A REVIEW AND A PRELEMINARY STUDY ON SELF-ABSORPTION CORRECTION IN GAMMA-SPECTROMETRY

Ahmad Saat*

ABSTRACT

The article gives a review on self-absorption problem encountered in gamma-ray spectrometry. Various correction methods for different experimental setups were discussed, in terms of their approaches and limitations. Assumptions used in deriving the correction models were also listed. Results of a preliminary study was also described..

INTRODUCTION

Gamma-rays spectrometry has been developed to become an important simple and fast non-destructive determination method. It is also an important complimentary part for neutron activation analysis and environmental studies (Nakamura & Suzuki (1983); Battiston et al. (1987)). Normally, a Ge(Li) or HPGe detector were used as the gamma-rays detector, which in turn coupled to an amplifier circuit and multichannel analyser and also a computer for futher evaluations. The analytical method depends on the evaluation of gamma peaks in the spectrum, and thus a better accuracy can be achieved if the evaluation was correct. However, a correct evaluation may be hampered by the self-absorption of gamma photons in the sample (Debertin & Ren (1989); Overwater et al. (1993)) as well as the difference in the response of the detector for gamma-rays that originate from different parts of the sample (Moens et al. (1981); Battiston et al. (1987); Overwater et al. (1993)) which might introduce systematic error in individual data (El-Daoushy and Garcia-Tenorio (1995)). The latter is known as geometry effect. The self-absorption depends significantly on the chemical (mineralogical) composition and the size (density) of the samples, as well as the gamma-rays energy itself. Figure 1 shows the absorption of gamma-ray in various medium as a function of energy, while Figure 2 illustrates the same absorption as a function of water-matrix source thickness, for five different energies.

* *Present address: Department of Applied Mathematics and Theoretical Physics, University of Liverpool, P. O. Box 147, Liverpool L69 3BX, UK .*

Before going futher, it is important to point out the distinction between *absorption* and *attenuation*, because many authors seem to be confused between the two terms. This was obvious when they sometimes interchanged the use of both terms in their description. In attenuation, both the interactions that involved scattering of gamma photons (without transfer of energy through Compton scattering) and the transfer of energy into the attenuation medium were taken into account. While, absorption is identified by the transfer of energy from the gamma photons to the absorber. Attenuation process is characterised by the *attenuation* coefficient of the medium, while absorption by the respective *absorption coefficient* of the absorber It is obvious that for a medium, the attenuation *coefficient* is larger than the absorption coefficient. The difference between the two coefficients is called *scattering coefficient* (Turner (1986)). However, in many of the publications encountered, the authors assumed no distinction between absorption and attenuation.

The effect of self-absoption is more important for low-energy gamma (<200 kev) (Bode et al. (1981); Dulinski & Dominik (1992)), particularly when applied to the analysis of low activity low energy photons of samples such as Pb-210 in sediments (Cutshall et al. (1983); Battiston et al. (1987); Dulinski & Dominik (1992)) and environmental samples (Nakamura & Suzuki (1983); Sanchez et al. (1991)), where sometimes voluminous (bulk) samples were used to increase the peak counts (Galloway, 1991). Thus for cases like this, the effect of self-absorption and geometry have to be corrected to get an accurate evaluation of the spectrum peaks. The influence of the absorption effect on the gamma spectrometry results will be more obvious when the standards used as comparison for the samples, or for plotting the efficiency curves for the detector system have a different composition and geometry from the samples (Moens et al, 1981 Battiston et al, 1987), while preparing an accurate standard source is a cumbersome and time consuming job (Moens et al.1981), and suitable standard materials rarely available (Zikovsky, 1989).

These effects have been realised since gamma spectrometry was first introduced into the determinative analytical circle. And many writtings and articles were published addressing this problem, among the earliest was by Dickens (1972). Most correction methods described are for specific sample geometries (shape), such as thick cylindrical (Dickens, 1972; Galloway,1991; Overwater et a, 1993), marinelli beakers (Nakamura & Suzuki, 1983; Debertin & Ren, 1989), disc shape (Cutshall et al. (1983); Battiston et al. (1987)), capsule (Bode et al. (1981) and small cylindrical vials (Dulinski & Dominik, 1992; Appleby et al. 1992); as well as specific detector geometry such as coaxial (Cutshal et al. 1983; Galloway 1991; Overwater et al. 1993), well-type (De Bruin et al. 1979; Appleby et al. 1992; El-Daoushy & Garcia-Tenorio 1995), and planar (Battiston et al. 1987), though some claim to be suitable for any sample shapes (Moens et al. 1981; Overwater et al. 1993). However, since gamma spectrometry is supposed to be a fast and simple method, any correction method proposed needs to be of simple usage and can be applied in routine

