MONTE CARLO SIMULATIONS FOR NON-DESTRUCTIVE ELEMENTAL ANALYSIS OF LARGE SAMPLES BY NEUTRON ACTIVATION ANALYSIS

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Abstract - Neutron Activation Analysis (NAA) is an established nuclear analytical technique with applications in a broad range of scientific and technological fields. Typically, conventional NAA involves analysis of small material portions of 10 to 100 μg in mass. However, analysis of samples of a larger mass implies a number of additional advantages such as (a) analysis of objects too precious to remove small parts from and (b) minimization of representative sampling problems of heterogeneous materials. In this work, a method to perform in depth, non-destructive, multi-element NAA of samples of large volume (up to 1 L) is presented. The Large Sample Neutron Activation Analysis (LSNAA) technique involves sample irradiation in the reactor's thermal neutron column and subsequent measurement of the induced radioactivity in the sample employing a HPGe based spectrometry system. Correction algorithms, to compensate for the effects of (a) thermal neutron self-shielding within the sample, during sample irradiation and (b) source geometry and gamma ray attenuation by the sample material during gamma ray counting, have been developed. The correction methods were based on Monte Carlo simulations of both the irradiation and counting facilities using the MCNP computer code. Calculations of thermal neutron self-shielding and gamma-ray detector efficiency for large samples representing industrial and archaeological materials are presented. LSNAA compliments and significantly extends the analytical tools available for in-depth, non-destructive, multi-element analysis of materials too precious to damage for sampling purposes (whole object analysis), representative sampling of heterogeneous materials, or analysis of samples of arbitrary shape. Potential applications of the technique are environmental protection, industrial waste, advanced technological materials, as well as cultural heritage and authentication studies.

Keywords: Monte Carlo method, neutron activation analysis, large samples.

1 Introduction

Neutron Activation Analysis (NAA) is an important tool for qualitative and quantitative multi-element analysis either as a stand alone method or as a complementary technique. NAA has found applications in a variety of fields such as the environment, industry and biology. Conventional NAA involves analysis of small samples, in the micro- to milligram range. However, application of NAA for non-destructive analysis of precious objects and analysis of highly heterogeneous mixtures such as solid waste necessitated enhancement of the technique to analysis of samples of a larger volume [1].

A LSNAA facility has been developed at the Institute of Nuclear Technology and radiation protection, NCSR 'Demokritos', to perform in depth, non-destructive, multi-element NAA of samples of volume up to 1 L [2]. The LSNAA technique involves sample irradiation in the reactor's thermal neutron column and subsequent measurement of the induced activity using a HPGe based spectrometry system. Correction algorithms, to compensate for the effects of (a) thermal neutron self-shielding within the sample, during sample irradiation and (b) source geometry and gamma ray attenuation by the sample material during gamma ray counting, have been developed based on Monte Carlo simulations of both the irradiation and counting facilities. The simulations were performed using the Monte Carlo method. In the present work the LSNAA technique is presented and the MCNP code is used to predict thermal neutron self shielding and gamma ray detector efficiency for large samples representing industrial and archaeological materials.

2 Experimental Facility

GRR-1 is a 5 MW, open pool type, research reactor, cooled and moderated by light water, employing beryllium reflectors at two opposing sides of the core. The LSNAA experimental facility consists of the graphite column neutron irradiation facility and a HPGe based gamma spectrometry system (Figure 1a,b). The sample

