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Evaluation of some research reactor parameters in Modified Irradiation Site of NIRR-1

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ABSTRACT

The evaluation of I_0 (α) and Q_0 (α) for Gold, Cupper, Thorium, Uranium and Potassium in Modified Irradiation Site of Nigeria Nuclear Reactor-1 (NIRR-1) was done using Instrumental Neutron Activation Analysis (INAA) and off-line gamma ray spectrometric technique. It was found that before the Cd-lined installation i.e. modification of the large irradiation site (A-3) of NIRR-1, the deviation parameter (α) was positive with a value of 0.024±0.002 indicating high neutron thermalization. However, after the Cd-line installation, the α-parameter was found to be negative with a value of -0.9274 ± 0.016 indicating poor thermalization and as a consequence achieved the purposed of the modification. Further correction for Q_0 to Q_0 (α) and I_0 to I_0 (α) for the nuclides; Au, Cu, Th, U, and K in the modified large outer irradiation channel A-3 were done. Before Cd-lined installation the Q_0 (α) values for Au was 15.05, for Cu was 0.88, for Th was 10.05, for U was 0.86 and for K was 90.37. However, after the Cd installation the Q_0 (α) values for Au increased to 139.64, for Cu increased to 4681.32, for Th increased to16661.89, for U increased to 6141.55 and for K it increased to 133177.92. Also, before Cd-lined installation (BF), I_0 (α) values for Au and Cu were 1485.410, 3.870 and after Cd-lined installation they increased to 13782.47 and 18631.64 respectively.

Introduction

Trace concentrations of elements in complex matrices such as rocks, soils and sediments are very important in both mineral exploitations and soil fertility mapping [5]. The analysis of these materials involves methods such as atomic absorption spectrometry (AAS), inductively coupled plasma-atomic emission spectrometry (ICP-AES) or mass spectrometry (MS). All of these methods require dissolution and sometimes subsequent chemical treatment of the sample to ensure full dissolution with the intrinsic risk of contamination [5]. However, the Instrumental Neutron Activation Analysis (INAA) method is used extensively to determine the concentrations of elements in some varieties of complex matrices such as rocks, soils and sediments.

INAA is an analytical technique which involves the use of neutrons. These neutrons are usually sourced from available nuclear research reactor. In this case, the neutrons were sourced from a nuclear reactor at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, with the code-named Nigeria Nuclear Reactor-1 (NIRR-1).

In the evaluation of thermal, epithermal and fast fluxes in the core of a research reactor and quantifying the concentrations of elemental contents in any given matrix, the nuclear data that are commonly used includes; thermal neutron cross sections and neutron resonance integrals. Other non-neutron nuclear data that are of significant contribution in the evaluations includes; isotopic compositions of target nuclides and radioactive half-lives, gamma-ray energies and intensities of reaction product nuclides. Specifically, in determination of elemental concentrations using INAA laboratory, the k_0 Neutron Activation Analysis (k_0-NAA) or k_0 Epithermal Neutron Activation Analysis (k_0-ENAA) techniques are used [13].

From the two equations (k_0-NAA and k_0-ENAA) published [13], the reliability of I_0 (α) and Q_0 (α) values affirmed the dependent on the deviation of the epithermal neutron spectrum from the 1/E shape (α) as essential parameters for accurate application of the k_0-IAEA program and k_0 standardization neutron activation analysis (NAA) technique.

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Among the various experimental methods available for determining the deviation shaping factor (α), the "Cd-covered multi-monitor" method was used in the modified permanent Cd-lined irradiation position A-3 of NIRR-1. The choice of the method was based on the fact that the site was modified for thermal neutron shielding using Cd-line of approximately 1.0 mm thickness.

The epithermal shaping (α) factor obtained experimentally was used to correct for resonance integrals (I_0) which is referred to as an integral of the cross section with a neutron flux density distribution proportional to the reciprocal of the neutron energy 1/E. The resonance integrals can be measured directly or calculated from measured resonance parameters. In the absence of resonance parameters, a resonance integral can be determined as a percentage of the cross section [7]. The Q_0 (α) defined as the ratio of resonance integral to thermal neutron capture cross section at neutron velocity of 2200 m/s for the nuclides is very paramount to k_0 standardization technique. This work is focused on the changes made when I_0 is corrected to I_0 (α) and Q_0 is corrected to Q_0 (α) specifically for the nuclides; Au, Cu, Th, U, and K in the modified permanent cadmium lined (A-3) irradiation channel of NIRR-1.

