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Research Article
Discrepancy in the k_0-values of ^{134}Cs, ^{152}Sm, ^{75}Se and Experimental Implementations in k_0 Standardization Techniques

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Abstract

Background and Objective: The k_0-users are faced with the challenge of the accurate the k_0-values of ^{134}Cs, ^{152}Sm and ^{75}Se produced by (n,γ) reaction are in k_0 standardization techniques. These values are used in determination of elemental concentrations via neutron activation analysis (NAA). The objective of this study was to evaluate the actual discrepancy of the k_0-values of ^{134}Cs, ^{152}Sm and ^{75}Se by using Certified reference materials (CRMs) for k_0-users. **Materials and Methods:** The CRMs SMELS I and III, NIST 1633b Coal fly Ash were used. These samples were irradiated using the Nigerian Research Reactor (NIRR-1) at half-full power (15 kW) with neutron flux of 5×10^{11} n/cm^{2}s for 7 h in the inner channel and evaluated following the established protocols. **Results:** The experimental and empirical calculated values of ^{134}Cs, ^{152}Sm and ^{75}Se were compared with recommended data. The experimental k_0 value of ^{152}Sm (103 keV) showed a large deviation while ^{134}Cs and ^{75}Se agreed with recommended values. **Conclusion:** The k_0 values of ^{134}Cs and ^{75}Se compared well with the recommended values while ^{152}Sm showed high deviation. The discrepancy using the empirical calculation was high for all the nuclides.

Key words: Neutron activation analysis, certified reference material, Nigerian research reactor, k_0-values, k_0 standardization

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The Nigeria Research Reactor-1 (NIRR-1) is a Miniature Neutron Source Reactor (MNSR) and a low power, which was designed to have one central control rod (CCR) that performs safety and regulatory functions and to serve as a neutron source[1]. The CCR are strong neutron absorbers that can be inserted or withdrawn from the reactor core. They are used to compensate for the excess reactivity necessary for long-term core operation and to adjust the power level of the reactor in order to bring the core to power, follow load demands and shutdown the reactor[2]. Jonah *et al.*[2,3] have standardized NIRR-1, via the relative and the k_0-standardization method for the use in Neutron Activation Analysis (NAA).

The implementation of the k_0-standardization concept in NAA involved the co-irradiation of the sample and a neutron fluence rate monitor to determine a composite nuclear constant[4,5], which eliminates the use of standards. When the composite nuclear constant is obtain accurately, these are used to evaluate the concentration of nuclides[6]. In the implementation of the k_0-standardization concept, the k-values, epithermal neutron flux shape factor (α) and sub-cadmium-to-epithermal neutron flux ratio (f) are important parameters used[7,8]. These parameters are dependent on each irradiation facility and must be determined for standardization and application purposes. In order to extend its utilization to include the k_0-standardization method, the following neutron spectrum parameters in inner and outer irradiation channels were determined by the Cd-ratio for multi-monitor method and published elsewhere[3]. The results were compared with the neutron spectrum parameters of other reactor facilities with similar core configuration such as the Slowpoke and Miniature Neutron Source Reactor facilities available in the literature of Kennedy *et al.*[4].

In the efforts to implement the k_0-standardization concept in NAA techniques in many laboratories worldwide, the problem of how accurate the k_0-values available in literature were in order to produce accurate results[9]. De Corte and Simonits[10] observed that some of the recommended k_0-values available in the Atomic and Nuclear Data Tables have discrepancies and Moens *et al.*[5], Simonits *et al.*[11] and De Corte and Simonits[10] revealed that the k_0-values in the Atlas of Neutron Resonances are inconsistent with the recommended data. In line with these observation, this study seek to determine the k_0-values of the following nuclides; ^{134}Cs, ^{152}Sm and ^{75}Se reported to be discrepant. The Synthetic Multi-element Standard (SMELS) recommended by the k_0-users for the validation of k_0-NAA method and the standard reference materials, NIST-1633b (Coal Fly Ash) were used[12,13].

The k_0 values of the discrepant nuclides; ^{134}Cs, ^{152}Sm and ^{75}Se have been obtained experimentally in this study using the NIRR-1 facilities. The experimental k_0 values results obtained in this study were in good agreement with the recommended values, except for ^{152}Sm whose deviation was 28%. Furthermore, there was a large deviation in the range of 18.29-39.53% from the results of this study compare to the empirical calculated values for all the nuclides and their respective peak energies. Hence, this study revealed the unreliability of the nuclear data and observed that the experimental determination of the k_0 values is necessary in order to use the data in k_0-standardization method.

