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Experimental assessment of the dependency of neutron self-shielding factor on neutron field and sample size

Kwame Gyamfi¹, Benjamin J. B. Nyarko^{1, 2}, Samuel A. Bamford², Owiredu Gyampo³ and Emmanuel Ampomah-Amoako¹ ¹Nuclear Reactors Research Centre, N.N.R.I., Ghana Atomic Energy Commission, Box LG 80, Legon-Accra, Ghana. ²Graduate School of Nuclear and Allied Sciences, University of Ghana, P.O. Box AE 1, Atomic, Accra, Ghana. ³Nuclear Application Centre, N.N.R.I., Ghana Atomic Energy Commission, Box LG 80, Legon-Accra, Ghana.

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ABSTRACT

The dependency of neutron self-shielding factor on neutron field and sample size in large sample neutron activation analysis was experimentally assessed for powdered leaves samples up to 5 g in mass. The measurements of the neutron flux depression inside the samples were used in determining the neutron self-shielding factor. The experimental results agreed with the theoretical estimation that neutron self-shielding factor is dependent on neutron flux level and sample size. Neutron self-shielding was found to increase with decreasing neutron flux level and increasing sample size and vice versa.

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Keywords

Neutron Self-Shielding; Neutron Activation Analysis; Attenuation Coefficient; Neutron flux.

Introduction

Large Sample Instrumental Neutron Activation Analysis (LSINAA) has evolved to be an excellent technique for the analysis of a wide variety of materials – having many advantages as well as giving accurate concentrations of many elements. During sample irradiation, a significant perturbation of the neutron field occurs due to neutron absorption and scattering within the sample matrix. Neutron field perturbation is among the principal sources of error in LS-INAA.

The magnitude of neutron self-shielding factor depends on the neutron energy, the size and shape of the sample, as well as the materials of the sample and surrounding medium (Tzika et al, 2004). Some materials contain high concentrations of elements that strongly absorb thermal and epithermal neutrons (Chilian et al, 2008), and a routine method is needed to correct the self-shielding effect. In large test portions, e.g., of kilogram size, neutron absorption and scattering result in substantial selfshielding, causing depression of the neutron flux within the sample compared to the periphery. Neutron self-shielding causes substantial changes in the neutron spectrum throughout the sample if the sample material also contains, for example, hydrogen (Overwater, 1994).

Neutron self-shielding factor, f_n for a sample is defined as follows (Tzika and Stamatelatos, 2004):

$$f_n = \frac{\Phi_{Av}}{\Phi_0}$$
[1]

where $\Phi_{A\nu}$ is an average flux in the sample (or the flux at the centre of samples that have relatively small volume) and will be always less than Φ_0 which is the incident flux. Using flux monitors, $\Phi_{A\nu}$ can be determined by using the equation [2].

$$\Phi_{Av} = \frac{AM}{wN_A\theta\sigma_{eff}} \frac{1}{(1 - e^{-\lambda t}ir)(e^{-\lambda t}d)} \frac{\lambda}{(1 - e^{-\lambda t}m)\gamma\epsilon}$$
[2]

Tele: +233243354886

E-mail addresses: kingbascillus@yahoo.co.uk

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where A = the activity (s⁻¹) measured at $t_m(s)$, σ_{eff} = the effective neutron capture cross section (cm⁻²), N_A = Avogadro's number (mol⁻¹), θ = the isotopic abundance, w = the mass of the sample (g), M = the atomic mass (gmol⁻¹), λ = the decay constant (s⁻¹) of the radioactive atom produced, t_{ir} = the irradiation time (s), t_d = the time in between irradiation and measurement (s), t_m = the measurement time (s), γ = the abundance of gamma-ray and \mathcal{E} = the photo-peak efficiency of the detector.

The present work has sought to study the dependency of neutron self-shielding factor on neutron field and sample size in large sample neutron activation analysis using two different sources of neutrons with different neutron flux; namely photoneutrons and an Americium-Beryllium neutron source for powdered leaves samples of up to 5 g in mass. This experimental assessment will provide an outlook for the estimation of neutron self-shielding corrections factors for large samples that will be analyzed in the facilities that were used.

