Development of a non-invasive method for the determination of the macroscopic neutron cross sections of a sample matrix in large-sample prompt-gamma neutron activation analysis

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In order to correct for neutron self-shielding in large-sample prompt gamma NAA, a method has been developed to determine the macroscopic scattering and absorption cross sections, i.e., Σ_s and Σ_a , using four Cu flux monitors placed around the sample. With Monte Carlo computations, the neutron densities throughout the sample and the resulting and the corresponding self-shielding factor as calculated from the Σ_a and Σ_s as obtained through the Cu monitors were compared to the true values. The derived Σ_a and Σ_s were found to be sufficiently accurate as long as $\Sigma_t = \Sigma_a + \Sigma_s$ was less than 0.6 cm^{-1} and Σ_s/Σ_t was greater than 0.1.

Introduction

If large-sample prompt-gamma neutron activation analysis (LS-PGNAA) is to determine accurate element fractions of large samples with an unknown homogeneous matrix composition, corrections have to be carried out for neutron self-shielding and gamma-attenuation. In large samples these corrections cannot be carried out separately.¹ For each position inside the sample the influence of the sample parameters for neutron self-shielding and gamma-attenuation has to be determined. The influence of the sample parameters for neutron self-shielding is expressed in a correction factor for neutron self-shielding. Another correction factor is determined for the influence of gamma-attenuation. After integrating the product of these correction factors over the sample volume, a correction factor for the entire sample is derived.

In order to calculate the local correction factors, the sample parameters with respect to gamma-attenuation and neutron self-shielding have to be determined. For gamma-attenuation, the attenuation coefficient as a function of gamma-energy must be known. For neutron self-shielding as occurring in a neutron beam, the macroscopic scattering and absorption cross sections (Σ_s and Σ_a), as well as the effective scattering mass (M_e) must be known. The influence of M_e is studied in a separate paper where it is found to be negligible for all practical purposes.²

In this paper a method is described to determine the sample material parameters Σ_s and Σ_a through flux monitoring outside the sample. First a Monte Carlo survey with our in-house computer program BUDA³ was carried out to determine the neutron density $n(r)$ at four positions outside the sample. For these four positions empirical relations were found depending on Σ_s and Σ_a . Similar relations were determined using

MCNP⁴ and published earlier.⁵ For that reason, the details of these relations are omitted from this paper, the interested reader is referred to the first author's Ph.D. Thesis.⁶

Secondly, using these relations, Σ_s and Σ_a can be determined from the $n(r)$ measured in those four positions. With these "derived" Σ_s and Σ_a , as well as with the "true" values, $n(r)$ within the sample material was simulated using BUDA and compared. This comparison was performed with the purpose of the methodology in mind, since possible discrepancies between the true and the experimental values of the parameters will not propagate in a simple way to the final analysis results.

Experimental

Neutron density distributions were simulated for 19 series of 54 hypothetical sample materials each. Within each series, the values of Σ_s and Σ_a varied from 10^{-6} to 10^4 cm^{-1} in 54 steps, but the

$$\Sigma_s/\Sigma_t = \Sigma_s/(\Sigma_a + \Sigma_s)$$

ratio was kept constant. This ratio ranged from 0.20 through 0.97 over the 19 series. All atoms in sample materials in these simulations are treated as rigidly bound, i.e., the scattering was isotropic in the laboratory system and the effect of thermal motion of the scattering atoms was absent.

A cylindrical PTFE (polytetrafluorethylene) container was modeled in all simulations with an outer height of 20 cm, an outer diameter of 10 cm and a wall thickness of 0.35 cm. A purely thermal, 2.54 cm diameter, homogeneous neutron beam with a Maxwell-Boltzmann energy spectrum at room temperature was modeled as impinging on the container perpendicular to the symmetry axis of the cylinder (Fig. 1). The negative

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Z-direction was towards the origin of the neutron beam, the Y-direction was parallel to the symmetry axis of the sample and the X-direction was perpendicularly to both other axes. The detector is thought to be placed at the positive X-axis.

Four copper foils were modeled outside the sample. The angles that the lines from the middle of the copper foils to the middle of the sample made with the neutron beam were 0, 45, 135 and 180 degrees. The foil at the position of 0 degrees was modeled between the sample and the neutron source.

A square copper foil with sides of 1 cm and a thickness of 0.025 cm in the Z-direction was modeled at 0 and 180 degrees. Two other copper foils with sides of 0.16 cm and a thickness of 0.98 cm in the Y-direction were situated at 45 and 135 degrees (Fig. 1). These last copper foils were modeled as rods, because of limited object shapes being available in BUDA. All four copper foils have the same volume. For each copper foil the probability of the absorption of a neutron was tallied until the imprecision of the tally of the copper foil at zero degrees was smaller than 0.5%.

With the approach described above four empirical relations were determined between $n(r)$ outside the sample as a function of Σ_a and Σ_s , using basically the same approach and methods as in the earlier paper.⁵

Application of the relations found

To test the relations found, new simulations were carried out with five series of nine sample materials. In each series the ratio Σ_s/Σ_t is kept constant at 0.01, 0.10, 0.50, 0.90 and 0.99, respectively. For all sample materials $n(r)$ was calculated.

