Calculation of Thermal Neutron Self- Shielding Correction Factors for Aqueous Bulk Sample Prompt Gamma Neutron Activation Analysis Using the MCNP Code

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Abstract

In this work thermal neutron self-shielding in aqueous bulk samples containing neutron absorbing materials is studied, for Bulk Sample Prompt Gamma Neutron Activation Analysis (BSPGNAA), using MCNP code. The code was used to perform three dimensional simulations of neutron source, neutron detector and sample of various material compositions. The MCNP model was validated against experimental measurements of the neutron flux performed using a BF_3 detector. Simulations were performed to predict thermal neutron self shielding in aqueous bulk samples containing neutron absorbing solutes. In practice, the MCNP calculations are combined with experimental measurements of the relative thermal neutron flux over the sample's surface, with respect to a reference water sample, to derive the thermal neutron self-shielding within the sample. The proposed methodology can be used in determination of the elemental concentration of unknown aqueous samples by BSPGNAA where knowledge of the average thermal neutron flux within the sample volume is required.

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1. Introduction

Increasing needs of material knowledge leaded to the development and application of nuclear analytical techniques. Between these methods, BSPGNAA has been extensively applied for the analysis of bulk samples in different industries, such as mineralogy, control process of cement, detecting of explosive material in airports, in vivo body composition animal studies, and nuclear fuel manufacturing process [1-3].
The PGNAA technique is based upon bombarding a sample by neutron and then measurement prompt gamma spectrum emitting from the elements in the sample after absorbing neutron. The gamma energy and its intensity determine respectively type of elements and its concentration. The rate of activation of elements is proportional to the average neutron flux over the sample volume. The description of neutron field perturbation is proportional to coefficients that are defined [4,5]. In this work it was used a relative method. Considering this

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subject, experiment is done in two steps. Measurements were first performed on aqueous bulk sample and the then on a reference sample of pure water. So to describe neutron field perturbation by the sample appropriate factors have been defined. The ratio of the average aqueous bulk sample flux is f_s. The ratio of the average aqueous surface sample flux to the average pure water surface sample flux is h_s. The defined factors are dependent to the neutron energy, shape and size of sample, material and surrounding media. Using these two factors, self shielding is calculated here by using the entity:

$$m_a = \left(\frac{AM}{\varphi_s I_\gamma \varepsilon N_{AV} f \sigma_a t \theta}\right)_a \qquad \textbf{(1)}$$

$\boldsymbol{M}$ = molar mass (kg mol^{-1})
$\boldsymbol{I_\gamma}$ = relative gamma-intensity
$\boldsymbol{\varepsilon}$ = full-energy peak detector efficiency
$\boldsymbol{N_{AV}}$ = Avogadro number (mol^{-1})
$\boldsymbol{\sigma_a}$ = microscopic absorption cross section (m^2)
$\boldsymbol{\theta}$ = abundance of the capturing isotope
$\boldsymbol{f}$ = self-shielding factor
φ_s = surface neutron flux (m^{-2} s^{-1})
$\boldsymbol{t}$ = irradiation/counting time (s)
$\boldsymbol{A}$ = net full-energy peak area
$\boldsymbol{m}$ = mass of any element in the sample (kg)

The concentration of element in PGNAA technique is determined. In the present work a relative method for calculating the concentration of unknown aqueous sample has been used.

$$\frac{m_H}{m_a} = \frac{\left(\frac{AM}{\varphi_s I_\gamma \varepsilon N_{AV} f \sigma_a t \theta}\right)_H}{\left(\frac{AM}{\varphi_s I_\gamma \varepsilon N_{AV} f \sigma_a t \theta}\right)_a} = \frac{\left(\frac{AM}{I_\gamma \varepsilon \sigma_a t \theta}\right)_H}{\left(\frac{AM}{I_\gamma \varepsilon \sigma_a t \theta}\right)_a} \times \frac{\overline{\Phi}_a}{\overline{\Phi}_H} = \frac{\left(\frac{AM}{I_\gamma \varepsilon \sigma_a t \theta}\right)_H}{\left(\frac{AM}{I_\gamma \varepsilon \sigma_a t \theta}\right)_a} \times f_s \qquad \textbf{(2)}$$

