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Application of Maxwellian Neutron Distribution Concept to the k₀-NAA standardization method using a miniature neutron source reactor

Aaron N. Adazabra^{1*}, Erwin. Alhassan¹, Benjamin J.B Nyarko²

¹Department of Applied Physics, Faculty of Applied Sciences, University for Development Studies, P. O. Box 24, Navrongo, Ghana. ²National Nuclear Research Institute, Ghana Atomic Energy Commission, P. O. Box LG 80, Legon – Accra, Ghana.

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ABSTRACT

Two widely used formalisms based on the cadmium ratio concept (with its associated problems) of the single comparator (k₀-NAA standardization) method are used for the multi - elemental analysis at Ghana Research Reactor - 1 (GHARR - 1) facility. This paper examines suitability of GHARR - 1 for reactor neutron activation analysis after the re configuration of the core by monitoring the stability of the flux parameters f and α over the course of nine months and the feasibility of using k_0 – NAA standardization method based on the Maxwellian neutron distribution concept at the facility for routine analysis. The concepts were validated by analyzing two reference materials. The concentrations of most of the elements were found to be within 10% of the certified values.

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Introduction

Instrumental neutron activation analysis (INAA) is a versatile, highly selective, sensitive and multielemental analytical method with good specificity, maintaining an important role in precise determination of elemental concentration. These outstanding features have provided the impetus for the method to be used as a robust analytical method for trace elemental determination even at very low levels (Abugassa et al., 1999). At present, INAA utilizing absolute (k_0) standardization is considered to be the most advanced and optimized version of the method (Salma and Zemplen - Papp, 1999). The k_0 – NAA standardization method also known as the single comparator method in neutron activation analysis (NAA) is based on the fundamental equation for the calculation of reaction rates (Adazabra, 2010). The calculation of the reaction rates is basic to nuclear reactor design. Absorption, capture, fission or scattering events all take place at rates that vary with the energy of the reacting particles. To simplify the calculations, the concept of effective cross section values has been introduced in various ways but all of them lack unity to various degrees (Nisle, 1963). For instance, the well known cadmium ratio concept used for the calculation of effective cross section values divides reacting nuclides into "1/v" and "non – 1/v" (n, γ) reactions leading to Hogdahl and Westcott formulations of the single comparator method (Westcott, 1960). There is therefore, the urgent need for an alternative simplified concept for the calculation of effective cross section values and hence reaction rates.

The form of a well - moderated neutron spectrum in a reactor is assumed to be the sum of a complete Maxwellian distribution plus an epithermal (non - Maxwellian) component with a low energy cut - off (Fastrup and Olsen, 1962). The Maxwellian neutron distribution concept assumes that the reactor neutron flux distribution can be represented by two

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major components, the Maxwellian and the 1/E (non -Maxwellian) components jointed by a transition region. An important feature of this concept is that, nowhere does the cadmium ratio appear in the formulation or the measurement of reactor neutron flux parameters (Nisle, 1963). Therefore, the problems associated with the cadmium ratio concept are completely avoided.

In general, the maximum nominal power of a Miniature Neutron Source Reactor (MNSR) such as Ghana Research Reactor - 1 (GHARR - 1) is approximately 30 kW which is equivalent to a thermal neutron flux of $1 \times 10^{12} \text{ n.cm}^{-2} \text{s}^{-1}$ in one of their inner irradiation channels (Balogun, 2003; Ahmed, 2004). In fact, the core configuration of these reactors are not altered until a burn – up of about 1% is achieved after the core is operated for over 10 years (Akaho and Nyarko, 2002; Nyarko et al., 2003). Consequently, there are no significant variations of the parameters of the neutron spectrum characteristics in the irradiation sites, which make the conditions ideal for the use of the k₀-NAA standardization method.

