Available online at www.sciencedirect.com
ScienceDirect
Nuclear Instruments and Methods in Physics Research A 580 (2007) 234–237
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

A self-sufficient and general method for self-absorption correction in gamma-ray spectrometry using GEANT4

S. Hurtado[a,*], M. Villa[a], G. Manjón[b], R. García-Tenorio[b]

[a]Servicio de Radioisotopos, CITIUS, Universidad de Sevilla, Reina Mercedes 4B 41012, Sevilla, Spain
[b]Departamento de Fisica Aplicada II, E.T.S.A, Universidad de Sevilla, Avda, Reina Mercedes 2 41012 Sevilla, Spain

Available online 16 May 2007

Abstract

This paper presents a self-sufficient and general method for measurement of the activity of low-level gamma-emitters in voluminous samples by gamma-ray spectrometry with a coaxial germanium detector. Due to self-absorption within the sample, the full-energy peak efficiency of low-energy emitters in semiconductor gamma-spectrometers depends strongly on a number of factors including sample composition, density, sample size and gamma-ray energy. As long as those commented factors are well characterized, the influence of self-absorption in the full-energy peak efficiency of low-energy emitters can be calculated using Monte Carlo method based on GEANT4 code for each individual sample. However this task is quite tedious and time consuming. In this paper, we propose an alternative method to determine this influence for voluminous samples of unknown composition. Our method combines both transmission measurements and Monte Carlo simulations, avoiding the application of Monte Carlo full-energy peak efficiency determinations for each individual sample. To test the accuracy and precision of the proposed method, we have calculated ^{210}Pb activity in sediments samples from an estuary located in the vicinity of several phosphates factories with the proposed method, comparing the obtained results with the ones determined in the same samples using two alternative radiometric techniques.

PACS: 29.30.Kv; 87.53.Wz

Keywords: Gamma spectrometry; Self-absorption; Efficiency calibration; Monte Carlo

1. Introduction

Gamma-ray spectrometry is, in general, a fast and simple method of measuring natural and man-made radionuclides in environmental samples. But, when low-energy gamma emitters are presented in the sample, self-absorption effects within the sample becomes significant. Factors like sample composition and sample size affect full-energy peak efficiency (FEP) and, by extension, the precision in the determination of the activities.

Many procedures have been suggested for correcting self-absorption effect regardless of high-cost experimental method for each sample. Semiempirical methods [1,2] are based on the transmission of a low-energy gamma-ray collimated beam through the sample in order to determine linear attenuation factor (μ). However, these methods are restricted to measurement geometries where detector–source distance is greater than their dimensions.

Monte Carlo (MC) methods calculate the efficiency reproducing totally any detector–source configuration [3] but usually exact sample composition is not known and additional techniques (TTPIXE, XRF, etc.) for its determination are required. An option consists of using a collimated beam [4], or a non-collimated one [5], to obtain μ and consequently FEP efficiencies via MC calculation.

In this work, we propose a general method for FEP determination in a wide energy range and any sample height in Petri dish geometry. First, we calculate FEP for different energies and air height using MC simulation, avoiding in this manner additional calculation for each sample. Afterwards, self-attenuation correction factors are calculated using a transmission experiment with non-collimated point sources for a wide energy range. The

*Corresponding author. Tel.: +34 954559750; fax: +34 954559753.
E-mail address: shurtado@us.es (S. Hurtado).

0168-9002/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2007.05.090

combination of both quantities gives us corrected FEP and consequently sample activity for any isotope and sample height.

2. Experimental procedure

Measurements were performed with a Canberra n-type Reverse electrode Germanium (ReGe) detector, with a relative photo-peak efficiency of 30% at 1332 keV. The preamplified signals from the detector are sent to a Canberra Spectroscopy Amplifier model 2020 and a Canberra ADC model 8701 connected to an AccuspecTM card. Gamma-ray peaks are analyzed using Genie2K.

Point sources used (^{241}Am, ^{137}Cs and ^{60}Co) for transmission experiment were supplied by PTB (Germany).

