

EFFICIENCY CALIBRATION OF A GAMMA-RAY SPECTROMETRY SYSTEM AND SELF-ABSORPTION MEASUREMENTS OF THE 46.5 keV GAMMA-RAY EMISSION FROM Pb-210 FOR THE QUANTITATIVE DETERMINATION OF RADIONUCLIDES IN SAMPLES

Marcelo Francis Máduar¹ and Pedro Miranda Junior²

¹ Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
mmaduar@ipen.br

² Instituto Federal de Educação, Ciência e Tecnologia de São Paulo (IFSP)
R. Pedro Vicente 625
01109-010 São Paulo, SP
pmirajr@gmail.com

Keywords: gamma-ray spectrometry, self-absorption, Pb-210, radionuclide determination.

ABSTRACT

Determination of activity concentration of gamma-ray emitting radionuclides in volumetric samples is usually accomplished with a hyper-pure Germanium semiconductor detector (HPGe) coupled to multichannel analyzer, amplifier and high voltage supply, comprising a gamma spectrometry system. Such a system was recently put into operation and calibrated by counting of radioactive sources with known activities of gamma-emitting radionuclides in the energy range required. Counting of 15 radioactive solutions, each containing known concentrations of 6 radionuclides, was performed. For the determination of efficiency points, 17 gamma lines from them were selected to achieve a calibration curve in the energy range from 30 to 1350 keV. The linear fit was done using composed polynomial functions in the form $y = f(x)$, $x = \ln(E)$, where E is the energy in keV, and $y = \ln(\epsilon)$ where ϵ is the counting efficiency. The validation of the fit was performed with a certified IAEA source in phosphogypsum matrix containing Pb-210 and Ra-226. Due to the difference of density and elemental composition of the certified material relative to those of the calibration sources, correction for attenuation of the 46.5 keV gamma-ray from Pb-210 by the sample itself was done, by adopting the procedure described by Debertin & Ren. The calibration curve fit adherence to the experimental data showed a correlation coefficient of $r = 0.9958$. The final results obtained with the methodology described for the certified material were acceptable according to the criteria adopted by the IAEA to evaluate precision and accuracy.

1. INTRODUCTION

Determination of activity concentration of gamma-ray emitting radionuclides in volumetric samples is usually accomplished with a hyper-pure Germanium semiconductor detector (HPGe) coupled to multichannel analyzer, amplifier and high voltage supply, comprising a gamma spectrometry system.

However, the viability of using such a system in quantitative determination of gamma-ray emitters in volumetric matrices depends heavily on the correct determination of the

relationship between the energies of the total absorption photopeaks and the efficiency of counting for a given geometric configuration of the sample/detector system, relationship thereafter named efficiency calibration.

A new gamma-ray spectrometry system was recently put into operation at the Environmental Radiometry Laboratory (LRA) at IPEN-CNEN/SP. In this work, the efficiency calibration of the system was performed and the curve of calibration was validated by using a certified material from IAEA. A correction for the attenuation of 46.5 keV gamma radiation from ^{210}Pb , by the sample of certified material itself, was also applied.

2. EXPERIMENTAL

Efficiency calibration was performed by counting radioactive sources with known activities of gamma-emitting radionuclides in the energy range required. Counting of 15 radioactive solutions, each containing known concentrations of ^{60}Co , ^{65}Zn , ^{106}Ru , ^{133}Ba , ^{134}Cs and ^{137}Cs was performed. For the determination of efficiency points, 17 gamma lines from these radionuclides were selected to achieve a calibration curve in the energy range from 30 to 1350 keV.

2.1. Counting of the Reference Radioactive Solutions

The laboratories of LRA participate routinely in the program of intercomparison exercises, known as PNI, for the evaluation of analytical performance of Brazilian metrology laboratories, coordinated by the Institute of Radioprotection and Dosimetry (IRD) [1].

In order to establish the efficiency vs. energy curve, undiluted samples from 3 editions of the PNI – 5 samples of each edition (August/2009, December/2009 and April/2010), totalizing 15 samples of gamma emitters in water – were counted with 200 000 s of live time.

