, 2010: Musa *et al.*, 2012).

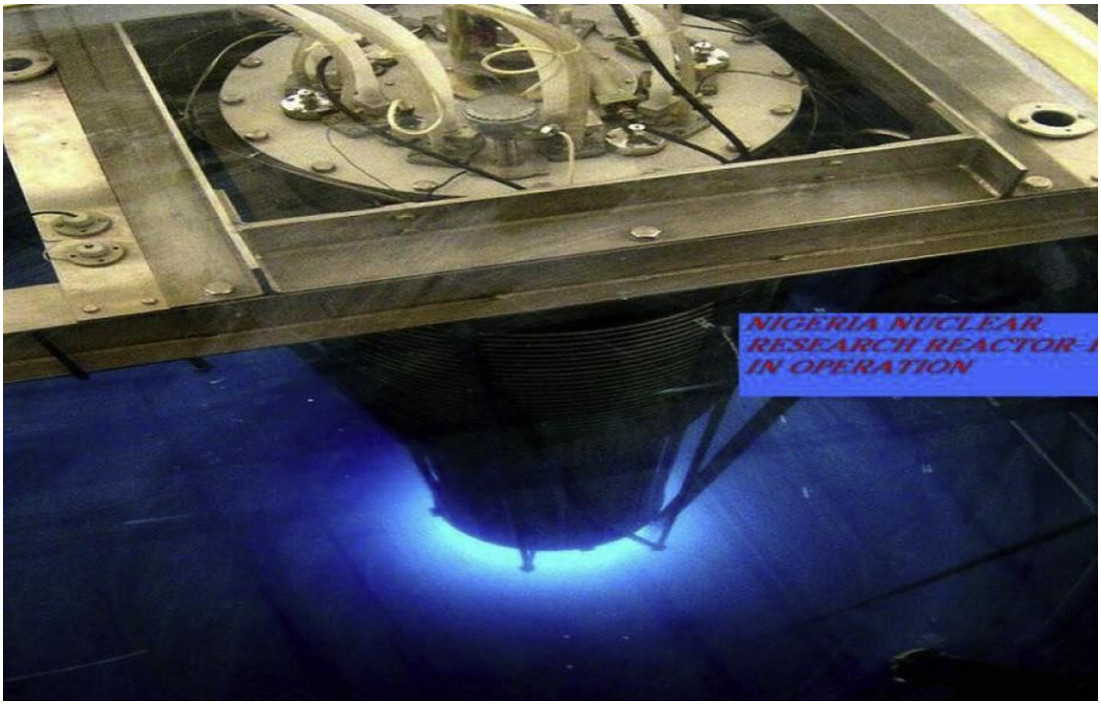

Figure 2: Nigeria Research Reactor-1 segmented view (Jonah *et al.*, 2012).

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DETERMINATION OF SPECIFIC ACTIVITY OF THE Al-0.1%Au THIN WIRE FLUX MONITOR

We examine the activity of the flux monitor in the irradiation site of the inner channel (A1) of NRR-1. The flux monitor (Al-0.1%Au thin wires) was washed with acetone dried for 2 minutes, and was weighed to be 0.0122 g, then placed in a 15.0 mm internal diameter polyethylene vials that has 1 mm wall thickness and a cotton all together. They were irradiated for 2 hours in A1 channel in a fixed position in inner irradiation channel with greater capacity than the sample volume using a pneumatic system of transfer using Rabbit system type "A".

The sample was allowed to cool down in a lead shield for lower activity to be obtained. A counting time of 10 minutes was used to monitor the ^{197}Au (n, γ) ^{198}Au reaction simultaneously. Every irradiation is conducted at a thermal power level of 3.0 kW, which is equivalent to the control console's preset thermal neutron flux value of 1.0 ×10^{11} $cm^{-2}s^{-1}$. ORTEC is an HPGe coaxial detector at 1332.5 keV of ^{60}Co gamma-ray of 25% relative efficiency and 1.8 keV energy resolution (Fedlix, 2015), which was considered for the study. The specific activity of the nuclide ^{198}Au in the epithermal flux monitor (Al-0.1%Au thin wire) was calculated from the nuclear data of equation (1) and can be used for the calculation of the epithermal neutron flux(φ_e) using equation (2) (De Sorte, 1972).

