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Collimated scanning for large sample neutron activation analysis of inhomogeneous samples

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Abstract A technique was developed for the identification of inhomogeneities in activity distribution and the correction of their effect on the interpretation of gamma spectrometry data in Large Sample Neutron Activation Analysis. The method was based on collimated gamma scanning using a germanium detector to obtain the activity pattern in the bulk sample and Monte Carlo simulations in order to correct the experimental data for the effect of the inhomogeneous activity distribution. The method was experimentally evaluated in the case of a large cylindrical reference sample of 2 L in volume containing quartz as matrix material and a known source of radioactivity and an excellent agreement was observed. The discussed technique improves the trueness of quantitative analysis of large samples with inhomogeneous activity distribution.

Keywords Large sample neutron activation analysis · Collimated scanning

Introduction

Neutron activation enables non-destructive elemental analysis of bulk samples up to several litres in volume. Large Sample Neutron Activation Analysis (LSNAA) experimental procedure involves neutron irradiation of the sample and subsequent measurement of the induced radioactivity using a gamma ray spectrometry system [1]. Corrections are required for self-shielding of the activating neutrons, self-attenuation of the gamma rays and the geometric factor during gamma counting [2, 3]. The distinct advantage of the technique is the potential for analysis of precious objects and artefacts that cannot be damaged for sampling purposes [4].

It has been shown that trueness of LSNAA depends on the inhomogeneity of the sample material. The uncertainties associated with the presence of inhomogeneities in a large sample have been investigated [5, 6]. The results of these studies suggested that some knowledge on the distribution of activity within the sample is necessary in order to perform accurate quantitative analysis. Baas et al. [7] developed a collimated scanning method for the evaluation of the presence of inhomogeneity and determination of the spatial distribution of radioactivity in the sample. The technique was successfully applied for testing trace element homogeneity in Brazilian coffee beans [8].

This work aims towards the development of a technique for the identification of inhomogeneous activity distribution within a sample and correction of the acquired gamma spectrometry data. The method was based on collimated gamma scanning to obtain the activity pattern in the sample and Monte Carlo simulations to correct the experimental data for the effect of the inhomogeneity in activity. In order to estimate how close the calculated activity to the reference value is, the relative bias and the Z-score were employed. Z-score was calculated as the difference between the evaluated and the reference value, divided by the combined uncertainties of the two. Z-score values are then compared to determined classification being $|Z| \leq 2$

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evaluated as satisfactory, $2 < |Z| \leq 3$ considered of questionable quality and $|Z| > 3$ considered unsatisfactory.

The present study contributes to the requirement for high quality analysis of bulk samples with inhomogeneous activity distribution and therefore extends the analytical capabilities of LSNAA in cultural heritage, waste characterization, geological and environmental studies.

Experimental

The experimental facility consisted of a germanium detector and its shielding, an adjustable parallel-hole collimator and a sample holder driven by stepper motor. The sample holder provided rotation, as well as vertical and horizontal movement capability. The facility provided also the option to perform gamma ray transmission measurements using a ^{152}Eu source in order to determine the effective linear attenuation coefficient of the sample material.

The coaxial germanium semiconductor detector was of 85% relative efficiency, 1.67 keV energy resolution (Full Width at Half Maximum-FWHM) at the 1332 keV ^{60}Co photo-peak and a peak-to-Compton ratio of 93:1. The thickness of the lead collimator was 10 cm and its aperture was 1 cm in diameter. The detector was surrounded by 5 cm lead shielding for background radiation reduction. The collimator to detector end cap distance was 4.5 cm.

^{60}Co and ^{137}Cs reference sources of 50.8 and 292.1 k Bq activity, respectively, were employed in order to evaluate the collimator performance. For both reference sources, measurements were performed in midair and for various source-to-collimator distances (SCD).

Moreover, measurements were performed using a reference cylindrical sample of 2 L in volume. The sample was composed of Quartz matrix (silicon dioxide powder, Sigma–Aldrich, Fluka 00653) in a cylindrical Perspex container of 12 cm in outer diameter, 20 cm in height and 0.3 cm in wall thickness. A Perspex disk of 11.4 cm in radius and 0.3 cm in thickness positioned at 5 cm sample height allowed for the introduction of active sources at specified locations within the sample. In the present study, an activated cobalt foil of 0.0234 g in mass and 23.95 k Bq in activity was introduced to produce the inhomogeneity. The foil was placed (a) on axis and (b) at 3 cm off axis as shown in Fig. 1. In both cases, measurements were performed with un-collimated and collimated detector configurations.

The bulk sample was scanned in vertical and horizontal steps of 1 cm and at four rotation steps (of 90 degrees each). The measurement time was 1 h for each step. The acquired spectra were corrected taking into consideration the corresponding gamma ray background spectra.

