Uncertainty and Detection Limit of Analysis Results in k_o-ENAA

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ABSTRACT

The accuracy of the α determination in the cadmium lined irradiation channel of the NIRR-1 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 in previous studies was tested by elemental analysis of the standard reference material NIST 1515 Apple leaves by the k_o-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 combined uncertainties and detection limits of the concentrations of Sm are relatively lower than those for Br in the NIST 1515 Apple leaves. This indicates that the cadmium lined irradiation channel of the NIRR-1 with the cadmium lining of thickness 1.00 mm and a low cut off energy of 0.55 eV may be preferable for the measurement of elements such as Sm and other elements with low resonance energies (less than 10 eV) in samples of materials. It is necessary to use moveable boron vials with a cut off energy of range up to 280 eV depending on thickness for epithermal activation of samples of materials in the cadmium lined irradiation channel of the NIRR-1 to improve the combined uncertainties and the detection limits of the trace elements such as Br and other elements with high resonance energies of more than 10 eV in the epithermal neutron region by the k_o-ENAA method.

Keywords— α values, cadmium lined irradiation channel, k_o-ENAA, uncertainty propagation and calculation, detection limit.

1. INTRODUCTION

The 30 kW Nigeria Research Reactor-1(NIRR-1) is one of the commercial miniature source research reactors(MNSRs) installed outside China. It was commissioned in 2004 and has been standardized for instrumental neutron activation analysis(INAA)[6]. In order to extend its utilization for the epithermal neutron activation analysis(ENAA) protocol, the cadmium(Cd) lined irradiation channel was installed in one of the large outer irradiation channels[9].

In the single comparator method of epithermal neutron activation analysis(k_o-ENAA), the samples of materials and the element used as the single comparator (usually Au) are co-rradiated with epithermal neutrons under a thermal neutron filter such as cadmium with a very large thermal neutron cross section of 1.991×10^4 barns. Consequently, the background interferences due to the high activities of the major and minor elements with low resonance integral to thermal neutron cross section ratios(Q_o) in the samples of materials such as Na, Cl, Al, Fe, Mn, Cr, Sc, Co and La in geological materials and Na, Cl, K, P, Zn, Fe, Mn and Cu in biological materials are greatly reduced. This lowers significantly the combined uncertainties and the detection limits of the trace elements of interest measured in the samples of materials such as Ag, As, I, Au, Ir, Sn, Cd, Mo, Se, Sb, Sm, In and Rb with high resonance integrals and with specific resonance energies in the epithermal neutron region(0.5 eV-1 MeV)[17]. The interfering background on the photopeaks of the elements of interest will therefore be due mainly to the Compton background below the analytical peaks of interest caused by the interaction of the high energy gamma-rays of the epithermal neutron induced radionuclides with high resonance integrals in the high pure germanium (HPGe) detector.

The combined uncertainty in the concentrations of elements in the samples of materials by the k$_o$-ENAA method are contributed mainly by random and systematic errors[2]. Random errors are mainly due to counting statistics of the activities of the nuclides of the elements measured in the sample of material and of the nuclide of the element used as the single comparator. Random errors influence the precision of the k$_o$-ENAA method. Systematic errors are contributed mainly by the uncertainties associated with the nuclear data of a number of parameters such as the k$_o$-factors, the resonance integral to thermal neutron cross section ratio (Q$_o$) and the effective resonance energy ($\bar{E}_r$) for each of the nuclides of elements in the sample of material and for the nuclide of the element used as the single comparator, the epithermal neutron shape factor(α) in the reactor irradiation site, the detector efficiency (ε_d) for the nuclides of elements of interest in the sample of material and the nuclide of the element used as the single comparator as well as the true coincidence corrections of the nuclides measured. Systematic errors influence the accuracy of the k$_o$-ENAA method.

