

Study of Efficiency Calibrations of HPGe Detectors for Radioactivity Measurements of Environmental Samples

S. Harb, K. Salahel Din and A. abbady

Physics department, Faculty of Science, South Valley University, 83523 Qena, Egypt.

Abstract

In this paper, we describe a method of calibrating of efficiency of a HPGe γ -ray spectrometry of bulk environmental samples (Tea, crops, water, and soil) is a significant part of the environmental radioactivity measurements.

Here we will discuss the full energy peak efficiency (FEPE) of three HPGe detectors it as a consequence, it is essential that the efficiency is determined for each set-up employed. Besides to take full advantage at γ -ray spectrometry, a set of efficiency at several energies which covers the wide the range in energy, the large the number of radionuclides whose concentration can be determined to measure the main natural γ -ray emitters, the efficiency should be known at least from 46.54 keV (^{210}Pb) to 1836 keV (^{88}Y). Radioactive sources were prepared from two different standards, a first mixed standard QCY40 containing ^{210}Pb , ^{241}Am , ^{109}Cd , and Co^{57} , and the second QCY48 containing ^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y , and ^{60}Co is necessary in order to calculate the activity of the different radionuclides contained in a sample.

In this work, we will study the efficiency calibration as a function of different parameters as:- Energy of gamma ray from 46.54 keV (^{210}Pb) to 1836 keV (^{88}Y), three different detectors A, B, and C, geometry of containers (point source, marinelli beaker, and cylindrical bottle 1 L), height of standard soil samples in bottle 250 ml, and density of standard environmental samples. These standard environmental sample must be measured before added standard solution because we will use the same environmental samples in order to consider the self-absorption especially and composition in the case of volume samples.

Introduction

In this paper is to design a simple, rapid and general method to find the FEPE curve in the calibrating matrix as a function of E_γ , the sample height, different detectors, different geometry and density for environmental samples (Hasse et al.[1], Pérez-Moreno et al.[2], Alfassi and Lavi [3], Hernandez [4]. For this, the only requirement is an active calibrating matrix with several γ -emissions in the energy range of interest; in our case 46.54-1836 keV. HPGe gamma detector is used for analysis environmental samples in which calibration is very important. To acquire the function ship between efficiency and energy in wider energy range. About 10 standard sources are now wed worldwide, among which, five nucleus' lifetimes are short. Since efficiency calibration is required to carry on once one week or two weeks, so spectrometry users have to pay much money for these sources. An efficiency calibration method has been developed to measure radioactivity of volume samples with HPGe detectors. In order to obtain correct results, the samples shall be counted under the same measuring conditions as those under which the system has been calibrated (Saegusa, et al.[5]).

For radioactivity measurement, the gamma-ray spectrometry with a high-purity germanium (HPGe) coaxial detector is widely used. In the method, a detection efficiency curve, that is, a set of photopeak efficiencies over the energy region of interest must be known in advance. The detection efficiency curve depends not only on a detection system but also on a sample shape and a sample matrix with different density and height of environmental samples.

Variability in the sample chemical composition is not major problem since mass attenuation coefficients (μ_m) differ only slightly from one environmental sample to another. This effect can be estimated using Monte-Carlo calculations [6, 7], or modeling [8].

Another method is the calibration of the spectrometers using well characterized artificial spiked matrices [9]. In this case, an efficiency curve can be experimentally determined and fitted for a large number of energies and matrices. For this purpose, the composition of the standards should approximate, as closely as possible, the density, attenuation factors and activity concentrations of the samples to be analyzed [10, 11].

Experimental

The calibration standard source must have physical dimensions, chemical composition, and density similar to the samples that will be analyzed, so that the deviation in the measured activity is minimized. With regard to the geometry, the deviation can be reduced almost to zero if the bottle has the same dimensions and the all heights are similar for the standard source and the samples. We have employed container in measuring the sample matrices considered in this work: 0.250 l. The 0.250 l bottle's dimensions are 6 cm diameter and 12 cm height. In order to obtain homogeneous sources, soil was sieved into 2mm.

Here we seek to generalize our method and we deal with different matrices. Therefore, we attempt to handle set-up characteristics which involve different magnitudes of both the geometrical and matrix effects. From the measurement of calibration sources, experimental efficiencies are calculated. The experimental efficiency at energy E_i for a given set of measuring conditions can be computed by equation 3.

The efficiency calibration of the HPGe detector available at the Zentrum für Strahlenschutz und Radioökologie (ZSR) Hannover University is presented.

Counting system

The work was carried out using three (indicated with A, B, and C) γ -ray detection made of high purity germanium (HPGe) in a vertical configuration cooled with liquid nitrogen. The detectors were properly shielded. The main technical specifications of these detectors are summarized in table 1.

