



The Accuracy of the α Determination in the Cadmium Lined Irradiation Channel of the NIRR-1 Using the ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{97}Zr - ^{95}Zr Monitor Combinations

F.T. Sheyin^{1,*}, S.A. Jonah¹ and U. Sadiq²¹Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria.²Department of Physics, Ahmadu Bello University, Zaria, Nigeria.**ARTICLE INFO****Article history:**

Received: 2 December 2016;

Received in revised form:

25 March 2017;

Accepted: 6 April 2017;

Keywords α value,

Cadmium covered multimonitor and triple methods,

Cadmium lined irradiation channel,

 k_0 -ENAA.**ABSTRACT**

In a previous study, the epithermal neutron shape factors (α) determined with the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr by the cadmium covered multimonitor method and the triple monitor ^{198}Au - ^{97}Zr - ^{95}Zr in the cadmium lined irradiation channel of the NIRR-1 were found to have relatively high and more reasonable values of -0.101 ± 0.019 and -0.106 ± 0.014 respectively. In this study, the α value determined with the triple monitor ^{198}Au - ^{99}Mo - ^{95}Zr that was obtained from the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr was found to be -0.114 ± 0.017 . The accuracy of the α determination with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr , ^{198}Au - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{99}Mo - ^{95}Zr was tested by elemental analysis of the standard reference material NIST 1515 Apple leaves by the k_0 -ENAA method using Al-0.1% Au thin foil as the single comparator. The concentrations of the elements Sm and Br with high resonance integrals determined in the NIST 1515 Apple leaves are in good agreement with the certified values. The monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr or the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr from the monitor set Au+Mo+Zr or only the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr from the monitor set Au+Zr can provide reliable values of α in the cadmium lined irradiation channel for elemental analysis of samples of materials by the k_0 -ENAA method in the NIRR-1.

© 2017 Elixir All rights reserved.

Introduction

The 30 kW Nigeria Research Reactor-1 (NIRR-1) is one of the commercial Miniature Neutron Source Reactors (MNSRs) installed outside China. It was commissioned in 2004 and has been standardized for instrumental neutron activation analysis (INAA) [11]. In order to extend its utilization for epithermal neutron activation analysis (ENAA) protocol, the cadmium (Cd) lined irradiation channel was installed in one of the large outer irradiation channels [15].

There has been the need for the use of various or different combinations of monitor sets that provide the most accurate determination of the epithermal neutron shape factor (α) that would give the most accurate results of the single comparator method of epithermal neutron activation analysis (k_0 -ENAA) in the cadmium lined irradiation channel of the NIRR-1 [17]. Consequently, the α value in the cadmium lined irradiation of the NIRR-1 was re-evaluated using different monitor combinations from the product nuclides of the monitor set Al-0.1% Au thin foil, Zr and Zn foils, Mo and Co thin wires irradiated in the cadmium lined irradiation channel of the NIRR-1. The α values obtained with the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr by the cadmium covered multimonitor method and with the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr were found to have relatively higher, more reasonable and comparable values of -0.101 ± 0.019 and -0.106 ± 0.014 respectively [17]. The first aim of this study was to re-assess the monitor combination

^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr for the possibility of obtaining a suitable triple monitor for α determination. The second aim of the study was to test the accuracy of the α value to be determined with this triple monitor and also the accuracy of each of the α values obtained with the ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{97}Zr - ^{95}Zr monitor combinations in the cadmium lined irradiation channel by elemental analysis of standard reference materials (SRMs) by the k_0 -ENAA method using the activity of ^{198}Au as the single comparator.

In the cadmium covered multimonitor method, a set of N suitable monitors are coirradiated under cadmium cover and the induced activities measured on an efficiency calibrated high purity germanium (HPGe) detector system. For elements having a cross section $\sigma(v) \propto 1/v$ in the range up to ≈ 1.5 eV, α can be found as the slope ($-\alpha$) of the straight line by plotting the graph of [3]

$$\log \frac{E_{r,i}^{-\alpha} \cdot (A_{sp,i})_{Cd}}{k_{o,Au(i)} F_{Cd,i} Q_o(\alpha)_i G_{e,i} \epsilon_{d,i}} \text{ versus } \log E_{r,i} \quad (1)$$

where

$k_{o,Au(i)} = k_0$ -factors for the i th monitor with respect to Au, resonance integral to thermal neutron cross section ratio in the actual irradiation site