procedures easily. In fact some authors proposed and described how the correction can be carried out easily using computer programmes (Zikovsky 1989; Jaegers & Landsberger 1990; El-Doushy & Garcia-Tenorio 1995) and some using Monte Carlo method coupled with a computer pogramme (Nakamura & Suzuki, 1983; Sanchez et al. 1991).

ASSUMPTIONS

In arriving at the correction factors for self-absorption of gamma photons, the following basic assumptions were made by many authors:

a. the radioactivity was distributed homogeneously within the sample.

b. the composition and density of the sample is uniform.

c. the sample placed coaxially to the detector.

d. the detector efficiency is energy dependent.

Although the method proposed by Overwater et al. (1993) involves no assumption with regards to sample size, shape, orientation, matrix composition or source-to detector distance, the verification was carried out using cylindrical sources.

Throughout most of the correction factor derivations, the absorption was assumed to follow correctly the Lambert (Bouguer) Law of Absorption

$$I = I_0 \exp(-\mu\rho\chi) \qquad (1)$$

I_0 the flux of incident photons, I that of the transmitted photons, x thickness of absorber and R (density of the absorber)

to obtain an expression for total mass absorption coefficient, μ (Bode et al. 1981; Nakamura & Suzuki 1983; Cutshall et al. 1983; Battiston et al. 1987; Appleby et al.1992), while for absorption of gamma photons by the sample itself, the relation for fraction of photons emitted out of the sample

$$I/I_0 = (1 - \exp(-\mu\rho\chi)/\mu\rho\chi \qquad (2)$$

was used (Bode et al. 1981; Cutshall et al. 1983; Jaegers and Landsberger 1990), assuming parallel trajectories of gamma photons and a sample with plane surface geometry. Hsu and Dowdy (1983) assumed that the mass attenuation coefficient is related to the interaction cross-section of the gamma-rays with the constituents in the samples concerned, and at low-energy (20 - 400 kev) the interaction is mainly due to photoelectric

effect, Compton and Rayleigh scattering (Bode et al. 1981). As for detection efficiency of the detector, the normal basic definition of the ratio between the count rate detected and the activity of the sample at that particular energy, was applied.

APPROACH

The approaches taken by most authors in deriving their self-absorption correction factors may broadly be categorised into:

a. using the basic concept of interaction, and following the fate of a gamma photon from the time of its emission at a certain location in the sample until being absorbed in the detector. Here, usually a vigorous mathematical modelling will be involved.

b. using a simple model, which is basically an approximation of the model from (a).

c. applying mathematical calculation using the Monte Carlo Method, usually used to solve the complicated calculation encountered in (a).

In many cases, the correction factor is incorporated in the efficiency determination.

The early analysis of self-absorption was described by Dickens (1972). He carried out a mathematical analysis on the absorption of neutron induced gamma-rays in large cylindrical samples, where the axes of the cylinder and detector were perpendicular to one another, and at a large source-detector distance. The relatively simple mathematical evaluations, begin by considering a cylindrical chord in the sample volume, described how to obtain an eaverageí gamma path length to compute the average attenuation in the sample using equation (1). His experimental results show that the model to be valid for energy greater than 0.5 Mev, with relatively large sample-detector distance, to correctly assumed parallel gamma-rays leaving the samples.

As mentioned earlier, a lenghty mathematical calculations will be involved when starting from the basics. For example, for samples in marinelli beakers, Debertin and Ren (1989) assumed a point detector located at the centre of the real detector, while the beaker was divided into three parts; a ring-shaped volume, a truncated cone and a disk-shaped upper part. Photons of energy E emitted from a volume element in the volume will tranverse through the sample (and experiencing attenuation) before depositing its energy to the point detector, with efficiency given by

$$\varepsilon_a(E) = C\,(E)\,\exp\,(-\mu(E)z_o)/z^2 \qquad (3)$$

where z^a is the distance travelled within the sample, and z the distance from the volume element to the point detector and C(E) is a proportional constant. The total efficiency was obtained by integrating the above equation through the whole sample volume V,

$$\varepsilon(E) = C\,(E)/V \exp\,(- \mu(E)z_o)/z^2\,dV \qquad (4)$$

They also described how the numerical integration of the above integral can be carried out. The ratio of the integral for standard to experimental sample will give the correction factor. In the description they also discussed the correction for coincidence-summing.