Table 1 Main technical specifications of the three detectors (A, B, and C) used in this study

Detector	Detector model	type	Relative efficiency (%)	Crystal length-diameter (mm)	Energy resolution (FWHM)	
					At 122 keV	At 1332 keV
A	GEM 50198-P	n-Type coaxial	50	82.0-64.5	0.950	1.95
B	GL2820R	n-Type planar	-	20-60	0.739	-
C	GX3018	p-Type coaxial	35.0	53.5-60	0.852	1.78

Source preparation

The preparation of the standards can be made using "in house procedures". This can be done by homogeneously incorporating of certified and traceable solutions of radionuclide into inactive matrices with the same composition and density as the sample to be assayed.

However as is the case for all analysis techniques, in order to carry out a quantitative analysis, γ -ray spectrometry requires standard samples to establish an experimental efficiency calibration which is so far the most accurate. Nevertheless, it can be laborious if numerous configurations are present (e.g.: various γ -ray detectors, geometries, density and sample shapes) [12].

The sources are traceable through the Physikalisch-Technische Bundesanstalt (PTB) and are mainly monoenergetic gamma ray emitters covering a wide range of gamma ray energies. Calibration sources for each matrix were obtained by spiking commercially available standard solutions: the QCY48 and QCY40 mixture containing. For measurements at the reference geometry, 10 point-like sources were prepared by pipetting a definite volume of standard solution over a hydrophobic filter four of them were made with the ²¹⁰Pb solution. Regarding the measuring set-ups, for the preparation of aqueous sources we first partially filled a bottle with soil and then added a specific volume of radioactive solution (see Table 2). Afterwards, the bottle is always filled to the same height. Soil sources were prepared by pipetting a volume of the solution onto the soil, and subsequent drying over 24 h at 40^oC. To ensure that the radionuclides were homogeneously distributed in the source, the sample was carefully mixed prior to measurement. Finally, filter sources were made in a similar way to the point-like sources. Table 2 shows the activities of the 10 radionuclides, their half-lives, the energies of their gamma ray emissions and their gamma ray emission probabilities.

Table 2 The radioactivity standard solution supplied by (ZSR)

Nuclides	Gamma-ray Energy (keV)	Gamma-ray per second	Emission probability Per gram	Uncertainty (%)	Half life time (d)
Pb-210	46.54	439	0.0425	2	8145
Am-241	59.54	1128	0.359	2.6	158047
Cd-109	88.03	649	0.0361	6.2	462.6
Co-57	122.1	627	0.856	1.5	271.4
Ce-139	165.9	685	0.7989	1.4	137.64
Sn-113	391.7	2093	0.64	4.1	115.09
Sr- 85	514	3927	0.984	2.5	64.84
Cs-137	661.7	2484	0.851	2	10958
Y-88	898	6343	0.937	1.6	106.6
Co-60	1173	3346	1	1.5	1924.9
Co-60	1333	3348	1	1.5	1924.9
Y-88	1836	6705	0.992	1.3	106.61

Here we describe a general method of calibrating the efficiency of a HPGe γ -ray spectrometer. On this basis, we establish a general function to describe the energy dependence of the efficiency for the particular geometry and source matrix. The method has been applied to 11 different experimental arrangements to provide efficiency calibrations over the range 46.54-1836 keV. This allows high precision measurements with environmental samples, which often have very low activities.

Efficiency calibration method

The basis of the method is described in detail in this section.

Experimental efficiency

After the bottle is filled to the desired volume (height or weight) with the homogenous soil. Finally, the bottles were hermetically closed with screw cap. As the soil itself has some natural radioactivity, we measured also soil alone in the same bottle. Calibration of the natural radioactivity of the soil was done by comparison with to the activity of the bottles with standard solution sources. Gamma-ray was analyzed with GW program at ZSR Fig. 1.