In general, if a quantity C with several variables $x_1, x_2, ..., x_n$ and with small independent uncertainties $\delta x_1, \delta x_2, ..., \delta x_n$ respectively is given as

$$C = x_1.x_2.......... .x_n \qquad (1)$$

Then the combined or total uncertainty δC in the quantity C can be obtained from the uncertainty propagation equation [14]

$$\left(\left(\frac{d\log C}{dC}\right).\delta C\right)^2 = \left(\left(\frac{d\log x_1}{dx_1}\right).\delta x_1\right)^2 + \left(\left(\frac{d\log x_2}{x_2}\right).\delta x_2\right)^2 + \ldots\ldots + \left(\left(\frac{d\log x_n}{dx_n}\right).\delta x_n\right)^2 \qquad (2)$$

which leads to the uncertainty propagation equation

$$\left(\frac{\delta C}{C}\right)^2 = \left(\frac{\delta x_1}{x_1}\right)^2 + \left(\frac{\delta x_2}{x_2}\right)^2 + \ldots\ldots + \left(\frac{\delta x_n}{x_n}\right)^2 \qquad (3)$$

where $\delta C =$ the combined uncertainty in the quantity

$C =$ the quantity

$\delta x_1, \delta x_2, \ldots\ldots, \delta x_n =$ small independent uncertainties

$x_1, x_2, ..., x_n =$ several parameters in the quantity C

The limit of detection is the minimum or lowest detectable concentration of an element of interest in a sample of material in a neutron activation analysis(NAA) procedure.In the k$_o$-ENAA method, the detection limits of elements of interest in a sample of material depend on the background interference resulting from the high Compton continuum below the analytical peak of interest in the pulse height spectrum caused by the interaction of the high energy gamma-rays of the epithermal neutron induced radionuclides with high resonance integrals in the high pure germanium (HPGe) detector. They are also much dependent on the nuclear data of the elements of interest such as the k$_o$-factors, the Q$_o$ and $\bar{E}_r$ values of each of the nuclides of the elements in the sample of material and of the nuclide of the element used as the single comparator, They also depend on the experimental conditions such as the composition of the sample of material, the epithermal neutron shape factor(α) and epithermal neutron flux in the reactor irradiation site, the irradiation, decay and counting conditions, the efficiency of the HPGe detector and the cut off energy of cadmium thermal neutron filter.

The detection limit of the element of interest in a sample of material by the k$_o$-ENAA method can be calculated using Currie's formula as [19]

$$L_{De} = \frac{(2.71 + 4.65.\sqrt{N_{be}}).C_e}{N_e} \qquad (4)$$

where $N_{be} =$ the Compton continuum background under the γ-ray peak of elements of interest in samples(counts)

$N_e =$ net photopeak counts of elements of interest in samples(in counts)

$C_e =$ the concentrations of the elements of interest in samples(in ppm)

$L_{De} =$ lower limit of detection of elements of interest in samples(in ppm)

In previous studies, following the re-assessment of the epihermal neutron shape factor(α) in the cadmium lined irradiation channel of the NIRR-1, the α values obtained with the monitor combinations ^{198}Au-^{99}Mo-^{97}Zr-^{95}Zr, by the cadmium covered multimonitor method and the monitor combinations ^{198}Au-^{97}Zr-^{95}Zr and ^{198}Au-^{99}Mo-^{95}Zr by the cadmium covered triple method were found to have relatively high and reasonable values of -0. 101±0.019, -0.106±0.014 and -0.114±0.017 respectively[10]-[11]. The accuracy of each of the α values was tested by elemental analysis of the standard reference material (SRM) NIST 1515 Apple leaves by the k_o-ENAA method using the Al-0.1%Au thin foil as the single comparator[11] The concentrations of the elements Sm and Br with high resonance integrals determined in the NIST 1515 Apple leaves were found to be in good agreement with the certified values[11].