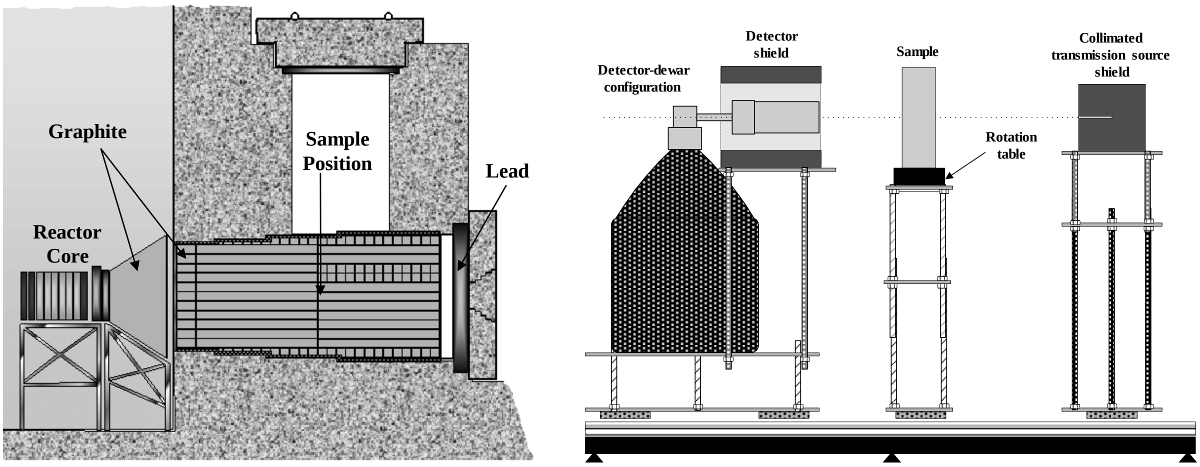

Figure 1 : Schematic representation of (a) the GRR-1 core and graphite column irradiation facility and (b) spectroscopy and transmission measuring facility (vertical cross-sections)

irradiation is performed at a position 240 cm from the reactor core surface, at the mid-height of the fuel elements where the thermal neutron flux (sub-cadmium) is about 5×10^{6} cm^{-2}s^{-1} and the thermal to non-thermal neutron flux ratio is about 300. The gamma ray counting system includes the detector and its shielding, the vertically adjustable sample holder and a collimated gamma-ray source. The gamma ray spectrometer consists of a HPGe detector of 85% relative efficiency for the 1.33 MeV γ ray energy of ^{60}Co, a digital signal processing data acquisition and analysis system. The sample is placed on a PC controlled rotation table. The system includes a transmission measurement option based on a collimated ^{152}Eu source used to asses the effective linear attenuation coefficient of unknown sample matrices prior to irradiation. The source is mounted in a lead shielding box offering a 0.5 cm in diameter and 10 cm in length opening for source collimation.

3 Correction methods

3.1 Monte Carlo simulations

The derivation of correction algorithms was based on explicit models of both the irradiation and counting facilities developed using the Monte Carlo code MCNP version 4C [3]. An MCNP model, including reactor core, thermal neutron column and sample, was developed in order to derive the thermal neutron flux distribution at the sample, during irradiation. Moreover, a detailed detector geometry configuration was modeled using data provided by the detector manufacturer to estimate the γ ray absolute peak efficiency of the detector, for the energy range between 0.05 MeV and 1.6 MeV, during counting. The Monte Carlo models have been presented elsewhere [2].

3.2 Neutron self-shielding

During sample irradiation, a significant perturbation of the neutron field occurs due to neutron absorption and scattering within the sample material. Neutron flux is perturbed not only inside the sample, but also in the graphite moderator in the vicinity of the sample. Self-shielding factor, f_n, has been defined as the ratio of the average flux, $\bar{\Phi}_V$, throughout the volume of the sample to the average flux, $\bar{\Phi}_S$, at the entire surface of the sample. Flux depression factor, h_n, has been defined as the ratio of the average flux, $\bar{\Phi}_S$, at the surface of the sample to the unperturbed flux, $\bar{\Phi}_R$, prior to the insertion of the sample. These factors depend on the neutron energy, the size and shape of the sample, as well as the materials of the sample and surrounding medium.

The calculated self-shielding and flux depression factors for cylindrical samples of radius 5 cm and height 20 cm are shown in Figure 2. The macroscopic thermal neutron absorption cross-sections of the materials ranged between 0.002 and 24 cm^{-1} and the scattering to total cross-section ratios between 0.01 and 0.98. The prediction of the thermal neutron self-shielding factor is based on experimental determination of the flux depression factor by measurement of the thermal neutron flux at the sample surface and at a reference position away from the sample using activation foils [4]. The proposed methodology can be applied for the analysis of samples with h_n>0.25. Moreover, the calculations can be extended to represent other sample sizes and shapes as well.