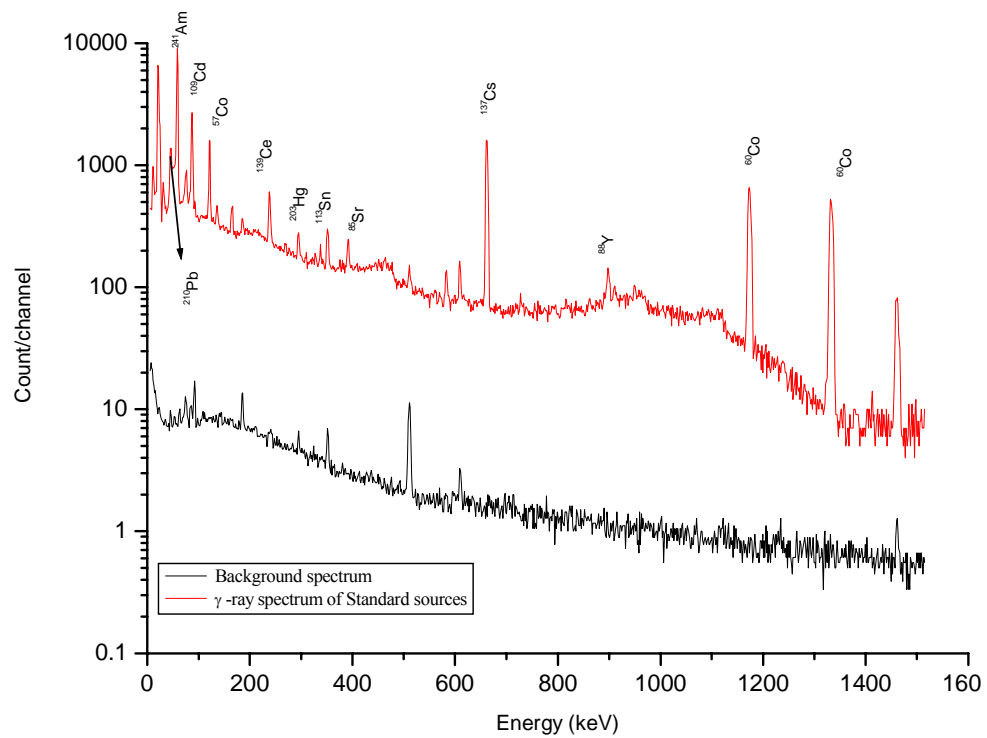


Fig. 1 γ -Ray energy calibration spectrum of a HPGe detector using several radioactive sources as indicated at the relevant peaks. Gamma ray spectrum of standard sources and background.

The dependence of the counting efficiency of our detector (E_{ff}) on the energy of the γ -ray photons (E_γ) in-house prepared standard of soil containing QCY48 is described quite well by the linear regression fitting.

Efficiency calculate and curves fitting

The absolute detector efficiency at that energy was then calculated by dividing the net count rate in the full-energy peak by the decay corrected gamma-ray-emission rate of the standard source. After the absolute detection efficiency was determined equation 3 for each calibration peak, a weighted least-squares fit was made to a polynomial expression of log of efficiency vs. log of energy. Efficiency curves were constructed from these full-energy-peak efficiencies.

$$\epsilon_{abs} = \frac{\text{Total number of counts recorded under the photo peak}}{\text{Total number of photons emitted by the standard sources}}$$

$$\epsilon_{abs} = \frac{\text{cps experimental}}{\text{cps theoretical}} \quad (1)$$

$$\epsilon_{abs} = \frac{\left(\frac{C}{S}\right)_{std} - \left(\frac{C}{S}\right)_{sample} - \left(\frac{C}{S}\right)_{BG}}{\left(\frac{C}{S}\right)_{theo.} \cdot \text{Exp}\left(-\ln(2) \cdot \frac{t}{t_{1/2}}\right)} \quad (2)$$

Where

$(C/S)_{std}$ Count of soil sample with standard solution

$(C/S)_{sample}$ Count of soil sample without standard solution

$(C/S)_{BG}$ Count of background

$(C/S)_{theo.}$ Counting of gamma ray of used standard solution (see table 2)

t the time of decay

$t_{1/2}$ half-life time of radionuclide

From the figure 2, we notice that the efficiency curve in the 45.54-1836 keV range shows two regions of different behavior because distinct attenuation and absorption processes dominate. At low energies the efficiency rises rapidly because of abrupt reduction in the attenuation in radioactive source, detector cap or inner dead layer. A maximum is reached for an energy value which depends on the detector and source characteristics. Above a few hundreds keV the efficiency decreases monotonically.

The procedure adopted for calculating the net area under the full energy absorption peak may influence the behavior of the efficiency curve, mainly at low gamma ray energies. In this region, a significant variation in the background counts can be observed comparing the plateaus located at the left and right sides of the peak. The results are compared with conventional linear polynomial fitting. The adopted procedure makes use of covariance analysis which is considered essential for complete description of all the uncertainties involved, as pointed out by Winkler [13], Geraldo and Smith [14].