However, the GHARR - 1 has been in operation for more than 10 years. Burn - up effects, poisons built up, addition of more beryllium shims in other to improve the reflection of more neutrons etc have altered the core configuration of this reactor. To determine if the neutron spectrum is still stable enough for routine k_0 –NAA standardization method based on the cadmium ratio concept, the neutron spectrum parameters, i.e., the epithermal neutron shape factor (α) and the thermal to epithermal flux ratio (f) were measured over a nine month period (Mumuni, 2008). These parameters were determined by a combination of bare zirconium and gold. A combination of bare zirconium and gold is known as a realistic mean for neutron flux characterization in routine basis (Lin et al., 2009). Therefore, considering the fact that, fast neutrons in MNSR's are negligible compared to the thermal neutrons, and they contribute very little

to (n, γ) reactions, the aim of this paper is to examine the feasibility of using the Maxwellian neutron distribution concept for the single comparator method by calculating from the activities of recommended neutron monitoring standards, the thermal (T_n) and epithermal (F_1) parts of the neutron spectrum in the Nisle neutron flux notation. Another objective was to re-evaluate the accuracy, precision and reproducibility of the single comparator method after the reconfiguration of the core using the cadmium ratio and the Maxwellian neutron distribution concepts.

Calculations

$k_0 - NAA$ standardization method

Consider an element *i* in a sample of mass *W*, with a specific count rate $A_{sp,i}$ and detector efficiency at its peak energy, $\varepsilon_p(E_i)$ that is co – irradiated with gold. If the mass of the element in the sample is m_i then, the measured concentration ρ , can be found by using the single comparator equation:

$$\rho = \frac{m_i}{W} = \frac{A_{sp,i}}{A_{sp,Au}} \cdot \frac{1}{k_o} \cdot EPI \cdot \frac{\varepsilon_p(E_{Au})}{\varepsilon_p(E_i)} \tag{1}$$

Where $A_{sp,Au}$ and ε_p (E_{Au}) are the specific count rate and detector efficiency for Au respectively,

 k_0 is a combination of nuclear constants relative to¹⁹⁸Au at its 412 keV gamma-line,

EPI is the index ratio of effective cross section values of Au relative to any element, i.

The cadmium ratio concept and the Maxwellian neutron distribution concept differ essentially by the way their effective cross – section factors are defined and hence, the calculation of their *EPI* values for various elements in any sample as shown in the sections below.

Cadmium ratio concept

Most radionuclides amenable to neutron activation analysis can be analysed using the Hogdahl convention of the cadmium ratio concept. Therefore, only this convention is discussed here. According to Hogdahl (1962) convention, the index ratio, *EPI* of an element in a sample with gold as the comparator in a real neutron flux spectrum $(1/E^{(1+\alpha)})$ is given by:

$$EPI_{H} = \frac{f + Q_{o,Au}(\alpha)}{f + Q_{o,i}(\alpha)}$$
(2)

Where $Q_{o,Au}(\alpha)$, $Q_{o,i}(\alpha)$ are the α – corrected resonance

integral of Au and activated element, i respectively, f is the neutron flux ratio and α is the epithermal neutron shape factor.

The resonance integral (Q_o) values are independent of irradiation channels, geometrical arrangements or counting parameters hence, they can be taken from published nuclear data. Detailed calculations of f, α and $Q_o(\alpha)$ are presented elsewhere (Akaho and Nyarko, 2002). However, Hogdahl convention, based on the cadmium ratio concept is restricted to only nuclides that follow the "1/v" (n, γ) reaction rates.

Maxwellian neutron distribution concept

The Maxwellian neutron distribution concept is also based on integrated reaction rates for the measurement of neutron flux parameters. Thus the rate of the reaction is given by the equation:

Rate of reaction = Rate of production – rate of radioactive decay. (7)

$$\Rightarrow \frac{dN_2(t_r)}{dt} = \text{Rate of production} - \text{Rate of radioactive decay}$$
(8)

Where N_2 is the number of radionuclides formed and $t_{\rm r}$ is the irradiation time

Rate of production =
$$\phi \sigma_1 N_1$$
 ⁽⁹⁾

Where N_1 is the number of stable nuclides isotopes exposed, σ_1 is the activation cross section of the stable nuclide for the

 (n, γ) reaction and ϕ is the thermal neutron flux

Rate of radioactive decay = $\lambda_2 N_2(t_r)$ (10)

Where λ_2 is the decay constant of the stable isotope. Hence

$$\frac{dN_{2}(t_{r})}{dt} = \phi \sigma_{1} \mathbf{N}_{1} - \lambda_{2} \mathbf{N}_{2}(\mathbf{t}_{r})$$
$$\Rightarrow \frac{dN_{2}(t_{r})}{dt} + \lambda_{2} \mathbf{N}_{2}(\mathbf{t}_{r}) = \phi \sigma_{1} \mathbf{N}_{1}$$
(11)

Equation [11] is in the form of a linear first order differential equation (Allan Jeffrey, 2002; John Bird, 2006) with the standard form: $\frac{dy}{dx} + P(x)y = Q(x)$ (12)

Where
$$P(x) = \lambda_2$$
, $Q(x) = \phi \sigma_1 N_1$, $y = N_2(t_r)$ and $x = t$.