MATERIALS AND METHODS

The Synthetic Multi-elemental Standards (SMELS) Type I and III was weighed and an amount was measured based on a phenol formaldehyde resin (Bakelite). The measured samples were spiked with 30 different materials as well as the NIST 1633b (Coal Fly Ash). The samples were wrapped in polyethylene films that have been cleaned appropriately and prepared for irradiation. The irradiation were performed using NIRR-1 operating at half-full power of about 15 kW with neutron flux 5×10^{11} n/cm^2 sec was performed in 2017[12,2].

The samples were irradiated for 7 h in the inner irradiation channel B-2. For the determination of ^{152}Sm, the samples were left for two days after irradiation before counting. Each of the samples were counted for 30 min using GEM-30195 HPGe Coaxial, vertical dipstick detector (ORTEC). This similar procedures is applied in the determination of the following elements; Na, K, As, Zn, Sb, Dy Br, Mo, La, Au, W, Ho, U, Ga, Lu, Ba, Yb.

For the determination of ^{134}Cs and ^{75}Se, the irradiated samples were kept for 15 days before measurement using the GEM-30195 HPGe. Each of the samples were counted for 60 min. This procedure is applied also in the determination of the elements; Sc, Ce, Co, Cr, Eu, Gd, Lu, Ba, Mo, Nd, Rb, Sb, Ta, Tb, Th, Yb, Zn, Cd, Fe, Sr, Ag, Hf, Ir, Hg, Zr, Te, Os.

Based on the regime of irradiation, all the irradiated induced activities were measured using the GEM-30195 HPGe Coaxial, vertical dipstick detector (ORTEC). The relative efficiency of the detector is 30% and the resolution is 1.95 Kev at 1.33 MeV for ^{60}Co. The gamma-ray acquisition system employed in this study consists of the MAESTRO emulation software compatible with Multi-channel Analyzer (MCA). This

software was used for the peak identifications and evaluations[14,15]. The efficiency of the GEM-30195 HPGe Coaxial detector as a function of source-detector geometries was determined using standard gamma ray sources[16], in order to determine the k_0-values. The full energy peak efficiency at 1 and 5 cm geometry of the HPGe detector was determined using gamma ray sources and an efficiency curve at the respective geometries were fitted to obtain the efficiencies of the discrepant nuclides.

The specific activity (A_{sp}) for the activation product of the element of interest was obtained using Eq. 1:

$$A_{SP} = \frac{N_p / t_c}{SDCW} \quad (1)$$

Where:

$S = 1 - e^{\lambda t_{irr}}$ = Saturated factor with t_{irr} = Irradiation time
$D = e^{-\lambda td}$ = Decay factor with t_d decay time
C = $1-e^{-\lambda tc}$ Counting time correction with t_c = counting time
$\lambda = \frac{In\,2}{T_{1/2}}$ = Decay constant
N_p = Net peak area
W = Weight of the measured samples

The equation for concentration calculation based on the k_0-standardization method by the use of Høgdahl convention for the so-called 1/v nuclides needs the neutron spectrum parameters[17], which are the shape factor of the epithermal neutron flux (α), approximated by a $1/E^{1+\alpha}$ distribution and the thermal-to-epithermal neutron flux ratio (*f*). Several author including De Corte *et al.*[13,18], Jonah *et al.*[2] and Rossbach and Blaauw[7] had enumerated the means of obtaining these neutron spectrum parameters based on the experimental methods which were required in both the Høgdahl convention and the Wescott formalism. However, with NIRR-1 and other similar reactors with stable neutron flux characteristics, it had been recommended that the Cd-ratio for multi-monitor method be performed for neutron spectrum monitoring. Moreover, this method had been observed as the most accurate for α-monitoring[3,19].

In this investigation, the neutron spectrum parameters (i.e., f and α) in the inner irradiation channel (B2) of NIRR-1 for NAA applications had been determined using the k_0-standardized method using Cu foil as the monitor and results published[3].

