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

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Abstract

In this paper bulk sample prompt gamma neutron activation analysis (BSPGNAA) was applied aqueous sample analysis using relative method. For elemental analysis of unknown bulk sample, gamma self shielding coefficient was required. Gamma self shielding coefficient depends on these quantities, gamma attenuation coefficient (μ), solid angle between detector and sample (Ω), full energy peak detection efficiency (ε), gamma transmission factor of source to detector (T) and correction factor in calculation of any aforementioned. Using relative technique, Ω was omitted. Relative detector efficiency was determined with internal mono standard method by measuring prompt gamma of H and Cl elements in the standard samples. Gamma mass attenuation coefficient of unknown samples was estimated by an approximative method and also by MCNP code calculation. The required Correction factor in gamma mass attenuation calculation was obtained by MCNP code. The proposed methodology can be used for the determination of the elemental concentration of unknown aqueous samples by BSPGNAA where knowledge of the gamma self shielding within the sample volume is required.

Keywords: *Prompt gamma neutron activation analysis, Gamma self shielding, Gamma mass attenuation, MCNP code, Unknown sample.*

1. Introduction

Prompt gamma-ray neutron activation analysis (PGNAA) is a non-destructive nuclear analytical technique for the determination of elements in a variety of samples. The method is based on the measurement of prompt gamma-ray emitted following neutron capture by the elements present in the sample. The energies of the gamma-ray are characteristic of the isotopes of the elements and their intensities are proportional to their concentration. Therefore, the prompt gamma-ray spectrum from a sample that carries quantitative as well as qualitative information of the elements present in the sample PGNAA is useful for the analysis of samples containing low *Z* elements, and elements like B, Cd, Si, P, S, Gd and Hg which have extremely good analytical sensitivity due to their large neutron absorption cross-section. However, this technique is also applicable to

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heavier elements. The detection limits for the later elements extend to ppb levels [1, 2].

In this work a point source was applied for neutron irradiation of sample. Therefore, bulk samples were used as increasing the rate of neutron absorption by target nucleus and decreasing the neutron irradiation time. In the voluminous samples neutron self shielding (f_n) and gamma self shielding (F_γ) phenomena arise [2, 3]. For elemental analysis of sample these coefficients should be determined. This work focused on gamma self shielding calculation. It is assumed that any prior information is not accessible for materials which can be solved in water. On the other hand, the combination and matrix of present elements in the sample were unknown. Some methods have already been developed for gamma and neutron self shielding measurements [4-7]. But in all of them the prior information should be accessible of the existence elements in sample. In paper [8] neutron and gamma self shielding has been determined for unknown samples, but there the samples have been activated by a neutron beam inside a reactor. These experiments were done in two stages through a relative method. Measurements were first performed on aqueous bulk samples and then on a reference sample of pure water. Using the following equation and the PGNAA technique the concentration of elements was determined.

$$\frac{m_a}{m_H} = \frac{\dfrac{A_a M_a}{t\varphi_s I_a N_A \sigma_a \theta_a f_n \varepsilon_a \Omega_a F_{\gamma a}}}{\dfrac{A_H M_H}{t\varphi_s I_H N_A \sigma_H \theta_H f_n \varepsilon_H \Omega_H F_{\gamma H}}} \qquad \textbf{(1)}$$

M = molar mass of element. (Kg mol^{-1})
I_γ = relative gamma-intensity
F_γ= gamma self shielding
N_A = Avogadro number (mol^{-1})
σ_a= microscopic absorption cross section (m^2)
θ = abundance of the capturing isotope
f_n = neutron self-shielding factor
φ_s = surface neutron flux (m^{-2} s^{-1})
t = irradiation/counting time (s)
A = net full-energy peak area
m = mass in the sample (Kg)
ε = full-energy peak detection efficiency
Ω = solid angle between detector to sample

Subscription (a) refers to the element of interest in the unknown sample and (H) refers to the Hydrogen in the reference sample. The quantities of f_n and φ_s were obtained using MCNP code calculation and measuring the thermal neutron flux on the surface of sample by BF_3 detector. The authors have already presented details of these calculations [9]. In the both stages of the experiment, detector-sample position was the same, so that Ω coefficient is the same in both numerator and denominator of equation 1. Gamma self shielding coefficients (F_γ) is defined in the following equation:

$$F_\gamma = \frac{T}{f_\gamma} \qquad \textbf{(2)}$$

In which T is transmission factor and f_γ is an improved coefficient required for T calculations. T is calculated for each material of the objects for the specified energies and used to determine the probability that the gamma reaches the detector without any interaction along a way and it is defined as equation (3):

$$T = \exp(-\mu x) \qquad \textbf{(3)}$$

In which μ is the gamma attenuation coefficient and x is the thickness of attenuating material. Furthermore, for unknown aqueous samples F_γ is determined by the said factor calculation.