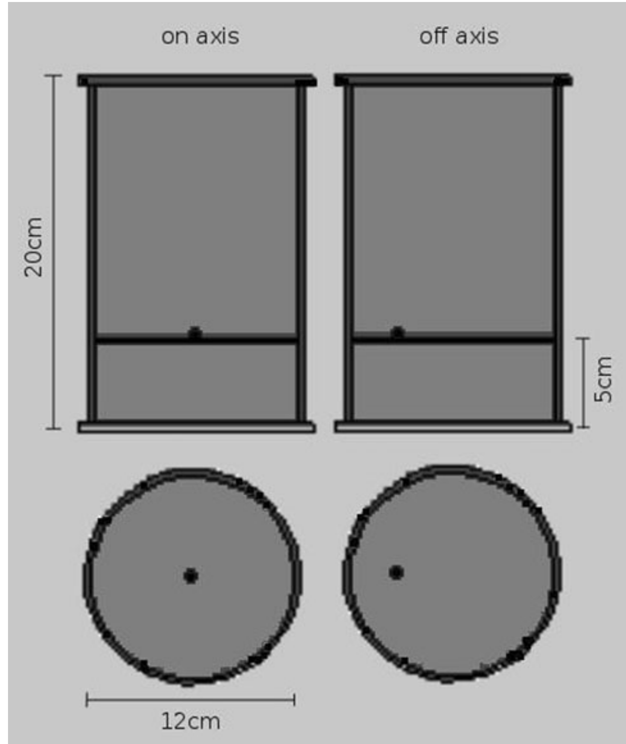

Fig. 1 Schematic representation of the bulk sample and source for the two cases studied (vertical and horizontal cross sections)

Simulations

Monte Carlo code MCNP5 [9] was employed to model the sample, detector, shielding and collimator configuration. Cross section data from the Evaluated Nuclear Data File (ENDF/B-VI) system were used. A schematic representation of the collimated detector and source model is shown in Fig. 2.

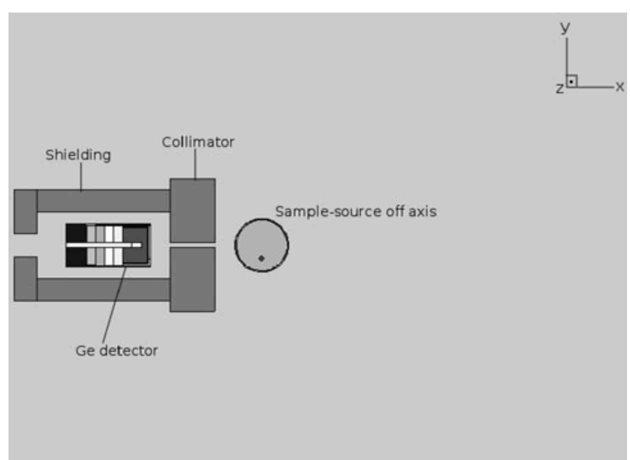

Fig. 2 Schematic representation of the collimated detector and source model (horizontal cross section)

The germanium crystal was modelled using data provided by the manufacturer. The detector active crystal volume was semi-empirically adjusted by comparison against experimental measurements performed using reference sources [10]. The MCNP pulse height tally (F8) was used to predict the detector response in terms of energy deposited in the active volume of the crystal in the specified energy bin and thus estimate the absolute Full Energy Peak (FEP) efficiency of the detector for the photon energies studied. FEP efficiency was calculated for the gamma ray energies of interest for vertical and horizontal steps of 1 cm and at four rotation steps (of 90° each). The relative statistical uncertainties of the computations were below 3%.

Results and discussion

Collimator performance

The collimator is used to limit the field of view of the detector so that gamma radiation from the source of interest can be measured in the presence of background radiation from other sources. Figure 3 shows the MCNP predicted FEP efficiency for a point source in midair as a function of off-axis distance for two SCD of (a) 10.5 cm and (b) 3 cm and gamma ray energies of 661 and 1332 keV. It can be seen that the spatial resolution of the system depends on photon energy and SCD. Relative Width at 50% of the maximum (RW50%) of the off-axis response function was 3.32 and 1.78 cm for SCD of 10.5 and 3 cm, respectively, in the case of 1332 keV photons. For the 661 keV photons, RW50% was 2.80 and 1.60 cm for SCD of 10.5 and 3 cm, respectively. Moreover, Relative Width at 90% of the maximum (RW90%) was found to be 2.00 and 1.10 cm for 1332 keV photons as well as 1.42 and 1.10 cm for 661 keV photons, at SCD of 10.5 and 3 cm, respectively. These results reflect the difference in the geometrical factor due to the source position, as well as the higher penetration properties of the higher energy photons through the collimator material. The off-axis response function results shown suggested that if 90% uniformity in response is required within the scanned sample volume, a scanning step of 1.1 cm is needed for the studied detector-collimator configuration. In this study, a scanning step of 1 cm was implemented.