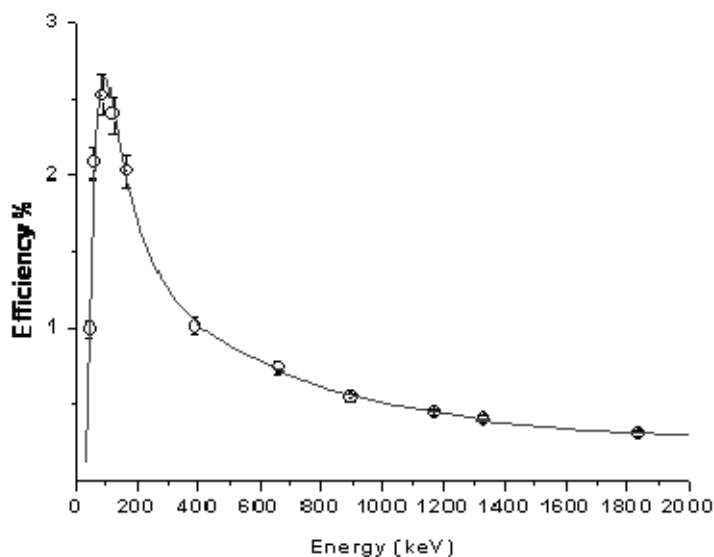


Fig. 2 Efficiency calibration curve obtained from the reference geometry bottle 250 ml with different gamma-ray energies.

1.1.1 Linear function

From the measurement of calibration sources, experimental efficiencies are calculated. The experimental efficiency at energy E for a given set of measuring conditions can be computed by:

$$\varepsilon_{\text{abs}} = a \times E^b \quad (3)$$

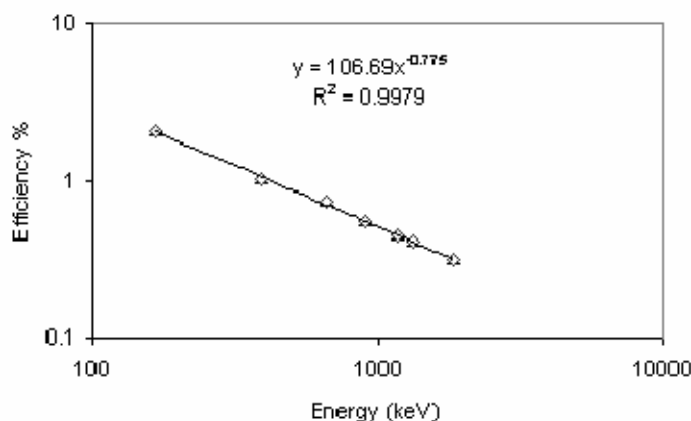


Fig.3 efficiency with energy

Where E is the energy of γ -ray and a, b are parameters from the Fig 3 are $a = 106.96$ and $b = -0.775$ for 270 g standard soil in bottle on detector A, and we used the equation 3 to determine relation between efficiency and energies higher than 165 keV. We used ε_{abs} for calculate the specific activity (in Bq kg^{-1}), $A_{E_{\gamma_i}}$ of a radionuclide i and for a peak at energy E_{γ} , is given by:

$$A_{E_{\gamma_i}} = \frac{NP}{t_c \cdot I_{\gamma}(E_{\gamma}) \cdot \varepsilon(E_{\gamma}) \cdot M} \quad (4)$$

where NP is the number of counts in a given peak area corrected for background peaks of a peak at energy E_{γ} , $\varepsilon_E(E_{\gamma})$ the detection efficiency at energy E_{γ} , t is the counting lifetime, $I_{\gamma}(E_{\gamma})$ the number of gammas per disintegration of this nuclide for a transition at energy E, and M the mass in kg of the measured environmental samples.

1.1.2 Nonlinear function

We used another treatment to obtain a relation between efficiency and energy and then used it to determine specific activities for nuclides less than 165 keV in the fig. 4, this treatment by Gray fit.

Once a sufficient number of data are acquired experimentally in the energy region of interest, a means of representing the efficiency as a function of energy should be chosen. Gray

fitting procedures are used to fit the efficiency data to an analytical expression. A generally accepted and simple expression for efficiency (ϵ) is as follows Gray [15] and Debertin[16].

$$\epsilon = \frac{P_1 + P_2 \cdot \ln(E) + P_3 \cdot \ln^2(E) + P_4 \cdot \ln^3(E) + P_5 \cdot \ln^5(E) + P_6 \cdot \ln^7(E)}{E} \quad (5)$$

where E represents energy in MeV. This expression is adequate for determining efficiency of gamma energies from 46.54 keV to 1836 keV.

