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SCATTERING AND ABSORPTION EFFECTS IN NEUTRON BEAM ACTIVATION ANALYSIS EXPERIMENTS

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We have investigated the effects of scattering and absorption in neutron beam activation analysis experiments, both by direct measurement and by Monte Carlo simulation. Significant sensitivity enhancements occur for thin disks placed at 45° to the beam but very much smaller effects occur for spheres. The agreement between measurement and calculation is generally good.

Introduction

About ten years ago, users of the University of Maryland-National Institute of Standards and Technology (UMd-NIST) neutron capture prompt gamma-ray activation analysis (PGAA) facility found a systematic error in some of their measurements.[1] They discovered that element sensitivities (count rates per unit mass) for hydrogenous materials were enhanced relative to values for nonhydrogenous materials. Recent experiments have shown that the degree of enhancement depends both on the H content of the sample and on its size and shape.[2]

Because of the large scattering cross-section of H (the bound atom value is 80 b), it was originally suspected that neutron scattering by H was somehow responsible for the observed enhancements. The neutron beam at the UMd-NIST facility is, however, well thermalized so that inelastic neutron scattering, including moderation of epithermal neutrons, is unlikely to be the principal explanation. The problem was recently reexamined[3] with Monte Carlo methods to calculate absorption probabilities in a variety of slab-shaped samples. Results of these calculations showed that sensitivity enhancements, comparable in magnitude with those observed experimentally, could be expected solely due to elastic scattering, i.e., scattering without change in energy; they also confirmed that the degree of enhancement depends on the scattering density and on the size of the sample.

Both neutron absorption and neutron scattering affect sensitivity by modifying the average fluence rate within a sample. In this paper we examine these effects restricting our attention to the special case of beam geometry. We describe PGAA experiments and compare them with Monte Carlo calculations.

For a pure absorber, reaction rates may be corrected using the self-shielding factor, f_A, which is readily calculated for simple geometries.[4] This quantity is a function of the macroscopic absorption cross section

$$\Sigma_A = \Sigma N_i \sigma_{Ai}, \qquad (1)$$

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where N_i is the number of atoms of element i per unit volume, σ_{Ai} is the absorption cross section per atom of element i, and the sum is over all elements in the material. It also depends on the shape and size of the sample. For a sphere of radius R, the self-shielding factor is

$$f_A^{(S)} = (3/x^3)[x^2/2 - 1 + \{1 + x\}e^{-x}], \quad (2)$$

where $x = 2R\Sigma_A$. For a flat plate of thickness t_P, normal to the beam, the appropriate expression is

$$f_A^{(P)} = (1/x)[1-e^{-x}], \quad (3)$$

where $x = t_P\Sigma_A$. For a disk of thickness t_D and radius R, inclined at an angle α to the beam (such that $\alpha = 0$ if the disk is normal to the beam), two approximations have been used.[5,6] Approximation 1 is to use the formula for a plate, eq. (3), substituting the effective thickness, $t^* = t_D/\cos(\alpha)$, for t_P. Approximation 2, in the spirit of reference 7, is

$$f_A^{(D)} = [Rf_A^{(P)} + t^* f_A^{(C)}] / [R + t^*], \quad (4)$$

where $f_A^{(P)}$ is evaluated with $x = t^*\Sigma_A$, and $f_A^{(C)}$ is the self-shielding factor for a cylinder of radius R whose axis is normal to the beam direction:

$$f_A^{(C)} = (2/x)[I_1(x) - L_1(x)]. \quad (5)$$

Here $x = 2R\Sigma_A$ and I_1 and L_1 are modified Bessel and Struve functions respectively.[8] In this paper we compare values of f_A for purely absorbing disks inclined at an angle of 45°, determined by the Monte Carlo method, with values obtained by using the two approximations.

Whereas neutron absorption decreases sensitivities, neutron scattering can either increase or decrease absorption reaction rates.[3] To account for the combined effects of scattering and absorption within the sample, the measured absorption reaction rate must be divided by a correction factor, f, to obtain the "ideal" reaction rate. The quantity f is simply the ratio of the average fluence rate within the sample to the incident fluence rate. For a pure absorber it is equal to f_A but in general it depends on the differential scattering cross section of the sample and cannot be simply calculated. To quantify f for typical matrices that scatter and absorb neutrons, we have performed Monte Carlo calculations and PGAA experiments on disks inclined at 45° to the beam and on spheres. The calculations for disks are new and provide reliable estimates of f for inclined disks which both scatter and absorb.