The samples were prepared by transferring 100 mL of each matrix solution, without dilution, to polyethylene flasks, that were sealed and counted. The gamma spectrometry counting system consists of a coaxial Be-layer HPGe detector from Canberra, model GX2518, with 25% relative efficiency, near 1.8 keV resolution at 1.33 MeV, and an integrated module DSA1000 with 4000 V supply and 8192 channels buffer. Counting areas were determined by analyzing the gamma spectra with the software Genie 2000 [2].

2.2. Efficiency vs. Energy Calibration

Efficiency values of the efficiency were determined according to the expression [3]

$$\varepsilon = \frac{C}{A \cdot t \cdot I} \quad (1)$$

where

ε is the counting efficiency, for the system and geometry considered, at the energy E (adim.)

C is the counting area of the peak corresponding to the gamma energy E (adim.)

A is the reference value for the activity in the sample of the radionuclide emitting the gamma with energy E (Bq)

t is the live time of counting of the sample (s)

I is the gamma emission probability per disintegration for the gamma transition with energy E (adim.)

For best results, usually a dual efficiency curve is calculated, in order to achieve a better fit over the entire energy range of interest. This approach consists in define 2 sets of points, one below and another above a crossover energy adequately chosen, and then perform 2 separate fittings, both employing the crossover point. In this work, this point was at 81.00 keV, corresponding to a gamma-ray from ^{133}Ba .

The linear fit was done using a polynomial function $y = f(x)$, $x = \ln(E)$, where E is the energy in keV, and $y = \ln(\varepsilon)$ where ε is the counting efficiency [3]. Formally, each partial fit with degree n is expressed as

$$\varepsilon(E) = \exp \left\{ \sum_{i=0}^n a_i (\ln E)^i \right\} \quad (2)$$

2.3. Activity determination

Once the efficiency vs. energy curve $\varepsilon(E)$ has been obtained, the activity per mass concentrations (A/m) of gamma emitters in matrices prepared in the same geometry can be obtained by

$$A/m = \frac{C}{\varepsilon(E) \cdot t \cdot I \cdot m} \quad (3)$$

where m is the quantity of sample in kg and A/m the concentration, in Bq/kg, of the radionuclide emitting the gamma with energy E .

2.4. Self-attenuation correction

When the sample has an apparent density near to that of the water, the analytical procedure ends at the point above and the values can be taken as the final results. Typically, this is the case in the radioactivity determination in liquid samples or low density pulverized material.

However, when the sample has apparent density above circa 1.2 g.cm^{-3} , or when its elemental composition significantly differs from that of the water, low energy gamma radiation can be

more effectively attenuated by the sample itself than would be by water [4]. Therefore, in these cases, the correction for self-attenuation must be performed for each energy of interest.

In this work, the correction for attenuation of the 46.5 keV gamma-ray from ^{210}Pb by the sample itself was achieved by adopting the procedure described by Debertin & Ren [4], which employs a collimated beam from ^{210}Pb point source positioned above the sample and above a blank sample of deionized water. Fig. 1 shows a vertical section of the experimental arrangement.

The steps of the procedure are:

- Measurement of the relevant parameters of the counting geometry: radius and height of the counting flask and distance from the bottom of the flask and the top of the detector case;
- Counting of the ^{210}Pb source placed at the top of a flask filled with deionized water and determination of the counting area (A_w) of the 46.5 keV photopeak.
- Counting of the same source, now placed at the top of the flask with the sample and determination of the counting area ($A_{m, \text{gross}}$).
- The liquid area (A_m) is obtained by subtraction between ($A_{m, \text{gross}}$) and the area corresponding to the sample measured alone, without the ^{210}Pb source, normalized for the counting time of ($A_{m, \text{gross}}$).
- Application of the auto-attenuation model [4] to determine the factor of attenuation due to the sample.