Overwater et al. (1993) approached this correction together with the source-geometry correction for voluminous sample. The photopeak efficiency of the voluminous source is based on the basic definition of the ratio of detected countrate to the total photon emission and incorporating the correction terms for self-attenuation and the effect of the change in solid angle of the points in the source. The correction factor was defined by the ratio of the calculated point source photopeak efficiency to the photopeak efficiency of the voluminous sample source. A Monte Carlo method of calculation was used to determine the photopeak efficiency, and performed by following a single photon through the complete source-detector system.

Bode et al. (1981) proposed quite a different approach for the correction of self-absorption of low energy photons that can be used routinely in Neutron Activation Analysis (NAA). They started by identifying the processes that caused the self-absorption of low photons energy; they are the photoelectric effect, Compton and Rayleigh scattering. Then the expression for the total mass attenuation coefficient of the sample was obtained by combining the general formula (function) for each of the process (neglecting Rayleigh scattering due to its low contribution). Thus a relation between mass absorption coefficient and energy only can be obtained. Through this relation the correction factor at any energy using the fraction of emitted photons from samples after absorption, as in eqn. (2), can be obtained.

A simple correction model using transmission through a planar cylindrical sample (100 cm3) was used by Cutshall et al. (1983) to determine the Pb-210 contents in sediment samples. They arrived at an expression for correction factor as $(\ln(I/I_o)\ /(I/I_o-1)$. Thus the Pb-210 contents was computed by the product of the correction factor, sample countrate and the counting efficiency at the peak concerned. Galloway et al.(1991) tried to improve the model by taking into account that the effective solid angle subtended by the sample at the detector is not constant, particularly at a short source-to-detector distance. They also showed that Cutshall model produced inaccurate results for low energy gamma.

Another simple correction model for low energy low gamma radiation that to be used in a well-type detector (thus cylindrical sample geometry) was proposed by Appleby et al. (1992), also based on the transmission fraction expression (2). By assuming that equal amount of radiation travelled inward and outward of a hollow cylindrical element of the sample, and using simple calculus, they arrived at approximated simple corrections; one for small mass sample and another for higher mass sample.

The use of empirical expression $\mu = c\lambda^n$, and then substituting into equation (1), to get the absorption coefficients as a function of energy only was adopted by Battiston et al. (1987) where λ is the wavelength of the photons in Armstrome unit. The correction factors thus obtained using equation (2); with x as the self-absorption effective thickness, which included the geometry effect. They proceeded by explaining the method of determining x using water as the comparing medium. The value of x is constant for the particular sample-detector setup. The model was evaluated using disk-shaped sources for energy range 40 - 120 kev.

An evaluation of self-absorption using internal standard for photon energies of 20 - 60 kev in samples of aquatic deposits was described by El- Daoushy and Garcia-Tenorio (1995). The internal standards were prepared by spiking samples of different mass with known amount of mixed radionuclides, and packed in identical volume containers. A plot of the slopes of transmittance-mass relation at different energy, against gamma energy, gives the self-absorption correction of the given experimental geometry, provided the sample mass is known. They also showed that liquid standards could be used for self-absorption studies of small sample volumes.

Hsu and Dowdy (1983) presented an empirical method for interpolating the values of mass attenuation coefficients for energies of 40 kev to 15 Mev. The polynomial used, which relates energy and μ is

$$\ln \mu_i = \sum_{J=0}^{N} A_j (\ln E_i)^j \qquad (5)$$

which was evaluated using a computer programme. A computer code was also developed by Jaegers and Landsberger (1990) to determined self-absorption fraction of gamma in low, medium and high-Z materials, for use in NAA. In their analysis, three sample geometries were considered: linear, cylindrical and spherical; where a labourious mathematical calculations were involved to obtain the total linear attenuation coefficients. The effect of K-edge absorption was also taken into account in the computer code.