Materials and methods

Calculations

The Monte Carlo calculations were performed with a computer code modeled on the neutron multiple scattering program MSCAT85.[9] The new program currently handles three sample geometries (sphere, disk and slab) and may be used to simulate experiments on samples placed in a beam or in an isotropic field of neutrons. In its "entry" mode, neutrons enter from the outside, their histories are tracked, and contributions to the reaction rate are accumulated. We have also made calculations in an "escape" mode where neutrons are born

within the sample, tracked through a number of collisions and finally allowed to escape. The results of "entry" and "escape" calculations are simply related, and the "escape" method has certain advantages. For simplicity, we have assumed (as in previous calculations) that there is no change in energy on scattering, and that the macroscopic scattering cross section, Σ_S (analogous to Σ_A), is isotropic. In this approximation the quantity f depends on the incident energy only to the extent that the total absorption and scattering cross sections are functions of energy.

Experiments.

The PGAA experiments were carried out using the UMd-NIST PGAA facility described in detail elsewhere.[10, 11] Gamma radiations were collected with a germanium detector (27% efficiency relative to a 7.6- x 7.6-cm NaI crystal) with a Nuclear Data (ND) 16K-channel analog-to-digital converter (ADC). The ADC was connected to a ND9900 workstation through an acquisition-interface module Ethernet multi-channel analyzer. Data reduction was performed using the workstation and a Digital Equipment Corporation VAX 11/730 computer with the ND software programs PEAK and PILEUP. Count rates were corrected for temporal fluence rate variations. Disks and spheres were prepared from tris-hydroxymethyl-aminomethane (THAM) which contains 9.15 (w/w) % H. Disks of a constant 1.27 cm diameter and ranging from approximately 0.1 to 1.3 cm in thickness were prepared with a commercial die. Eight spheres ranging in diameter from 0.48 to 1.27 cm were prepared with dies designed and fabricated at NIST.[6] The average measured density for disks and spheres of this material was 1.25 ± (0.10) g/cm^3. All samples were packaged in bags formed from Teflon film. Irradiation times were generally less than 30 min.

Results and discussion

Pure absorber

In PGAA experiments, prompt gamma-rays are collected during sample irradiation and samples are placed at 45° with respect to the neutron beam to minimize the combined effects of neutron and gamma attenuation. The determination of self-shielding factors for disks in this

Table 1.

Correction factors for purely absorbing disks inclined at 45° to a neutron beam

$R\Sigma_A$	$t\Sigma_A$	Approximation #1	Approximation #2	Average of approximations	Monte Carlo results
0.5	0.25	0.842	0.882	0.862	0.858
	0.5	0.717	0.831	0.774	0.770
	0.75	0.616	0.808	0.712	0.716
	1	0.535	0.796	0.666	0.684
1.0	0.25	0.842	0.863	0.853	0.850
	0.5	0.717	0.782	0.750	0.744
	0.75	0.616	0.733	0.675	0.667
	1	0.535	0.702	0.619	0.612

geometry is not straightforward and, as previously stated, two approximations and a Monte Carlo method were used to quantify f_A. Results for selected values of $R\Sigma_A$ and $t\,\Sigma_A$ are shown in Table 1. The Monte Carlo data indicate that approximation 1 is an underestimate and approximation 2 an overestimate of f_A. In all cases, the average of the two approximations compares well with the Monte Carlo value.

Scattering and absorption.

Materials generally possess non-zero values of Σ_S. For materials with low H content, the approximation $f = f_A$ is adequate but this is not the case if Σ_S is substantial. For disks and spheres of THAM, H sensitivities were determined by PGAA. Sensitivities for spheres of different sizes were identical within the errors associated with counting statistics. In addition, the average sensitivity value for the eight spheres, 1.11 (counts·sec^{-1}·mg^{-1}), was essentially identical to the average sensitivities for spheres of three other materials of differing H content[6] and it was therefore assumed that the correction factor for these spheres was approximately unity. Sensitivities for disks were normalized to the average value for spheres to yield correction factors for disks. The PGAA data for disks and spheres are presented in Fig. 1. In the same figure we also show Monte Carlo results for disks with $\Sigma_A = 0.0375$ cm^{-1}, the value for THAM obtained from eq. 1 using values of σ_A for neutrons with a velocity of 2200 m/s. Since the value of Σ_S for THAM is much less well known, we show results for three representative values (3.0, 4.5 and 6.0 cm^{-1}); these values of Σ_S were computed with an equation analogous to eq. 1 with H atom scattering cross-sections of 40, 60 and 80 b.