In this study, the method of the uncertainty propagation and calculation of the combined uncertainties of the concentrations of the elements Sm and Br determined in the NST 1515 Apple leaves by the k_o-ENAA based on the MS Excel spread sheet is fully presented. The detection limits of the concentrations of the elements Sm and Br determined in the NIST 1515 Apple leaves based on the irradiation and counting conditions of the NIST 1515 Apple leaves activated with epithermal neutrons in the cadmium lined irradiation channel of the NIRR-1 are also presented.

2. MATERIALS AND METHODS

In this work, the calculation of the combined uncertainties and detection limits of the elements of interest in the analysis results in k_o-ENAA was based on the results of the elemental analysis of the standard reference material(SRM) NIST 1515 Apple leaves by the k_o-ENAA method using the Al-0.1%Au thin foil as the single comparator and also using the α values obtained in the cadmium lined irradiation channel of the NIRR-1 in previous studies 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 [10]-[11].The standard reference material 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 $5x10^{11}$ $n.cm^{-2}.s^{-1}$ for 6 hours. The induced activities on the elements of interest in the NIST 1515 Apple leaves and the Al-0.1%Au thin foil were measured under the same counting conditions at a distance of 2 cm from the efficiency calibrated P-type GEM 30195 HPGe coaxial detector system after appropriate decay and counting periods. The energy resolution of the system is 1.95 keV for the 1332 keV peak of ^{60}Co 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 distance of 2 cm from the detector over the energy range of 59.54-1408 keV using the set of IAEA standard sources ^{241}Am, ^{152}Eu, ^{226}Ra, ^{137}Cs, ^{60}Co and ^{22}Na. A detailed description of the measured full energy peak efficiency curves and the theoretical fitting function to the experimental efficiency curves is given elsewhere[8].

The concentrations of the elements determined in the NIST 1515 Apple leaves were calculated by the k_o-ENAA method using the activity of ^{198}Au as the single comparator by the Hogdhal convention with the Microsoft Excel spread sheet from the expression[3]

$$C_{s(Cd)} = \frac{\left(\dfrac{N_{p,s}/t_c}{SDCW}\right)_s}{\left(\dfrac{N_{p,Au}/t_c}{SDCw_{Au}}\right)_{Au}} . \frac{F_{Cd,Au}.Q_o(\alpha)_{Au}.\varepsilon_{d,Au}}{k_{o(Au).F_{Cd,s}.Q_o(\alpha)_s}.\varepsilon_{d,s}} \qquad (5)$$

where the $k_{o(Au)} -$ factor is a compound nuclear constant of the nuclide in the sample of material with respect to Au which is independent of the irradiation and counting conditions and is defined theoretically in the k_o-ENAA method as [12]

$$k_{o(Au)} = \frac{M_{Au}\theta_s \varepsilon_{\gamma,s}\sigma_{o,s}}{M_s \theta_{Au}\varepsilon_{\gamma,Au}\sigma_{o,Au}} \qquad (6)$$

M_{Au} = atomic mass of gold(Au)

M_s = atomic mass of element of interest in the NIST 1515 Apple leaves

θ_s = isotopic abundance of the element of interest in the NIST 1515 Apple leaves

θ_{Au} = isotopic abundance of gold(Au)

$\varepsilon_{\gamma,s} = \gamma -$ray intensity of the element of interest in the NIST 1515 Apple leaves

$\varepsilon_{\gamma,Au} = \gamma -$ray intensity of ^{198}Au

$\sigma_{o,s}$ = thermal neutron cross section of elements of interest in the NIST 1515 Apple leaves

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

$F_{Cd,s}$ = the cadmium epithermal neutron transmission factor for elements of interest in the NIST 1515 Apple leaves

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

$Q_o(\alpha)_{Au}$ = corrected resonance integral to thermal neutron cross section ratio for ^{198}Au in the cadmium lined irradiation channel

$Q_o(\alpha)_s$ = corrected resonance integral to thermal neutron cross section ratios for nuclides of elements of interest in the NIST 1515 Apple leaves in the cadmium lined irradiation channel

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

$\varepsilon_{d,s}$ = detector efficiency for the $\gamma-$ray energies of the nuclides of elements of interest in the NIST 1515 Apple leaves

