



Nuclear and Radiation Physics

Elixir Nuclear & Radiation Phys. 105 (2017) 46102-46106

Elixir
ISSN: 2229-712X

Assessment of α in the Cadmium Lined Irradiation Channel of the NIRR-1 Using Different Monitor Combinations.

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ARTICLE INFO

Article history:

Received: 17 November 2016;

Received in revised form:

25 March 2017;

Accepted: 6 April 2017;

Keywords

α value,
Cadmium covered
Multimonitor method,
Cadmium lined irradiation
Channel.

ABSTRACT

The epithermal neutron shape factor(α) in the permanent cadmium lined irradiation channel of the Nigeria Research Reactor-1(NIRR-1) was re-evaluated using the four monitor combinations ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr , ^{198}Au - ^{97}Zr - ^{95}Zr - ^{65}Zn , ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr - ^{99}Mo in the monitor set Al-0.1% Au thin foil, Zr and Zn foils, Mo and Co thin wires irradiated for α determination by the cadmium covered multimonitor method. The monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr was found to give a relatively higher and more reasonable value of α of -0.101 ± 0.019 . Also the value of α determined using only the three monitor combination ^{198}Au - ^{97}Zr - ^{95}Zr was found to be -0.106 ± 0.014 and is comparable with the α value for the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr . The negative values of α in both determinations indicate a hardened epithermal neutron spectrum in the cadmium lined irradiation channel. They are comparable with the value of -0.137 ± 0.018 previously obtained. The values of the epithermal neutron flux(Φ_e) and comparator factor ($F_{c,\text{Au}}$) of $(4.80\pm 0.04)\times 10^9$ and $(1.38\pm 0.01)\times 10^3$ respectively using the activity of ^{198}Au for the monitor combination ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr are comparable with the Φ_e and $F_{c,\text{Au}}$ values of $(4.76\pm 0.04)\times 10^9$ and $(1.37\pm 0.05)\times 10^3$ respectively for only the three monitors ^{198}Au - ^{97}Zr - ^{95}Zr . The α , Φ_e and $F_{c,\text{Au}}$ values can be well determined in the cadmium lined irradiation channel of the NIRR-1 using only the Au+Mo+Zr or Au+Zr monitor combinations.

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1. 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)[10]. 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 [14].

The single comparator method of epithermal neutron activation analysis(k_0 -ENAA) is useful for the determination of the concentrations of elements with high resonance integrals which strongly absorb neutrons of specific resonance energies in the epithermal neutron energy region(0.5 eV-1 MeV) in many types of samples of materials in a reactor irradiation site. The determination of the epithermal neutron shape factor (α) is important for the correction of the resonance integrals to thermal neutron cross section ratios of nuclides in the $\frac{1}{E^{1+\alpha}}$ epithermal neutron flux distribution of

the irradiation channel to ensure the accuracy of the method.

The determination of the α parameter in a reactor irradiation site is based on the types of and response of the selected activation nuclei (or set of monitors) having resonance peaks for neutron capture at different neutron energies in the epithermal neutron region [7].

Therefore, the α value in a particular irradiation channel may depend not only on the epithermal neutron flux

distribution, irradiation and counting conditions but also on the set of monitors selected for α determination. Thus, there is a need for the use of various or different combinations of monitor sets that provide the most accurate determination of the neutron flux parameters and consequently the most accurate k_0 -ENAA results in a reactor irradiation site.

After the installation of Cadmium lined irradiation channel in the NIRR-1, the value of the epithermal neutron shape factor (α) was determined by the cadmium covered multimonitor method using the monitors Al-0.1% Au, Zr and Zn thin foils and Mo thin wire and was found to be -0.137 ± 0.018 . [14]. The epithermal neutron flux was found to be $(4.51\pm 0.09)\times 10^9 \text{ n.cm}^{-2}.\text{s}^{-1}$. The value of α determined was successfully applied for elemental analysis of some legume samples by the k_0 -ENAA method using Al-0.1% Au thin foil as the single comparator [15]. The aim of this study was to re-evaluate the α value in the Cd lined irradiation channel for different combinations of the monitors in the monitor set Al-0.1% Au thin foil, Zr and Zn thin foils, Mo and Co thin wires irradiated for α determination by the cadmium covered multimonitor method. The combination of monitors to be chosen for α determination will be such that they fulfill the condition that their effective resonance energies $E_{r,i}$ have

a uniform distribution ranging from low to high over the whole epithermal neutron energy region [3].