It is a fact that gold (^{197}Au) with the well-known nuclear characteristics, as a primary standard monitor, is generally used for the cross sections measurements [3], [11]. The nuclear reaction ^{63}Cu (n, γ) ^{64}Cu emitting gamma peak line at 1346 keV having half-life ($T_{1/2}$) of 12.7 hours has been employed for the investigation of flux stability of NIRR-1 through specific activity ratios [9]. However, Thorium, uranium and potassium are very essential in archaeometry studies [1]

Experimental Procedure

The following sets of monitors were used to characterize the neutron irradiation facility: Al-0.1% Au, Zr with purity of 99.7329 %; Zn with purity of 99.9872 %. Each of these foil monitors has a thickness varying from 0.1mm to 0.254 mm. These elements have cross sections and resonance energies with Q_0-values ranging from low to high and are used both for irradiation position characterization and for thermal, epithermal and fast flux evaluations. The monitors were carefully weighed using the analytical balance called Mettlar EA 240 and on the same day in order to eliminate the systematic error due to weighing of small masses. Known amount of the monitors were cut into roughly equal pieces and rolled spirally. A set of these four monitors were prepared and irradiated in the Cd-lined channel A-3 packed together in a standard polyethylene vial. All the samples were irradiated for 3 hours at maximum thermal power of 15.5 kW. The irradiated samples were allowed to decay with an appropriate cooling time. Within same day of irradiation all the activated monitors were counted at distance of 15 cm for first counts. After one day of cooling, all the monitors were counted at 2 cm for second counts. Details of the experimental procedures established at centre for energy research and training (CERT), Ahmadu Bello University, Zaria has been published [8]

Efficiency Calibration

The ORTEC detector (GEM-30195) that was employed has the following dimensions; crystal diameter 58.8 mm, crystal length 76.3 mm, end cap to crystal 3 mm, aluminum absorbing layer 1.27 mm, inactive germanium layer 0.7 mm, and the top cover diameter 72.4 mm were entered in the permanent database of the k_0-IAEA program as well as the quoted activity (Bq) and manufactured date for ^{137}Cs and ^{152}Eu obtained from certificates. The background was counted for 3600 seconds for proper background separation by the software. The single-radionuclide source ^{137}Cs was measured for 1800 sec. at 17 cm for peak-to-total curve estimation. The mixed peaks (energies) source ^{152}Eu was measured at the two listed distances of 15 cm and 2 cm from the detector end cap for 3600 seconds. The measurements were performed such that the peak statistics in each main peak areas of the radionuclide were better than 0.5% [8]. The detailed analysis and interpretation of the efficiency data generated has been published [10].

Calculation And Results

The determination of the thermal to epithermal neutron flux parameter *f* is described elsewhere by Njinga et al., [9] and epithermal shape factor for the Cd-lined channel A-3 were determined using Table 1 simultaneously with the calculated specific activity. The epithermal shape factor (α) for the Cd-lined channel A-3 was determined using Cd-covered multi-monitor technique.

Table 1: Monitors and relevant nuclear data require in the work

Monitors	$\bar{E}_r$, eV	Q_0	Half-life	Eγ keV	F_{cd}
Au-197	5.7±7.1	15.7±1.8	2.7±0.10d	411.8	0.991
Zr-94	6260.0±4.0	4.6	64.0±0.01d	724.2 756.7	1.0
Zr-96	338.0±2.1	231.0	16.4±0.100h	657.9 743.3	1.0
Zn-64	2560.0±10.0	1.9±4.9	244.0±0.08d	1115.5	1.0
Zn-68	590.0±10.0	3.2±1.4	13.8±0.150h	438.6	1.0

$\bar{E}_r$,= effective resonance energy obtained from literature [2], [4], [6]