$\overline{\Phi}_a$ = average thermal neutron flux in the aqueous sample and $\overline{\Phi}_H$ = average thermal neutron flux in the pure water sample, where (H) indice refer to the amount of Hydrogen in the reference sample and (a) indice refer to the amount of the element of interest in the unknown sample. The ratio $(f\varphi_s)_a/(f\varphi_s)_H$ is equivalent to coefficient f_s, where f is thermal neutron self shielding in sample.
Self-shielding correction methods for large sample irradiation have been proposed [6-8]. The difference is that the others have used from a thermal neutron beam while in the present work there is used a fast neutron point source that its neutrons thermalize in the sample medium.

2 .materials and methods

2.1. Experimental

The experimental work was performed at the Isfahan Nuclear Science & Technology Research Institute. For this experiment, four liters of pure water in a cylindrical container of radius 19cm and height of 17.2cm are used. For neutron bombarding it is used a ^{252}Cf source with a $(1.7\times10^{8}\,n/cm^{2}s)$ flux is used. The source is put in the center of cylindrical sample. The experimental samples are listed in Table 1.

Table1. The experimental samples

Samples
10 g/L NaCl
15 g/L NaCl
20 g/L NaCl
5 g/L H_3BO_3 + 20 g/L NaCl
10 g/L H_3BO_3 + 20 g/L NaCl
15 g/L H_3BO_3 + 20 g/L NaCl

The cylindrical samples are irradiated for 1000 s. Measurement and analysis of the prompt gamma spectra are performed using a HPGE based spectroscopy system and Maestro software with detector being immersed in a boron shield. The measurement of neutron flux has been done by BF_3 detector which was tangential with the surface of the sample. The BF_3 gas counter proportional with an outer diameter of 14 mm, an effective length of 99 mm, containing 96 percent ^{10}B enriched BF_3 gas at the pressure of 71.5 kPa, was used for neutron detection. The efficiency of the system is about 47%. This detector is used to measure the slow neutron flux.

2.2. Simulations

MCNP version 4C was used in neutron transport mode with cross-section data from endf/bvi library [9]. The MCNP code enabled a detailed three-dimensional modeling of the actual source and geometry configuration including point source and sample. (F4) and (F2) respectively refer to Track Length estimate tally and surface flux tally. These two parameters were used for prediction of average flux over a cell in units of cm^{-2} per source neutron. The relative errors of the computations were kept below 10%. All statistical tests for the estimated answers were passed. f_s and h_s coefficients were predicted for cylindrical aqueous bulk samples.

3. Results and discussion

3.1. MCNP Code Verification

To evaluate the accuracy the calculations were compared with experimental data. After the code verified, then all calculations at other points for other quantities were confirmed. In this experiment there was a comparison between the relative flux measured by BF_3 detector and relative flux calculated by (F2) MCNP tally. The relative flux in both of them is the ratio of the average flux at the surface of the aqueous sample to the average flux at the surface of pure water which is defined as the coefficient h_s. First the flux at the surface of the sample including water, salty water, and boric acid solution is measured. These solutions are simulated by MCNP code. The factor h_s is determined in both experimental and simulation. In Figs. 1 and 2 the quantity h_s is drawn versus concentration of salt and boric acid in the water. There is a good agreement between experiment and simulation. Based on this the simulations with code were verified, allowing these simulations to be used in other kinds of calculations.