$Q_o(\alpha)$ = corrected resonance integral to thermal neutron cross section ratio of nuclides in actual irradiation site

$(A_{sp,i})_{Cd}$ is the specific activity of the ith monitor

$$(A_{sp,i})_{Cd} = \left[\frac{N_p / t_c}{SDCW} \right]_{Cd}$$

monitor

the Cd index denotes Cd cover irradiation ,

N_p = Net photopeak area (counts) of each monitor

irradiated in Cd lined channel,

$S = (1 - e^{-\lambda t_i})$ = saturation factor for each monitor

$D = e^{-\lambda t_d}$ = decay factor for each monitor

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

counting for each monitor

λ = decay constant, t_i = irradiation time, t_d = decay time,

t_c = counting time

w = mass of element in sample of thin foil or wire monitors irradiated(g)

F_{Cd} = cadmium epithermal neutron transmission factors for the ith nuclide

$\epsilon_{d,i}$ = full energy peak detector efficiency of the ith nuclide,

$\epsilon_\gamma = \gamma$ - ray intensity,

$i=1,2,\dots,N$ = number of nuclides

$\bar{E}_{r,i}$ = average resonance energy of the ith monitor,

$G_{e,i}$ = correction factor for epithermal neutron self shielding of ith monitor

The left hand term of Eq. 1 is itself a function of α , and thus an iterative procedure can be applied with square regression analysis to fit the experimental data to the straight lines for every iterative step.

Mathematically, the final α result of this iteration procedure is identical with solving for α from the equation [3]

$$\alpha + \frac{\sum_{i=1}^N \left\{ \log \bar{E}_{r,i} - \frac{\sum_{i=1}^N \log \bar{E}_{r,i}}{N} \right\} \left\{ \log T_i - \frac{\sum_{i=1}^N \log T_i}{N} \right\}}{\sum_{i=1}^N \left[\log \bar{E}_{r,i} - \frac{\sum_{i=1}^N \log \bar{E}_{r,i}}{N} \right]^2} = 0 \quad (2)$$

where

$$T_i = \frac{\bar{E}_{r,i}^{-\alpha} (A_{sp,i})_{Cd}}{k_{o,Au(i)} F_{Cd,i} Q_o(\alpha)_i G_{e,i} \epsilon_{d,i}}$$

$k_{o,Au(i)}$ = k_o -factors for the ith monitor with respect to Au

$\bar{E}_{r,i}$ = average resonance energy of the ith monitor,

$i=1,2,\dots,N$ = number of nuclides

2. Materials and methods

The set of monitors Al-0.1%Au thin foil, Zr and Zn foils, Mo and Co thin wires were weighed, arranged in a small polyethylene thin film bag and then put inside a 25 cm³ polyethylene vial. They were covered with cotton wool and the top of the polyethylene vial properly sealed with a celotape. The characteristics of the detector foils and wires used are shown in Table 1.

The set of monitors in the 25 cm³ polyethylene vial were irradiated simultaneously in the Cd lined irradiation channel of the NIRR-1 at a thermal neutron flux of 5×10^{11} n.cm⁻².s⁻¹ for 3 hours. The induced activities on the irradiated set of monitors were measured using the full energy peak efficiency calibrated P-type GEM 30195 HPGe coaxial detector system at the distance of 2cm from the detector. The energy resolution of the system is 1.95 keV for the 1332 keV peak of ⁶⁰Co and the relative detector efficiency is 30%. The full energy peak efficiency of the P-type GEM 30195 HPGe coaxial detector was measured at the distances 2cm and 15cm from the detector over the energy range 59.54 -1408 keV using the set IAEA standard sources ²⁴¹Am, ¹⁵²Eu ²²⁶Ra, ¹³⁷Cs, ⁶⁰Co and ²²Na. A detailed description of the measured full energy peak efficiency curves and the theoretical fitting function to the experimental efficiency curves is given elsewhere [14].

