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# Towards a methodology for bulk sample neutron activation analysis

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**Abstract**

The main challenge in large sample neutron activation analysis (LSNAA) is the determination of neutron self-shielding and gamma ray self-attenuation corrections. After these corrections are determined, the analysis proceeds as in normal neutron activation analysis (NAA), as if the sample were infinitely small. In this paper, these corrections are calculated using the MCNP code for different standard sample geometries with different diameters. Modelling studies for LSNAA using an external neutron beam were performed. An analytical formula for the correction factors for neutron self-shielding and gamma ray self-attenuation is derived. The correction factors as well as flux parameters are calculated analytically. The analytical formula is verified using the MCNP code. All of the calculated parameters were tabulated and graphed. From the calculated data, other unknown material parameters could be obtained based on tabulated data or graphs. This method is a direct and easy method to perform large sample neutron activation analysis without complex calculations. In addition, for the user who does not have good experience with codes such as MCNP, she/he can use the chart or the tabulated information to define their unknown sample with the required information for the LSNAA experiment.


*Keywords:* Large sample neutron activation analysis; Neutron self-shielding corrections; Gamma ray self-attenuation corrections; NAA

## 1. Introduction

All of the multielemental analysis methods (instrumental neutron activation analysis (INAA)) [1], and inductively coupled plasma atomic emission spectrometry (ICP-AES), inductively coupled plasma mass spectrometry (ICP-MS) [2], etc. involve studying a small portion of material (a few milligrams of solids or a few millilitres of liquids) (see Table 1). The current trend is to use even smaller test samples, such as in total reflection X-ray fluorescence (XRF) spectrometry, solid-state atomic absorption spectrometry (AAS), and laser-ablation ICP [3].

The obtained information in the case of XRF is from the surface layers, which represent a few milligrams, making the use of quantities larger than required to prepare the target meaningless [4].

The limitation to the size of the sample is one of the biggest problems facing the analyst when dealing with a large sample. For example, soils, rocks, plant material, etc. can be more easily and representatively sampled at quantities on the order of hundreds of grams to kilograms than at quantities of less than 1 g because a sample is considered as "representative" only if it can present

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Table 1
Sizes of the samples and analytical portions handled in several multielement analysis techniques [4].

| Analysis technique | Solid material mass used or prepared to test portion | Volume used as test portion |
|---|---|---|
| Atomic absorption spectroscopy (AAS) gas furnace | Typically 1–2 g dissolved; | 10–20 μL |
| Atomic absorption spectroscopy (AAS) flame | Maximum approximately 10 g | 1–2 mL |
| Inductively coupled plasma spectroscopy (ICP) | Typically 1–2 g dissolved; maximum approximately 10 g | Approximately 500 μL |
| X-ray fluorescence spectroscopy (XRF) | 10 g | |
| Instrumental neutron activation analysis (INAA) | Typically approximately up to 500 mg; in some cases, up to 30 g | 1–50 mL |

the average properties of the material, environment, or population to which it belongs.

Representativeness is a priori preserved when (i) the sampling is performed according to specific, certified norms or when (ii) a truly homogeneous material is sampled [4].

## 2. Large sample neutron activation analysis

A few phenomena require more attention in large sample neutron activation analysis (LSNAA) than in normal NAA (which uses samples varying from micrograms to a maximum of 0.5 g) because these phenomena usually have only an insignificant impact on the degree of accuracy of the results in normal NAAA [5]. In large samples, e.g., of kilogram size, neutron absorption and scattering result in substantial self-shielding, causing depression of the neutron flux at the centre of the sample compared to the periphery. Neutron self-thermalisation may cause substantial changes in the neutron spectrum throughout the sample if the sample material also contains, for example, hydrogen.

Similarly, the gamma-radiation of the activation products deep inside in the sample will be more strongly absorbed and scattered before leaving the sample than the radiation resulting from, e.g., the surface of the sample; moreover, the absorption and scattering increase rapidly at lower gamma-ray energies. This effect is denoted as gamma-ray self-attenuation. Additionally, a sample of 1 kg cannot be considered as a more-or-less "point source" during counting at normal sample–detector distances of, e.g., 10–30 cm, resulting in a corresponding different response of the detector for the gamma-radiation.

Other methods for standardisation have been proposed as well; these methods are primarily based on a priori available information on the (gross) composition of the object, e.g., using Monte Carlo simulations [6] or neutron transport codes [7] ("fixed point iteration method"). Degenaar [8] developed a method in which no a priori information is used and the neutron self-shielding is estimated on the basis of the attenuation and scattering of the neutron beam measured outside the sample. Baas developed a method for Neutron Activation Analysis of Inhomogeneous Large Samples. In this method, he considered the large sample as a large number of small samples, and the same detector area is divided into small portions to be in line with his assumed subsamples [9].

Other methods have also been proposed for Prompt Gamma Neutron Activation Analysis (PGNAA) using isotopic neutron sources, such as 252Cf or 241Am (Be) [10] and Pu–Be [11]. PGNAA is used for analysing large solid samples, including irregularly shaped meteorite samples [12,13]. Archaeological objects, such as bronzes, were analysed by this method [14].

### 2.1. Large sample neutron activation analysis calculations

The basic measurement equation of NAA by which the mass of the unknown element is calculated directly demonstrates the fact that the technique does not set a priori constraints on the mass of the sample analysed:

$$A_0 = \emptyset_{\text{th}}\sigma_{\text{eff}}\frac{N_{\text{AV}}\theta_m}{M}(1-e^{-\lambda t_{ir}})e^{-\lambda t_d}\frac{(1-e^{-\lambda t_m})}{\lambda}\gamma\varepsilon \quad (1)$$

where $A_0$ is the area of the relevant peak in the gamma-ray spectrum, $\emptyset_{\text{th}}$ is the thermal neutron fluence rate ($\text{cm}^{-2}\,\text{s}^{-1}$), $\sigma_{\text{eff}}$ is the effective absorption cross section ($\text{cm}^2$), $N_{\text{Av}}$ is Avogadro's number ($\text{mol}^{-1}$), $\theta$ is the isotopic abundance. $m$ is the mass of the irradiated element (g), $M$ is the atomic mass number ($\text{g mol}^{-1}$), $\lambda$ is the decay constant of the radioisotope formed ($\text{s}^{-1}$), $t_{ir}$ is the irradiation duration (s).

Eq. (1) could be used in LSNAA as in Eq. (2) after calculating the ratio of

- The neutron self-shielding inside the sample,
- The gamma-ray self-attenuation inside the sample.