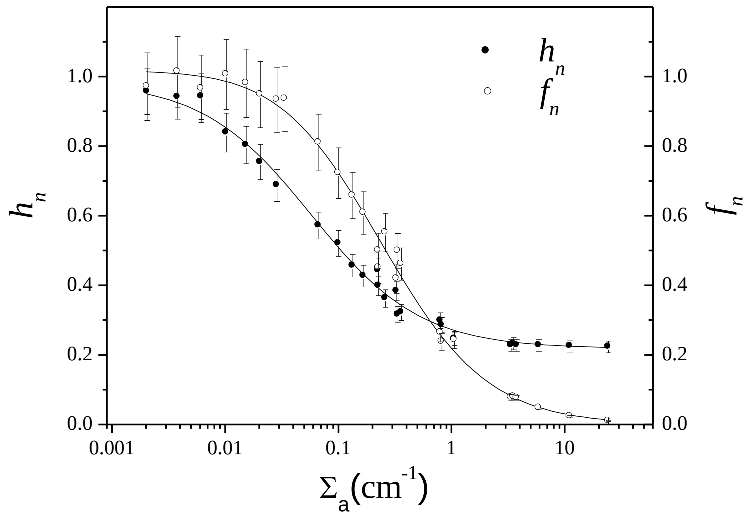

Figure 2 : Thermal neutron flux depression factor (h_n) and self-shielding factor (f_n) as a function of the macroscopic thermal neutron absorption cross-section (Σ_a) of cylindrical samples 5 cm in radius and 20 cm in height

3.3 Counting efficiency of volume source

Gamma-ray counting efficiency requires correction for both the extended source geometry and the gamma ray self-absorption and scattering by the sample material. The volume source efficiency correction factor, f_γ, was defined as the ratio of the volume source photopeak efficiency to the point source photopeak efficiency, located at the centre of the sample, for a given photon energy and source to detector distance [5]. A semi-empirical relationship between the correction factor f_γ and a parameter $\mu \cdot r \cdot \mathrm{H}/(r+\mathrm{H})$, where r and H are the radius and height of the sample, and μ the apparent attenuation coefficient of photons in the sample was proposed [2].

The calculated correction factors, f_γ, for cylindrical samples representing a wide range of materials, are shown in Figure 3. The densities and mass attenuation coefficients of the simulated materials ranged from 0.93 to 11.35 g·cm^{-3} and 0.049 to 0.395 cm^2·g^{-1}, respectively. The radius and height of the simulated samples ranged between 4.3 and 7.5 cm and 9 to 30 cm, respectively. The distance of the source centre of mass to the detector end cup was 25cm. The apparent attenuation coefficient can be determined experimentally by a series of gamma transmission measurements.

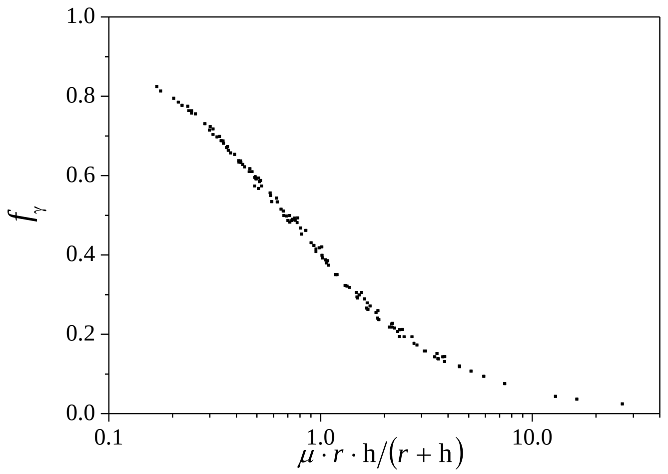

Figure 3 : Predicted volume source efficiency correction factor, f_γ, as a function of the dimensionless variable $\mu \cdot r \cdot \mathrm{h}/(r+\mathrm{h})$

4 Industrial and archaeological materials

The Monte Carlo models were used to predict the thermal neutron flux perturbation and gamma ray detector efficiency of large samples representing industrial and archaeological materials. Plexiglas, iron and copper were selected to represent an industrial material range while iron and ceramic compositions were assumed to represent typical archaeological objects. Samples of three geometrical shapes, slab (10×10×20 cm^3) cylindrical ($r = 7.5$ cm, $h = 20$ cm) and cylindrical cell ($r_1 = 6.0$ cm, $r_2 = 7.5$ cm, $h = 20$ cm) were considered. The 'source to detector' distance was assumed to be 25 cm.

Figure 4 shows a comparison of the thermal neutron flux distributions within slab samples of Plexiglas, iron and copper, as estimated by MCNP and measured by gold foils. The unperturbed thermal neutron flux distribution in the graphite moderator, without the presence of the sample, is also shown. Neutron flux perturbation is well predicted by the code and a very good agreement between estimated and experimental values for all test materials can be observed.

Figure 5 shows the predicted thermal neutron flux distribution, as a function of distance from the graphite column front, for the (a) ceramic and (b) iron samples. The unperturbed thermal neutron flux distribution in the graphite moderator, without the presence of the sample, is also shown. It can be observed that the neutron flux perturbation for the iron samples is higher than that of the ceramic material samples. These results reflect the higher macroscopic thermal neutron absorption cross-section of iron as compared to that of the ceramic material. On the other hand, the hollow cylinders (cylindrical cells) showed a less pronounced self-shielding effect and therefore a much better uniformity of thermal neutron flux in the sample volume.