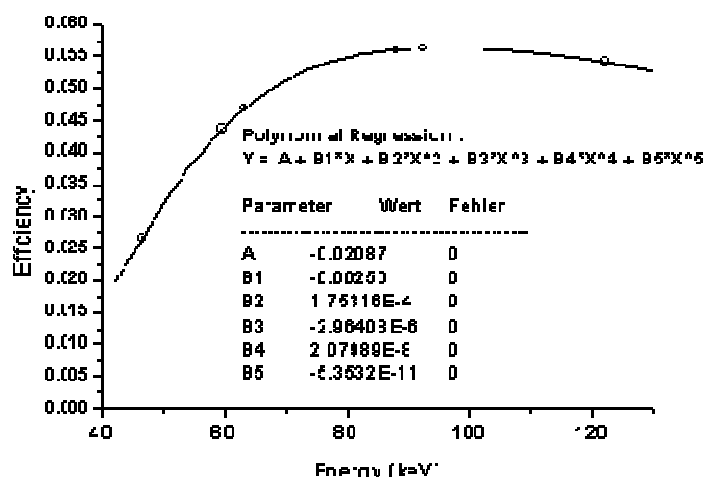


Fig. 4 efficiency - low energy by using Gary fit.

The dependence of the efficiency on the radiation energy was determined experimentally and by using the Gray and Ahmad fit function [16]

2 Results and discussion

2.1 Efficiency curve for standard sample

The use of extended sources in γ -ray spectrometry improves the sensitivity of detection thus enabling the measurement of low-activity environmental samples. To obtain reliable measurements of radionuclide activity, the knowledge of the detector absolute peak efficiency in the counting conditions is required. This becomes a complex problem whenever there are many types of matrices and geometries involved in the measurements because the count rate depends on the characteristics of the matrix, the source geometry and the source-detector configuration [15]. As a consequence, it is essential that the efficiency is determined for each set-up employed. Besides, to take full advantage of γ -ray spectrometry, a set of efficiencies at several energies is necessary in order to calculate the activity of the different radionuclides contained in a sample. Efficiency calibration as a function of energy provides the efficiency value at any energy within a given range, with the energy range covered being dependent on the application. The wider the range in energy, the larger the number of radionuclides whose concentration can be determined. To measure the main natural γ -ray emitters, the efficiency should be known at least from 46:54 keV (^{210}Pb) to 1836 keV (^{88}Y).

Efficiency- geometry curves of calibration source

Fig. 5 show efficiency curves for the soil source and measured for different geometry point source, marinelli beaker and bottle we can see that clearly the efficiency for point source higher than for marinelli and bottle.

Table: 3 shows the experimental results of the peak efficiency for different geometry.

Nuclides	Energy [keV]	Efficiency		
		point Source	Marinelli beaker	Bottle 1 L
Am-241	59.9	0.0622	0.0184	0.0052
Cd-109	88.4	0.1076	0.0472	0.0094
Co-57	122.4	0.1443	0.0517	0.0111
Ce-139	166.2	0.157	0.0515	0.0131
Zn-113	392.1	0.1057	0.032	0.0079
Sr-85	514.4	0.0704	0.0275	0.0078
Cs-137	661.9	0.0548	0.0239	0.0060
Y-88	898.2	0.0404	0.0172	0.0049
Co-60	1173.2	0.0327	0.0146	0.0042
Co-60	1332.3	0.0273	0.0134	0.0039
Y-88	1835.5	0.0188	0.0107	0.0032

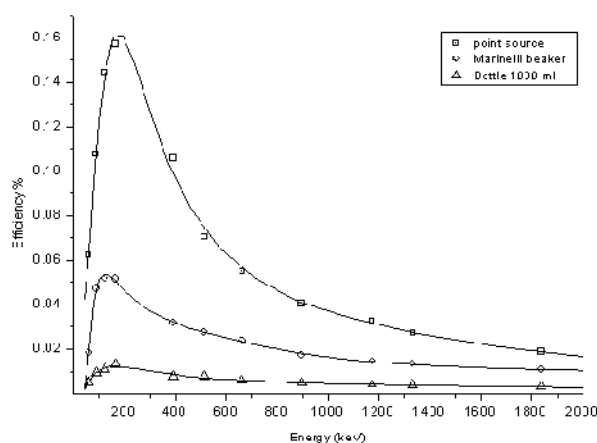


Fig. 5 variation of efficiency-energy with different geometry of standard source

Efficiency- density curves of calibration source

The efficiency calibration of HPGe detector is more complex for energies below 165 keV. The peak efficiency calibration, in this energy range, does depend strongly on the composition and density of the source. The efficiency calibration is, therefore, done relative to a known source composition. In order to avoid this problem the soil of standard must be the same soil which wanted to measure them.

