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Application of an empirical expression for full energy peak efficiency for the estimation of volumetric efficiency in large sample neutron activation analysis

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ABSTRACT

An estimation of volumetric efficiency in large sample neutron activation analysis through the application of an empirical expression for the full energy peak efficiency was done. The gamma-ray self-attenuation correction factor and the geometric correction factor based on which the volumetric efficiency is finally estimated were experimentally and theoretically determined respectively. Within the limits of mass range of the sample investigated, there was no significant difference between the photo-peak efficiency and the volumetric efficiency. The result of the study therefore establishes that the mass (volume) of the test portions does not have significant effect on the efficiency of elemental detection when geometrical correction is considered.

Introduction

In recent years, Large Sample Instrumental Neutron Activation Analysis (LS-INAA) has been acknowledged as an excellent technique among the various nuclear analytical techniques that are in operation. It has many advantages in improving the element determination capacity of Instrumental Neutron Activation Analysis (INAA) particularly at small and medium size research reactors. On the other hand, a few phenomena need more attention in LS-INAA than in normal NAA (Overwater, 1994). One of these phenomena is volumetric peak efficiency. An irregular response of the detector is among the principal sources of error in LS-INAA.

To obtain accurate results from gamma-ray spectroscopy, a correction factor that takes into consideration the source geometry as well as the gamma ray self-absorption and scattering by the sample material has to be applied. A sample of, say, 1 kg cannot be considered anymore as a ‘‘point source’’ during counting at normal sample–detector distances of, e.g., 10–30 cm, resulting in a corresponding different response of the detector for the γ-radiation.

Volumetric Efficiency Determination

The full energy peak efficiency $\varepsilon(E)$ of an HPGe detector for a coaxial cylindrical source is defined as the quotient of the number of detected photons in a peak $N_{\det}$, and the total number of emitted photons in the same peak N_{emit}, both per unit time interval (Haase et al., 1995; Debertin and Helmer, 1988). This efficiency (which is a dimensionless fraction) is related to specific source–detector geometry and a particular peak analysis procedure. Then, the full-energy-peak efficiency is defined as

$$\varepsilon(E) = \frac{N_{\det}}{N_{emit}} \qquad [1]$$

When using this expression for cylindrical sources, two effects need to be taken into account: the self-attenuation of the photons in the source, and the effect of the solid angle covered by the detector for each volume element. For the simpler case of uniform activity planar disk sources, Helmer and Chatani independently proposed a mathematical relation between the peak efficiencies of disks sources of radius R and that of point sources (Helmer, 1983; Chatini, 1999). In particular, Helmer proposed the following expression (Aguiar and Galiano, 2004):

$$\varepsilon_a(E) = \varepsilon_p(E)\frac{d^2}{R^2} In\left(1+\frac{R^2}{d^2}\right)dx \qquad [2]$$

Where dx is the elemental thickness of the disk source, $\varepsilon_a(E)$ is the disk source peak efficiency and $\varepsilon_p(E)$ is the peak efficiency of a point source on the detector axis, both at distance d from the detector as illustrated in figure 1.

The peak efficiency, $\varepsilon_p(E)$ can be calculated using the relations

$$In(\varepsilon_p(E)) = a_0(z) + a_1(z)In(E) \qquad E_\gamma > 130$$

[3]

$$\varepsilon_p(E) = \sum_{i=0}^{2} a_i(z)E^i \qquad E_\gamma \le 130$$

[4]

where, a_0, a_i and a_1 are the corresponding coefficients for the energy polynomial with respect to the source-detector distance (Osae et al, 1999).

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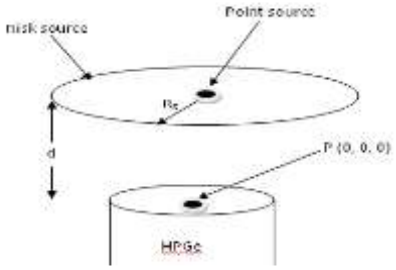

Figure 1 Geometry of disk source of infinitesimal thickness, which must be integrated longitudinally in order to produce the cylindrical volume source. Equivalent point source is located at center of disk.