Detection of inhomogeneity

Figure 4 shows the detector response function along z axis for the two source cases studied for sample-centre to collimator distance of 10.5 cm, detection angle of 0° and 1332 keV photons. The distance of 10.5 cm was chosen to accommodate the diameter of the large sample. From Fig. 4 it can be derived that the 90% of the signal originates from the layer corresponding to sample height (5.0 ± 1.0) cm and (5.0 ± 0.5) cm for the on and off axis source cases, respectively. The activity distributions obtained at sample height of Z = 5 cm for the two source cases studied are shown in Fig. 5a (source on axis),

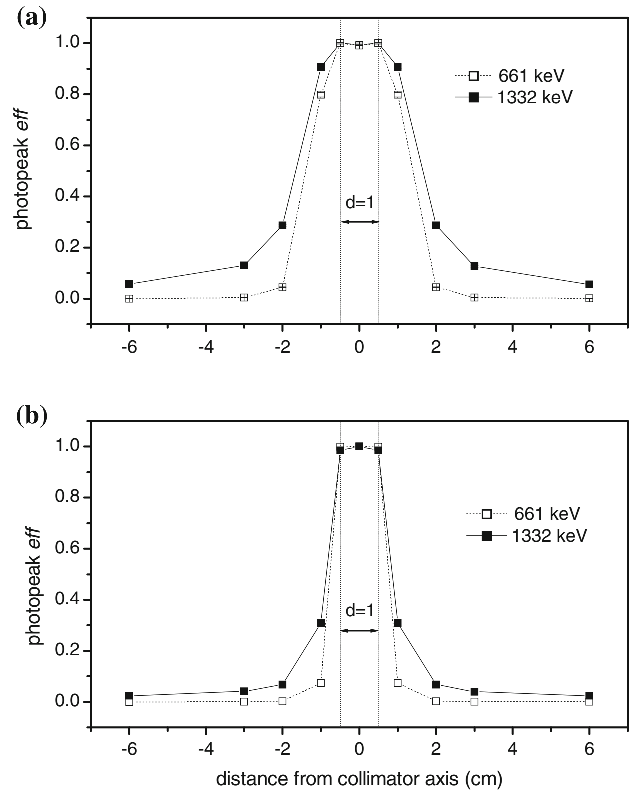

Fig. 3 MCNP predicted photopeak efficiency as a function of lateral distance from collimator axis for SCD of **a** 10.5 and **b** 3 cm for 661 and 1332 keV photons

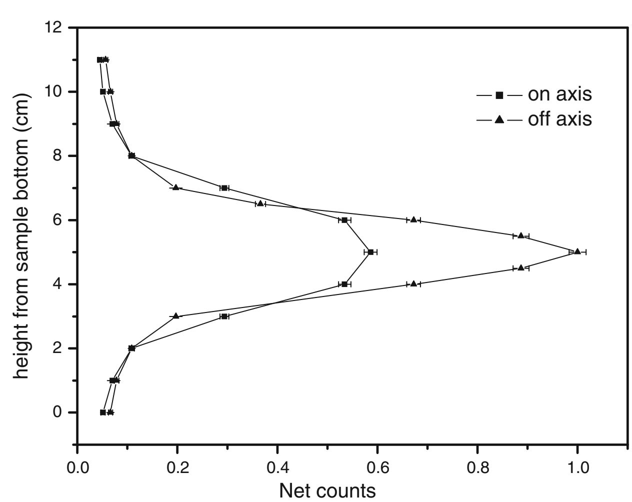

Fig. 4 Detector response along z axis for sample-centre to collimator distance of 10.5 cm and 1332 keV photons