Due to some contrasting results obtained by other authors between measured and calculated procedures in semi-empirical absorption model of gamma in marinelli beakers, Sanchez et al. (1991) proposed a method via a Monte Carlo calculation, based on the simulated determination of gamma detection efficiency. The method incorporated the efficiency

variations produced by different self-absorption of samples, using simulation program which allows to obtain a wider energy range of detector response. Nakamura and Suzuki(1983) also used a Monte Carlo calculation, via a computer code called PEAK, to determine the peak efficiency of a Ge(Li) and a HPGe detector for voluminous samples. They described in detail the steps taken to incorporate the geometry and self-absorption effect in the calculations.

RESULTS AND LIMITATIONS

Most authors carried out the validation of their models either by comparing with the experimental results of gamma-rays spectrometry, or with other methods of determination, such as wet radioanalytical analysis (Cutshall et al. (1983)) or alpha-spectrometry (Battiston et al. (1987)). Within the limitations of the models, the results were quoted to be in good agreements within several percents; ranging from 1% (Debertin and Ren (1989)) to as high as about 8% (Battiston et al. (1987)). Besides the basic universal assumptions mentioned earlier, some authors included other limitations on their models. For example, Dickens (1972) model was for gamma of energy greater than 0.5 Mev, while Battiston et al. (1987) showed that for energy greater than about 120 kev, the correction was not necessary.

Restrictions were also imposed on samples to be used. Hsu and Dowdy (1983) model needs more than one polynomials if the samples contained high-Z materials, to account for the K- and L-absorption edges. While the results of Appleby et al. (1992) showed a less good agreement for half-full cylindrical vials, due to the relatively higher end effects. The model of El-Daoushy and Garcia-Tenorio (1995) produced best results for relatively low mass ((50g) samples. Although, the model proposed by Overwater et al. (1993) was for voluminous sources, the source shape should be able to be described by a mathematical function.

As mentioned in the earlier section, most of the models proposed were for the specific detector geometry (type), such as planar, well or coaxial detectors. Other limitations was imposed on the source-detector distance, which need to be large (Dickens (1972)), so that parallel gamma-rays were assumed to be emitted from the sample. However, some models did not impose this restriction (Cutshall et al. (1983); Battiston et al. (1987); Overwater et al. (1993).The model proposed by Galloway (1991) showed that this distance was a parameter that depended on energy, since it was estimated to be the distance between the face of the source to the mean depth of photon detection in the detector.

PRELIMINARY STUDY ON A SIMPLE MODEL

A simple model was developed to carry out a self-absorption study on circular disc samples coupled to a planar detector. Gamma-rays from the disc samples will traverse through different distances and angles within the samples before being detected by the detector. Using simple mathematical manipulations and estimations, the unattenuated fraction of gamma-rays eminated after being self-absorbed by the sample could be modelled simply as

$$I/I_0 = \exp(-k\mu m) \qquad (6)$$

where μ (m^2kg^{-1}) is the mass attenuation coefficient for the particular gamma energy and sample composition, m (kg) is the mass of the sample, while k (m^{-2}) is a geometrical factor for the sample-detector arrangements.

The model was tested on two HPGe planar detectors (PS1 and GMX3), using various masses of standard gamma source mixed homogeneously into plain flour and compressed into circular disc of 4.5 cm diameter. The standard gamma source containing U-238 and U-235 produced various gamma-rays with energy ranging from 63 kev to 1001 kev. These energies and sample masses provide a wide range of μm values.

Figure 3 and Figure 4 illustrated the results of I/I_0 versus μm for the two detectors. The experimental results were found to agree quite closely to the simple model. The values of the geometrical factors for the detectors are 0.0544 and 0.0851 for PS1 and GMX3 respectively. This results of the initial study show that the simple model could be usede as the first estimate of self-absorption correction for routine gamma-ray spectrometry.

CONCLUSIONS

The exhaustive number of models proposed by many authors regarding the correction for self-absorption, reflect that this problem is far from being solved. Most models are peculiar for the authors experimental setup. What is most needed is a model that possesses the characteristics of (a) suitable for any experimental setup, (b) easily incorporated in a routine analysis, (c) universally applicable for any sample shapes, density and composition. And, a particular emphasis should be on the absorption of low energy gamma-photons, where the effect is more dominant.