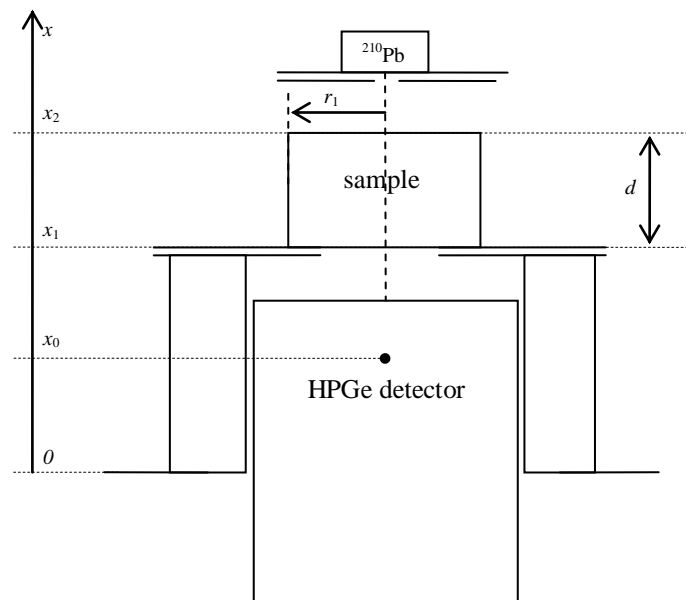


Figure 1. Vertical section of the experimental setup with the geometric parameters (adapted from [4]).

The attenuation coefficient of the sample matrix is obtained by

$$\mu_m = \mu_w + \frac{1}{d} \cdot \ln \left(\frac{A_w}{A_m} \right) \quad (4)$$

The factors F for the water (F_w) and for the sample matrix (F_m) are obtained by evaluating numerically the integral below, successively replacing μ by μ_w and μ_m .

$$F = \int_{x_1}^{x_2} \int_0^{r_1} \exp \left[-\mu \cdot \frac{\sqrt{r^2 + (x - x_0^2)}(x - x_1)}{x - x_0} \right] \cdot \frac{2\pi r}{r^2 + (x - x_0^2)} dr dx \quad (5)$$

The numerical integrations were performed with the aid of the numerical analysis software GNU Octave [5]. The corrected activity is then calculated by

$$A_{corr} = A \cdot \frac{F_w}{F_m} \quad (6)$$

For the geometry under study, the parameters are: $x_1 = 3.2$ cm, $x_2 = x_1 + 3.8 = 7.0$ cm, $r_1 = 2.9$ cm, $x_0 = 1.0$ cm and $d = 3.8$ cm. Attenuations coefficient for water at $E = 46.5$ keV is $\mu_a = 0.2382$ cm⁻¹.

3. RESULTS AND DISCUSSION

3.1. Determination of efficiency curve

Table 1 shows the gamma lines chosen for the determination of the efficiency points.

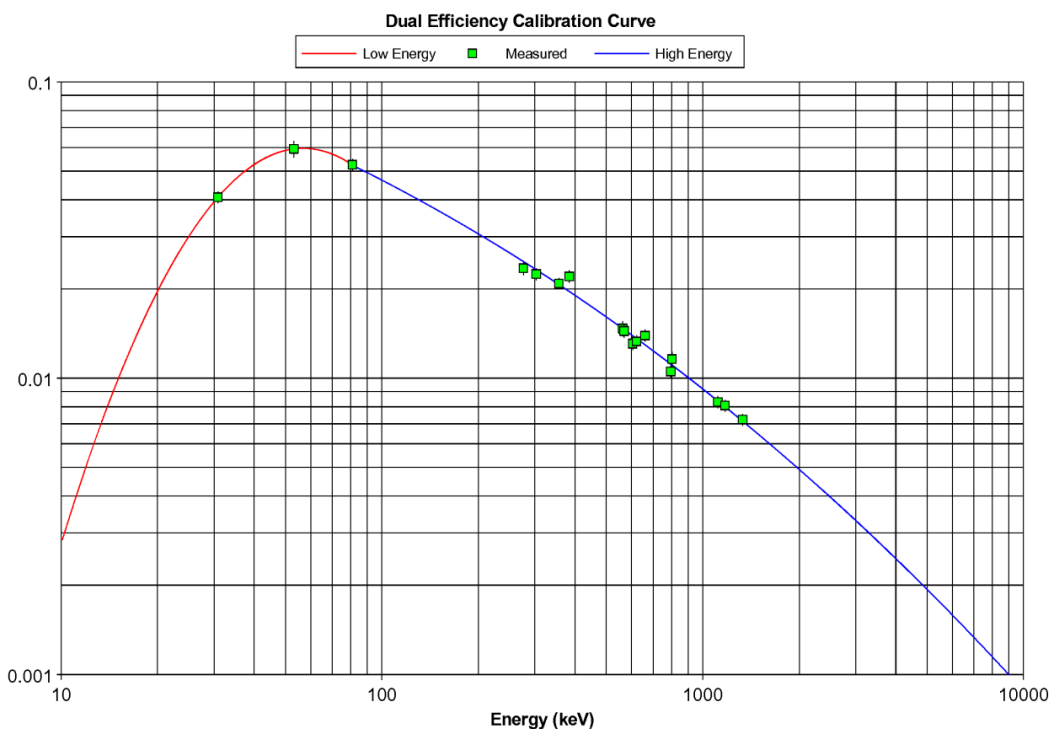
The dual efficiency curve was determined according to the item 2.2 and the parameters are shown in Table 2. Fig. 2 shows the experimental points and the plot of the dual fit.