2. Materials and methods

2.1. PGNAA System

This experiment was carried out in Isfahan Nuclear Science & Technology Research Institute (NSTRI), Reactor and Accelerators Research and Development School, Atomic Energy Organization of Iran. Point neutron source ^{252}Cf with 1.7×10^8 (n/cm^{2}s) flux and average energy 2.348 MeV was used for neutron irradiation of sample. Neutrons should be thermalized by moderator such as graphite or water between source and bulk sample. But this method caused thermal neutron flux to be reduced in spot of the analyzed elements. Another solution is that neutrons were thermalized in the bulk sample material itself. Accordingly, a ^{252}Cf point source was located in the center of cylindrical container with 9.1 cm and 17.2 cm, radius and height, respectively. Neutrons thermalized a long passing material and absorb with existent elements in sample. Gamma-ray detection was done using P type HPGe detector. The relative efficiency and resolution of the detector at 1333 keV ^{60}Co was 13.8 % and 2.5 keV respectively. Detector connected to a multi channels analyzer (MCA) with 4000 channels. The MCA was energy calibrated using ^{60}Co and ^{137}Cs sources. To protect the detector from neutrons, it was covered with one centimeter boron as shield. Prompt gamma spectra were analyzed using Mastro software and subsequently the areas under different peaks were obtained. Detector was located in 25 cm of sample. Schematic diagram of system used is shown in Fig. 1. Experiments were done in two stages. In the first stage measurements were carried out in pure water as a reference sample and in the next stage the unknown aqueous sample. Three standard salt water samples with concentrations 10, 15 and 20 gr/L NaCl were used as unknown samples. Each sample was irradiated and measured in 1000s.

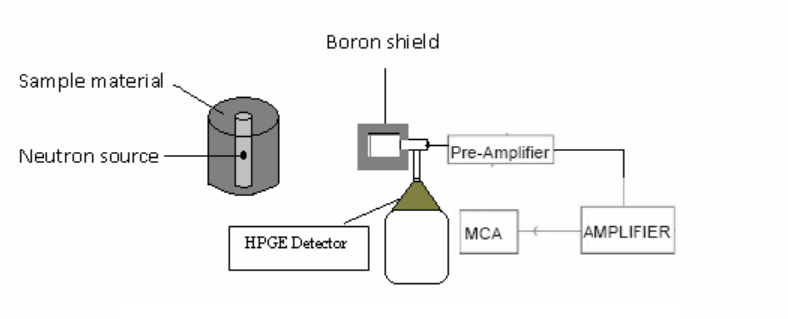

Fig.1. Geometrical setup of experiment

2.2. Simulations

MCNP version 4C software was applied to simulate the interaction between gamma and neutron with material [10]. Library cross section endf/bvi was used in MCNP4C software. The code enables us to perform for three dimension simulation of source, sample and detector in varied configuration of geometries. MCNP code is useful for PGNAA experiment simulation because it follows gamma and neutron particle. To measure the gamma flux F5 tally was used. The parameter F5 refers to a deterministic estimate of the flux at a point in space in units of cm^{-2} per source gamma. Using F5 tally enables us to measure the totally reached flux to the detector and the uncollided flux which passes through distance between source and detector without any interaction.
The relative errors of the computations were kept below 5% and all statistical tests for the estimated answers were passed. μ and fγ coefficients were obtained with simulation by MCNP code in this work.

3. Results and discussion

3.1. The Transmission Calculation

Total attenuation coefficient (μ) is the sum of the probability of photoelectric effect, Compton scattering and pair production. μ is the probability of gamma interaction in unit of length. Total gamma mass attenuation (μ_m) is given by equation (4) and its unit is (cm^2/g).

$$\mu_m = {}^{\mu}/_{\rho} \qquad (4)$$

In which ρ is density and μ is gamma attenuation coefficient. If single energy gamma-ray enters a material with thickness of x, a fraction of it exp (-μx) without any interaction will reach to the detector. μ is calculable using this equation. In this work μ has been calculated using MCNP code and totally assumed geometry was simulated by the code. This geometry contains a flat plate source that produces beam of single energy gamma-ray is shown in Fig. 2. Initially, a point

detector (F5 tally) was located in front of the source and the primary flux (I_0) was estimated. Then attenuating material with thickness of x was put in front of the source and the uncollided flux (I) was estimated. For a certain material the amount of μ is calculated through the equation (5).

$$I = I_0 \exp(-\mu x) \quad (5)$$

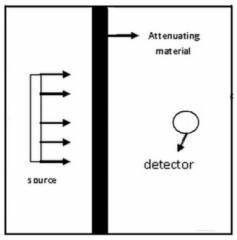

Fig. 2. The assumed geometry simulated by MCNP code containing a source, an attenuating material and a detector (F5 tally).

