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# Dependence of thermal and epithermal neutron self-shielding on sample size and irradiation site

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**Abstract**

Analytical expressions recently developed for calculating thermal and epithermal neutron self-shielding for cylindrical samples used in neutron activation analysis were verified using three different irradiation sites of a SLOWPOKE reactor. The amount of self-shielding varied by less than 10% from one site to another. The self-shielding parameters varied with the size of the cylinder as $r(r+h)$, for $h/r$ ratios from 0.02 to 6.0, even in slightly non-isotropic neutron fields. A practical expression, based on the parameters of the neutron spectrum and the well-known thermal neutron absorption cross-section and the newly defined epithermal neutron absorption cross-section, is proposed for calculating the self-shielding in cylindrical samples.


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## 1. Introduction

The $k_0$ standardization method, widely used for multi-element neutron activation analysis (NAA) with large research reactors, was adapted for small reactors like SLOWPOKE with highly stable neutron fluxes [1]. Regardless of the reactor type, for large samples and for samples with high concentrations of neutron absorbing elements, the equation used in $k_0$ neutron activation requires the knowledge of thermal and epithermal self-shielding factors. Recent studies [2–4] indicate that both self-shielding factors, $G_{th}$ and $G_{ep}$, can be expressed by a sigmoid function with a parameter depending on the nuclide and the geometry and the composition of the sample. The sigmoid function was established for mono-element objects irradiated in an isotropic neutron field, conditions not applicable in the real NAA situation. Thus, in a previous paper [5], we developed an experimental method to determine the epithermal self-shielding as a function of the amount of the element present in the sample, and the experimental parameter of the thermal and epithermal sigmoid self-shielding functions was determined.

In our the previous work [5], measurements were done only for 1 mL cylindrical samples irradiated in the inner irradiation site of the SLOWPOKE reactor where the thermal neutron field is very nearly isotropic. In the present work, we verify experimentally the validity of the theoretical sample geometry dependence of the expressions for thermal and epithermal self-shielding. We also verify whether the magnitude of the self-shielding varies with the size of the irradiation site and the surrounding neutron reflecting materials.

## 2. Theory

### 2.1. General

In a typical reactor neutron spectrum, the activity $A$ of a given nuclide produced by the (n, $\gamma$) reaction upon irradiating a sample containing an amount $m$ of the

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element is given by

$$A = \frac{mN_{Av}\theta}{M_{at}}\sigma_{th}\varphi_{th}(G_{th} + G_{ep}Q_0/f)(1 - e^{-\lambda t_i}) \quad (1)$$

where $N_{Av}$ is Avogadro's number, $\theta$ is the isotopic abundance, $M_{at}$ is the atomic mass, $\sigma_{th}$ is the thermal neutron activation cross-section, $Q_0 = I_0/\sigma_0$, the ratio of resonance integral to $2200\,m\,s^{-1}$ cross-section, $f = \varphi_{th}/\varphi_{ep}$, $\varphi_{th}$ and $\varphi_{ep}$ are the unperturbed thermal and epithermal fluxes, $\lambda$ is the decay constant and $t_i$ is the irradiation time. In practice, the unperturbed thermal and epithermal fluxes may be measured by simultaneously irradiating flux monitors near the absorbing sample, taking into account flux gradients and the fact that the absorbing sample may perturb the nearby fluxes outside the sample. For short irradiations, assuming the unperturbed thermal and epithermal fluxes are constant with time, they may be measured by irradiating flux monitors before or after the absorbing sample and at the same position. $G_{th}$ and $G_{ep}$ are the thermal and epithermal self-shielding factors for the given (n, $\gamma$) reaction. These definitions of $\varphi_{th}$,$\varphi_{ep}$, $G_{th}$ and $G_{ep}$ are consistent with those used in Monte-Carlo simulations [2–4] where $\varphi_{th}$ and $\varphi_{ep}$ are found by simulating a sample of infinite dilution. $G_{th}$ and $G_{ep}$ depend on sample geometry and they are equal to unity for sufficiently dilute samples. The thermal self-shielding factor depends on the amounts of all elements in the sample that absorb thermal neutrons. The epithermal self-shielding factor will be influenced by nuclides other than the nuclide being activated (isotopes of the same element or isotopes of the different elements present in the sample), only if these nuclides absorb neutrons at resonance energies that overlap with the resonances that activate the nuclide in question.