The experimental k_0-values was determined by imputing the evaluated specific activity using Eq. 2 as written in the Hogdahl formalism:

$$k_0 = \frac{A_{SP(analyte)} \times f + Q_{0(Au)}(\alpha) \times \varepsilon_{(analyte)}}{A_{SP(Au)} \times f + Q_{0(analyte)}(\alpha) \times \varepsilon_{(Au)}} \quad (2)$$

Where:

$$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_{th}}$$

is the ratio of resonance integral to thermal cross-section corrected for non-ideal epithermal neutron flux:

$$f = \frac{\phi_{th}}{\phi_{epi}}$$

is the thermal to epithermal flux ratio and ε is the full energy peak detector efficiency. The determination of the resonance integral $I_0(\alpha,)$ for the non-ideal epithermal neutron flux have been done elsewhere[2].

The values of the thermal and epithermal fluxes were obtained using Eq. 3 and 4:

$$\phi_{th} = \frac{N_{p\,(flux\,monitor)} \times \lambda \times M}{S \times D \times \left(1 - e^{-\lambda t_c}\right) \times \bar{\sigma}_f \times \theta \times I_\lambda \times N_A \times w \times \varepsilon \times c} \quad (3)$$

$$\phi_{epi} = \frac{N_{p\,(flux\,monitor)} \times \lambda \times M}{S \times D \times \left(1 - e^{-\lambda t_c}\right) \times I_0(\alpha) \times \theta \times I_\lambda \times N_A \times w \times \varepsilon \times c} \quad (4)$$

Where:

θ = Isotopic abundance
c = Concentration of foil monitor (Cu)
I_λ = Gamma abundance
w = Weight of foil monitor
$\bar{\sigma}_f$ = Average cross section for fast neutron
M = Atomic weight
N_A = Avogadro's number
N_p = Net peak area of the foil monitor

The empirical calculated k_0-values of each of the nuclides of interest were determined using the Eq. 5:

$$k_0 = \frac{M_{(Au)} \times \theta_{(analyte)} \times \gamma_{(analyte)} \times \sigma_{th(analyte)}}{M_{(analyte)} \times \theta_{(Au)} \times \gamma_{(Au)} \times \sigma_{th(Au)}} \quad (5)$$

Where:

σ_{th} = Thermal absorption cross section for neutron (velocity 2200 m sec^{-1})
γ = Gamma-ray intensity

The values of these parameters were obtained from literature Glascock[20] and Blaauw[21].

RESULTS AND DISCUSSION

The k_0-values of the following discrepant nuclides; ^{134}Cs, ^{152}Sm and ^{75}Se have been re-measured (experimentaly) using Certified Reference Materials; Synthetic Multi-element Standard (SMELS) I and III, NIST 1633b Coal fly Ash. The experimental deviation (%) and empirical deviation (%) from the recommended values of De Corte and Simonits[10] were presented in Table 1. As shown, the sources of the uncertainties in the results of both the recommended and experimental results may be due to peak analysis or from determination of neutron flux parameters f and α which are used in the evaluation process.

From the results in Table 1, the k_0 value of ^{152}Sm obtained in this study experimentally showed the highest percentage deviation of 28.14% compared with the recommended values of De Cort and Simonits[10]. The ^{134}Cs k_0 value at the peak energy of 802 keV showed the second highest deviation of 4.38% compared with recommended values. The other peak lines of ^{134}Cs (562, 569 and 602 keV) showed slight deviation of 3.38, 0.27 and 1.89%, respectively.

The percentage deviation of the nuclides as a function of their respective peak lines for each of the studied nuclides was shown in Table 1. Table 1 showed how the experimental values and the empirical calculated values deviated (%) from the recommended values. As indicated in Table 1, the deviation of the empirical calculated from the recommended were as follows; ^{152}Sm at 103 keV (39.52%), ^{75}Se at 136 and 401.7 keV were 24.55 and 18.29% and for ^{134}Cs at 562, 569, 602 and 802 keV were 18.34, 21.16, 19.19 and 22.16%, respectively.