$\varepsilon_{d,Au}$ = detector efficiency for the $\gamma-$ray energy of ^{198}Au

W_s = total amount of sample of NIST 1515 Apple leaves irradiated in the cadmium lined irradiation channel

w_{Au} = weight of the sample of Al-0.1%Au thin foil irradiated in the cadmium lined irradiation channel

C_s = concentrations of the elements in the NIST 1515 Apple leaves irradiated in the cadmium lined irradiation channel

$N_{p,s}$ = net photopeak area of nuclide of element of interest in the NIST 1515 Apple leaves

$N_{p,Au}$ = net photopeak area of ^{198}Au

$D = e^{-\lambda t_d}$ = decay factor of ^{198}Au or nuclides of elements of interest in the NIST 1515 Apple leaves

$S = (1 - e^{-\lambda t_i})$ = saturation factor for ^{198}Au or nuclides of elements of interest in NIST 1515 Apple leaves

$C = \frac{\left(1 - e^{-\lambda t_c}\right)}{\lambda t_c}$ = correction factor for decay during counting for ^{198}Au or nuclides of elements of interest in NIST 1515 Apple leaves

λ = decay constant, t_i = irradiation time,

t_d = decay time, t_c = counting time

The corrected resonance integral to thermal neutron cross section ratio of an isotope in the non-ideal $\frac{1}{E^{1+\alpha}}$ epithermal neutron energy region is given as [2]

$$Q_o(\alpha) = \frac{Q_o - 0.429}{\bar{E}_r^{\,\alpha}} + \frac{0.429}{(2\alpha + 1)E_{Cd}^{\alpha}} \qquad (7)$$

where $\bar{E}_r$ = effective resonance energy

E_{Cd} = cadmium cut of energy (0.55 eV)

α = value of epithermal neutron shape factor in reactor irradiation channel

$Q_o(\alpha)$ = resonance integral to thermal neutron cross section ratio of nuclides in the non-ideal $\frac{1}{E^{1+\alpha}}$ epithermal neutron spectrum

Q_o = resonance integral to thermal neutron cross section ratio of nuclide in the ideal $\frac{1}{E}$ epithermal neutron spectrum

The nuclear data for the k_o-factors, Q_o, $\bar{E}_r$ and F_{Cd} values for the nuclide ^{198}Au used as the single comparator and of the nuclides of the elements of interest determined in the NIST 1515 Apple leaves are shown in Table 1.

Table 1 Nuclear data for ^{198}Au and nuclides of elements of interest in NIST 1515 Apple leaves

Nuclide	E_r (eV)	$\bar{E}_r$ (eV)	k_o	Q_o	F_{Cd}
^{198}Au	411.8	5.65±0.4	1	15.7±0.28	0.991
^{153}Sm	103.2	8.53±0.09	$2.31\text{x}10^{-1}$ $\pm 9.24\text{x}10^{-4}$	14.4±0.3	1
^{82}Br	776.5	152±13.98	$2.76\text{x}10^{-2}$ $\pm 2.21\text{x}10^{-4}$	19.3±0.58	1

Table 1 shows the nuclear data of the nuclide ^{198}Au used as the single comparator and of the nuclides determined in the NIST 1515 Apple leaves. The nuclear data for the k_o-factors, Q_o and $\bar{E}_r$ values and the associated uncertainties of the ^{198}Au and the nuclides determined in the NIST 1515 Apple leaves were obtained from the literature[4]-[5]. The net photopeak areas and associated standard uncertainties of the nuclide ^{198}Au and of the nuclides determined in the NIST 1515

Apple leaves were measured experimentally with the gamma-ray acquisition system of the P-type GEM 30195 HPGe coaxial detector system that consists of the Maestro Multichannel Analyzer (MCA) emulation software card coupled to the detector via electronic nuclear instrumentation modules manufactured by Ortec. The efficiencies of the gamma-ray energy of the nuclide ^{198}Au used as the single comparator and of the nuclides determined in the NIST 1515 Apple leaves were calculated by the author using mathematical fitting functions described elsewhere[8]. The corrected resonance integrals to thermal neutron cross sections ratios $Q_o(\alpha)$ of ^{198}Au and of the nuclides determined in the NIST 1515 Apple leaves were calculated by the author using Eq. 7. The concentrations of the elements determined in the NIST 1515 Apple leaves by the k_o-ENAA method using Eq. 5 are shown in Table 2.