If one considers the cylindrical source as

$$\sum_{d}^{d+h} disc\ \ sources$$

Equation [2] can then be integrated from d to $d+h$ as shown in figure 2(a) below.

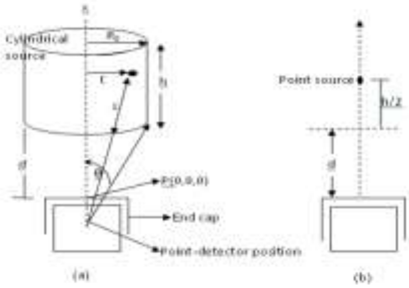

Figure 2: (a) Geometry of the cylindrical source obtained by integrating the disk source from d to $d+h$. (b) Position at $\frac{h}{2}+d$ with respect to the detector, where the point source must be located to determine $\varepsilon_p(E)$

This leads to a generation of a geometric correction factor, f_g for thepeak efficiency

$\varepsilon_{vol}(E)$ for a cylinder as follows (Aguiar and Galiano, 2004):

$$\varepsilon_{vol}(E)=\varepsilon_p(E)\int_d^{d+h}\frac{x^2}{R_s^2}In\left(1+\frac{R_s^2}{x^2}\right)dx \quad [5]$$

Which upon integration yields

$$\varepsilon_{vol}(E)=\varepsilon_p(E)\left(\frac{2}{3h}\right)\left[h-R_s\arctan\left(\frac{d+h}{R_s}\right)+\frac{(d+h)^3}{2R_s^2}\times In\left(1+\frac{R_s^2}{(d+h)^2}\right)+R_s\arctan\left(\frac{h}{R_s}\right)-\frac{d^2}{2R_s^2}In\left(1+\frac{R_s^2}{d^2}\right)\right]$$

[6]

Equation [6] represents the theoretical (peak) efficiency for the cylindrical source which is just the summation of all disks of radius R_s from d to $d+h$.

If we now refer to $\varepsilon_{vol}(E)$ as the theoretical efficiency $\varepsilon_t(E)$, then

$$\varepsilon_t(E)=\varepsilon_{vol}(E)=\varepsilon_p(E)\,f_g \quad [7]$$

Where

$$f_g=\left(\frac{2}{3h}\right)\left[h-R_s\arctan\left(\frac{d+h}{R_s}\right)+\frac{(d+h)^3}{2R_s^2}\times In\left(1+\frac{R_s^2}{(d+h)^2}\right)+R_s\arctan\left(\frac{h}{R_s}\right)-\frac{d^2}{2R_s^2}In\left(1+\frac{R_s^2}{d^2}\right)\right]$$

[8]

Equation [7] determines the volumetric efficiency for the cylinder under the assumption it is in a vacuum, or a non-attenuating gas such as air. f_g can then be interpreted as a geometric volume elements of the cylinder with respect to point P(0,0,0).

Finally, the theoretical, attenuation-corrected volumetric efficiency for the cylinder source is given by the expression:

$$\varepsilon_t(E)=\varepsilon_p(E)\,f_g\,f_{att} \quad [9]$$

The gamma-ray attenuation correction factor f_{att} is expressed as

$$f_{att}=\frac{I(\mu,R,h,d)}{I(R,h,d)} \quad [10]$$

Where

$$I(\mu,R,h,d)=\int_0^R\int_0^h\frac{\exp[1-(\mu x)+(\mu x)^2/2]}{r^2+(x+d)^2}rdxdr \quad [11]$$

$$I(R,h,d)=\int_0^R\int_0^h\frac{r}{r^2+(x+d)^2}dxdr \quad [12]$$

d, *r* and *x* are the source-to-detector distance, sample radius and sample thickness respectively.