Table 1. Experimental points of the efficiency vs. energy curve

Radionuclide	Energy (keV)	Efficiency	Uncertainty
^{133}Ba	30.85	4.08×10^{-2}	0.17×10^{-2}
^{133}Ba	53.16	5.94×10^{-2}	0.36×10^{-2}
^{133}Ba	81.00	5.25×10^{-2}	0.22×10^{-2}
^{133}Ba	276.40	2.35×10^{-2}	0.11×10^{-2}
^{133}Ba	302.85	2.25×10^{-2}	0.10×10^{-2}
^{133}Ba	356.02	2.09×10^{-2}	0.09×10^{-2}
^{133}Ba	383.85	2.21×10^{-2}	0.10×10^{-2}
^{134}Cs	563.23	1.47×10^{-2}	0.08×10^{-2}
^{134}Cs	569.32	1.44×10^{-2}	0.07×10^{-2}
^{134}Cs	604.70	1.31×10^{-2}	5.56×10^{-4}
^{106}Ru	621.84	1.33×10^{-2}	5.83×10^{-4}
^{137}Cs	661.66	1.39×10^{-2}	6.00×10^{-4}
^{134}Cs	795.84	1.05×10^{-2}	4.51×10^{-2}
^{134}Cs	801.93	1.16×10^{-2}	6.40×10^{-4}
^{65}Zn	1115.55	8.29×10^{-3}	3.58×10^{-4}
^{60}Co	1173.24	8.07×10^{-3}	3.48×10^{-4}
^{60}Co	1332.50	7.23×10^{-3}	3.12×10^{-4}

Table 2. Efficiency curve parameters

Parameters a_i for $E \leq 81 \text{ keV}$	Parameters a_i for $E > 81 \text{ keV}$
$a_0 = -1.945 \times 10^1$	$a_0 = -1.578 \times 10^0$
$a_1 = +8.233 \times 10^0$	$a_1 = -1.140 \times 10^{-1}$
$a_2 = -1.019 \times 10^0$	$a_2 = -3.889 \times 10^{-2}$
	$a_3 = -1.419 \times 10^{-3}$



Datasource: C:\GENIE2K\CAMFILES\Pni\2009_Dezagua 01 teste 2010 nova curva2.CNF
 $\ln(\text{Eff}) = -1.945e+001 + 8.233e+000 \cdot \ln(E) - 1.019e+000 \cdot \ln(E)^2$
 $\ln(\text{Eff}) = -1.578e+000 - 1.140e-001 \cdot \ln(E) - 3.889e-002 \cdot \ln(E)^2 - 1.419e-003 \cdot \ln(E)^3$

Figure 2. Dual efficiency calibration curve for the gamma spectrometry system for the sample geometry of 100 mL flask.