Materials and methods Sample preparation

Two sets of powdered leaves of *Moringa oleifera* (biological sample) of mass ranging from 0.5 g to 5 g (i.e., 0.5 g, 1.0 g, 2.0 g, 3.0 g, 4.0 g and 5.0 g) were prepared. The samples were placed into irradiation vials (diameter 1.6 cm and height 5.5 cm) and heat-sealed. Pin-shaped aluminium-gold flux monitors of diameter of approximately 0.1 cm were fixed in the middle of each sample that was irradiated for the measurement of the flux depression at the middle of the sample.

Sample irradiation

Sample irradiation for LSINAA of one of two sets of samples was performed using neutron flux of 9.47×10^4 n/cm².s¹ from an Americium-Beryllium neutron source irradiation facility which has an activity of 20 Curies and the neutron emission is quoted as 2.2×10^6 ns⁻¹Ci⁻¹ and the source

strength is calculated as $4.4 \times 10^7 \text{ ns}^{-1}$ (Osea and Amoh, 1999; Asamoah et al, 2011) at the National Nuclear Research Institute (NNRI) of the Ghana Atomic Energy Commission (GAEC). Figure 1 (Asamoah et al, 2011) shows the schematic vertical cross-section of the ²⁴¹Am-Be neutron source facility. The other set of samples was irradiated using photoneutrons from a low power miniature neutron source reactor (MNSR) irradiation facility (see figure 2) (Ampomah-Amoako et al, 2011) at the Research Reactor-1 (GHARR-1) Centre. Ghana The photoneutrons were obtained in the reactor during the shutdown state of the reactor. The measured thermal neutron flux of the photoneutrons that was used for the irradiation ranged from ranged from 6.22×10^7 to 5.10×10^8 ncm⁻²s⁻¹. Samples were transferred into the ²⁴¹Am-Be source as displayed in Figure 1 below. The irradiation capsule was tied to a rope which was lowered to the irradiation site 2. The other set of samples were transferred into the reactor via the pneumatic transfer system at a pressure of 0.6 MPa. The samples were irradiated for 72 hours.



Figure 1: Schematic vertical cross section view of the ²⁴¹Am-Be neutron source facility at NNRI (Asamoah et al, 2011). This representation is not in scale



Figure 2: Vertical cross section of GHARR-1 reactor (Ampomah-Amoako et al, 2011)

Sample counting

At the end of the irradiation, the samples were removed from the reactor and the ²⁴¹Am-Be neutron source and the flux detectors were removed and placed on the coaxial high purity germanium (HPGe) semi-conductor γ -ray detector (Canberra) and the γ -activity of the ¹⁹⁸Au (t_{1/2} = 2.695 days; E_{γ}= 411.8 keV) was measured. Measurement time (10 hours) depended on the amount of the induced radioactivity. A plexiglass source support was mounted on the detector during the measurement in order to ensure easy and reproducible source positioning (De Corte et al, 1987). The ORTEC MAESTRO-32 γ -spectroscopy software was used for γ –spectrum acquisition.

Neutron Self-shielding

The incident flux is the measured flux before the sample was sent into the irradiation site and the average flux is the flux that was measured inside the sample via the flux monitors after irradiation. Using some measured parameters the flux monitors and other parameters and constants, equation [2] was used to determine the average flux and equation [1] was further used in estimating the neutron self-shielding factor in the samples.

Results and discussion

Tables 1 and 2 show the neutron self-shielding factors obtained for the samples irradiated with the photoneutrons and the ²⁴¹Am-Be neutron source, respectively. For both sets of samples (see Tables 1 and 2), the one irradiated with the photoneutrons and that irradiated with the ²⁴¹Am-Be neutron source, the neutron self-shielding factor decreases towards zero as the sample mass (size) is increased. The same trend of variation is observed in Table 3 which is the result of a similar experiment (Gyampo, 2008). Neutron self-shielding factor of one (unity) means that the neutron flux at the centre of the sample is the same as the incident flux or the flux at the periphery, thus there is no self-shielding within the sample.

The farther the self-shielding factor moves from unity toward zero the greater the neutron self-shielding in the sample and vice versa. The increase in the sample size increases the probability of absorption and scattering of the neutrons in the sample causing the depression of the neutron flux inside the sample.

Figure 3 shows the trend of variation of the neutron selfshielding with the level of neutron flux and the mass (size) of the sample. At the two different neutron flux levels, there is not much increase in the self-shielding from the 0.5 g to 1.0 g. However, as the mass increases from 1.0 g to 5.0 g, there is a steady increase in the self-shielding corresponding to the increase in mass (size) of the sample. This shows that neutron self-shielding in the sample investigated becomes appreciable at sample mass $\geq 1 g$.