For determination of non-ideality of the neutron spectrum (α-value), a set of N monitors is irradiated in the Cd-lined irradiation channel, the so called Cd-covered multi-monitor method and the induced activities are measured on a detector with well known efficiency curve. With all the monitors obeying the $\sigma(v)$ versus $1/v$ dependence, the mathematical expression given in Equation 1 can be solved by an iterative procedure for a set of N monitors and the α-value is found as the root of the equation [12]

$$\alpha+\frac{\sum_{i=1}^{N}\left[\left(\log\bar{E}_{r,i}-\frac{\sum_{i=1}^{N}\log\bar{E}_{r,i}}{N}\right)\left(\log\frac{\bar{E}_{r,i}^{-\alpha}(A_{sp,i})_{Cd}}{k_{0,Au}(i)\varepsilon_{p,i}F_{Cd,i},Q_{0,i}(\alpha)G_{e,i}}-\frac{\sum_{i=1}^{N}\log\frac{\bar{E}_{r,i}^{-\alpha}(A_{sp,i})_{Cd}}{k_{0,Au}(i)\varepsilon_{p,i}F_{Cd,i},Q_{0,i}(\alpha)G_{e,i}}}{N}\right)\right]}{\left(\sum_{i=1}^{N}\log\bar{E}_{r,i}-\frac{\sum_{i=1}^{N}\log\bar{E}_{r,i}}{N}\right)^2}=0 \qquad 1$$

Alternatively, the α-value can be obtained from the plot of $\log T_a$ versus $\log T_b$.

where $T_a=\frac{\bar{E}_{r,i}^{-\alpha}(A_{sp,i})_{Cd}}{k_0(i)\varepsilon_{p,i}F_{Cd,i},Q_{0,i}(\alpha)G_{e,i}}$ and $T_b=\bar{E}_{r,i}$

The specific activity $A_{sp,i}$ is defined as;

$$A_{sp}=\frac{N_p/t_m}{wSDC} \qquad 2$$

where w = weight of foil monitors, S = saturated factor, D = decay factor, C = counting factor, t_m = measuring time, N_p = net peak count corrected for pulse losses.

where i denotes the i^{th} monitor, N the number of monitors used, $\bar{E}_{r,i}$ the effective resonance energy, F_{cd} the Cd-transmission factor for epithermal neutrons, $G_{e,i}$ the epithermal neutron self- shielding factor, $A_{sp,i}$ is the specific activity of the i^{th} monitor irradiated in the Cd-line, $\varepsilon_{p,i}$ is full energy peak detector efficiency, $k_0(i)$ is k_0 value of the i^{th} monitor relative to Au.

The $Q_{0,i}=I_0/\sigma_0$ defined as the ratio of resonance integral to thermal neutron capture cross section at neutron velocity of 2200m/s for the i^{th} monitor where corrected from $Q_{0,i}$ to $Q_{0,i}$ (α) as shown in Equation 3 [12];

$$Q_{o,i}(\alpha)=\left[\frac{Q_{0,i}-0.429}{\bar{E}_{r,i}^{\alpha}}+\frac{0.429}{(2\alpha+1)E_{Cd}^{\alpha}}\right]E_{\alpha}^{\alpha} \qquad 3$$

Also, the resonance integral ($I_{0,i}$) for the i^{th} monitor where corrected from $I_{0,i}$ to $I_{0,i}$ (α) as shown in Equation 4 [12];

$$I_{0,i}(\alpha)=\left(\frac{I_{0,i}-0.429\sigma_{th}}{\bar{E}_{r,i}^{\alpha}}+\frac{0.429\sigma_{th}}{(2\alpha+1)E_{Cd}^{\alpha}}\right)E_{\alpha}^{\alpha} \qquad 4$$

where $E_{\alpha}^{\alpha}=1^{\alpha}$ is the 1 eV arbitrary energy, σ_{th} =thermal flux and $E_{Cd}^{\alpha}=0.55^{\alpha}$ is the effective cadmium cutoff energy.

The nuclear data for the four monitors used were F_{Cd} (^{198}Au) = 0.991, and $G_{e,i}$ (^{95}Zr) = 0.983 where F_{Cd} is the Cd-transmission factor for epithermal neutrons and $G_{e,i}$ is the epithermal neutron self-shielding factor for the i^{th} monitor. The results of the evaluated α-values are shown in Table 2. However, the value of thermal to epithermal flux ratio, (f) after Cd-line installation Table 2 was performed using the k_0- IAEA software since the theoretical value was given zero due to the cut-off of the thermal neutrons by the Cd-liner.