Similar trends are evident in the PGAA and Monte Carlo data shown in Fig. 1. Correction factors decrease with increasing disk thickness for disks ranging in thickness from

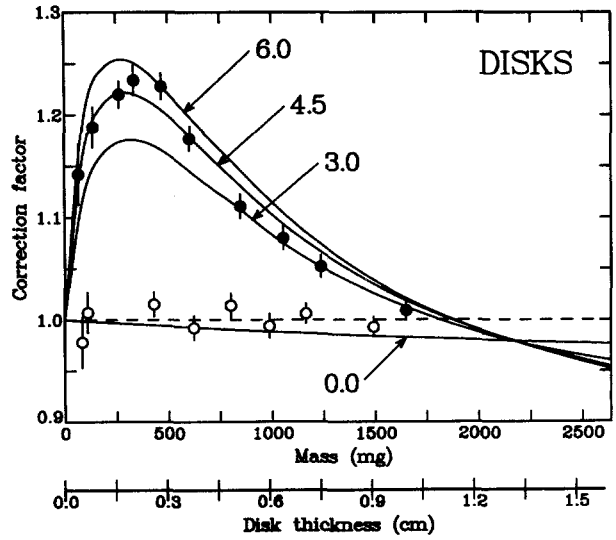

Figure 1. Correction factors for disks determined from Monte Carlo calculations with $\Sigma_A = 0.0375$ cm^{-1} for several values of Σ_S (0, 3, 4.5, and 6 cm^{-1}), and by PGAA for THAM disks (•) and spheres (○).

approximately 0.2 to 1.5 cm and decrease with decreasing disk thickness for very thin samples ($t < 0.1$ - 0.2 cm), indicating that correction factors are largest for relatively thin samples. The Monte Carlo results show a maximum correction factor for disks of 0.15 - 0.20 cm thickness whereas the PGAA data show a maximum at 0.20 - 0.25 cm. In Fig. 1 we also show Monte Carlo results for $\Sigma_S = 0$ cm^{-1}, i.e., the self-shielding factor f_A. Clearly $f > f_A$ for disks of THAM ranging in thickness from 0.0 to roughly 1.3 cm whereas $f < f_A$ thereafter. (That the correction factor for a 1.27 cm diameter, 1.27 cm thick disk is approximately equal to f_A is not unexpected as this sample shape closely resembles a sphere.) The maximum error in assuming that $f = f_A$ for relatively thin disks (if $\Sigma_S = 6$ cm^{-1}) is approximately 25%.

Correction factors for spheres of THAM, determined from Monte Carlo calculations[12] (with $\Sigma_A = 0.0375$ cm^{-1}), are shown in Fig. 2; again each curve represents a different value of Σ_S. Note that for a pure scatterer $f = 1$, independent of Σ_S. For spheres that both scatter and absorb, $f < f_A$ so that enhanced sensitivities should not be observed for spherical samples.

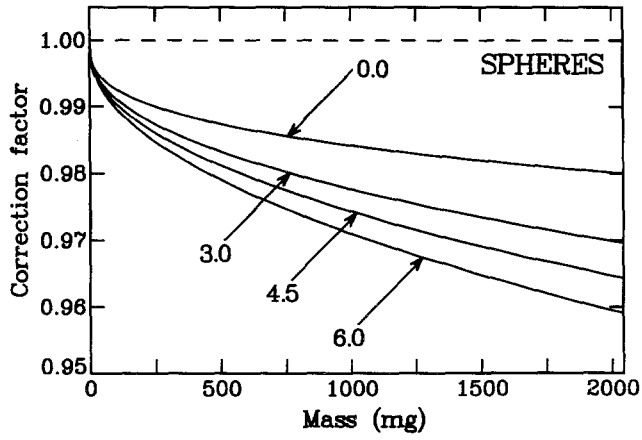

Figure 2. Correction factors for spheres determined using Monte Carlo calculations with $\Sigma_A = 0.0375$ cm^{-1}, for several values of Σ_S (0, 3, 4.5 and 6 cm^{-1}).

The error associated with assuming that $f = f_A$ is about 1% for a 0.5 g sphere and about 2% for a 1.0 g sphere (if $\Sigma_S = 6$ cm^{-1}). This amount of error is of the same order as uncertainties associated with counting statistics for spheres for which H sensitivities were determined by PGAA. Thus a comparison of PGAA data with Monte Carlo results for spheres would not be useful.

Conclusions

This work clearly demonstrates the importance of scattering effects in neutron beam activation analysis experiments, particularly when the matrix contains large amounts of strong scatterers such as hydrogen. The results of this study demonstrate that corrections as large as 25% may be required for 0.2 cm disks of typical biological samples. Our conclusion is that the

effects of neutron scattering and absorption within such matrices can be significant even when sample dimensions are relatively small. Other factors being equal, correction factors for spheres are much less than for disks.

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