The product nuclides ¹⁹⁸Au, ⁹⁷Zr and ⁹⁹Mo were counted for 3600 seconds within one day after irradiation of the α monitors. The product nuclides ⁶⁵Zn, ⁹⁵Zr and ⁶⁰Co were counted for 72000 seconds after 14.91-14.99 days. The photopeak areas of the radionuclides found in the spectra were obtained from the gamma-ray acquisition system of the P-type GEM 30195 HPGe coaxial detector system that consists of the Maestro Multichannel Analyser (MCA) emulation software card coupled to the detector via electronic nuclear Instrumentation modules manufactured by Ortec. The nuclear data of the product nuclides ¹⁹⁸Au, ⁹⁷Zr, ⁹⁵Zr, ⁹⁹Mo, ⁶⁵Zn and ⁶⁰Co are shown on Table 2.

Table 1. Description of the activation detector foils and wire for α determination.

Element	Material description	Diameter (cm)	Mass(g)
Au	Al-0.1%Au thin foil, 0.1mm thick,IRMM-530	0.8	0.0132
Zn	99.95% Zn foil, 0.025mm thick, Goodfellow	0.8	0.0083
Zr	99.8% Zr foil, 0.125mm thick, Goodfellow	0.8	0.0446
Mo	thin wire		0.0168
Co	thin wire		0.0105

Table 2. Nuclear data for the product nuclides of monitors irradiated for α determination in the Cd lined irradiation channel.

Monitor	Half life	E_γ (keV)	\bar{E}_r (eV)	Q_o	factors $k_{o,A}$	F_{Cd}
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ ($G_e=1$)	2.695d	411.8	5.65	15.7	1	0.991
$^{96}\text{Zr}(n,\gamma)^{97}\text{Zr}$ ($G_e=0.973$)	16.74h	743.4	338	251.6	1.24×10^{-5}	1
$^{94}\text{Zr}(n,\gamma)^{95}\text{Zr}$ ($G_e=0.983$)	64.02d	724.2 +756.7	6260	5.31	2.00×10^{-4}	1
^{99}Mo	65.94h	140.51	241	53.1	5.27×10^{-4}	1
^{65}Zn	243.9d	1115.55	2560	1.908	5.72×10^{-3}	1
^{60}Co ($G_e=0.520$)	5.27y	1173.24	136	1.993	1.32	1

Table 2 shows the nuclear data for the product nuclides of the monitor set Al-0.1% Au thin foil, Zr and Zn foils, Mo and Co thin wires irradiated for α determination in the Cd lined irradiation channel of the NIRR-1. The nuclear data \bar{E}_r , Q_o , F_{Cd} , G_e and $k_{o,Au}$ -factors were obtained from the literature [4], [6]. The efficiencies ϵ of the gamma-ray energies of the product nuclides of the monitors were calculated by the authors using mathematical fitting functions described elsewhere [14]. The specific activities $A_{sp(Cd)}$ for each of the product nuclides of the monitors were also calculated by the authors. In Table 2, the two energies of the ^{95}Zr nuclide have been summed (724.24 keV + 756.7 keV). This nearly doubles the useful counts of the two γ -ray lines thus improving the counting statistics of the ^{95}Zr nuclide in view of the relatively low resonance integral (0.3 barns) for epithermal activation [18]. The specific activity of the ^{95}Zr nuclide was calculated using the sum of the activities of the 724.2 keV and 756.7 keV γ -lines.

It has been reported recently that following the re-evaluation of the α in the cadmium lined irradiation channel of the NIRR-1 by the cadmium covered multimonitor method using four different monitor combinations from Table 2, the α values determined with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr by the cadmium covered multimonitor method and with the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr were found to have relatively higher, more reasonable and comparable values of -0.101 ± 0.019 and -0.106 ± 0.014 respectively [17]. The α values determined with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{97}Zr - ^{95}Zr as well as the epithermal neutron fluxes and comparator factors using the activity of ^{198}Au in the two monitor combinations are shown in Table 3.

In this study, we were interested in re-assessing the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr for the possibility of obtaining a suitable triple monitor combination for α determination. To achieve this, it was necessary that the three monitors to be chosen from the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr should fulfill the condition that their effective resonance energies $\bar{E}_{r,i}$ are uniformly distributed

ranging from low to high over the whole epithermal neutron energy region [3]. Therefore, the isotope ^{97}Zr with high Q_o value (251.6) was excluded from the monitor combination

leaving only the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr with suitable spread on their \bar{E}_r values (5.65 eV, 241 eV and 6260 eV). The α value was determined with the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr by solving Eq.1 by the iterative linear regression method based on MS Excel spread sheet. Starting with step 1 by setting $\alpha=0$ resulted in the α value of -0.107 ± 0.010 . After a four step iteration, the final value of α was found to be -0.114 ± 0.017 .