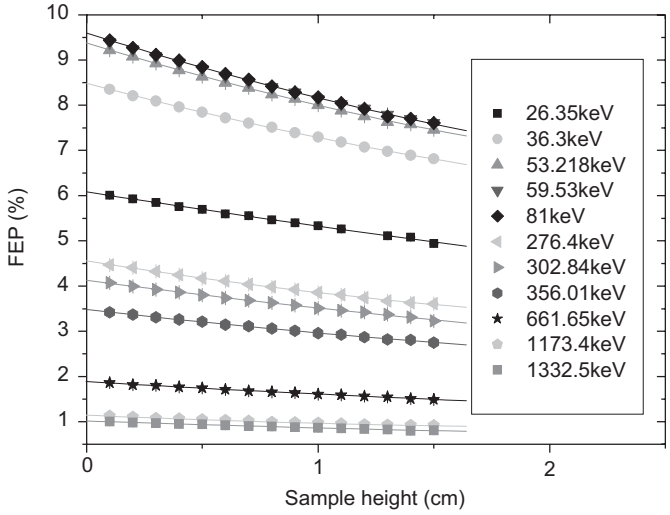

Fig. 1. FEP (ε_0) calculated (MC) for different sample height and emission energy.

The sediments samples from Huelva estuary were dried and introduced into Petri dish in order to measure its height (t) carefully.

3. Monte Carlo simulation

GEANT4 code (v.8.1) was used to simulate the detector–sample configuration. The internal dimensions of the detector were optimized using a well-checked procedure [6] in order to obtain accurately FEP for any energy and measurement geometry.

4. Self-absorption correction method

The corrected FEP is calculated for every energy using

$$\varepsilon = f\varepsilon_0 \tag{1}$$

where ε_0 is FEP calculated using MC simulation of an empty Petri dish for different sample heights, and f is the self-absorption correction factor. ε_0 is fitted using a polynomial of order two in function of sample height (t) [7] allowing us to obtain FEP (ε_0) for every energy (Fig. 1).

Self-absorption correction factors (f) are calculated experimentally, for each real sample using

$$f = (1 - \exp(-\mu t))/(\mu t) \tag{2}$$

where $\mu(E)$ is the linear attenuation coefficient that depends on the energy and is calculated using a transmission experiment and t is the sample height.

The Petri dish containing each sample is located at 1.1 cm above detector window and the point sources (^{241}Am, ^{137}Cs and ^{60}Co) are located on top of each sample. Then $\mu(E)$ is calculated using following expression for every emission:

$$\ln(I_0/I) = \mu t \tag{3}$$

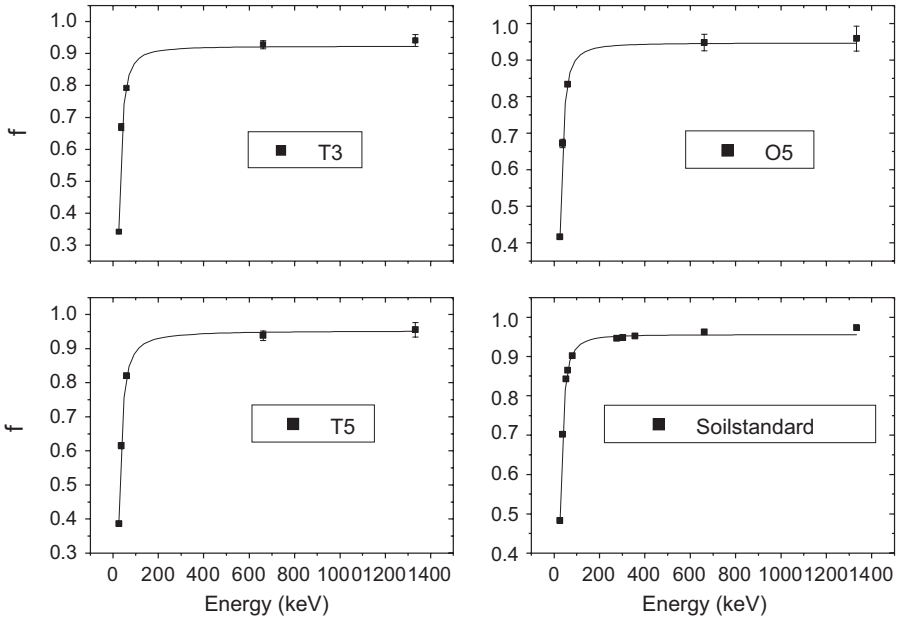

Fig. 2. Transmission factor (f) fitted to Eq. (4) for whole energy range and sediment sample (T3, O5, T5 and standard soil composition).

where I_0 is the net count rate in the peak at energy E without sample and I is the net count rate with sample. Finally self-attenuation correction factor, for each real sample, is calculated using Eq. (2) and non-linear weighted fitted for whole energy range to

$$f = a + b/E + c/E^2. \quad (4)$$

This allows us to correct self-absorption effect for each sample and emission energy (see Fig. 2). In order to check the accuracy of Eq. (4) to represent f, the μ for a standard soil composition is extracted from XCOM database [8] and used to calculate f for a height of 1 cm. The equation fit shows a good agreement (see Fig. 2).