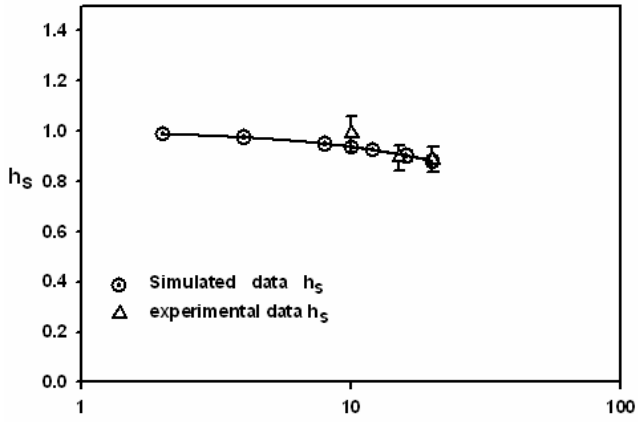

Fig.1. The h_S as a function of concentration of salt.

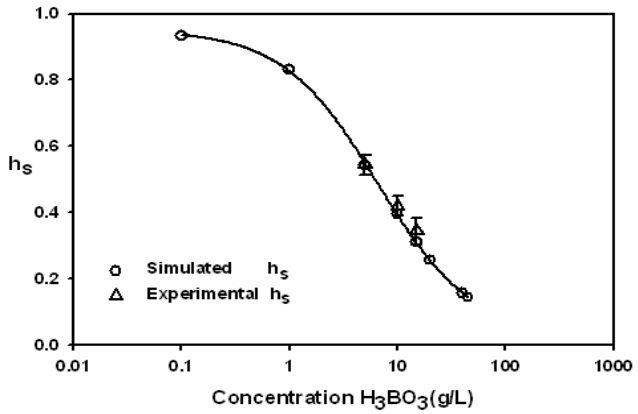

Fig. 2. The h_S as a function of concentration of boric acid.

3.2. Calculation of Self Shielding Coefficient for Thermal Neutrons

^{252}Cf is an isotropic neutron source with an average fission spectrum 2.348 MeV. Due to existence neutron self-thermalization, when neutrons travel a short distance about 5.77cm, they slow down and then leading to thermal neutron flux. A sample of pure water was used as a calibration sample. The average bulk flux was calculated in a pure water sample, $\overline{\Phi_v}(H_2O)$, and aqueous bulk samples, $\overline{\Phi_v}(sample)$, using simulation by MCNP code. Then the value of f_s was given by

$$f_s = \frac{\overline{\Phi_v}(sample)}{\overline{\Phi_v}(H_2O)} \qquad \textbf{(3)}$$

The calculations of f_s were done by simulations of 13 assumptive samples using MCNP code. Absorption and scattering macroscopic cross sections are respectively defined as Σ_a and Σ_s. Using these macroscopic cross-sections, the relative absorption and scattering macroscopic cross-sections are respectively given by

$$\Sigma_{ar} = \frac{m \times \Sigma_a}{1000 + m} \qquad \Sigma_{sr} = \frac{m \times \Sigma_s}{1000 + m} \qquad \textbf{(4)}$$

In these equations "m (g/L)" is the mass of dissolved material in the water. Using simulations, two factors f_s and h_s were found versus relative absorption macroscopic cross section of dissolved material in the water. The results are shown in the Figs. 3 and 4. Σ_{ar} gives the absorption macroscopic cross-section

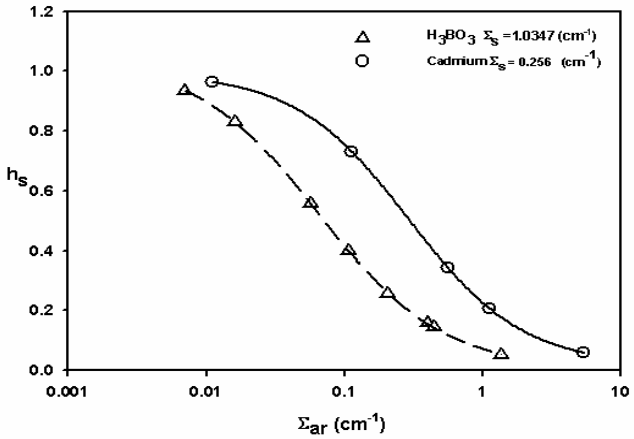

Fig.3 . h_s coefficients as a function of the samples relative macroscopic thermal neutron cross section (maximum relative error < 10%)