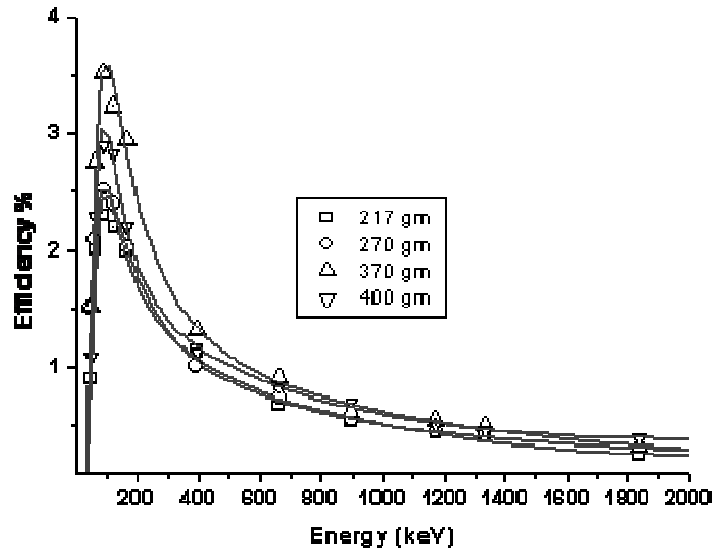


Fig. 6 Variation of efficiency-energy curves with densities of soil source for a constant volume.

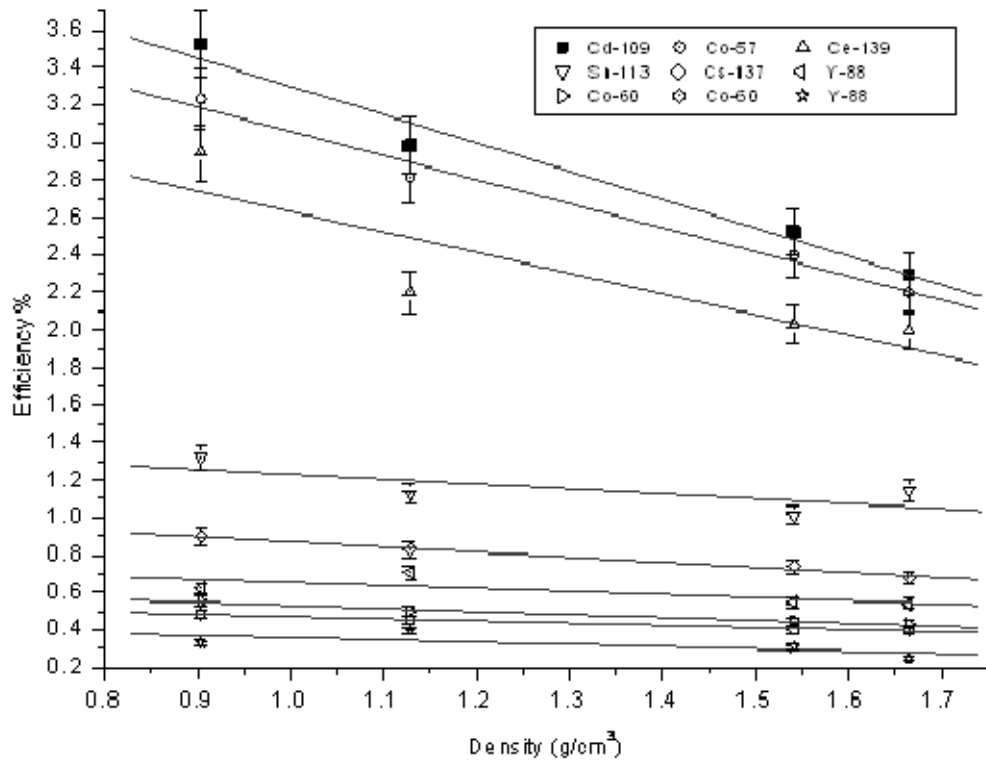


Fig. 7 Variation of the peak efficiency as a function of the density ρ for a standard soil sample in bottle 250 ml measured by detector (A) considered and various photon energies.

The present data presented in Figs. 6 and 7, it can be noticed that for the ^{40}K (1460.8 keV), whose energy lies between the bottom two lines of the illustration, the variation in the efficiency is insignificant. For the energy interval from the ^{60}Co to the ^{88}Y , a horizontal line is obtained for the whole density range, from 0.9 to 1.75 g/cm³. So, for the mentioned work F.L. Melquiades [10], no variation in the counting efficiency due to the sample densities from 0.9 to 1.75 g/cm³ for that energy range was evident. On the other hand, another work, Park et al. [17], shows a smooth inclination in the straight lines for the ^{60}Co and the ^{88}Y energies showing that, in fact, there is a dependence of efficiency on the sample density for this energy range.