From the α value obtained, the epithermal neutron flux in the Cadmium lined irradiation channel at the preset power of the NIRR-1 was determined using the peak area of the 411.8 keV line of ^{198}Au in the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr from the equation

$$\phi_e = \frac{(A_{sp})_{Au} \cdot M_{Au}}{I_o(\alpha)_{Au} \cdot N_A \theta_{Au} \gamma_{Au} \cdot F_{Cd} \epsilon_d} \tag{3}$$

A_{sp} = specific activity for the ^{198}Au nuclide

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

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

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

α = measured epithermal neutron shape factor in the Cd lined channel

M = atomic weight of gold

θ = isotopic abundance for gold, γ_{Au} = absolute gamma-ray intensity for ^{198}Au

ϵ_d = photopeak efficiency of the detector for ^{198}Au

The comparator factors $F_{c,Au}$ using the activity of ^{198}Au in the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr was calculated from the equation [4]

$$F_{c,Au} = \frac{N_A \cdot \theta_{Au} \cdot \sigma_{o,Au} \cdot \gamma_{Au}}{M_{Au}} \cdot \phi_e \cdot 10^{-6} \tag{4}$$

where N_A = Avogadro's number, M = molar mass, θ = isotopic abundance, σ_o = the (n, γ) thermal neutron cross section, γ = the absolute gamma-ray intensity

Table 3 . Results of neutron spectrum parameters in the Cd lined irradiation channel.

Monitor combination	α	Φ_e (n.cm ⁻² .s ⁻¹)	$F_{c,Au}$
¹⁹⁸ Au- ⁹⁹ Mo- ⁹⁷ Zr- ⁹⁵ Zr	-0.101±0.019	(4.80±0.04)×10 ⁹	(1.38±0.01)×10 ³
¹⁹⁸ Au- ⁹⁷ Zr- ⁹⁵ Zr	-0.106±0.014	(4.76 ±0.04)×10 ⁹	(1.37±0.05)×10 ³
¹⁹⁸ Au- ⁹⁹ Mo- ⁹⁵ Zr	-0.114±0.017	(4.69±0.04)×10 ⁹	(1.35±0.01)×10 ³

ϕ_e = calculated epithermal neutron flux using ¹⁹⁸Au in each monitor combination

The epithermal neutron shape factors(α), the epithermal neutron flux and comparator factor determined with the three monitors ¹⁹⁸Au-⁹⁹Mo-⁹⁵Zr in the cadmium lined irradiation channel of the NIRR-1 are shown on Table 3.

3. Results and Discussions

Table 3 shows the values of the epithermal neutron shape factors(α), the epithermal neutron fluxes and comparator factors determined in the cadmium lined irradiation channel of the NIRR-1 using the monitor combinations ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr, ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr and ¹⁹⁸Au-⁹⁹Mo-⁹⁵Zr. It can be observed from Table 3 that the value of α determined with the triple monitor ¹⁹⁸Au-⁹⁹Mo-⁹⁵Zr is comparable with the α values obtained with the monitor combinations ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr and ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr. The values of α for the monitor combinations in Table 3 are all negative. This indicates a hardened (poorly thermalized) epithermal neutron spectrum in the cadmium lined irradiation channel of the NIRR-1.

From Table 3, the values of the epithermal neutron fluxes calculated using the activity of ¹⁹⁸Au in Al-0.1% Au thin foil in the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr, ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr and ¹⁹⁸Au-⁹⁹Mo-⁹⁵Zr monitor combinations are(4.80±0.04)×10⁹, (4.76 ±0.04)×10⁹ and (4.69±0.04)×10⁹ respectively. These values are comparable with the theoretical value of 5.05×10⁹ n.cm⁻².s⁻¹ in the outer irradiation site A2 of the NIRR-1 based on the assumption that the thermal neutron flux value in the outer irradiation sites are 50% of the value in the inner irradiation sites [13]. The thermal to epithermal neutron flux ratio in the outer irradiation site A2 of the NIRR-1 has been found to be approximately 49.5±0.96 [13]. It should be noted that the channel A2 is now the channel A3 that has been lined with the 1.0 mm thick Cd-liner.