An HPGe detector can be treated as a point detector since the energy of the photons is absorbed after these have penetrated a certain distance into the crystal. Based on this idea, some investigators have proposed the concept of a point–detector position as indicated in figure 2(a) (Noguchi et al., 1981; Zikovsky and Chah, 1988). Figure 2(b) illustrates the position where a point source needs to be located in order to properly determine $\varepsilon_p(E)$. The idea that it is possible to determine $\varepsilon_t(E)$ by multiplying $\varepsilon_p(E)$ by a geometric correction factor is valid as long as the source–detector system exhibits unchanging efficiencies for the working angles (Aguiar and Galiano, 2004). It is important to perform this type of geometric analysis since implicit in Equation [7] is the fact that the point source can be located anywhere inside the cylindrical volume to be integrated.

Experimental evaluations and calculations

Gamma-ray self-attenuation correction

A transmission experiment was performed to determine the attenuation coefficient of the sample for the correction of the gamma-ray self-attenuation occurring during counting of samples after irradiation. A multi-gamma-ray emitting source, Europium (^{152}Eu) was used in this work.

The sample was placed between the detector and a transmission source (^{152}Eu). The beam of photons from the transmission source was collimated through a pinhole in a lead shield to create a nearly pencil beam geometry and allowed to pass through the sample to the detector.

This was used for the calculation of the effective linear gamma-ray attenuation coefficient (µ) of the sample used for this work by using the relation:

$$I=I_0e^{-\mu x} \quad [13]$$

where x, I and I_0 are the thickness of the sample, gamma-ray intensity without attenuation and attenuated gamma-ray intensity respectively.

The correction factor for the gamma-ray self-attenuation in the sample at a given gamma-ray energy at a fixed geometry for the case of a cylinder, coaxially positioned with the detector was then calculated using equation [10].

Peak efficiency and volumetric efficiency calculations

The peak efficiencies of the elements present in the sample (taking the sample as a point source) on the detector axis were calculated at a source-to-detector distance of 1.2 cm using equations [3] and [4] with the corresponding coefficients for the energy polynomial.

The attenuation-corrected volumetric efficiencies of the gamma-ray energies of the elements were further calculated using equation [9].

Results And Discussion

The evaluation of the volumetric efficiency depends on the photo-peak efficiency and the geometric correction factor. The geometric correction factor tries to bring the 'large sample' back to a 'point source'. Table 3 shows the calculated values of the geometric correction factors for different sample mass. It can be seen that the geometric correction factor increases as the sample mass increases. As the sample mass increases, there is a much deviation from having a point source geometry. Therefore the geometric correction factor increases with increasing mass to bring it back to point source geometry.

It can be observed from figure 3 that the volumetric efficiency does not vary much from the photo-peak efficiency. Volumetric efficiency of sample masses of 0.5 g and 5.0 g and photo-peak efficiency for some elements of interest were plotted. Both photo-peak efficiency and volumetric efficiencies decrease exponentially as energy increases. There is a good agreement (comparison) between the photo-peak efficiency and the volumetric efficiency. This is so because of the dependency of the volumetric efficiency on the geometric correction factor.

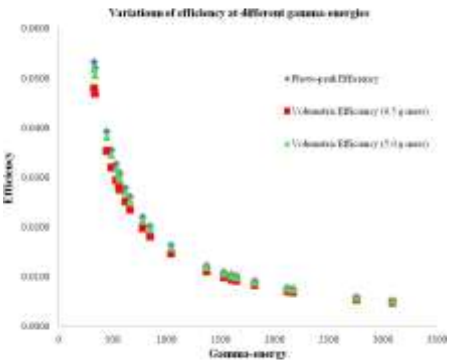

Figure 3 Efficiency as a function of gamma-energy. A comparison of volumetric efficiency of sample masses of 0.5 g and 5.0 g and photo-peak efficiency for some elements of interest.