2.1 Calculation of the combined uncertainty

In Eq. 5, all the time variables such as irradiation, decay and counting time and half life are assumed to have negligible contributions to the combined uncertainties of the concentrations of elements in the NIST 1515 Apple leaves. The uncertainty components of S, D and C are also assumed to have negligible contributions to the combined uncertainties of the concentrations of elements in the NIST 1515 Apple leaves.

The main contributions to the combined uncertainties in the analysis results in k_o-ENAA are statistical counting errors and the estimated systematic uncertainties. The statistical counting errors are contributed by the standard errors of the net photopeak areas of the nuclide ^{198}Au used as the single comparator and of the nuclides of the element determined in the NIST 1515 Apple leaves. The true coincidence corrections were not carried out in this work. Therefore, the systematic uncertainties are due mainly to the nuclear data of the parameters k_o-factors, the Q_o and $\bar{E}_r$ values, the detection efficiency(ε_d) of the gamma-ray energy of the nuclide ^{198}Au used as the single comparator and of each of the nuclides determined in the NIST 1515 Apple leaves. Making use of Eqs. 2 and 3, the combined uncertainties of the concentrations of elements determined in the NIST 1515 Apple leaves by the k_o-ENAA method were calculated from Eq. 5 as the square root of the summation of the statistical counting errors and the estimated systematic uncertainties of the nuclide ^{198}Au and of the nuclides of the elements determined in the NIST 1515 Apple leaves with the MS Excel spread sheet using the uncertainty propagation equation [3], [13]-[14]

$$\frac{\sigma^2_{C_e}}{C_e^2} = \frac{\sigma^2_{N_{p,Au}}}{N^2_{p,Au}} + \frac{\sigma^2_{N_{p,s}}}{N^2_{p,s}} + \frac{\sigma^2_{\varepsilon,Au}}{\varepsilon^2_{Au}} + \frac{\sigma^2_{\varepsilon,s}}{\varepsilon^2_{s}} + \frac{\sigma^2_{k_{o-Au(s)}}}{k^2_{o,s(Au)}} + \frac{\sigma^2_{Q_o(\alpha),Au}}{Q_o^2(\alpha)_{Au}} + \frac{\sigma^2_{Q_o(\alpha),s}}{Q_o^2(\alpha)_{s}} \qquad (8)$$

where C_e = concentrations of elements in NIST 1515 Apple leaves

$N_{p,Au}$ = measured net photopeak area of 411.8 keV energy of ^{198}Au

$\sigma_{N_{p,Au}}$ = standard deviation in measure net peak area of 411.8 keV energy of ^{198}Au

$N_{p,s}$ = net photopeak areas of γ-ray energies of nuclides of elements of interest in NIST 1515 Apple leaves

$\sigma_{N_{p,s}}$ = standard deviation in net peak areas of γ-ray energies of nuclides of elements of interest in NIST 1515 Apple leaves

ε_{Au} = efficiency of 411.8 keV energy of ^{198}Au

$\sigma_{\varepsilon,Au}$ = standard deviation of the efficiency of 411.8 keV energy of ^{198}Au

ε_s = efficiency of γ-ray energy of nuclides of elements of interest in NIST 1515 Apple leaves

$\sigma_{\varepsilon,s}$ = standard deviation of the efficiency of γ-ray energies of nuclides of elements of interest n NIST 1515 Apple leaves

$Q_o(\alpha)_{Au}$ = corrected resonance to thermal neutron cross section ratio for ^{198}Au