Table 2: Flux Parameters obtained

Reactor Irradiation Channel Parameters	Irradiation channel; A-3 (BF)	Irradiation channel; A-3 (AT)
Thermal to epithermal flux ratio, (f)	49.5 ± 0.10	(1.9 ± 0.020) E-05
Deviation of the epithermal neutron flux distribution from the ideal 1/E, law, α	0.024 ± 0.002	-0.9274 ± 0.016

BF = Before Cd-line installation, AT = After Cd-line installation

This α-values obtained in Table 2, was used to correct for Q_0 (α) using Equation 1 for the following nuclides, Au, Cu, Th, U, and K and I_0 (α) for the following nuclides, Au and Cu in the modified permanent large outer cadmium lined (A-3) and the unmodified outer irradiation position (B-4) and results indicated in Table 3, 4, 5, 6 and 7. Also, the Q_0 (α) and I_0 (α) values for the inner irradiation channels (A-1, B-2, and B-3) were validated and the results also shown in Table 3, 4, 5, 6 and 7. However, the f-value of the channels inner irradiation channel (A-1, B-2, and B-3) and outer irradiation channel B-4 of NIRR-1 has been described elsewhere [9].

Table 3: Q_0 (α) values obtained for Au

Irradiation Positions	A-1	A-3 , BF	A-3, AT		B-2	B-3	B-4
Pro-Isotope	^{198}Au	^{198}Au	^{198}Au		^{198}Au	^{198}Au	^{198}Au
Reaction	^{197}Au (n γ)	^{197}Au (n γ)	^{197}Au (n γ)		^{197}Au (n γ)	^{197}Au (n γ)	^{197}Au (n γ)
Q_0	15.7	15.7	15.7		15.7	15.7	15.7
Q_0 (α)	17.0	15.1	139.6		17.2	17.1	14.9

Table 4: Q_0 (α) values obtained for Cu

Irradiation Positions	A-1	A-3, [BF]	A-3, [AT]	B-2	B-3	B-4
Pro-Isotope	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu
Reaction	^{63}Cu (n γ)	^{63}Cu (n γ)	^{63}Cu (n γ)	^{63}Cu (n γ)	^{63}Cu (n γ)	^{63}Cu (n γ)
Q_0	1.1	1.0	1.0	1.1	1.1	1.1
Q_0 (α)	1.4	0.9	4681.3	1.5	1.4	0.9

Table 5: Q_0 (α) for Th, K and U

Irradiation Positions	A-3, BF	A-3, AT	A-3, BF	A-3, AT	A-3, BF	A-3, AT
Pro-Isotope	^{233}Th	^{233}Th	^{42}K	^{42}K	^{239}U	^{239}U
Reaction	^{232}Th (n γ)	^{232}Th (n γ)	^{41}K (n γ)	^{41}K (n γ)	^{238}U (n γ)	^{238}U (n γ)
Q_0	11.5	11.5	0.9	0.9	103.4	103.4
Q_0 (α)	10.1	16661.9	0.9	6141.6	90.4	133177.9

Table 6: I_0 (α) values obtained for Au

Radiation Positions	A-1	A-3, BF	A-3, AT	B-2	B-3	B-4
Pro-Isotope	^{198}Au	^{198}Au	^{198}Au	^{198}Au	^{198}Au	^{198}Au
Reaction	^{197}Au(n γ)	^{197}Au(n γ)	^{197}Au(n γ)	^{197}Au(n γ)	^{197}Au(n γ)	^{197}Au(n γ)
I_0	1549.6	1549.6	1549.6	1549.6	1549.6	1549.6
I_0 (α)	1680.8	1485.4	13782.5	1698.8	1686.8	1472.4

Table 7: $I_0(\alpha)$ values obtained for Cu

Irradiation Positions	A-1	A-3, BF	A-3, AT	B-2	B-3	B-4
Pro-Isotope	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu	^{64}Cu
Reaction	^{63}Cu(n γ)	^{63}Cu(n γ)	^{63}Cu(n γ)	^{63}Cu(n γ)	^{63}Cu(n γ)	^{63}Cu(n γ)
I_0	4.4	4.4	4.4	4.4	4.4	4.5
$I_0(\alpha)$	5.6	3.9	18631.6	5.8	5.7	3.8

Discussion

The specific activities of the respective monitor foils were calculated using the expression in equation 2. The data obtained together with the detector efficiency and nuclear data of monitor reactions were substituted in equation 1. The alfa value in the Cd-lined channel was determined by solving equation 1 and by means of an iterative procedure based on MS-EXCEL 'solver' spreadsheet utilities. In conformity, the alfa value was also obtained from the plot of $\log T_a$ versus $\log T_b$ and results displaced in Table 2.