Volumetric efficiency increases with increase in the sample mass as it can be seen from figure 4. It is observed figure 4 that the rate of variation in the volumetric efficiency with increasing sample mass is approximately the same for different energy levels. There is a gradual increase in volumetric efficiency till the mass gets to 3.0 g, after the 3.0 g mass there is not significant increase. At this point (mass $\geq$ 3.0 g) the increase assumes a kind of plateau shape. Within the mass range of the sample investigated, it can be observed that the mass of test portions does not have much significant effect on the efficiency of elemental detection when geometrical correction is done.

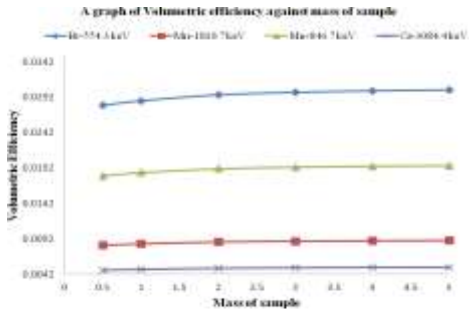

Figure 4 Volumetric efficiency as a function of sample mass for some isotopes of different gamma-energies

Conclusion

Within the mass range of the sample investigated, there was no significant difference between the photo-peak efficiency and the volumetric efficiency. The result of the study therefore establishes that the volume or mass of the test portions does not have significant effect on the efficiency of elemental detection when geometrical correction is well factored into the evaluation processes.

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Table 1: Coefficient for the energy polynomial, $a_i(z)$ for the different z positions (Osae at al, 1999)

Sample to detector distance (z), cm	Coefficient of energy polynomial $a_i(z)$				
	Gamma-ray energy, E ≤ 130 KeV			Gamma-ray energy, E ≥ 130 KeV	
	$a_0(z)\times10^{-2}$	$a_1(z)\times10^{-3}$	$a_2(z)\times10^{-6}$	$a_0(z)$	$a_1(z)$
1.2	7.968	1.500	-9.931	3.032	-1.029
2.2	5.188	1.060	-6.821	2.466	-0.999
3.5	2.863	0.566	-3.593	1.924	-0.998
4.7	2.109	0.342	-2.163	1.537	-0.991
6.0	1.398	0.265	-1.639	1.163	-0.979
7.2	0.998	0.210	-1.295	0.658	-0.941

Table 2: Coefficients of the z polynomial a_{ij} , corresponding to each of the energy polynomial, $a_1(z)$ (Osae at al, 1999)

Coefficients of the energy polynomial $a_1(z)$	Coefficients of the z polynomial a_{ij}				
	a_{i0}	a_{i1}	a_{i2}	a_{i3}	a_{i4}
	Gamma-ray energy, E ≤ 130 KeV				
a_0 (z)	0.1336	-5.536×10^{-2}	9.653×10^{-3}	-7.123×10^{-4}	1.501×10^{-5}
a_1 (z)	2.066×10^{-3}	-3.535×10^{-4}	-1.105×10^{-4}	3.352×10^{-5}	-2.267×10^{-6}
a_2 (z)	1.352×10^{-5}	-2.480×10^{-6}	-6.484×10^{-7}	2.067×10^{-7}	1.411×10^{-8}
	Gamma-ray energy, E ≥ 130 KeV				
a_0 (z)	41.93	-24.16	6.27	-0.7614	3.462×10^{-2}
a_1 (z)	1.120	-0.1145	3.705×10^{-2}	-4.544×10^{-3}	1.984×10^{-4}

Table 3 Table of values of geometric correction factors for various sample masses.

Geometric correction factor	Mass of sample
0.900	0.5 g
0.921	1.0 g
0.948	2.0 g
0.960	3.0 g
0.967	4.0 g
0.971	5.0 g