In Fig. 3 the μ_m as a function of Z is plotted for three different energies. The dashed lines are drawn using XCOM software and the points were produced by MCNP code [11]. Fig.3 shows the coincidence between computations data of XCOM software with calculated data by MCNP code. This implies the applied method is completely correct. In Fig. 3 the μ_m for high energy gamma is plotted against Z. This figure illustrates that the μ_m was approximately constant relative to Z variation. The above method determines the μ for any known sample but here it is determined for unknown sample. Furthermore, an approximated method was used to estimate for an unknown aqueous sample. As seen in Fig. 3 for high energy the μ_m calculation may be possible using the average amount of Z equal to 10.

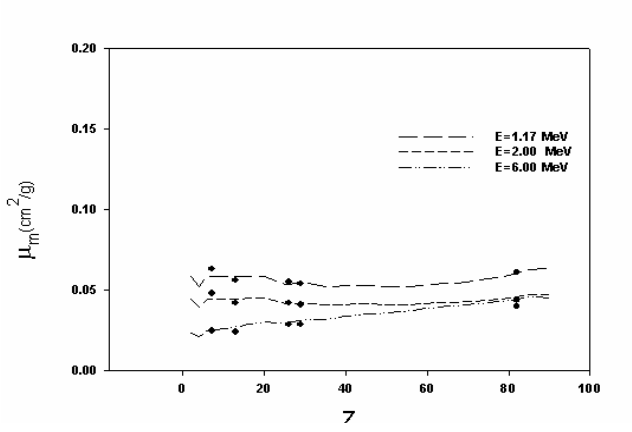

Fig. 3. The mass attenuation coefficient (μ_m) is plotted as a function of atomic number (Z). The dashed curves represent the results corresponding to XCOM computation.

This estimation is applicable when the error in the transmission coefficient calculation is less than 10 percent. The μ_m coefficient was estimated using the above error and then μ was determined from μ_m time ρ. Finally T was calculated using the both real and estimated μ. With comparison of both T, the interval less than 10 percent error was determined. A 2000 (Kg/m^3) density was used which is appropriate range for aqueous samples that those thickness is equal to cylinder radius. The limitation of atomic number was shown in Table 1 (in energies between 1-10MeV and for samples with density under 2000Kg/m^3). In energy 10MeV the range of atomic number that can be used is more limited than other energy ranges (4<Z<20).

Table 1.

E(MeV)	Z_{min}	Z_{max}
1	6	70
2	4	90
5	2	30
7	4	20
10	4	20

A deep glance shows that for density less than 2000(Kg/m^3), except Rb and Cs the atomic number of every element lies between 2 to 20 (Fig. 4). On the other hand for aqueous samples with density less than 2000 (Kg/m^3) the range of atomic number is between 2 to 20.

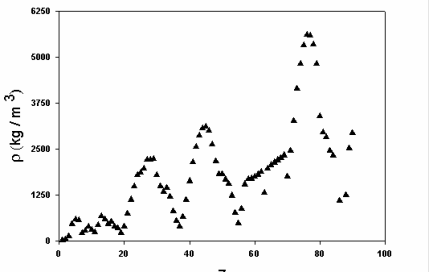

Fig. 4. Density of materials as a function of the atomic number.

Note that the aqueous samples are perfectly homogeneous and the emitted neutrons are isotropic. After neutron absorption the whole volume of the sample will active and then emit prompt gamma-ray. As a result, the aqueous sample will act as a volume gamma source. The fact is that the pass way for emitted gamma in the container could not be determined exactly. Anyway the pass way for the gamma-ray could not be out of 0 to 9.1 cm.
We suppose that the pass way for the gamma ray in the container to be 9.1 cm. By this assumption an error will appear in calculating of T factor. So f_γ is used to improve the created error in the calculation. This correction coefficient was calculated by MCNP code. The calculation with MCNP code contains two

simulation stages. In first stage a volume homogeneous-gamma source was simulated using cylinder container (9.1 cm radius and 17.2 cm height). Also in this stage of simulation it is assumed that there is not any attenuation material and the gamma-ray reached the detector without collision (I_0). In the next stage the cylinder was filled with attenuating material and the uncollided flux of gamma-ray, (I), was calculated by F5 tally. Assuming the pass way for gamma ray in the material is 9.1cm, therefore the coefficients I, I_0 and μ will satisfy the equation $I=I_0 exp\ (-9.1\ \mu)$. A simple calculation was shown that this assumption was not satisfying the above equation, and then a correction coefficient was entered in the equation so that both side of equation were equal.

$$I = \frac{I_0 \exp(-\mu x)}{f_\gamma(\mu)} \qquad (6)$$

With various simulations the attenuation material was changed and f_γ was calculated in every case. The simulations show that f_γ is a function of μ. In Fig. 5 f_γ versus μ is plotted. As seen in Fig. 5, increasing the μ decrease the f_γ to zero and decreasing the μ increase the f_γ to unity. The estimated f_γ using sigma plot software is given by equation (7).