The distribution of the k_0 values as obtained experimentally in this study[9] and including the empirical calculated and the recommended k_0 values[10] were shown in Fig. 1. As observed, the deviation between the recommended, experimental and the calculated k_0 values was large for all the peak lines of ^{134}Cs and ^{152}Sm. Moens *et al.*[5] also reported this similar observation of large deviations that appeared in the comparison of the calculated k_0-values with recommended k_0-values. According to Moens *et al.*[5], it revealed the unreliability of the nuclear data. Moens *et al.*[5] observed that the experimental determination of the k_0

Table 1: Empirical calculation and measured k_0-values compared with the recommended values and the measured k_0-values

Formed Isotopes	Energy (KeV)	Recom. k_0-values	Expt. k_0-values	Empirical cal. k_0-values	Measured k_0-values	Expt. deviation from recom. values (%)	Empirical cal. deviation from recom. values (%)
^{152}Sm	103.00	$(2.31\pm0.4)\times10^{-1}$	$(1.66\pm0.1)\times10^{-1}$	3.82×10^{-1}	-	28.14	39.52
^{75}Se	136.00	$(6.76\pm1.1)\times10^{-3}$	$(6.88\pm1.1)\times10^{-3}$	8.96×10^{-3}	-	-1.78	24.55
^{75}Se	401.73	$(1.43\pm0.8)\times10^{-3}$	$(1.38\pm0.7)\times10^{-3}$	1.75×10^{-3}	-	3.50	18.29
^{134}Cs	562.00	$(4.14\pm1.7)\times10^{-2}$	$(4.00\pm1.4)\times10^{-2}$	5.07×10^{-2}	$(3.93\pm1.7)\times10^{-2}$	3.38	18.34
^{134}Cs	569.00	$(7.34\pm1.5)\times10^{-2}$	$(7.32\pm1.5)\times10^{-2}$	9.31×10^{-2}	$(7.11\pm1.7)\times10^{-2}$	0.27	21.16
^{134}Cs	602.00	$(4.76\pm2)\times10^{-1}$	$(4.67\pm1.8)\times10^{-1}$	5.89×10^{-2}	$(4.57\pm1.7)\times10^{-1}$	1.89	19.19
^{134}Cs	802.00	$(4.11\pm2)\times10^{-2}$	$(3.93\pm1.8)\times10^{-2}$	5.28×10^{-2}	$(4.11\pm1.9)\times10^{-2}$	4.38	22.16

Source: De Corte and Simonits[10] and St-Pierre and Kennedy[9]

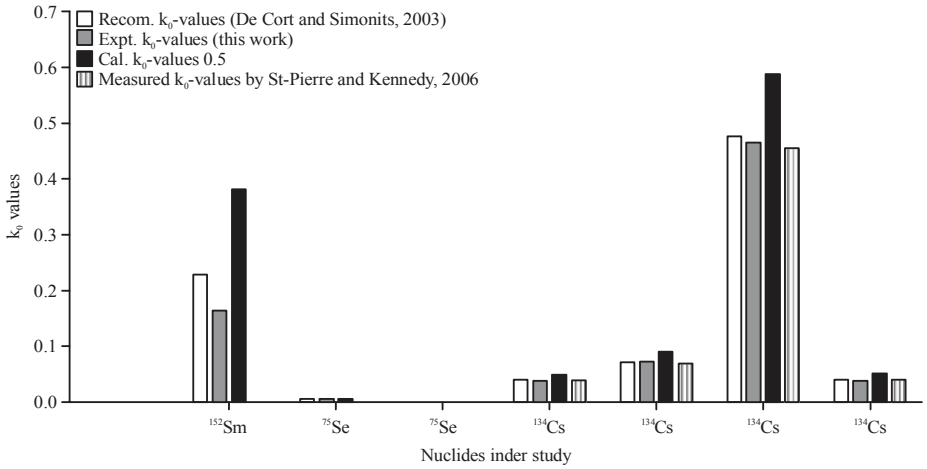

Fig. 1: k_0 value distributions

Source: De Corte and Simonits[10] and St-Pierre and Kennedy[9]

values should be done in order for the data to be use in k_0- standardization computations. This was in agreement with this study as it was cleared that the discrepancy between the calculated k_0 values and the recommended values were very high.

The distribution of the k_0 values between the recommended and the experimental k_0 values have been plotted as shown in Fig. 2 to show how many of the data fully agreed with each other.

As revealed in Fig. 2, the results of this work compared well with the recommended data for most of the nuclides except for ^{152}Sm whose recommended k_0-value of 2.31×10^{-1}. The experimental value obtained was 1.66×10^{-1}, which represent a deviation of 28% as seen in Table 1. The reason may be due to the discrepant nuclear data used in the calculation of k_0-value for this nuclide.