$\sigma_{Q_o(\alpha)_{Au}}$ = uncertainties of corrected resonance integral to thermal neutron cross section ratio for Au

$Q_o(\alpha)_s$ = corrected resonance to thermal neutron cross section ratio for nuclides of elements of interest in NIST 1515 Apple leaves

$\sigma_{Q_o(\alpha)_s}$ = standard deviation of corrected resonance integral to thermal neutron cross section ratio for nuclides of elements of interest in NIST 1515 Apple leaves

$k_{o(Au)}$ = k_o-factors for Au

$k_{o,s(Au)}$ = k_o-factors for elements of interest in NIST 1515 Apple leaves

$\sigma_{k_{o,s(Au)}}$ = uncertainties of k_o-factors for elements of interest in NIST 1515 Apple leaves

It was not possible to calculate the uncertainties of the efficiency values of the γ-ray of ^{198}Au and of the nuclides of the elements determined in the NIST 1515 Apple leaves calculated with the mathematical fitting function. Therefore, the uncertainties in the efficiencies of the γ-ray energy of ^{198}Au and of the nuclides of elements determined in the NIST 1515 Apple leaves were calculated from the expression [1]

$$\frac{\sigma_\varepsilon^2}{\varepsilon^2} = \frac{\sigma_{N_p}^2}{N_p^2} \qquad (9)$$

where ε = efficiency of γ-ray energies of nuclides

σ_ε = uncertainty in efficiency of γ-ray energies of nuclides

N_p = net peak areas of nuclides

σ_{N_p} = standard deviation of net peak areas of nuclides

In the k_o-ENAA method, the concentrations of elements of interest in the samples of materials irradiated with epithermal neutrons at a reactor irradiation site are those with high resonance integrals and which strongly absorb neutrons of specific resonance energies in the epithermal neutron energy region (0.5 eV- 1 MeV). When the resonance integrals of the nuclides are much higher than the thermal neutron cross sections ($I_o >> \sigma_o$), that is for nuclides with high Q_o values, Eq. 7 used for the calculation of the $Q_o(\alpha)$ values of ^{198}Au and of the nuclides determined in the NIST 1515 Apple leaves can be approximated by the equation [2]

$$Q_o(\alpha) = Q_o(\bar{E}_r)^{-\alpha} \qquad I_o >> \sigma_o \qquad (10)$$

where $Q_o(\alpha)$ = resonance integral to thermal neutron cross section ratio of nuclides in the non-ideal $\frac{1}{E^{1+\alpha}}$ epithermal neutron spectrum

Q_o = resonance integral to thermal neutron cross section ratio of nuclides in the ideal $\frac{1}{E}$ epithermal neutron spectrum

α = value of epithermal neutron shape factor in reactor irradiation channel

$\bar{E}_r$ = effective resonance energy

The Q_o(α) values of ^{198}Au used as the single comparator and of the nuclides ^{153}Sm and ^{82}Br determined in the NIST 1515 Apple leaves calculated from Eq. 7 and Eq. 10 using the nuclear data of nuclides in Table 1 and the α values obtained in previous studies 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 [10]-[11] are shown in Table 2.

Making use of Eqs. 2 and 3, the uncertainties in the corrected resonance integral to thermal neutron cross section ratio of ^{198}Au and of the nuclides ^{153}Sm and ^{82}Br determined in the NIST 1515 Apple leaves were calculated from Eq. 10 using the uncertainty propagation equation [14]

$$\frac{\sigma^2_{Q_o(\alpha)}}{Q_o^2(\alpha)} = \frac{\sigma^2_{Q_o}}{Q_o^2} + \left|-\alpha\right|^2 . \frac{\sigma^2_{\bar{E}_r}}{\bar{E}_r^{\,2}} \qquad (11)$$

where

$Q_o(\alpha)$ = resonance integral to thermal neutron cross section ratio of ^{198}Au or nuclides of elements of interest in NIST 1515 Apple leaves in Cd lined channel