The solution of equation (12) is obtained by multiplying it throughout by an integrating factor, $\mu(x)$ expressed as:

$$\mu(x) = \exp\left\{\int P(x)dx\right\}$$
(13)

The solution of equation (11) therefore, leads to equation (14). Details of this solution are presented in appendix A. Hence, (14)

$$P_{x}(F_{1},T_{n}) = \left\lfloor \frac{A}{N_{1}\sigma} \right\rfloor_{x} / \left\lfloor \frac{A}{N_{1}\sigma} \right\rfloor_{1/v}$$

Thus, $P_x(F_1,T_n)$ may be calculated from activation measurements by use of equation (14).

The index ratio *EPI* in terms of Nisle neutron flux notation is therefore expressed as:

$$EPI_{N} = \frac{\left[P_{Au}\left(F_{1}, T_{n}\right)\right]}{\left[P_{i}\left(F_{1}, T_{n}\right)\right]}$$
(15)

Experimental

Sample Preparation

Ten replicate of a standard reference material (SRM) obtained from National Institute of Standards and Technology (NIST), Oyster Tissue (1566b) and another ten replicate of lichen – 336 reference material obtained from International Atomic Energy Agency (IAEA), were accurately weighted directly into pre – clean 2.0 mL polyethylene vials which were then capped and heat – sealed. The Au standard solution used as both an ultimate comparator in the k_0 – NAA standardization method and a suitable "1/v" (n, γ) neutron monitor, was also pipette into a 2.0 mL polyethylene vials, capped and heat sealed. The standard had a certified purity of > 99.999% and a concentration of 1000 ppm. To obtain reproducible geometry, all vials were half filled. All the vials were then placed into 7.0 mL polyethylene irradiation vials which were again capped and heat sealed.

Sample irradiation, Counting and analysis

The standards were irradiated in the isotropic neutron field of the inner irradiation site 2 of Ghana Research Reactor -1(GHARR -1) facility. The irradiated standards after appropriate decay periods were assayed for gamma activities using a gamma-ray spectrometry system. By means of the MCA card, the spectra intensities were accumulated for a preset time. Care was taken to account for the counting losses by keeping the dead time around 10% at the start of counting of the samples. The counting times were chosen not to exceed 0.2 times the half-life of the radionuclide of interest. The self-shielding correction factor for thermal (G_{th}) and epithermal (G_{epi}) neutrons were drastically reduced by keeping the volumes of the standards very small and hence, were assumed to be negligible. The full peak energy efficiency (Moens et al., 1981; IAEA, 2007) determination at this position was achieved by using the IAEA mixed standard radionuclide solution. The efficiency parameters were obtained by fitting the efficiency curve with polynomial function (Osae et al., 1999).

Results and Discussion

A Maxwellian neutron distribution concept was developed for the quantification of elements based on the single comparator (k₀ - NAA standardization) method using Ghana's MNSR (GHARR - 1). One uniqueness of this concept is the absence of cadmium filters in its formulation as shown in Appendix A, Hence, the requirement of epithermal neutron shape (deviation) factor α , cadmium transmission factor F_{cd} , cadmium ratio R_{cd} etc, which come with their associated uncertainties, are entirely unnecessary in the quantification of element(s). To validate the stability and reproducibility of the neutron flux in the irradiation channels of GHARR - 1, which makes it ideal for neutron action analysis, after the re - configuration of the core, conventional INAA based on the cadmium ratio of the single comparator method was also performed. The specific count rates (A_{sp}) values of Au was determined weekly over a nine month period and the uncertainty of the measurement was found to be less that 1.5% confirming constant neutron flux. Therefore, for stable and reproducible neutron flux, the gold comparator conditions and sensitivities can be determined once for quantification using any of the two concepts.