The sample activity that would have been produced if self-shielding had been negligible is obtained by dividing the sample's measured activity by its effective self-shielding factor, $G_{eff}$. This factor is defined as the ratio between the reaction rate per atom in the real sample and in a similar and infinitely dilute sample. Using this definition, the effective self-shielding factor is obtained by dividing the activity of Eq. (1) with the activity calculated with the same equation when $G_{th} = 1$ and $G_{ep} = 1$. Thus, $G_{eff}$ is given by

$$G_{eff} = \frac{G_{th} + G_{ep}Q_0/f}{1 + Q_0/f}. \quad (2)$$

Experimentally, $G_{eff}$ is proportional to the specific activity of the nuclide in the irradiated sample, which may be obtained from the area of the detected peak divided by the element concentration in the sample and by the detection efficiency and corrected for the decay time. The proportionality factor between $G_{eff}$ and the specific activity is found by irradiating sufficiently dilute samples.

### 2.2. Thermal self-shielding

For almost all nuclides, the thermal cross-section of radiative capture is a slowly varying function of neutron energy, proportional to $E^{-1/2}$. Hence, the thermal neutron self-shielding factor for any nuclide can be expressed by the same function of the amount of the element and the element's thermal neutron absorption cross-section, $\sigma_{abs}$. In the literature [7,8] we find different forms for this function, but, in all of them, the thermal self-shielding factor dependence on the sample geometry is the same. In the particular case of cylindrical samples of a given concentration, it varies with sample size according to the factor $rh/(r + h)$, where $r$ is the radius and $h$ is the height. If the self-shielding is expressed as a function of the total amount of the element $m$, then, for a given $m$, as the sample size increases, the sample is diluted and the self-shielding decreases. It then varies with the size of the cylinder according to the factor $1/r(r + h)$.

This study will use results from a recent paper of Martinho et al. [4] that propose a universal sigmoid function for thermal neutron self-shielding. Simplifying and rewriting their expression for $G_{th}$ as a function of the amount of the element in the sample of a given geometry [5], one obtains

$$G_{th} = \frac{1.00}{1 + (m/m_{th})^{0.964}} \quad (3)$$

where $m$ is the amount of the element. For cylindrical samples, the parameter $m_{th}$ varies with the radius and height of the cylinder as $r(r + h)$ and can be estimated from the thermal neutron absorption cross-section as follows:

$$\frac{1}{m_{th}} = \frac{k_{th}N_{Av}\sigma_{abs}}{r(r + h)M_{at}}(\sigma_{tot}/\sigma_{abs})^{0.15} \quad (4)$$

where $\sigma_{tot}$ is the element total thermal neutron cross-section (scattering plus absorption) and $k_{th}$ is a parameter which may depend on the irradiation site, but is independent of the nuclide and the sample composition. In Eq. (4), the factor involving neutron scattering is usually close to unity and can be neglected, especially for elements with high absorption cross-sections.

### 2.3. Epithermal self-shielding

The estimation of $G_{ep}$ as a function of element concentration and sample geometry has always been a difficult task because each nuclide has a different and sometimes complex resonance pattern; in nuclear reactor studies, it is usually done by the Monte-Carlo method. Such calculations are time consuming and impractical for routine NAA where the concentration of the absorbing element is not known in advance; indeed that is the aim of the analysis. The most common approach has been to avoid the problem by diluting the samples or by irradiating in a neutron spectrum with a very small epithermal component. Thus, the question of a practical solution to the problem of epithermal self-shielding in NAA has not been investigated systematically.