In the cadmium covered multimonitor method, a set of N suitable α 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(- α) of the straight line by plotting the graph of [3]

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

$$\log \frac{E_{r,i} \cdot (A_{sp,i})_{Cd}}{k_{o,Au(i)} F_{Cd,i} Q_o(\alpha)_i G_{e,i} \varepsilon_{d,i}}$$

where

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

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

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

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

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

$\varepsilon_\gamma = \gamma$ – 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

$$T_i = \frac{E_{r,i} \cdot (A_{sp,i})_{Cd}}{k_{o,Au(i)} F_{Cd,i} Q_o(\alpha)_i G_{e,i} \varepsilon_{d,i}}$$

$k_{o,Au(i)}=k_o$ -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.

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

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 [13].

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.

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	$k_{o,Au}$ facors	F_{Cd}
$^{197}Au(n,\gamma)^{198}Au$ ($G_e=1$)	2.695d	411.8	5.65	15.7	1	0.991
$^{96}Zr(n,\gamma)^{97}Zr$ ($G_e=0.973$)	16.74h	743.4	338	251.6	1.24×10^{-5}	1
$^{94}Zr(n,\gamma)^{95}Zr$ ($G_e=0.983$)	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

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 Annalyser (MCA) emulation software card coupled to the detector via electronic nuclear Instrumentation modules manufactured by Ortec. The nuclear data of the product nuclides ^{198}Au , ^{97}Zr , ^{95}Zr , ^{99}Mo , ^{65}Zn and ^{60}Co are shown in Table 2.

Table 2 shows the nuclear data for the product nuclides 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], [5]. 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 [13]. The specific activities $Asp(Cd)$ for each of the product nuclides of the monitors were also calculated by the authors. In Table2, the two energies of the ^{95}Zr nuclide have been summed(724.24 keV + 756.7 keV). This nearly doubles theuseful 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 [16]. The specific activity of the ^{95}Zr nuclidewas calculated using the sum of the activities of the 724.2 keV and 756.7 keV γ -lines.

From Table 2, the product nuclides considered for the determination of α by cadmium covered multimonitor method were the four monitor combinations : ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr , ^{198}Au - ^{97}Zr - ^{95}Zr - ^{65}Zn , ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr - ^{99}Mo . The α value for each of the four combinations of monitors was determined 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 respective α values of -0.095 ± 0.011 , -0.152 ± 0.006 , -0.181 ± 0.007 and -0.189 ± 0.007 . After a four step iteration, the final values of α for the ^{198}Au - ^{99}Mo - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{97}Zr - ^{95}Zr - ^{65}Zn monitor combinations were found to be -0.101 ± 0.019 and -0.167 ± 0.015 respectively. After a three step iteration, the final values of α for the ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr and ^{198}Au - ^{60}Co - ^{97}Zr - ^{95}Zr - ^{99}Mo monitor combinations were found to be -0.188 ± 0.016 and -0.195 ± 0.015 respectively.

From the determined α values obtained for each of the four monitor combinations, 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 from the equation

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

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 factor $F_{c,Au}$ using the activity of ^{198}Au in each of the four combinations of monitors was calculated from the equation [4]

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

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

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

The determined values of epithermal neutron shape factor(α), the epithermal neutron fluxes and comparator factors using the four combinations of the monitors 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 factor(α) and the epithermal neutron flux determined in the Cadmium lined irradiation channel of the NIRR-1. As can be seen, the values of α for the four combinations of monitors are negative.

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- ⁶⁵ Zn	-0.167±0.015	(4.28±0.05)×10 ⁹	(1.23±0.01)×10 ³
¹⁹⁸ Au- ⁶⁰ Co- ⁹⁷ Zr- ⁹⁵ Zr	-0.188±0.016	(4.12±0.06)×10 ⁹	(1.19±0.02)×10 ³
¹⁹⁸ Au- ⁶⁰ Co- ⁹⁷ Zr- ⁹⁵ Zr- ⁹⁹ Mo	-0.195±0.015	(4.07±0.06)×10 ⁹	(1.17±0.02)×10 ³

The negative values of α indicate a hardened (poorly thermalized) epithermal neutron spectrum in the Cd lined irradiation channel of the NIRR-1.