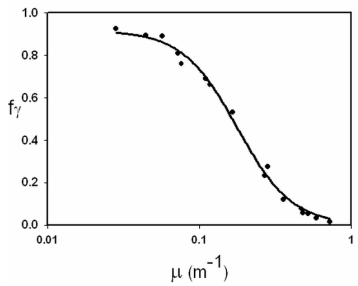

Fig. 5. f_γ as a function of attenuation coefficient μ.

$$f_\gamma = 5.01236 \times 10^{-3} + (0.917172 - 5.01236 \times 10^{-3})/(1 + (\mu/0.177423)^{2.4146}) \qquad (7)$$

The equation (8) is generally used to determined the gamma self shielding of unknown sample.

$$F_\gamma = \frac{T}{f_\gamma} = \frac{exp(-\mu \times 9.1)}{f_\gamma} \qquad (8)$$

In the first step μ_m of unknown sample was estimated in specified high energy gamma-ray and then μ was calculated from μ_m times ρ. f_γ was calculated by inserting μ in equation (7). Finally gamma self shielding (F_γ) was calculated according to equation (8).

3.2. Calculation of relative energy peak efficiency

PGNAA required a gamma-ray detection efficiency curve covering a wide energy range, i.e. from 500 keV to 10 MeV. The internal mono-standard method is currently used to determine the concentration of other elements present in the known sample [5]. The existing mass of elements in the samples is clear to us. Replacing the mass and other characteristics of elements into the equation (9), the $\varepsilon_{a_E}/\varepsilon_{H_{2.22}}$ was exactly calculated.

$$\frac{\varepsilon_{a_E}}{\varepsilon_{H_{2.22}}}=\frac{\dfrac{A_{a_E}M_a}{t\varphi I_a N_A \sigma_a \theta_a f_n F_{\gamma a}\,\Omega m_a}}{\dfrac{A_{H_{2.22}}M_H}{t\varphi I_H N_A \sigma_H \theta_H f_n F_{\gamma H}\,\Omega m_H}}=\frac{I_H\,\sigma_H\theta_H\,F_{\gamma H}m_H\times A_{a_E}M_a}{I_a\sigma_a\theta_a F_{\gamma a}\,m_a\times A_{H_{2.22}}M_H} \qquad (9)$$

Subscriptions (a) and (E) refer to the element of interest in the known sample and its energy peak respectively, and subscriptions (H) and (2.22) refer to the Hydrogen in the known sample and its energy peak respectively. The equation (9) was calculated by using the equations (1) and (2). In the method the elements as well as the sample are irradiated and measured under the same conditions, so that φ, t, N_A and f_n cancels out. Using equations (3) and (8) T and F_γ can be obtained respectively. The obtained relative efficiency calibration using γ-rays from aqueous solution of NaCl is given in Fig. 6. The ratio of $\varepsilon_{a_E}/\varepsilon_{H_{2.22}}$ is plotted as a function of energy using the software Sigma-plot and finally the equation (10) is extracted (see Fig. 6).

$$\frac{\varepsilon_{a_E}}{\varepsilon_{H_{2.22}}}=\exp\left[1.2514\ \exp(-0.1152\times E)+110.8375\ \exp(-3.6852\times E)\right] \qquad (10)$$

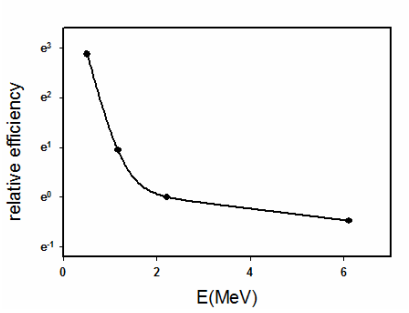

Fig. 6. The relative full energy γ-ray detection efficiency calibration curve from aqueous solution of NaCl.

Conclusion

In the present work a simple method for determination of gamma self-shielding for unknown aqueous bulk sample was developed. In the method a simplified Monte-Carlo simulation to estimate mass attenuation coefficient, μ_m, and the transmission coefficient, T, was presented. Of advantages of the method is sample geometry flexibility provided by the MCNP input geometry and an extension of the useful range of material applications to gamma absorbing materials. The internal mono-standard method was used to determine the curve of relative efficiency. Further, a method for calculating gamma self shielding of the unknown aqueous sample was presented. In spite of a relative method in aqueous bulk samples was used, but determination of gamma self shielding is extendable for an absolute method in other types of samples in the form of solids or solutions.

Acknowledgements

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