$\sigma_{Q_o(\alpha)}$ = uncertainties of corrected resonance integral to thermal neutron cross section ratio of ^{198}Au or nuclides of elements of interest in the NIST 1515 Apple leaves in the Cd lined channel

Q_o = resonance integral to thermal neutron cross section ratio of ^{198}Au or nuclides of elements of interest in NIST 1515 Apple leaves in the ideal $\frac{1}{E}$ epithermal neutron spectrum

α = epithermal neutron shape factor determined in the Cd lined channel using each of the monitor combinations

$\bar{E}_r$ = effective resonance energy of ^{198}Au or of nuclides of elements of interest determined in the NIST 1515 Apple leaves

$\sigma_{\bar{E}_r}$ = standard deviation of effective resonance energy of ^{198}Au or nuclides of elements of interest in the NIST 1515 Apple leaves

The uncertainties in the $Q_o(\alpha)$ values of the nuclides ^{153}Sm and ^{82}Br determined in the NIST 1515 Apple leaves and of ^{198}Au used as the single comparator calculated using Eq. 11 are shown in Table 2. The results of the concentrations of the elements determined in the NIST 1515 Apple leaves by the k_o-ENAA method and the calculated combined uncertainties are shown in Table 3.

2.2 Calculation of the detection limits

The determination of the detection limits of the concentrations of the elements Sm and Br in the NIST 1515 Apple leaves was based on the irradiation, decay and counting conditions of the NIST 1515 Apple leaves irradiated in the cadmium lined irradiation channel of the NIRR-1 as mentioned earlier. The detection limits of the concentrations of the elements Sm and Br determined in the NIST 1515 Apple leaves were calculated using Eq. 4. The results of the calculated detection limits of the concentrations of Sm and Br in the NIST 1515 Apple leaves in the cadmium lined irradiation channel of the NIRR-1 are shown in Table 3.

3.RESULTS AND DISCUSSIONS

Table 2, comparison of $Q_o(\alpha)$ values calculated from Eq. 7 and Eq. 10

Nuclide	^{198}Au-^{99}Mo-^{97}Zr-^{95}Zr monitors α=-0.101±0.019		^{198}Au-^{97}Zr-^{95}Zr monitors α= -0.106±0.014		^{198}Au-^{99}Mo-^{95}Zr monitors α= -0.114±0.017	
	$Q^{o}(\alpha)$ from Eq. 7	$Q_o(\alpha)$ from Eq. 10	$Q_o(\alpha)$ from Eq. 7	$Q_o(\alpha)$ from Eq. 10	$Q_o(\alpha)$ from Eq. 7	$Q_o(\alpha)$ from Eq. 10
^{198}Au	18.70	18.70 ±0.36	18.86	18.86 ±0.36	19.12	19.13 ±0.37
^{153}Sm	17.85	17.88 ±0.37	18.05	18.07 ±0.38	18.36	18.39 ±0.38
^{82}Br	31.85	32.06 ±4.39.	32.65	32.87 ±4.51	33.08	34.22 ±4.69

Table 2 shows the $Q_o(\alpha)$ values and the calculated uncertainties for the nuclides ^{153}Sm and ^{82}Br determined in the NIST 1515 Apple leaves and of ^{198}Au used as the single comparator calculated from Eq. 7 and Eq. 10 using the nuclear data of the nuclides in Table 1 and the $Q_o(\alpha)$ values obtained 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. The results in Table 2 show that the $Q_o(\alpha)$ values calculated from the approximate formula in Eq. 10 are in good agreement with the $Q_o(\alpha)$ values calculated from Eq. 7. Therefore, in practice, Eq. 7 can be replaced with Eq. 10 for the calculation of the $Q_o(\alpha)$ values of the nuclides with high resonance integrals that are involved in the k_o-ENAA method. This enables the uncertainties in the $Q_o(\alpha)$ values of the nuclides with high resonance integrals involved in the k_o-ENAA method to be easily calculated.