Figure 6 shows the predicted γ ray detector efficiency as a function of γ ray energy for (a) slab samples composed of Plexiglas, iron and copper and (b) cylindrical and cylindrical cell samples, consisted of ceramic material and iron. The detector efficiency depends on the intrinsic efficiency of the germanium detector, the geometrical factor, as well as the γ ray attenuation properties of the sample material for the γ ray energy of interest. From Figure 6a it can be observed that the γ ray efficiency for the Plexiglas slabs is higher than that of the iron slabs. The lower efficiency over the energy range examined is observed in the case of copper slabs due to higher attenuation of gamma rays in copper as compared with iron and Plexiglas. From Figure 6b it can be observed that the γ ray efficiency for the ceramic material samples is higher than that of the iron samples for the γ ray energy range examined. Moreover, the γ ray efficiency for the cylindrical cells is higher than that of the solid cylinders, due to the lower γ ray self-attenuation effect within these geometrical shapes.

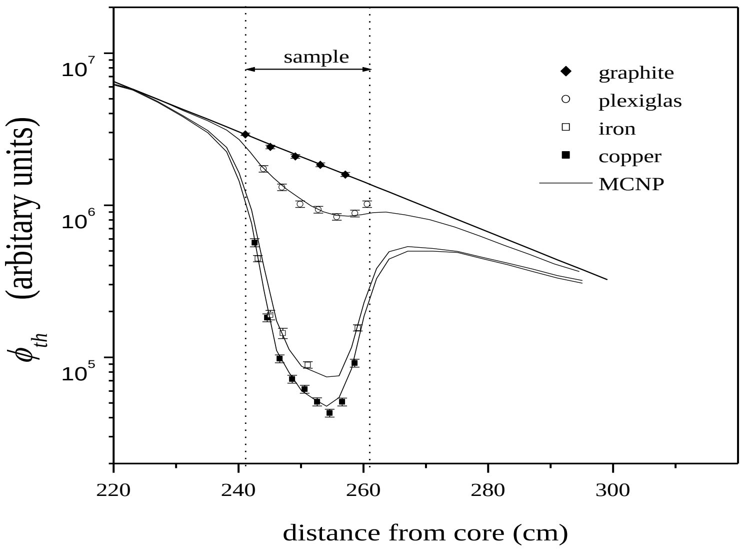

Figure 4 : Thermal neutron flux distribution in slab samples composed of Plexiglas, iron and copper, as a function of distance from the reactor core

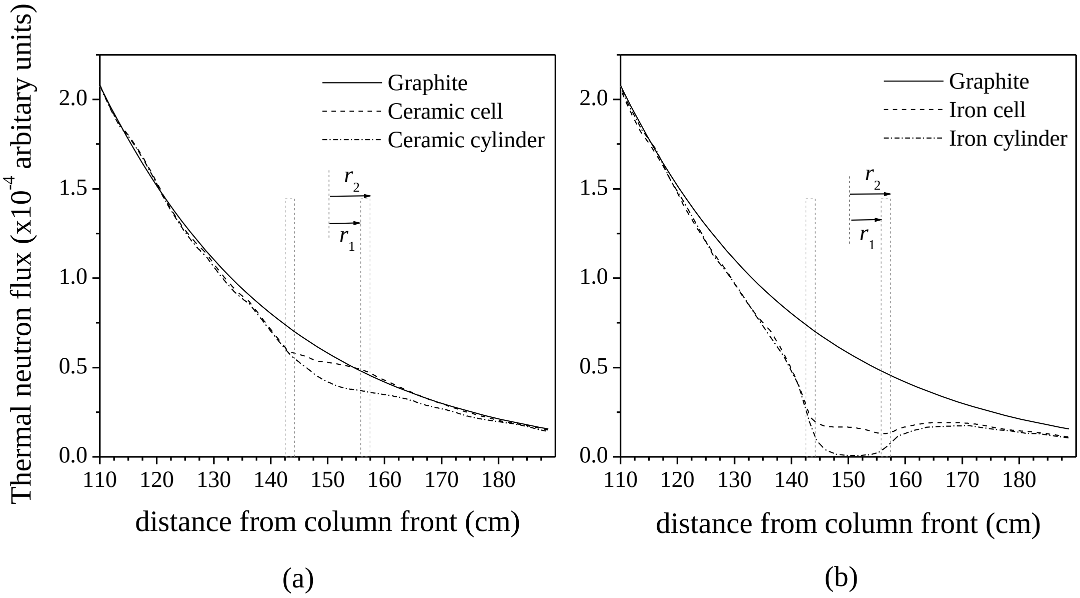

Figure 5 : Predicted thermal neutron flux distribution as a function of distance in the presence of cylindrical cell and cylindrical samples composed of (a) ceramic and (b) iron