Debertin and Jianping [15] show how from a calibration of a detector with a Marinelli beaker filled with one material the calibration for another density can be calculated. However, it is best to calibrate experimentally the beaker for the required density. This is the reason why it is so important to be able to prepare a calibrated Marinelli secondary standard in each laboratory. In papers Lavi [18], they described a procedure for preparation of a homogenous mixture of natural thorium oxide (ThO₂) (^{232}Th at secular equilibrium with its daughters) with either CaCO₃ or dried milk and described the preparation of similar radioactive sources with various densities in order to study the dependence of the efficiencies of γ counting of a Ge detector on the density of the matrix for 450 and 1000 ml Marinelli beakers.

Efficiency- height curves of calibration source

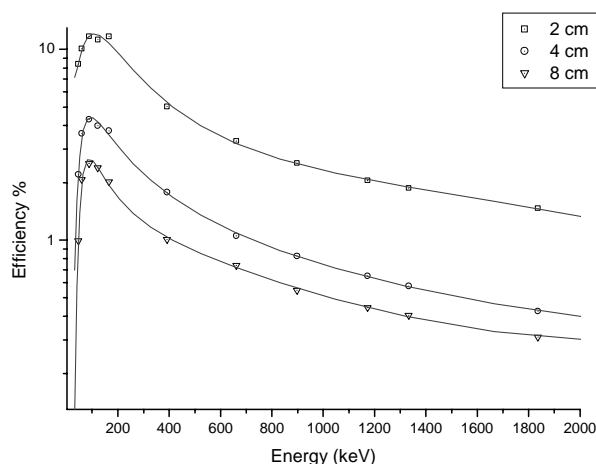
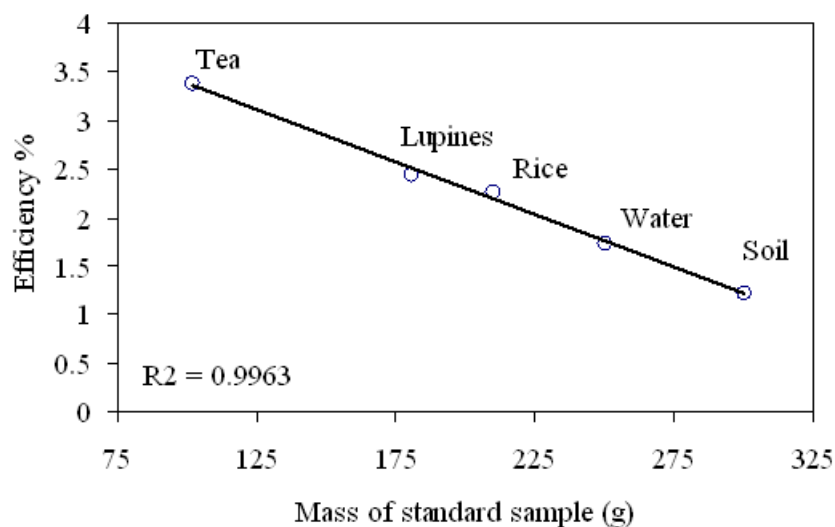


Fig. 8 variation of efficiency-energy curves with heights of soil sample (bottle 250 ml) source for constant density.

We have estimated the full energy peak efficiency in the calibration sample by using different sample heights: ϵ_{abs} was determined for heights ranging from 2 to 8 cm, with intervals of 2 cm. In Fig. 8, we have plotted the different values of ϵ_{abs} ; together with the function obtained by least square fitting of $\epsilon_c(h)$ data. We have selected a polynomial function of order two due to its simplicity and the good results supplied [19, 20].

Efficiency- masses curve of calibration source

We have estimated the full energy peak efficiency in the environmental samples by using



different samples(Tea, Lupines, Rice, Water and Soil): ϵ_{abs} was determined for masses ranging from 102 to 300 g in constant bottle(250 ml) for 45.54 keV (Pb-210).

Fig. 9 variation of efficiency curve with different density environmental samples in constant volume (bottle 250 ml) for Pb-210.

In Fig. 9, we have plotted the different values of ϵ_{abs} and different masses of different environmental samples for Pb-210.

Conclusion

The absolute full-energy peak efficiency of high purity germanium (HPGe) detectors has been measured between (46,54 and 1836) keV using the environmental samples matrix standard with both single-photon emitting nuclides mixed standard QCY40 containing ^{210}Pb , ^{241}Am , ^{109}Cd , Co^{57} , and the second QCY48 containing ^{241}Am , ^{109}Cd , Co^{57} , ^{139}Ce , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y , and ^{60}Co .

From the results the efficiency depends on the gamma ray energy, geometry, density, height of soil sample and characterization of detectors.