The results of the mean measured elemental concentrations based on the two concepts (i.e. the cadmium ratio and Maxwellian neutron distribution) for reference materials, NIST 1566 b Oyster Tissue and IAEA - 336 lichen are shown in Table and Table 2 respectively. The average elemental 1 concentrations were taken from ten replicate measurements. The accuracy and precision of the two concepts were assessed by simultaneous activation of the two reference materials under the sample experimental conditions with their precisions calculated as the percentage relative standard deviation of the ten measurements. Comparison of the mean measured elemental concentrations for the concepts and the reported (recommended or informed) elemental concentrations, (i.e. % the percentage deviations of the mean measured concentrations) of the reference materials are shown in Figs 1 and 2 for 19 different elements.

As seen in the Tables, the mean measured elemental concentrations of the reference materials are in good agreement with the corresponding reported values. It is evident from both Fig 1 and Fig 2 that in most of the cases the deviations from the reported elemental concentrations is within 10%. Thus in a typical irradiation channel of a low power research reactor such as Ghana's MNSR, which has about 90-95% of their total neutron flux been thermal neutrons, the Maxwellian neutron distribution concept is particularly a versatile tool for the calculation of effective cross section values for routine analysis based on the single comparator method.

Element	Cadmium ratio concept	Maxwellian Neutron Distribution concept	Certified Values			
Ba	9.1 ± 0.4	9.8 ± 0.2	8.6 ± 0.3			
Ca	870 ± 30	795 ± 36	840 ± 20			
Cl	5210 ± 105	5350 ± 85	5140 ± 100			
Cu	72.4 ± 0.9	77.8 ± 0.5	71.6 ± 1.6			
Κ	6610 ± 100	6430 ± 120	6520 ± 90			
Mg	1012 ± 42	1115 ± 32	1090 ± 23			
Mn	18.1 ± 1.1	17.9 ± 1.8	18.5*			
Na	336 ± 44	345 ± 46	330 ± 53			
Rb	2.98 ± 0.03	3.5 ± 0.1	3.26 ± 0.145			
S	720 ± 79	660 ± 90	690*			
Sr	7.3 ± 0.2	7.7 ± 0.07	6.8 ± 0.2			
V	0.53 ± 0.03	0.55 ± 0.02	0.58 ± 0.03			
Zn	1470 ± 67	1380 ± 150	1424 ± 46			

Table 1. Analysis of NIST SRM Oyster Tissue (1566b) in mg/kg by INA A

* Recommended or non - certified values

Table 2. Analysis of IAEA -336, lichen reference material in mg/kg by INAA

Element	Cadmium ratio	Maxwellian Neutron	Recommend Values	
	concept	Distribution concept		
Al	690 ± 30	720 ± 45	680 (21)*	
As	0.741 ± 002	0.761 ± 0.01	0.693 (10) ^a	
Ba	5.9 ± 0.9	5.6 ± 1.6	6.4 (20)*	
Br	13.3 ± 0.8	12.3 ± 1.2	12.9 (16)	
Ca	2770 ± 180	2800 ± 100	2600 (27)*	
Cl	2000 ±98	1850 ±77	1920 (13)*	
Cu	3.54 ± 0.02	3.72 ± 0.06	3.55 (17)	
Fe	440 ± 23	430 ± 14	426 (9)	
Κ	1880 ± 59	1930 ± 87	1840 (8)	
Mg	620 ± 22	650 ± 17	610 (28)*	
Na	310 ± 23	300 ± 34	320 (9)*	
Р	635 ± 33	590 ± 62	610 (25)*	
Rb	1.87 ± 0.20	1.94 ± 0.07	1.72 (9)	
Sm	0.109 ± 0.003	0.102 ± 0.008	0.106 (15)	
V	1.42 ± 0.11	1.39 ± 0.30	1.5 (11)*	
Zn	33.7 ± 0.18	32.1 ± 0.20	31.6 (13)	

* Non certified values, ()^a % relative standard deviation



Fig. 1. Comparison of the accuracy of the two concepts based on the single comparator method using NIST SRM Oyster Tissue (1566b)



Fig. 2. Comparison of the accuracy of the two concepts based on the single comparator method using IAEA – 336 lichen reference material

Conclusion

The results obtained in Table 1 and Table 2 shows that GHARR – 1 still has relatively stable and well – moderated (i.e. there is better thermalization) neutron spectra which can largely be represented by the Maxwellian neutron distribution. This confirms the fact that, MNSR such as GHARR – 1 is well suited for instrumental neutron activation analysis. The accuracy and precision were evaluated for the elements analyzed. The precision was calculated for ten replicate measurements and was found to be with 14%. However, the mean concentrations of most of the measured elements were within 10% of the certified values. This work has therefore, demonstrated the capability of applying the Maxwellian neutron distribution concept to the well – known k_0 - NAA standardization method for the quantification of multielemental samples.