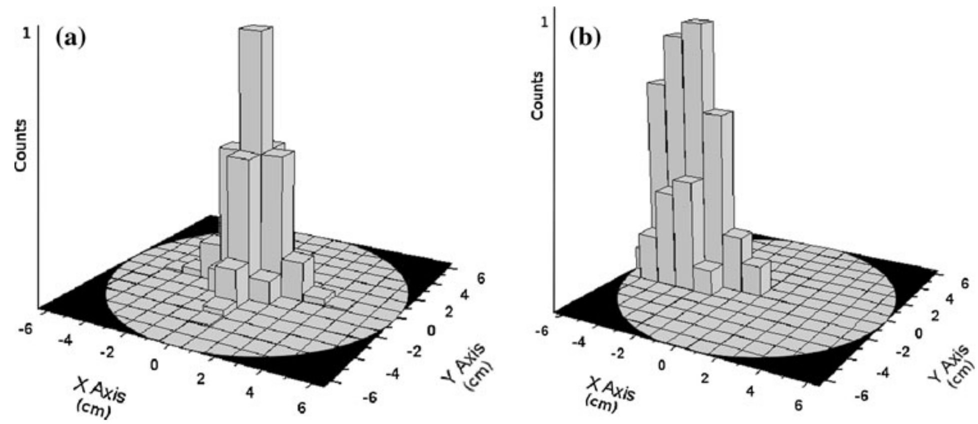

Fig. 5 Activity distribution obtained for the two source cases studied **a** on axis and **b** off axis

b (source off axis). Both patterns were taken for horizontal steps of 1 cm and four rotation angles (0, 90, 180 and 270°). It can be observed that an activity "hot spot" is evident in both figures, corresponding to the actual position of the activated foil in each case.

Data correction

The experimentally determined gamma ray emission patterns indicated that an activity "hot spot" of 1332 keV photons was present within the sample. The activity was detected at sample height of Z = 5 cm at voxels corresponding to (a) x = 0 cm y = 0 cm (on axis) and (b) x = −3 cm y = 0 cm (off axis). The obtained activity distribution was used as input source for MCNP runs in order to predict the detector response for the studied detector, collimator and sample configuration. Detector FEP efficiency was calculated for the following two cases:

(1) Homogeneous activity distribution and an un-collimated detector
(2) Activity distribution as shown in Fig. 5a (on axis), b (off axis) and a collimated detector

In the first case, a homogeneous cylindrical volume source with dimensions equal to those of the large sample was assumed. In the latter case, a weighted spherical activity distribution of 3 cm radius was employed to model the source, with its centre position and associated weighting factors derived from the experimentally determined activity patterns (shown in Fig. 5). Subsequently, the calculated FEP efficiency was applied on the gamma spectrometry data in order to provide the source activity in each case.

Table 1 shows the evaluated total activity, the reference ^{60}Co activity, their ratio and Z-score for each of the cases studied, along with their combined standard uncertainties. It is noted that the combined standard uncertainties include all identified contributing sources related to nuclear data, experimental procedure and simulations. From Table 1 it can be observed that ignoring the activity inhomogeneity (case 1) resulted in a bias of 25 and 10% in the sample activity value for the cobalt source on and off axis positions, respectively. Moreover, Z-score has a value of −5.91 and −2.41 for the on and off axis cases, respectively, indicating unsatisfactory and questionable agreement between the results. However, the knowledge of the activity pattern as obtained from collimated scanning and its employment in the calculation of the FEP efficiency of the detector resulted in an excellent evaluation of the activity within the sample when it was applied on the collimated scanning spectrometry data (case 2). In this case, the ratio of evaluated to reference activity was 1.00 ± 0.05 and 1.03 ± 0.06 for the on and off axis source positions, respectively. In addition, the calculated Z-score

Table 1 Evaluated total activity, reference ^{60}Co activity, their ratio and Z-score for each of the cases studied along with their combined standard uncertainties

Source	Collimator	Distribution	Reference activity (kBq)	Evaluated activity[a] (kBq)	Activity ratio	Z-score
On axis	No	Homogeneous	23.95 ± 1.01	17.96 ± 0.08	0.75 ± 0.03	−5.91
	Yes	From Fig. 5a		24.06 ± 0.64	1.00 ± 0.05	0.09
Off axis	No	Homogeneous		21.51 ± 0.10	0.90 ± 0.04	−2.41
	Yes	From Fig. 5b		24.75 ± 0.92	1.03 ± 0.06	0.59

[a] Average calculated activity over four angles

values of 0.09 and 0.59 for the on and off axis source cases respectively, suggest a satisfactory agreement.

Conclusions

A technique for the identification of inhomogeneity in activity distribution within a sample as well as the correction of the inhomogeneity effect in LSNAA was discussed. The method was based on collimated gamma scanning using a germanium detector to obtain the activity pattern in the bulk sample and Monte Carlo simulations in order to correct the experimental data for the effect of the inhomogeneous activity distribution. The results of the calculations were combined with the acquired gamma spectrometry data in order to evaluate the activity of the sample. The calculated activity was compared against the reference one and an excellent agreement was observed in the case where the inhomogeneity distribution was taken into account and applied on the experimental data. Further work is required on determination of detection limits, optimization of scanning procedure and image reconstruction algorithms in order to extend the capabilities of the technique towards accurate neutron induced gamma ray tomography [11] with unique applications in cultural heritage, waste characterization, geological and environmental studies.

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