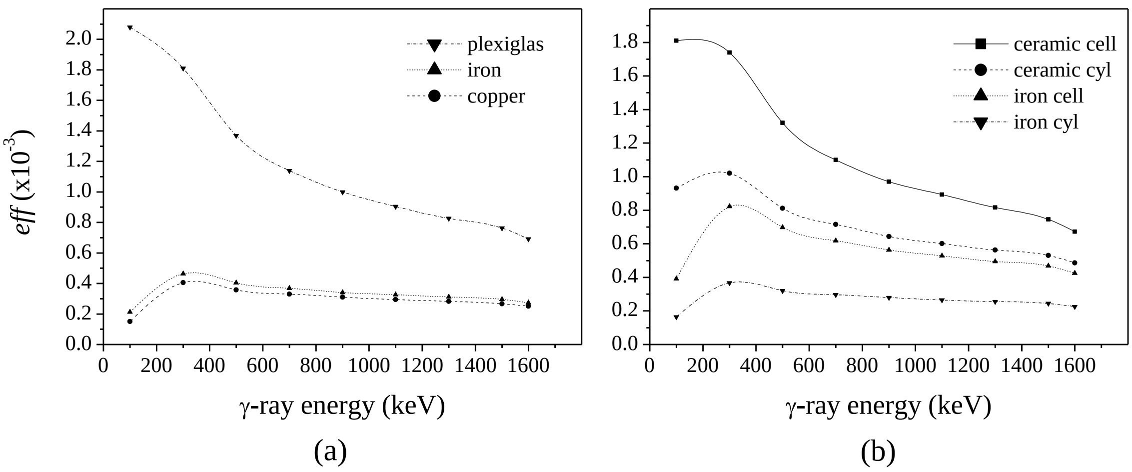

Figure 6 : Predicted detector efficiency as a function of gamma ray energy for (a) slab samples composed of Plexiglas, iron and copper and (b) cylindrical cell and cylindrical samples composed of ceramic and iron

5 Conclusions

Elemental analysis of large volume samples is of a great importance in several scientific fields. LSNAA enables non-destructive analysis of whole objects (up to several litters in volume) providing excellent sampling in depth. In comparison, other established non-destructive analytical methods, such as X-ray fluorescence or analytical techniques based on charged particle irradiation, analyze superficial layers of the sample and thus can provide limited information over the whole volume of the object of interest. As the neutron is a matter penetrating probe, this application of NAA is unique in the characterization of kilogram amounts of metallic, plastic, computer waste or other heterogeneous mixtures of materials. Moreover, non-destructive analysis is a requirement when analyzing precious archaeological objects which can not be damaged for sampling purposes. In addition, representative sampling and minimization of sample contamination is of great importance in the analysis of environmental samples, where "hot spots" or other type of inhomogeneities are often encountered (i.e. filters or substrates, non-homogenous sediment, compost material, electronic waste material). Furthermore, the knowledge of major and trace elements concentrations in tissues, whole organs or even the total body of animal carcasses significantly assists bio-medical studies related to metabolism, nutrition and toxicology.

The application of LSNAA requires consideration for the effects of thermal neutron self-shielding during sample irradiation and γ ray attenuation within the sample and detector efficiency over the volume source during counting. These factors depend on the irradiation facility, the γ ray detection system employed and the sample characteristics. Several calibration techniques have been proposed such as the comparative method, the internal standard method and theoretical modeling of the irradiation and measurement processes. In this work, Monte Carlo simulation of the irradiation and counting facilities using the MCNP code was applied. Advantages of this approach are a better representation of the actual neutron and γ ray fields and flexibility in the geometrical representation of the sample. Slab samples composed of plexiglas, iron and copper, to represent industrial materials, as well as samples of ceramic and iron of a cylindrical and cylindrical cell shapes, representing archaeological objects, were examined. However the computations can easily be extended to any size, shape and material of interest. The latter is important for the analysis of objects with a non regular shape.

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