The accuracy of each of the α values determined using the three monitor combinations in Table 3 was tested by elemental analysis of the standard reference material NIST 1515 Apple leaves by the k_o-ENAA method in the cadmium lined irradiation channel of the NIRR-1 using the activity of ¹⁹⁸Au as the single comparator. The standard reference materials NIST 1515 Apple leaves was irradiated together with the standard Al-0.1% Au thin foil as the single comparator in the Cd lined irradiation channel at a preset thermal neutron flux of 5×10¹¹ n.cm⁻².s⁻¹ for 6 hours. The induced activities on the elements of interest in the samples of NIST 1515 Apple leaves and Al-0.1% Au thin foil were measured under the same counting conditions at the distance of 2cm from the efficiency calibrated P-type GEM 30195 HPGe coaxial detector system after appropriate decay and counting periods.

The concentrations of the elements determined in the NIST 1515 Apple leaves were calculated by the k_o-ENAA

method with the MS Excel spread sheet from the expression [4]

$$C_{s(Cd)} = \left[\frac{\left(\frac{N_{p,s}/t_c}{SDCW} \right)_s}{\left(\frac{N_{p,Au}/t_c}{SDCW_{Au}} \right)_{Au}} \right]_{Cd} \cdot \left[\frac{F_{Cd,Au} \cdot Q_o(\alpha)_{Au} \cdot \epsilon_{d,Au}}{k_{o(Au)} F_{Cd,s} Q_o(\alpha)_s \epsilon_{d,s}} \right] \tag{5}$$

where the $k_{o,Au}$ factor is a compound nuclear constant of the nuclide in the sample of material with respect to Au

M_{Au} = atomic mass of gold

M_s = atomic mass of the element of interest in the sample matrix

θ_s = isotopic abundance of the element of interest in the sample matrix

θ_{Au} = isotopic abundance of gold,

$\epsilon_{\gamma,s}$ = γ -ray intensity of the element of interest in the sample matrix,

$\epsilon_{\gamma,Au}$ = γ -ray intensity of gold,

$\sigma_{o,s}$ = thermal neutron cross section of elements of interest in the sample matrix,

$\sigma_{o,Au}$ = thermal neutron cross section for Au,

$F_{Cd,Au}$ = the cadmium epithermal neutron transmission factor for gold

$F_{Cd,s}$ = the cadmium epithermal neutron transmission factor for elements in sample matrix,

$Q_o(\alpha)_{Au}$ = corrected resonance integral to the thermal neutron cross section ratios for gold in the Cd lined irradiation channel

$Q_o(\alpha)_s$ = corrected resonance integral to the thermal neutron cross section ratios for the elements in sample matrix in the Cd lined irradiation channel

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

$\epsilon_{d,s}$ = detector efficiency for γ -ray energy of nuclide in sample matrix

$\epsilon_{d,Au}$ = detector efficiency for γ -ray energy of ¹⁹⁸Au

W_s = total amount of sample irradiated under cadmium cover

C_s = concentration of the unknown element in sample matrix under cadmium cover(ppm)

w_s = calculated unknown weight of element in sample matrix under cadmium cover
 w_{Au} = weight of the sample of gold foil irradiated under Cd cover
 $N_{p,s}$ = Net photopeak area of radionuclide of interest in sample matrix under Cd cover
 $N_{p,Au}$ = Net photopeak area of ^{198}Au under Cd cover,
 $D = e^{-\lambda t_d}$ = decay factor for gold or elements of interest in the sample matrix under Cd cover,
 $S = (1 - e^{-\lambda t_i})$ = saturation factor for gold or elements of interest in sample matrix under Cd cover,
 $C = \frac{(1 - e^{-\lambda t_c})}{\lambda t_c}$ = correction factor for decay during counting for gold or elements in sample matrix
 λ = decay constant, t_i = irradiation time, t_d = decay time, t_c = counting time