$$A_{sp(Cd)}=\left(\frac{N_P/t_c}{SDCw}\right) \quad (1)$$

$$\phi_e = \left(\frac{N_P/t_c}{SDCw}\right).\frac{M}{N_A\,\theta I_\gamma I_o(\alpha)F_{Cd}\,\varepsilon_d} \quad (2)$$

where $A_{Cd} = \frac{N_p}{t_c}$ = Activity of ^{198}Au measured in an irradiation channel lined with Cd

N_p = Net photopeak area (counts) of ^{198}Au

$S = (1 - e^{-\lambda t_i})$ = Saturation factor (3)

$D = e^{-\lambda t_d}$ = Decay factor (4)

$C = \left(\frac{1-e^{-\lambda t_c}}{\lambda t_c}\right)$ = Correction factor for decay during counting for ^{198}Au

$N = \left(\frac{N_A\,\theta w I_\gamma}{M}\right)$ = Total number of nuclei/atom in ^{197}Au

N_A = Avogadro's number= 6.023×10^{23} atoms/mol

$I_o(\alpha)$ = Corrected resonance integral for gold in the Cd lined irradiation channel (in barns)

F_{Cd} = Cadmium epithermal neutron transmission factor for Au=0.991

α = Measured epithermal neutron shape factor in the Cd lined irradiation channel

$w = W \times C$ = Actual mass of gold in the sample of Al-0.1% Au foil irradiated(g),

W = Mass of Al-0.1%Au thin foil irradiated

C = Percentage of Au present in Al-0.1% Au thin foil

M = Atomic weight of gold

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θ = Isotopic abundance for gold,

I_γ = The abundance of γ-ray energy of ^{198}Au

ε_d = Photopeak efficiency of the detector

λ = Decay constant, t_i=Irradiation time, t_d = Decay time , t_c= Counting time

RESULTS AND DISCUSSION

The specific activity of the flux monitor Al-0.1%Au thin wire of 0.0122 g was calculated, 2 hours irradiation time in the inner cadmium irradiation channel A1 and counting time of 30min was considered, and using Excel Sheet 2007, specific activity of 5.0 x 10^8counts/s g using the experimental parameter of net peak value of 1310462, and correction factors. Saturation factor "S" and Decay factor "D" as shown in Table 2 was found using equation (3) and (4) respectively. The epithermal flux value which can be derived from the specific activity of 5.0 x 10^8 counts/s. g found is in good agreement with previous works of (Njinga *et al*., 2011; Fedlix, 2015) with a value of 5.34 x 10^8 counts/s.g using different flux monitor (Al-0.1% Au foil). Therefore, the value found in this study shows that the epithermal channel A1 of the NIRR-1 remain suitable for ENAA after the reactor's fuel conversion from HEU to LEU. This implies that the values of $Q_0(\alpha)$ and $I_0(\alpha)$ will show stable trend along the inner and outer channels. Our results equally shows that the neutron flux distributions have not been affected by the installation of the cadmium lined in the inner channel A1.

Table 2: Experimental nuclear parameter for specific activity and epithermal neutron flux determination

Reaction	Isotopic Abundance	$t_{1/2}$ (s)	γ (keV)	Np (count)	t_m (s)	w (g)	S	D	M	Np/t_m count/s	A_{sp} (counts/s.g)
Au^{197} (n, γ) Au^{198} wire	1	2.33E+05	411.83	1310462	1800	0.0122	0.01066	0.63042	0.0122	728.0344	5E+08

CONCLUSION

The activity of ^{198}Au in the Al-0.1% Au wire irradiated for 2 hours in the inner channel of A1 of NIRR-1 was found to be 5.0×10^8 n/cm^2.s which can be used to measure the epithermal neutron flux in the Cd lined irradiation channel, and was discovered to be in good accord with the values established in related research on the NIRR-1's Cd channels after its conversion to LEU. This present study has equally re-established the fact that NIRR-1 has a stable neutron flux despite its fuel conversion, which is one of its hallmarks. It is important to emphasize that this study, though performed after the installation of cadmium line in the thermal region of the rector and its conversion to LEU, the Epithermal Neutron Activation Analysis will consequently improve, thereby improving the analysis of key trace elements (such as I, Cd, Ag, Sn, Ba, Th, U, among others) and other radioactive isotope in biological, environmental and geological samples.

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