2.3. Determination of the ^{210}Pb and ^{226}Ra activity concentrations

The validation of the fit was performed with a IAEA reference material [6], consisting of a phosphogypsum matrix containing ^{210}Pb and ^{226}Ra . The source was prepared in the 100 mL polyethylene flask, sealed and set apart for a minimum 30-day period to achieve ^{226}Ra / short-lived decay products radioactive equilibrium. After that, the sample was measured (live time = 200 000 s) and analyzed employing the efficiency curve described above, using the Genie 2000 software. Determination of ^{210}Pb activity employed the 46.5 keV gamma line; for ^{226}Ra , gamma lines of 295 and 352 keV by ^{214}Pb and 609 keV by ^{214}Bi were used.

Due to the difference of density and elemental composition of the certified material relative to those of the calibration sources, correction for attenuation of the 46.5 keV gamma-ray from ^{210}Pb was performed as described earlier. The attenuation coefficient at this energy, for the reference material matrix, was found to be $\mu_m = 0.564 \text{ cm}^{-1}$ resulting in a correction factor $F_w/F_m = 1.45$.

Table 3 shows the final results for activity concentrations. The corresponding IAEA reference values and uncertainties [6] are also shown.

Table 3. Results of the activity concentration obtained in this work and corresponding reference values from IAEA.

	Results obtained (Bq/kg)		Reference values (Bq/kg)	
	Activity	Uncertainty ^a	Activity	Uncertainty ^a
²¹⁰ Pb	796	50	680	58
²²⁶ Ra	807	9	780	62

a. Confidence interval: 68% (1 σ)

The application of the IAEA criteria [7] for evaluation of the performance of the analytical results is outlined in Table 4. The estimator P, that evaluates the claimed measurement uncertainty, is defined [7] as

$$P = \sqrt{\left(\frac{u_{target}}{value_{target}}\right)^2 + \left(\frac{u_{reported}}{value_{reported}}\right)^2} \quad (7)$$

Table 4. Performance evaluation by applying IAEA criteria.

	Relative bias	Estimator P	Trueness	Precision	Final score
²¹⁰ Pb	17.11%	7.61%	A	A	A
²²⁶ Ra	3.48%	4.13%	A	A	A

A: acceptable

3. CONCLUSIONS

The calibration curve fit adherence to the experimental data showed a correlation coefficient of $r = 0.9958$. The final results obtained with the methodology described for the certified material were acceptable according to the criteria adopted by the IAEA to evaluate precision and accuracy.

The need for auto-attenuation corrections for low gamma energies such as that from the ²¹⁰Pb became evident, as they led to results 45% greater than the uncorrected figures.

ACKNOWLEDGMENTS

The authors acknowledge the collaboration of the LRA colleagues in the counting and preparation of samples for the measurements.

REFERENCES

1. L. Tauhata, M.E.C. Vianna, A.E. Oliveira, A.C.M Ferreira, C.C.S. Conceição, “Metrological capability of the Brazilian laboratories of analyses of radionuclides in environmental samples”, *Appl. Radiat. Isot.*, **56**, pp. 409-414 (2002).
2. Canberra Inc., “Genie™ 2000 Gamma Analysis Software”, http://www.canberra.com/pdf/Products/SW_pdf/5gamma.pdf (2006).
3. G. F. Knoll, *Radiation Detection and Measurement*, 3rd ed., John Wiley & Sons, USA (2000).
4. K. Debertin and Ren Jianping, “Measurement of the activity of radioactive samples in Marinelli beakers”, *Nucl. Instr. Meth. Phys. Res.*, **A278**, pp. 541-549 (1989).
5. Octave Documentation, <http://www.gnu.org/software/octave/docs.html> (2011).
6. International Atomic Energy Agency, “Reference Material IAEA 434: Naturally Occurring Radionuclides in Phosphogypsum”, IAEA, Vienna, 2010 (IAEA/AQ/17).
7. International Atomic Energy Agency, “Worldwide Open Proficiency Test: Determination of Naturally Occurring Radionuclides in Phosphogypsum and Water – IAEA-CU-2008-03”, IAEA, Vienna, 2010 (IAEA/AQ/18).