The total uncertainties in the concentrations of elements in the NIST 1515 Apple leaves by the k_0 -ENAA method were calculated from Eq.5 using the MS Excel spreadsheet from the uncertainty propagation equation [19]

$$\frac{\sigma_{C_e}^2}{C_e^2} = \frac{\sigma_{N_{p,Au}}^2}{N_{p,Au}^2} + \frac{\sigma_{N_{p,s}}^2}{N_{p,s}^2} + \frac{\sigma_{\epsilon_{Au}}^2}{\epsilon_{Au}^2} + \frac{\sigma_{\epsilon_{s}}^2}{\epsilon_{s}^2} + \frac{\sigma_{k_{o,Au(s)}}^2}{k_{o,s(Au)}^2} + \frac{\sigma_{Q_o(\alpha)_{Au}}^2}{Q_o^2(\alpha)_{Au}} + \frac{\sigma_{Q_o(\alpha)_s}^2}{Q_o^2(\alpha)_s} \quad (6)$$

where C_e = concentration of elements of interest in NIST 1515 Apple leaves by k_0 -ENAA

σ_{C_e} = standard deviation in concentrations of elements of interest in NIST 1515 Apple leaves by k_0 -ENAA
 $N_{p,Au}$ = measured net photopeak area of 411.8 keV energy of ^{198}Au
 $\sigma_{N_{p,Au}}$ = standard deviation in measured net peak area of 411.8 keV energy of ^{198}Au
 $N_{p,s}$ = net photopeak areas of elements of interest in NIST 1515 Apple leaves
 $\sigma_{N_{p,s}}$ = standard deviation in net peak areas of elements of interest in NIST 1515 Apple leaves
 ϵ_{Au} = efficiency of 411.8 keV energy of ^{198}Au
 $\sigma_{\epsilon_{Au}}$ = standard deviation of the efficiency of 411.8 keV energy for ^{198}Au
 ϵ_s = efficiency of γ -ray line of elements of interest in NIST 1515 Apple leaves

$\sigma_{\epsilon_{s}}$ = standard deviation of the efficiency of γ -ray lines of elements of interest in NIST 1515 Apple leaves
 $Q_o(\alpha)_{Au}$ = corrected resonance integral to thermal neutron cross section ratio for Au
 $\sigma_{Q_o(\alpha)_{Au}}$ = standard deviation of resonance integral to thermal neutron cross section ratio for Au
 $Q_o(\alpha)_s$ = corrected resonance integral to thermal neutron cross section ratio for elements of interest in NIST 1515 Apple leaves
 $\sigma_{Q_o(\alpha)_s}$ = standard deviation of resonance integral to thermal neutron cross section ratio for elements of interest in NIST 1515 Apple leaves
 $\sigma_{k_{o,s}}$ = standard deviation for $k_{o(Au)}$ - factors for elements of interest in NIST 1515 Apple leaves
 $k_{o,s(Au)}$ = factors for elements of interest in NIST 1515 Apple leaves

The accuracy of the concentrations of elements determined in the NIST 1515 Apple leaves by the k_0 -ENAA method was estimated by calculating the percentage difference between the measured concentrations of elements and the certified values in the NIST 1515 Apple leaves from the equation

$$A(\%) = \frac{|C_m - C_r|}{C_r} \cdot 100 \quad (7)$$

where C_m = measured concentrations of elements in NIST 1515 Apple leaves

C_r = certified values of concentrations of elements in NIST 1515 Apple leaves

$A(\%)$ = percentage relative accuracy or error of concentrations of elements in NIST 1515 Apple

The results of the concentrations of elements found in the NIST 1515 Apple leaves determined by the k_0 -ENAA method, the calculated uncertainties and accuracies are shown on Table 4.

From Table 4, the elements Sm and Br were better determined in the NIST 1515 Apple leaves by the k_0 -ENAA method because they have high resonance activation integrals compared to their thermal neutron cross sections and strongly absorb neutrons with specific resonance energies in the epithermal neutron energy region. The results of the concentrations of elements in NIST 1515 Apple leaves by k_0 -ENAA using the α values determined with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr , ^{198}Au - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{99}Mo - ^{95}Zr are in good agreement with the certified values. This indicates the reliability of the α values determined in the cadmium lined irradiation channel of the NIR-1 with these monitor combinations.

Table 4 . Results of Concentrations of Elements in NIST 1515 Apple leaves by k_0 -ENAA using α values of different monitor combinations.