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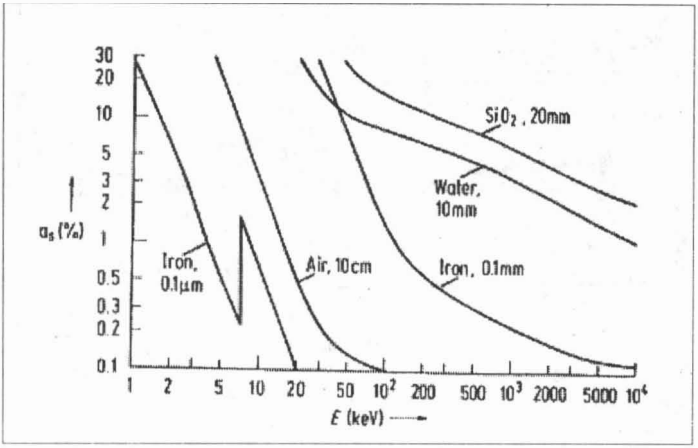

Figure 1. Percentage self-attenuation of photons in sources of various matrix as a function of energy for cylindrical source geometry, with respective thickness shown (from Debertin and Helmer (1988)).

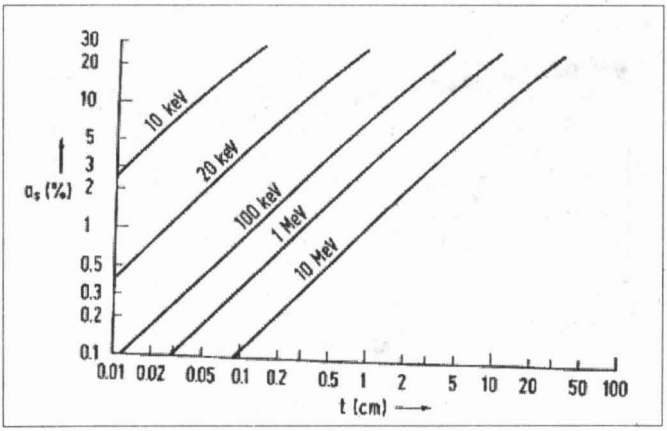

Figure 2. Percentage self-attenuation of photons in water-matrix cylindrical source, as a function of source thickness, for various energies (from Debertin and Helmer (1988)).

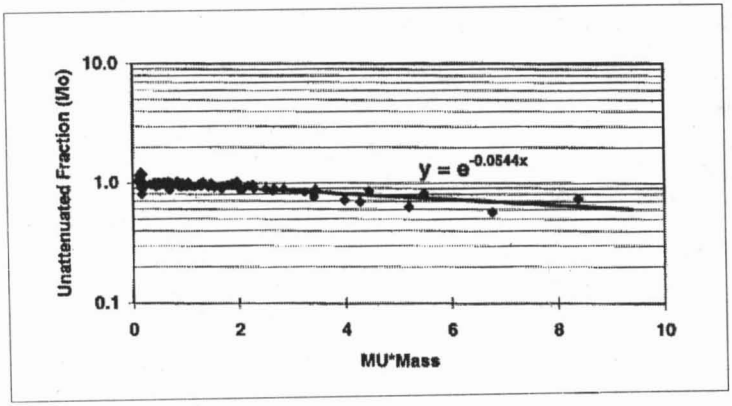

Figure 3: Unattenuated gamma-ray fraction against (m for planar detector PS1.

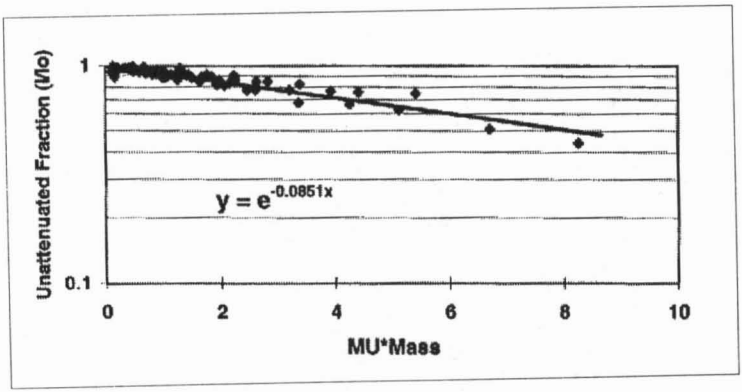

Figure 4: Unattenuated gamma-ray fraction against (m for planar detector GMX3.