Some remarks must be commented for this procedure. If non-collimated point sources are used the photons reaching the germanium crystal should travel a larger distance (t_{eff}) than t within the sample [5]. For that reason, MC simulations of various sample composition (SiO_2, $CaCO_3$, Fe_2O_3, $CaSO_42(H_2O)$) and heights have been made and using Eq. (3) a weighted linear fit through origin of $\ln(I_0/I)$ and μ is made obtaining t_{eff} from the fitted slope. If $t = 1$ cm (common for sediment samples), we have obtained that $t_{\text{eff}} = 1.03 \pm 0.05$ cm, so it could be concluded that t_{eff} is almost identical to t (see Fig. 3) and for this detector–source configuration that effect is negligible.

Additionally, we have simulated radioactive decay of isotopes (^{241}Am, ^{137}Cs and ^{60}Co) contained into point sources in order to check coincidence-summing effects, finding that they are also negligible for this measuring geometry.

Once obtained corrected FEP (ε) for each sample using Eq. (1) a non-linear least-square fit to a log–log function (see Fig. 4) is made in order to calculate sample activity A:

$$A = \frac{N - B}{\varepsilon p T m} \quad (5)$$

where N is the number of counts in the peak at energy E, B is the number of background counts, p is the photon emission probability, T is the measuring time, m the sample mass, and ε is corrected FEP calculated as shown before.

In order to check accuracy of the proposed method some sediment samples from Huelva estuary have been measured by gamma-ray spectrometry in order to obtain ^{210}Pb activity via its low-energy emission of 46.5 keV. This activity has been also measured using Liquid Scintillation Counting (LSC) [9] and alpha spectrometry. The results

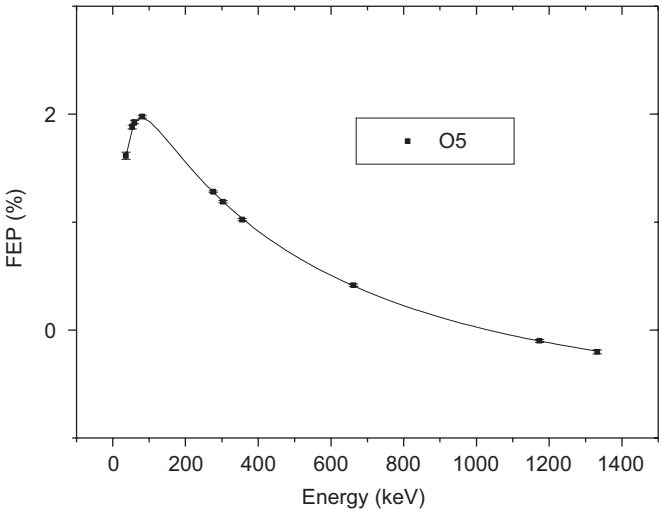

Fig. 4. FEP log–log fit for sediment sample O5.