Element	Certified value (mg.kg ⁻¹)	¹⁹⁸ Au- ⁹⁹ Mo- ⁹⁷ Zr- ⁹⁵ Zr combination (mg.kg ⁻¹)	A(%)	¹⁹⁸ Au- ⁹⁷ Zr- ⁹⁵ Zr combination (mg.kg ⁻¹)	A(%)	¹⁹⁸ Au- ⁹⁹ Mo- ⁹⁵ Zr combination (mg.kg ⁻¹)	A(%)
Sm	3.00	3.15±0.14	5.00	3.14±0.14	4.67	3.13±0.14	4.33
Br	1.80	1.89±0.82	5.00	1.86±0.77	3.33	1.86±0.81	3.33

The percentage deviation between the measured concentrations and certified values of Sm and Br in the NIST 1515 Apple leaves by k_0 -ENAA using the α value of the ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr monitor combination are 5%. The percentage deviation between the measured concentrations and certified values of Sm and Br in the NIST 1515 Apple leaves by k_0 -ENAA using the α value of the ^{198}Au - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{99}Mo - ^{95}Zr monitor combinations are less than 5% compared to the results obtained with the ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr monitor combination.

Under the specific irradiation and counting conditions employed in this work, the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr or the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr from the monitor set Au+Mo+Zr and the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr from the monitor set Au+Zr can provide reliable values of α in the cadmium lined irradiation channel for elemental analysis of samples of materials by the k_0 -ENAA method in the NIRR-1.

4. Conclusions

Following the assessment of the epithermal neutron shape factor (α) in the cadmium lined irradiation channel of the NIRR-1 using different monitor combinations from the monitor set Al-0.1%Au thin foil, Zr and Zn foils, Mo and Co thin wires irradiated, the α values obtained with the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr by the cadmium covered multimonitor method and the triple monitor ^{198}Au - ^{97}Zr - ^{95}Zr were found to have relatively high and more reasonable values of -0.101 ± 0.019 and -0.106 ± 0.014 respectively [17]. In this study, the α value determined with the triple monitor ^{198}Au - ^{99}Mo - ^{95}Zr that was obtained from the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr was found to be -0.114 ± 0.017 . The negative values of α indicates a hardened (poorly thermalized) epithermal neutron spectrum in the cadmium lined irradiation channel of the NIRR-1.

The values of the epithermal neutron fluxes and comparator factors using the activity ^{198}Au in the triple monitors ^{198}Au - ^{99}Mo - ^{95}Zr were found to be comparable with the values obtained with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{97}Zr - ^{95}Zr .

The accuracy of the α determination with the monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr , ^{198}Au - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{99}Mo - ^{95}Zr was tested by elemental analysis of the standard reference material NIST 1515 Apple leaves by the k_0 -ENAA method using Al-0.1% Au thin foil as the single comparator. The concentrations of the elements Sm and Br with high resonance integrals determined in the NIST 1515 Apple leaves are in good agreement with the certified values. The monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr or the three monitors ^{198}Au - ^{99}Mo - ^{95}Zr from the monitor set Au+Mo+Zr or only the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr from the monitor set Au+Zr can provide reliable values of α in the cadmium lined irradiation channel for elemental analysis of samples of materials by the k_0 -ENAA method in the NIRR-1.

To improve the precision and accuracy and also extend the number of determinable elements with higher resonance energies of more than 10 eV in the epithermal neutron region by k_0 -ENAA, it is necessary to use moveable boron shielded vials with a cut-off energy of range up to 280 eV depending on thickness for epithermal activation of samples of materials in the Cd lined irradiation channel of the NIRR-1.

Acknowledgements

The authors are thanking the management of the Centre for Energy Research and Training, Ahmadu Bello University, Zaria and members of staff of the Nuclear Science and Technology Section, Centre for Energy Research and Training, Ahmadu Bello University, Zaria for their assistance in the experimental work of this study.