As shown in Table 2, before the Cd-lined installation, the α-value was positive in the A-3 with a value of 0.024±0.002. This positive values of α indicates that the neutron spectrum was softened before the installation in channel A-3 (i.e.high thermalization). However, after the Cd-line installation, in A-3 the α-value was found to change from positive 0.024±0.002 (high thermalization) to a negative value of -0.9274 ± 0.016 showing that the neutron spectrum is hardened (approximately zero thermalization). This result was expected as the Cd-liner completely cuts off the thermal component of the neutron spectrum in the Cd-lined irradiation channel [9].

Results presented in Table 3 & 4, of $Q_0(\alpha)$ parameter shows a great agreement for A-1, B-2 and B-3 with $Q_0(\alpha)$-values of 17.030, 17.210 and 17.090 for Au and 1.430, 1.480 and 1.440 for Cu respectively because of similarities in channel configurations (all inner irradiation channel). However, this value was 14.920 for Au and 0.940 for Cu in B-4 outer irradiation channel. Also results in Table 6 & 7 of $I_0(\alpha)$ for A-1, B-2 and B-3 were1680.81, 1698.770 and 1686.780 for Au and 5.610, 5.790 and 5.670 for Cu respectively shows a great agreement between the channels of similar configurations. But for B-4 the outer irradiation channel, the $I_0(\alpha)$ value for Au was 1472.390 and Cu was 3.770.

From Table 3 & 4, the $Q_0(\alpha)$-values for Au and Cu in A-3 before Cd-lined installation was 15.1 and 0.9 respectively while Table 9 and 10 shows $I_0(\alpha)$-values for Au and Cu to be 1485.4 and 3.9 respectively before installation. After the installation, these $Q_0(\alpha)$ values increased drastically to 139.6 and 4681.3 while for $I_0(\alpha)$ it increased to 13782.5 and 18631.6 respectively. However, the interferences of (n, γ) reactions of matrix elements occurs mainly with thermal neutrons of low resonance integral to thermal cross section ratio such as ^{23}Na (n, γ) ^{24}Na with Q_0 = 0.59, ^{41}K (n, γ) ^{42}K with Q_0 = 0.87 and ^{46}Ca (n, γ) ^{47}Ca with Q_0 = 1.3. Thus, the installation of Cd-line increase these values as indicated in Table 3, 4, 5, 6, and 7. From Table 5, the $Q_0(\alpha)$-values for Th/Pa, K and U/Np increased as follows; for ^{233}Th, rose from 10.1 to 16661.9, for ^{42}K, rose from 0.9 to 6141.6 and for ^{239}U, it rose from 90.4 to 133177.9. These increased in $Q_0(\alpha)$-values implies reduction in the interferences of Th/Pa, K and U/Np and allowed almost all (n, γ) activation of the matrix to be induced by epithermal neutrons thereby allowing the determination of these elements in A-3 accurately.

Conclusion

The $Q_0(\alpha)$ and $I_0(\alpha)$-values for^{197}Au (n γ) ^{198}Au, ^{63}Cu (n γ) ^{64}Cu, ^{232}Th (n γ) ^{233}Th, ^{41}K (n γ) ^{42}K and ^{238}U (n γ) ^{239}U have been determined using instrumental neutron activation analysis technique to provide fundamental data for research on nuclear engineering, nuclear archaeometry and nuclear physics. The result obtained showed low interferences in the matrices during irradiation attributed to poor thermalization for α value of -0.9274 ± 0.016 unlike before the installation, the α value was observed to be 0.024±0.002 indicating high thermalization of the neutron spectrum. Thus, the installation of Cd-line in A-3 has been achieved to remove the thermal neutron component in the large Cd-lined irradiation channel (A-3). Therefore, the k_0-Epi-Cd-NAA and FNAA techniques will be applied accurately and the (n, γ) activation of Th, K and U by epithermal neutrons will be useful for archaeological studies.

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