Appendix A

From equations (11), (12) and (13) $\mu(x)$ is expressed as:

$$\mu(x) = e^{\lambda_2 t} \tag{A.1}$$

Therefore, multiplying equation (11) by the term $e^{\lambda_2 t}$, gives

the expression,
$$e^{\lambda_2 t} \cdot \frac{dN_2(t_r)}{dt} + e^{\lambda_2 t} \cdot \lambda_2 N_2(t_r) = \phi \sigma_1 N_1 \cdot e^{\lambda_2 t}$$
(A. 2)
But $e^{\lambda_2 t} \cdot \frac{dN_2(t_r)}{dt} + e^{\lambda_2 t} \cdot \lambda_2 N_2(t_r) = \frac{d[N_2(t_r) \cdot e^{\lambda_2 t}]}{dt}$
(A. 3)
Thus $d[N_2(t_r) \cdot e^{\lambda_2 t}] = \phi \sigma_1 N_1 \cdot e^{\lambda_2 t} dt$
(A. 4)

Integrating equation (A. 4) within the limits of zero to infinite on the Left Hand Side (LHS) and zero to time, t on the Right Hand Side (RHS) gives the expression:

$$\int_{0}^{\infty} d\left[N_{2}(t_{r}) \cdot e^{\lambda_{2}t}\right] = \phi \sigma_{1} N_{1} \cdot \int_{0}^{t} e^{\lambda_{2}t} dt$$
(A. 5)
(A. 6)

$$\Rightarrow N_2(t_r) \cdot e^{\lambda_2 t} = \frac{\varphi \sigma_1 \cdot \mathbf{v}_1}{\lambda_2} \cdot \left[e^{\lambda_2 t} \right]_0^t$$

Hence
$$N_2(t_r) \cdot e^{\lambda_2 t} = \frac{\phi \sigma_1 N_1}{\lambda_2} \cdot \left[e^{\lambda_2 t} - 1 \right]$$
 (A. 7)

$$\therefore N_2(t_r) = \frac{\phi \sigma_1 N_1}{\lambda_2} \left[1 - e^{-\lambda_2 t} \right]$$
(A. 8)

Thus, during irradiation the production of N_2 (t_r) nuclides from N_1 nuclides is given by

$$N_2(t) = \frac{N_1 \phi \sigma_1}{\lambda} \left(1 - e^{-\lambda t} \right) \tag{A.9}$$

After irradiation ceases,
$$\frac{dN_2(t)}{dt} = -\lambda N_2(t)$$
 (A. 10)
At saturation, $e^{-\lambda t} = 0$ in equation (A. 9). This

saturation,
$$e^{-\lambda t} = 0$$
 in equation (A. 9). This is

equivalent to saying that at saturation the decay rate given in equation (A. 10) is equal to the production rate used to deduce equation (A. 9). Therefore, equation (A. 9) may be substituted into equation (A. 10). The result is $\lceil dN(t) \rceil$ (A. 11)

result is
$$\left[\frac{dN_1(t)}{dt}\right]_{sat} = N_1 \phi \sigma_1 = A_x$$
 (A. 11)

Where A _x is the specific count rate of a nuclide x. Hence $[A/\sigma]_x = [N_1\phi]_x$ (A. 12)

Similarly, for a 1/v absorber, $[A/\sigma]_{1/\nu} = [N_1\phi]_{1/\nu}$ (A. 13)

Dividing equation (A. 12) by equation (A. 13) and rearranging gives:

$$\frac{\left[A/N_{1}\sigma\right]_{x}}{\left[A/N_{1}\sigma\right]_{1/v}} = \frac{\left[\phi\right]_{x}}{\left[\phi\right]_{1/v}}$$
(A. 14)
But the ratio on the right is just P(E, T). Therefore

But the ratio on the right is just $P(F_1,T_n)$. Therefore, $P_x(F_1,T_n) = \left[\frac{A}{N_1\sigma}\right]_x / \left[\frac{A}{N_1\sigma}\right]_{1/\nu}$ (A. 15)

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