References

- [1] G. Hasse, D. Tait and A. Wiechen “ determination of full energy peak efficiency for cylindrical volume sources by the use of a point source standard in gamma-spectrometry” Nuclear Instruments and Methods in Physics Research A 361 (1995) 240-244
- [2] J. P. Pérez-Moreno, E. G. San Miguel, J.P. Bolívar, J. L. Aguado “A comprehensive calibration method of Ge detectors for low-level gamma-spectrometry measurements” Nuclear Instruments and Methods in Physics Research A 491 (2002) 152–162
- [3] Z. B. Alfassi, N. Lavi “The dependence of the counting efficiency of Marinelli beakers for environmental samples on the density of the samples” Applied Radiation and Isotopes 63 (2005) 87–92
- [4] F. J. Hernandez “Optimization of environmental gamma spectrometry using Monte Carlo methods” Ph.D. thesis, Uppsala University, Sweden. 2002.

- [5] J. Saegusa, Katsuya Kawasaki, Akira Mihara, Mitsuo Ito, Makoto Yoshidaa "Determination of detection efficiency curves of HPGe detectors on radioactivity measurement of volume samples" *Applied Radiation and Isotopes* 61 (2004) 1383–1390
- [6] M. Jurado Vargas, A. Fernandez Timon, N. Cornejo Diaz, D. Pérez Sánchez "Monte Carlo simulation of the self-absorption corrections for natural samples in gamma-ray spectrometry" *Applied Radiation and Isotopes* 57 (2002) 893–898
- [7] M. Jurado Vargas, N. Cornejo Dazb, D. Pérez Sánchezab "Efficiency transfer in the calibration of a coaxial p-type HPGe detector using the Monte Carlo method" *Applied Radiation and Isotopes* 58 (2003) 707–712
- [8] T. Vidmar, M. Korun, A. Likar, R. Martinčič "A semi-empirical model of the efficiency curve for extended sources in gamma-ray spectrometry" *Nuclear Instruments and Methods in Physics Research A* 470 (2001) 533–547
- [9] M. S. Al-Masri, A. Aba, A. Al-Hamwi, A. Shakhashiro "Preparation of in-house reference soil sample containing high levels of naturally occurring radioactive materials from the oil industry" *Applied Radiation and Isotopes* 61 (2004) 1397–1402.
- [10] F.L. Melquiades, C.R. Appoloni "Self-absorption correction for gamma spectrometry of powdered milk samples using Marinelli beaker" *Applied Radiation and Isotopes* 55 (2001) 697–700
- [11] E.G. San Miguel, J.P. P!erez-Moreno, J.P. Bolivar, R. Garc!ya-Tenorio, J.E. Martina "²¹⁰Pb determination by gamma spectrometry in voluminal samples (cylindrical geometry)" *Nuclear Instruments and Methods in Physics Research A* 493 (2002) 111–120
- [12] M. I. Abbas, Younis S. Selim "Calculation of relative full-energy peak efficiencies of well-type detectors" *Nuclear Instruments and Methods in Physics Research A* 480 (2002) 651–657
- [13] Winkler, G., 1998. "On the role of covariances for uncertainty estimates in radioactivity measurements" *Appl. Radiat. Isot.* 49 (9–11), 1153.
- [14] Geraldo, L.P., Smith, D.L., 1990. "Covariance analysis and fitting of germanium gamma-ray detector efficiency calibration data" *Nucl. Instrum. Methods A* 290, 499.
- [15] K. Debertin and R. G. Helmer; "Gamma and x-ray spectrometry with semiconductor detectors" Elsevier science publishers Amestrdam (1988).
- [16] P. W. Gray, A. Ahmed "Linear classes of Ge(Li) detector efficiency functions" *Nuclear Instruments and Methods in Physics Research A* 237, 577-589, (1985).
- [17] Park, T.S., Jeon, W.J., 1995. Measurement of radioactivity samples in Marinelli beakers by gamma- ray spectrometry. *J. Radioanal. Nucl. Chem.* 193, 133–144.
- [18] N. Lavia, Z. B. Alfassi, "Development and application of Marinelli beaker standards for monitoring radioactivity in Dairy-Products by gamma-ray spectrometry" *Applied Radiation and Isotopes* 61 (2004) 1437–1441
- [19] M. J. Daza, B. Quintana M. Garca Talavera, F. Fernandez "Efficiency calibration of a HPGe detector in the [46.54-2000] keV energy range for the measurement of environmental samples" *Nuclear Instruments and Methods in Physics Research A* 470 (2001) 520-532
- [20] M. S. Dias, V. Cardoso, V.R. Vanin, M.F. Koskin "Combination of nonlinear function and mixing method for fitting HPGe efficiency curve in the 59–2754 keV energy range" *Applied Radiation and Isotopes* 60 (2004) 683–687