The relationship between the experimental and the empirical calculated k_0 values and how far it deviated (%) from the recommended values was illustrated in Fig. 3. As seen in Fig. 3, the deviation from the empirical calculated values were high for all the nuclides in this studies. This showed that much effort should be emphasized on the experimental determination of the k_0 values before using the data in the k_0 standardization method. It also revealed that using the empirical calculation will lead to a large discrepancy in the determination of elemental concentrations.

As revealed in this study, with the exception of ^{152}Sm all the studied nuclides had good agreement with the recommended values. In comparison between this study and that measured k_0 values of ^{134}Cs for all the peak lines were in good agreement. It was observed that the uncertainties of the k_0 values of ^{134}Cs for all the peak lines were higher compared to this study except for gamma line of 602 and 802 keV (^{134}Cs). The reason for the difference may be coming from the type of detector used in each of the cases. Hence, the results of this work showed very good agreement with the recommended data and the measured value. This showed the accuracy of the adopted irradiation and analytical protocols employed in the laboratory for the determination of elemental concentrations and facilities used for this work.

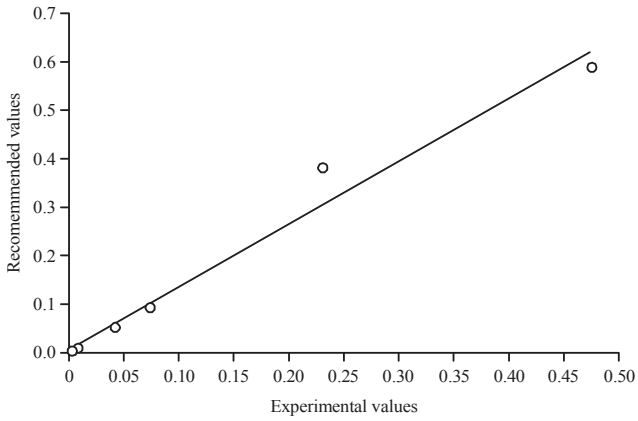

Fig. 2: A plot of recommended k_0 values and the experimental values

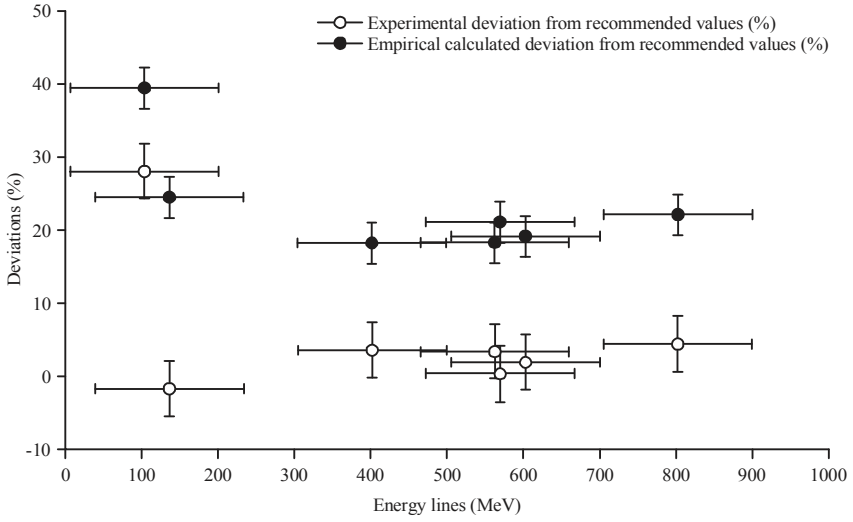

Fig. 3: Deviation (%) distribution of the experimental values from the empirical calculated k_0-values of each of the nuclides of interest

CONCLUSION

It is concluded that experimental determination should be performed rather than empirical calculations for these discrepant nuclides before applying the data in the k_0 standardization technique. The data obtained in this study will aid in elemental determination of concentrations and the established protocols will be applied in the determination of other nuclides; Sc, Ce, Co, Cr, Eu, Gd, Lu, Ba, Mo, Nd, Rb, Sb, Ta, Tb, Th, Yb, Zn, Cd, Fe, Sr, Ag, Hf, Ir, Hg, Zr, Te, Os in author's laboratory.

SIGNIFICANCE STATEMENTS

This study revealed the discrepancy observed in the nuclear data libraries used in determination of elemental concentration using k_0 standardization methods. This study uncover critical areas and shows high discrepancy using empirical calculation of the k_0 values employed in this field. Thus, this new development emphasis the implementation of experimental determination of the k_0 values for the standardization methods.

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