Table 3 Results of concentrations and detection limits of elements in NIST 1515 Apple leaves by the k_o-ENAA method using α values of different monitor combinations

Element	Certified value mg.kg^{-1}	^{198}Au-^{99}Mo-^{97}Zr-^{95}Zr monitors		^{198}Au-^{97}Zr-^{95}Zr monitors		^{198}Au-^{99}Mo-^{95}Zr monitors	
		mg.kg^{-1}	L_{De}	mg.kg^{-1}	L_{De}	mg.kg^{-1}	L_{De}
Sm	3.0	3.15 ±0.14	0.20	3.14 ±0.14	0.20	3.13 ±0.14	0.20
Br	1.8	1.89 ±0.82	0.98	1.86 ±0.77	0.96	1.86 ±0.81	0.96

From Table 3, the elements Sm and Br were better determined in the NIST 1515 Apple leaves by the k_o-ENAA method because they have high resonance integrals compared to their thermal neutron cross sections and strongly absorb neutrons with specific resonance energies in the epithermal neutron region. The results of the concentrations of Sm and Br in the NIST 1515 Apple leaves by the k_o-ENAA method using Al-0.1%Au thin foil as the single comparator 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 NIRR-1 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 for application of the k_o-ENAA method in the cadmium lined irradiation channel of the NIRR-1.

From Table 3, it can be observed that the combined uncertainties in the concentrations of Sm ($\bar{E}_r$ =8.53 eV) are relatively lower than the combined uncertainties of the concentrations of Br($\bar{E}_r$ = 152 eV) in the NIST 1515 Apple leaves. Also the detection limits of the concentration of Sm are relatively lower than those for Br in the NIST 1515 Apple leaves. This suggests that the cadmium lined irradiation channel of the NIRR-1 with the cadmium lining of thickness of 1.00 mm and a cut off energy 0.55 eV may be preferable for the measurement of elements such as Sm and other elements with high resonance integrals but which have low resonance energies(less than 10 eV) in samples of materials[17]-[18]. For elements with high resonance integrals and high resonance energies(more than 10 eV), it may be 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. Boron with a very large thermal neutron cross section of 3.84x10^3 barns will provide additional filtering of thermal neutrons and the lower epithermal neutron region (1-10 eV) which may improve the combined uncertainty and the detection limits of the trace elements such as Br and other elements in samples of materials with high resonance integrals and high resonance energies above 10 eV in the epithermal neutron region [15], [16]-[18].

4. CONCLUSIONS

The accuracy of the α values obtained in the cadmium lined irradiation channel of the NIRR-1 using the monitor combinations ^{198}Au-^{99}Mo-^{97}Zr-^{95}Zr, ^{198}Au-^{97}Zr-^{95}Zr and ^{198}Au-^{99}Mo-^{95}Zr in previous studies was tested by elemental analysis of the standard reference material NIST 1515 Apple leaves by the k_o-ENAA method using the 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 combined uncertainties and detection limits of the concentrations of Sm and Br in the NIST 1515 Apple leaves by the k_o-ENAA method using the Al-0.1%Au thin foil as the single comparator were calculated with the MS Excel spread sheet. The relatively low combined uncertainties and detection limits of the concentrations of Sm compared to those of Br in the NIST 1515 Apple leaves suggests that the cadmium lined irradiation channel of the NIRR-1 with the cadmium lining of thickness 1.00 mm and a low cut off energy of 0.55 eV may be preferable for the measurement of elements with high resonance integrals but which have low resonance energies (less than 10 eV) such as Sm in samples of materials. [17]-[18]. To improve the combined uncertainties and the detection limits of the concentrations of elements such as Br and other elements with high resonance integrals and high resonance energies (more than 10 eV) in the epithermal neutron region, it may be 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 cadmium lined irradiation channel of the NIRR-1[15]-[16], [18].

ACKNOWLEDGMENT

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

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