In the next step, for each sample material, experimental values of Σ_s and Σ_a were calculated by fitting the $n(r)$ at the positions of the copper foils with the four empirical relations found before, using least-squares fitting.

Using both the true and the experimental values, simulations were carried out to investigate the influence of the inaccuracy of the determination of Σ_a and Σ_s on $n(r)$. The sample material was divided in $10 \times 20 \times 10$ voxels in respectively the X, Y and Z direction. Each voxel had the dimensions $1 \times 1 \times 1$ cm³. Some voxels were not in the cylindrical sample material, but, since no neutrons were absorbed in the sample material at those voxels, this has no consequences.

The results were expressed as a function of d and r . Quantity r is the distance from the middle of the voxel to the central axis of the beam ($X, Y = 0, 0$), quantity d is the depth of the middle of the voxel with respect to the point where the beam enters the sample material (Fig. 2).

Fig. 1. Setup geometry of the simulation: the PTFE bottle being irradiated with a neutron beam and the position of the copper flux monitors

Fig. 2. Illustration of quantities r and d in the sample; r is the distance from the middle of the voxel to the middle of the beam ($X, Y=0,0$); d is the depth of the middle of the voxel with respect to the point where the beam enters the sample material

Three parameters were determined to test the method of the determination of Σ_s and Σ_a on $n(r)$ inside the sample material. These were in order of importance: the average neutron density in the sample material (represented by a generalized self-shielding factor f), the d coordinate of the centre of mass (d_{CM}) of $n(r)$ and the average neutron distance from that centre of mass (Δr and Δd). With parameters d_{CM} , Δd and Δr it can be shown whether the shape and position of $n(r)$ are properly reproduced, the relevant aspect being the part of $n(r)$ seen by the detector, and the detector and collimator being placed at the positive X axis.

The generalized self-shielding factor applies to objects that are not completely illuminated by the neutron beam and is defined as:

$$f = \frac{RV}{\sigma_{a,0} N v_0 n V} \quad (1)$$

where f is the self-shielding factor, R is the capture rate density, $\text{m}^{-3} \cdot \text{s}^{-1}$; V is the sample volume, m^3 ; $\sigma_{a,0}$ is the microscopic absorption cross section, m^2 , at neutron velocity $v_0 = 2200 \text{ m/s}$, n is the neutron density, m^{-3} , and N is the atom density, m^{-3} .

The d_{CM} parameter was calculated with Eq. (2). The average neutron distance from the center of mass in the d direction, Δd , was calculated with Eq. (3). The value for Δr can be calculated with the same equation in which d is replaced by r :

$$d_{CM} = \frac{\sum_v n_a d}{\sum_v n_a} \quad (2)$$

$$\Delta d = \frac{\sum_v n_a |d - d_{CM}|}{\sum_v n_a} \quad (3)$$

where n_a is the total number of neutrons absorbed; d is the depth of the position where the neutron absorption occurs, m, Δd is the average neutron distance to CM in the d -direction, m; and v is the voxel index number.

Results

In Table 1, example results are shown for the $\Sigma_s/\Sigma_t = 0.50$ test series. As can be seen, the cross sections are reproduced quite well at this setting, except when the total cross section exceeds 2 cm^{-1} .

Self-shielding factors f for all simulated sample materials, as computed by BUDA, are compared in Table 2 for the true and derived Σ_s and Σ_a . As can be seen, the corresponding f_t and f_d , and thus the average neutron density in the sample, agree with each other to within 4% for sample materials with a Σ_t smaller than 0.6 cm^{-1} and a Σ_s/Σ_t of 0.10 or larger.

The differences in the position of the center of mass of the neutron density distribution on the Z axis, as expressed in d_{CM} can be seen in Fig. 3. The differences are smaller than 0.1 cm for all sample materials with Σ_t smaller than 0.6 cm^{-1} and Σ_s/Σ_t larger than 0.01. Such differences are small as compared to realistic collimator opening sizes, which are typically in the order of a few cm.

In Figs 4 and 5, the differences between Δd and Δr are shown as calculated with the true and derived Σ_s and Σ_a . As can be seen in Fig. 4, the difference in Δd is smaller than 0.2 cm for Σ_t smaller than 0.6 cm^{-1} and Σ_s/Σ_t larger than 0.01. As can be seen in Fig. 5 the difference in Δr is smaller than 0.2 cm for almost all sample materials as long as Σ_t is smaller than 0.6 cm^{-1} and Σ_s/Σ_t larger than 0.01.

Discussion

Inaccuracies in the reconstructed neutron flux distribution will affect the elemental mass fractions obtained as analysis results in the end.

A first-order source of error is the average neutron density in the sample, being linearly proportional to the count rates. This average is represented by the self-

shielding factor f , and the results indicate that accuracies of better than 4% can be realized in this respect if Σ_t is smaller than 0.6 cm^{-1} and Σ_s/Σ_t is 0.10 or larger.