References

- [1] J. Daindith,, A Dictionary of Chemistry, 6th edition, Oxford University Press, pp 574, 2008.
- [2] K. Debertain and R.G. Helmer, , Gamma and X-ray Spectrometry with Semiconductor Detectors, Elsevier Science Publishing Co. , INC., New York, USA, pp. 206-223, 1988
- [3] F. De Corte, K. Sordo-El Hammami, L. Moens, A. Simonits, A. De Wispelaere, and J. Hoste, "The accuracy and precision of the α -determination in the $\frac{1}{E^{1+\alpha}}$ epithermal reactor neutron spectrum," J. Radioanal. Nucl. Chem. vol.62, no. 1-2, pp. 209-255, 1981.
- [4] F. De Corte and A. Simonits, Vade Mecum for k_0 -users, addendum to the KAYZERO/SOLCOL software package (Version 3.0), DSM Research Geleen (NL), R94/11492, pp.4, 1994.
- [5] F. De Corte, K., A. Moens and A. Simonits, Role of the $\frac{1}{E^{1+\alpha}}$ epithermal neutron flux distribution in the k_0 -method α -mapping of the reactor Thetis, BuLL. Soc. Chim., Belgium, vol. 90, no.4, pp.317-324, 1981.
- [6] F. De Corte and A. Simonits, "Recommended nuclear data for use in the k_0 - Standardization of neutron activation analysis," Atomic Data and Nuclear Data Tables. vol.85, pp.47-67, 2003.
- [7] D. De Soete, R. Gijbels and J. Hoste, , Neutron Activation Analysis, John Wiley and Sons Ltd, New York, USA, pp.67-71, pp.502-508, 1972.
- [8] M.S. Dias, V. Cardoso, M.F. Koskinas and I.M. Yamazaki, "Determination of neutron spectrum shape parameter α k_0 -NAA methodology using covariance analysis," Appl. Rad. Isotopes, vol. 68 pp. 592-595, 2010.
- [9] H.M. Dung and Y. Cho, "A simple method for α determination," J. Radioanal. Nucl. Chem, vol.257, no. 3, pp. 573-575, 2003.
- [10] S.A. Jonah, G.I. Balogun, I.M. Umar and M.C. Mayaki, Neutron spectrum parameters in irradiation channels of the nigerian research reactor-1(NIRR-1) for the k_0 -NAA standardization, J. Radioanal. Nucl. Chem. vol. 266, no. 1, pp. 83-88, 2005.
- [11] S.A. Jonah, I.M. Umar, M.O.A. Oladipo, G.I. Balogun, and D.J. Adeyemo, "Standardization of NIRR-1 irradiation and counting facilities for instrumental neutron activation analysis," Appl. Rad. Isotopes, vol.64, pp. 818-822, 2006.
- [12] W.Liyu, CIAE/WINSPAN, Multi-purpose Gamma-ray Spectrum Analysis Software, China Institute of Atomic Energy, Beijing, China, 2004.

- [13] U. Sadiq, S.A. Jonah, R. Nasiru, Y.I. Zakari, "Neutron spectrum parameters in two irradiation channels of the nigeria research reactor-1(NIRR-1) for use in k_0 -NAA," Bayero J. Pure and App. Sciences, vol.3, no.1, pp.220-223, 2010..
- [14] F.T. Sheyin, Characterization of the Cadmium Lined Irradiation Channel of the Nigeria Research Reactor 1(NIRR-1) and its Application for Elemental Determination in Some Legumes, Ph.D. Thesis, Ahmadu Bello University, Zaria, Nigeria, 2015.
- [15] F.T. Sheyin, M.O.A. Oladipo, S.A. Jonah and U. Sadiq, "Determination of α in the cadmium lined irradiation channel of the NIRR-1 for use in k_0 -ENAA," World Journal of Nuclear Science and Technology, **vol. 5**, pp. 233-240, 2015a, <http://dx.doi.org/10.4236/wjnst.2015.53023>
- [16] F.T. Sheyin, M.O.A. Oladipo, S.A. Jonah and U. Sadiq, "Elemental analysis of some nigerian food legumes by k_0 -ENAA and INAA," World Journal of Nuclear Science and Technology, **vol.5**, pp. 241-251, 2015b,<http://dx.doi.org/10.4236/wjnst.2015.53024>
- [17] F.T. Sheyin, S.A. Jonah, U. Sadiq, Assessment of α in the cadmium lined irradiation channel of the NIRR-1 using different monitor combinations, in press
- [18] A. Simonits, F. De Corte and J. Hoste, "Zirconium as a multi-isotopic flux ratio monitor and a single comparator in reactor neutron activation analysis," J. Radioanal. Chem. vol. 31, pp.467-486, 1976.
- [19] J.R. Taylor, An Introduction to Error Analysis, 2nd ed., University of Science Books, Sausalito, California, USA, pp. 45-91, 1997.