From Table 3, it can be observed that the neutron spectrum parameters α , Φ_e and $F_{c,Au}$ of the four different monitor combinations are considerably different. This may be due to differences in nuclear data due to the presence of both isotopes with high and low Q_0 nuclides in the different combinations of the monitors. It can be observed that where the number of low Q_0 isotopes in a monitor combination are more than one, the α value for the monitor combination becomes more negative or lower. This suggests that the number of low Q_0 isotopes have influence on the α values of the four monitor combinations. From Table 3, the α value (as well as the epithermal neutron flux and comparator factor) for the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr with a suitable spread on their effective resonance energies (5.65 eV, 241 eV, 338 eV and 6260 eV) is relatively more reasonable and higher than the values for the other three monitor combinations. This suggests that a relatively more reliable value of α can be obtained in the cadmium lined irradiation channel of the NIRR-1 with a monitor combination that consists of three isotopes with high Q_0 values such as ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr and a single isotope such as ⁹⁵Zr with a relatively low Q_0 value (5.31) and a very large effective resonance energy (6260 keV).

The α value was also determined using only the three monitor combination ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr with a suitable spread on their \bar{E}_r values (5.65 eV, 338 eV and 6260 eV). The α value for the three monitor combination ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr was determined by solving Eq.1 by the iterative linear regression method based on MS Excel spread sheet. After a four step iteration, the α value was found to be -0.106±0.014. The result of α for only the three monitors ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr are comparable with the value of α for the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr. The negative values of α using the ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr and ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr monitor combinations indicate a hardened (poorly thermalized) epithermal neutron spectrum in the cadmium lined irradiation channel of the NIRR-1. They are comparable with the α value of -0.137±0.018 previously obtained [14]. The values of the epithermal neutron flux and comparator factor using the activity of ¹⁹⁸Au in the three isotope combination ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr were found to be (4.76 ±0.04)×10⁹ and (1.37±0.05)×10³ respectively. These values are comparable with the values of the epithermal neutron flux and comparator factor of (4.80±0.04)×10⁹ and (1.38±0.01)×10³ respectively for the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr.

Further more, 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 and the triple monitor ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr compare well 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 [12]. 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 [12]. 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.

Under the specific irradiation and counting conditions employed in this work, the monitor combinations Au+Mo+Zr or Au+Zr can provide more reliable values of α by the cadmium covered multimonitor and cadmium covered triple methods respectively as well as the epithermal neutron fluxes and comparator factors in cadmium lined irradiation channel of the NIRR-1.

4. Conclusions

The epithermal neutron shape factor (α) was re-evaluated in the permanent cadmium lined irradiation channel of the Nigeria Research Reactor-1 (NIRR-1) using the four monitor combinations ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr, ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr-⁶⁵Zn, ¹⁹⁸Au-⁶⁰Co-⁹⁷Zr-⁹⁵Zr and ¹⁹⁸Au-⁶⁰Co-⁹⁷Zr-⁹⁵Zr-⁹⁹Mo in the monitor set Al-0.1% Au thin foil, Zr and Zn foils, Mo and Co thin wires irradiated for α determination by the cadmium covered multimonitor method. The monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr with suitable spread on their \bar{E}_r values (5.65 eV, 241 eV, 338 eV and 6260 eV) was found to give a more reliable value for α of -0.101±0.019. Also the value of α determined using only three monitor combination ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr with suitable spread on their \bar{E}_r values (5.65 eV, 338 eV and 6260 eV) was found to be -0.106±0.014 and is comparable with the α value for the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr. The negative values of α in both determinations indicate a hardened epithermal neutron spectrum in the cadmium lined irradiation channel. They are comparable with the value of -0.137±0.018 previously obtained [14]. The values of the epithermal neutron flux and comparator factor using the activity ¹⁹⁸Au in the monitor combination ¹⁹⁸Au-⁹⁹Mo-⁹⁷Zr-⁹⁵Zr were found to be (4.80±0.04)×10⁹ and (1.38±0.01)×10³ respectively. These values are comparable with the epithermal neutron flux and comparator values of (4.76 ±0.04)×10⁹ and (1.37±0.05)×10³ respectively for the three monitors ¹⁹⁸Au-⁹⁷Zr-⁹⁵Zr.

The values of the epithermal neutron shape (α), epithermal neutron flux and comparator factor can be well determined in the cadmium lined irradiation channel of the NIRR-1 using only the Au+Mo+Zr or Au+Zr monitor combinations.

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.

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