The detector will be placed at a distance of 10 cm or more from the centre of mass of the sample material, and the collimator opening size is likely to be in the order of cm. In a first-order approximation it is clear that if the center of mass of the neutron flux distribution d_{CM} remains in front of the collimator opening, no change is expected in the measured element mass fraction, since the total detector efficiency remains virtually the same for all positions of d_{CM} in front of the collimator, as well as for positions shielded by the collimator.

The contribution of the higher-order moments of the spatial neutron flux distribution, expressed in Δd and Δr , to the final error of the elemental mass fractions, will in part be due to gamma-attenuation. The error in the gamma attenuation correction due to an error in Δr can be calculated for a sand sample, with a density of $2330 \text{ kg}\cdot\text{m}^{-3}$. The gamma-attenuation coefficient for 1, 2 and 5 MeV is 0.0634 cm^{-1} , 0.0447 cm^{-1} and 0.0297 cm^{-1} , respectively. Assuming an average sample thickness of 5 cm and an erroneous Δr of 2.2 instead of 2.0 cm, the influence on the final element mass fraction will then be respectively 1.2%, 0.8%, and 0.6%.

Table 1. Example results of true and derived cross sections at $\Sigma_s/\Sigma_t = 0.50$

True				Derived		
Σ_a, cm^{-1}	Σ_s, cm^{-1}	Σ_t, cm^{-1}	Σ_a, cm^{-1}	Error, %	Σ_s, cm^{-1}	Error, %
0.005	0.005	0.01	0.0049	6	0.0052	6
0.01	0.01	0.02	0.0102	4	0.0103	6
0.03	0.03	0.06	0.0303	3	0.0313	3
0.05	0.05	0.1	0.0482	1	0.0533	1
0.1	0.1	0.2	0.093	2	0.104	2
0.3	0.3	0.6	0.275	2	0.280	2
0.5	0.5	1	0.425	3	0.464	3
1	1	2	0.95	6	1.08	9
3	3	6	1.9	10	2.1	10

The errors are one standard deviation.

Table 2. Difference in self-shielding factors for different sample materials

Σ_t, cm^{-1}	$\frac{f_t - f_d}{f_t}$				
	$\Sigma_s/\Sigma_t=0.01$	$\Sigma_s/\Sigma_t=0.10$	$\Sigma_s/\Sigma_t=0.50$	$\Sigma_s/\Sigma_t=0.90$	$\Sigma_s/\Sigma_t=0.99$
0.01	-1.2%	-0.6%	0.1%	0.4%	0.01%
0.02	-2.5%	-1.2%	-0.1%	0.5%	0.6%
0.06	-6.3%	-1.4%	-0.1%	1.9%	0.5%
0.1	-11%	-1.9%	0.9%	0.7%	1.3%
0.2	-27%	-3.7%	2.8%	1.4%	1.9%
0.6	-119%	-43%	7.3%	-1.6%	-5.9%
1	-138%	-125%	15%	1.9%	-1.9%
2	83%	15%	-0.7%	8.0%	-36%
6	63%	11%	-1.3%	-12%	-211%

f_t was calculated with the true Σ_s and Σ_a , f_d was calculated with the derived Σ_s and Σ_a .

Fig. 3. Difference in d co-ordinate of CM of the neutron distribution calculated with the true Σ_s and Σ_a ($d_{CM,t}$) and with the derived Σ_s and Σ_a ($d_{CM,d}$) versus the true Σ_t for the sample material simulated

Fig. 4. The difference between the width of the neutron distribution in the d-direction calculated with the true Σ_s and Σ_a (Δr_t) and with the derived Σ_s and Σ_a (Δr_d) versus the true Σ_t for sample materials simulated with both the derived and the true Σ_s and Σ_t

Fig. 5. The difference between the width of the neutron distribution in the r -direction calculated with the true Σ_s and Σ_a (Δr_I) and with the derived Σ_s and Σ_a (Δr_I) versus the true Σ_t for sample materials simulated with both the derived and the true Σ_s and Σ_t

An error in Δd implies neutrons being captured behind the collimator that were supposed to be captured in front of the opening or vice versa. This effect can be minimized to arbitrarily small levels by using a generous collimator opening. If count rates turn out to be too high as a result, the detector could be moved back with the collimator.

In this work, the sample material is assumed to be homogeneous, even though large-sample activation analysis is useful mostly in case of inhomogeneous materials. If the matrix is homogeneous but the element to be determined is not, at the 1 g level, the method proposed here would be of use. Also, the effects of inhomogeneity can be reduced by rotating the sample during the measurement, so that a volume average would be obtained. Such procedures are applied in the real analysis as performed with the existing system for large-sample activation analysis in Delft.

Conclusions

With the proposed methodology, LS-PGNAA results that are accurate to within 4% can be expected as long as Σ_t is smaller than 0.6 cm^{-1} and Σ_s/Σ_t larger than 0.1. By

far the majority of sample materials will satisfy these criteria.

The 4% mentioned is due to discrepancies between true and reconstructed average neutron densities in the sample, the other discrepancies being of much less importance. In practice, the inaccuracy of the methodology for gamma self-absorption correction will constitute an additional contribution to the inaccuracy budget.

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