Figure 3: Neutron self-shielding factor as a function of sample mass for both ²⁴¹Am-Be neutron source irradiation and photoneutron irradiation methods.

The experiment establishes a relationship between the neutron self-shielding and the mass of the sample, for the specific matrix studied, that follows the equations [3] and [4] for photoneutron and ²⁴¹Am-Be neutron source irradiation, respectively. Moreover, it verifies the theoretical estimation that the magnitude of the neutron self-shielding factor depends on the size of the sample.

 $f_n = -0.003m^3 + 0.046m^2 - 0.248 + 1.181$ [3] $f_n = -8 \times 10^{-5}m^3 + 0.007m^2 - 0.071m + 0.585$ [4] where f_n is the neutron self-shielding factor and m is the mass of sample.

Comparing the values obtained for self-shielding factors for both irradiation methods in Tables 1 and 2, it can be seen that the self-shielding factors for the ²⁴¹Am-Be neutron source

irradiation are far from unity than in the photoneutron irradiation for the same sample mass.

The neutron self-shielding factors (see Table 3) measured in a powdered biological sample (Cassava - Manihot esculenta, whose density compares well with the powdered leaves of Moringa oleifera) of mass ranging from 0.5 g to 5.0 g using the thermal neutron from GHARR-1 (Gyampo, 2008) did not deviate farther from one (unity) as the mass (size) of the sample increases compared with the experiments involving the photoneutrons and the ²⁴¹Am-Be neutron source. This observation could be due to the fact that neutron absorption and scattering are dominant when low flux neutrons are used than when high flux neutrons are used. The photoneutron has an average flux of about 2.86×10^8 ncm⁻²s⁻¹, which is higher than the flux of the ²⁴¹Am-Be neutron source used $(9.74 \times 10^4 \text{ ncm}^{-2}\text{s}^{-1})$. The flux of the thermal neutrons that was used by Gyampo (2008) is quoted as 5.6×10^{11} ncm⁻²s⁻¹. It can then be validated from the experiments that neutron self-shielding is neutron flux dependent. Thus, for the same matrix, the kind of relationship that exits between the neutron self-shielding and the level of neutron flux that is used for irradiation is that of an inverse relationship (variation), the higher the neutron flux, the smaller the neutron self-shielding in the irradiated sample and vice versa.

Conclusion

Previous studies have estimated the dependency of neutron self-shielding on neutron energy and the size of the sample (Chilian et al, 2008; Tzika and Stamatelatos, 2004). The dependency of neutron self-shielding factor on the level of neutron field and the mass (size) of sample within the sample matrix has been evaluated experimentally. The experimental results agreed with the theoretical estimation that neutron selfshielding varies inversely with neutron field, and there is a direct variation between the sample mass (size) and the neutron selfshielding. However, the issue of neutron self-shielding factor on the mass of sample and the neutron field used need to be further investigated experimentally.

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Mass of sample (g) Neutron self-shielding factor (f_n) Average Neutron flux in the sample (ϕ_{Av}) 0.5 2.44×10^{7} 0.989 1.0 7.37×107 0.977 2.0 2.59×108 0.847 3.13×10^{8} 3.0 0769 1.61×108 4.00.729 1.40×108 5.0 0.702

Table 1: Neutron self-shielding factor for photoneutron irradiation at GHARR-1

Table 2: Neutron self-shielding factor for - Am-Be neutron source irradiation	Table 2: Neutron self-shielding f	actor for ²⁴¹ Am-Be	neutron source irradiation
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Mass of sample (g)	Average Neutron flux in the sample (ϕ_{Av})	Neutron self-shielding factor (f_n)
0.5	5.17×10^4	0.531
1.0	5.08×10^4	0.521
2.0	4.58×10^4	0.470
3.0	4.23×10^4	0.434
4.0	3.99×10^4	0.409
5.0	3.87×10^4	0.398

 Table 3: Neutron Self-shielding Factor for normal thermal neutron irradiation at GHARR-1 (Gyampo, 2008)

Mass of sample (g)	Average Neutron flux in the sample (ϕ_{Av})	Neutron self-shielding factor (f_n)
0.5	5.500×10^{11}	0.981
1.0	5.393×10 ¹¹	0.961
2.0	4.786×10 ¹¹	0.853
5.0	4.571×10^{11}	0.814