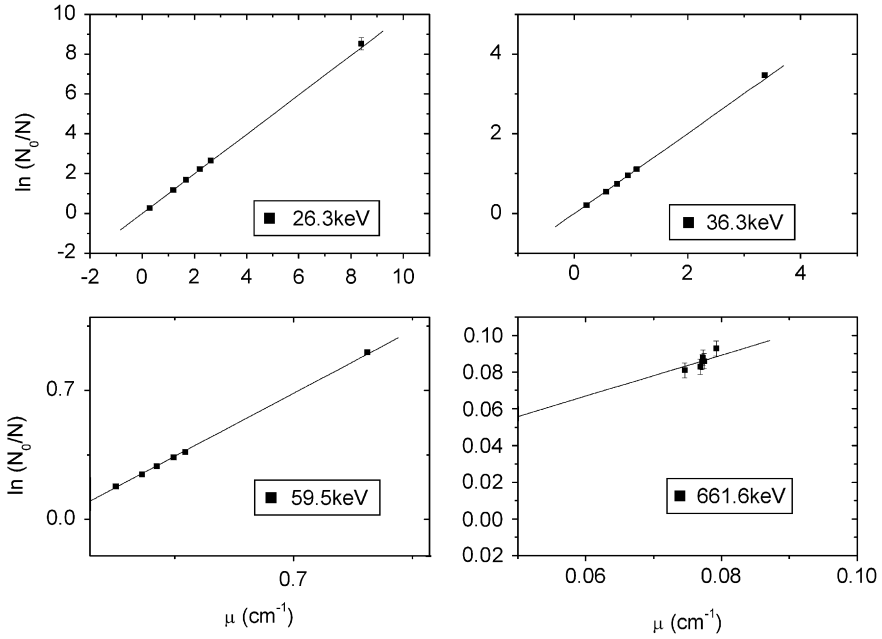

Fig. 3. Calculation of t_{eff} for a Petri dish with 1 cm of sample height.

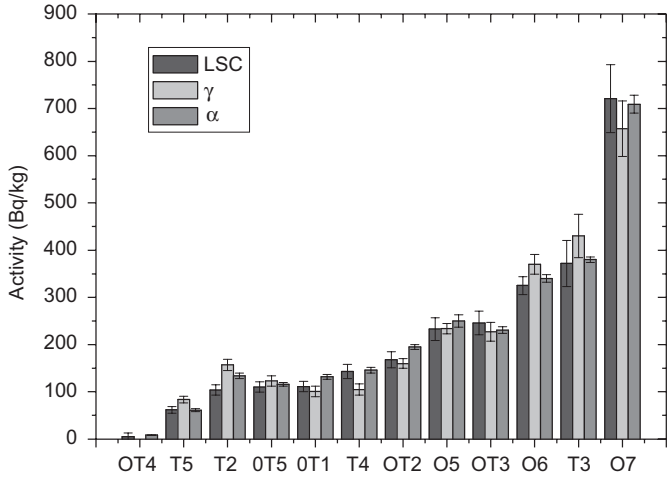

Fig. 5. ^{210}Pb activity for sediment samples measures with three methods: Liquid Scintillation Counting (LSC), gamma and alpha spectrometry.

showed (see Fig. 5) a good agreement between the three measuring methods.

5. Conclusions

A self-sufficient method for measurement of the activity of low-level gamma-emitters in voluminous samples by gamma-ray spectrometry with a coaxial germanium detector has been proposed. Only an extra transmission experiment per sample is needed for FEP determination regardless of sample height and emission energy. And MC simulations have run only one time for each measuring configuration.

The accuracy and precision of the proposed method has been checked calculating ^{210}Pb activities in sediment samples from an estuary located in the vicinity of several phosphates factories. In these samples, the ^{210}Pb levels have been previously determined independently using two alternative radiometric techniques.

Acknowledgments

Financial support from the Spanish Security Council (CSN) is specially acknowledged.

References

[1] N.H. Cutshall, I.L. Larsen, C.R. Olsen, Nucl. Instr. and Meth. A 206 (1983) 309.
[2] S. Joshi, Intern. J. Radiat. Appl. Inst. A 40 (1989) 691.
[3] O. Sima, D. Arnold, Appl. Radiat. Isot. 47 (1996) 889.
[4] R.M.W. Overwater, P. Bode, J.M. de Goeij, Nucl. Instr. and Meth. A 324 (1993) 209.
[5] M. Garcia-Talavera, V. Peña, Appl. Radiat. Isot. 60 (2004) 227.
[6] S. Hurtado, M. García-León, R. García-Tenorio, Nucl. Instr. and Meth. A 518 (2004) 764.
[7] E.G. San Miguel, J.P. Perez-Moreno, J.P. Bolivar, R. Garcıa-Tenorio, J.E. Martin, Nucl. Instr. and Meth. A 493 (2002) 111.
[8] M.J. Berger, J.H. Hubbell, S.M. Seltzer, J.S. Coursey, D.S. Zucker, XCOM: Photon Cross Section Database, version 1.2, National Institute of Standards and Technology, Gaithersburg, MD, 1999, ⟨http://physics.nist.gov/xcom⟩.
[9] M. Villa, G. Manjón, M. García-León, Nucl. Instr. and Meth. A 496 (2003) 413.