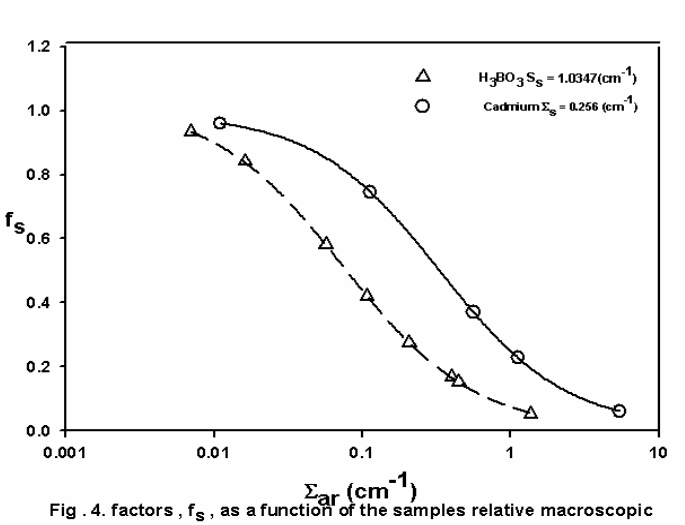

Fig . 4. factors , f_s , as a function of the samples relative macroscopic thermal neutron cross section (maximum relative error < 10%)

in an aqueous medium. Figs 3 and 4 show that coefficients f_s and h_s are close to unity for weak values of Σ_{ar} and tend to zero for strong Σ_{ar}. The value of h_s, can be experimentally determined by BF_3 detector at the surface of the sample and at the surface of a standard sample. Then, the relative macroscopic thermal neutron absorption cross-section of the unknown sample can be estimated using the MCNP calculated data (Fig. 3) for the appropriate macroscopic cross- section of sample. Finally, the f_s can be predicted from the data shown in Fig. 4 as a function of the sample relative macroscopic thermal neutron absorption cross-section. In both Figs. 3 and 4 the difference between solid and dash lines is due to the two various values of Σ_s for dissolved cadmium and boric acid in water. Thus this method cannot be used for calculating absorption or scattering cross-sections. In the present work h_s and f_s values for the eight samples presented in the Table 2 were calculated by MCNP code. Then the value of f_s was calculated by replacing the value of h_s in both dashed and solid graphs. From the comparison of data in Table 2, very good agreement (with a relative error less than 10%) was observed. Using the formalism and the explanation presented in the introduction it is possible to calculate the thermal neutron self- shielding.

Table 2. Comparison between simulated and calculated values for f_s

Material is solved in water	Simulation f_s	Calculation of cadmium graph f_s	Calculation of H_3BO_3 graph f_s
10(g) NaCl	.929	.934	.936
15(g) NaCl	.898	.908	.906
20(g) NaCl	.877	.881	.886
20(g) NaCl +5 (g) H_3BO_3	.553	.569	.574
20(g) NaCl +5 (g) H_3BO_3	.408	.423	.424
20(g) NaCl +5 (g) H_3BO_3	.323	.338	.329
50(g) Eu	0610.	.0560	.0590
50(g) Hf	.637	.663	.664

4. Conclusions

When a bulk sample is irradiated with a neutron source, the self-shielding phenomena occur. Analysis can be carried out based on the relative method where information about the sample material to be analyzed has to be known. Because one of the aims in the development of a methodology in this work is that material is to be analyzed quantitatively and without *a priori* information on the composition, other methods cannot be used, as a whole or partially. In the present work by using Monte Carlo simulations and also neutron flux measurements at the surface of the sample a method has been developed for neutron self-shielding calculations.

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