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HPGe Detector Efficiency Calibration for Extended Sources in the 50–1400 keV Energy Range

L. VENTURINI[1] and V. R. VANIN[2]

[1]IPEN-CNEN/SP, SPA, CP 11049, CEP 05422-970, São Paulo, Brazil and [2]Instituto de Física, USP, CEP 20516-970, São Paulo, Brazil

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A non-linear function is proposed to fit the experimental efficiency data for HPGe detector in the 50–1400 keV energy range for extended sources: 100 mL flask and 850 mL Marinelli beaker. The experimental data were corrected for coincidence summing effect. Covariances were taken into account. Simulation showed that the fitted data are Gaussian like distributed and the fit presents no appreciable bias.

Introduction

The efficiency calibration is of considerable importance in many applications of the gamma-ray spectrometry as, for example, environmental and liquid effluents monitoring. To present a suitable calibration function for extended samples and the procedure for fitting the experimental data is the subject of this paper.

The activity of a radionuclide in a sample measured by gamma-ray spectrometry is usually determined as

$$A_0 = \frac{C}{I_\gamma \cdot \epsilon \cdot t} \tag{1}$$

where: A_0, activity; C; full energy absorption peak net counts corresponding to the energy E; I_γ, gamma-ray intensity; ϵ, counting system efficiency at the energy E for the assayed geometry; and t, counting time.

The efficiency ϵ means the ratio between the corrected number of pulses registered in the full energy absorption peak and the number of photons emitted by the sample. This efficiency takes into account the absorption and attenuation factors involved in the gamma-ray detection for a given sample geometry. The net counts C must be corrected for secondary detection effects.

When I_γ is well known and C can be well determined, the efficiency value given by a fitted curve will be the main source of uncertainty in the calculated activity. The experimental data for efficiency calibration can be obtained by measuring calibrated commercially available radioactive solutions. Equation (1) also allows the efficiency evaluation at the energy of each gamma transition for each standard radionuclide. The choice of the activities in the calibrated standard samples is very important to avoid pulse pile-up and spectrum analysis difficulties. These activities must be high enough to allow good counting fractional precision and low enough to avoid peak distortions and complicated spectrum analysis. The use of radionuclides such as ^{60}Co, ^{133}Ba and ^{134}Cs, can require summing effect corrections. For such radionuclides, the greater the solid angle the higher the probability of registering the gamma-rays emitted in cascade out of the total energy absorption peaks. This is a typical secondary detection effect when measuring samples placed near the detector.

The efficiency curve in the 50–1400 keV energy range shows two regions where different gamma-ray absorption and attenuation processes predominate. At lower energies the efficiency increases fast as the energy increases until a maximum value is reached. The location of this maximum depends on both detector and sample characteristics. In this region the efficiency is limited by the self absorption of the photons in the sample and by the photon attenuation and absorption in the detector dead-layers and case. After reaching the maximum the efficiency decreases with the energy increase. In this region the mean free path of the photons in the detector active volume and the self absorption of photons in the sample determine the efficiency.

These two regions in the efficiency curve should be described by an energy dependent curve which properly represents the experimental data. Several curve fitting techniques have already been analyzed by Seymour *et al.* (1988) who has summarized their main advantages and problems. The Least Squares Method (LSM) and the non-linear methods seem to be

the most suitable ones for treating experimental data with nuclear or spectral effects.

In measuring geometries such as Marinelli beakers and flasks the coincidence-summing and the self absorption effects must be considered since a great portion of the sample is placed near the detector active volume.

This paper intends to describe the efficiency in the 50–1400 keV energy range by a continuous, differentiable and non-linear energy dependent function. The experimental data treatment has been done by using the Least Squares Method and the matrix formalism from Eadie *et al.* (1971) and Kendall and Stuart (1979). Correlations between the experimental data have been also accounted for and the experimental efficiency values were corrected for the summing-effect. The interactive algorithm used was that suggested by Marquardt (1963). The bias of the estimates were verified by simulation.

Experimental

The detector used was a 52.0 mm dia and 44.1 mm length HPGe Ortec detector, model GEM-15190 which has about 90 cm^3 active volume. The electronic system includes a spectroscopy amplifier with pile-up rejection. The amplifier time constant was set to 3 μs. A 4096 multichannel analyzer was used to store the spectra measured in the 40–2800 keV energy range. The spectra analysis was performed by the computer code Spectrum (Vandenput and Venturini, 1979). The measurements were performed for two water counting geometries: 100 mL polyethylene flask of about 5 cm dia and 860 mL Marinelli beaker with 850 mL of water, hereafter referred as 100 and 850 mL geometries, respectively. The calibration standards were prepared by diluting Amersham calibrated radioactive solutions of ^{57,60}Co, ^{133}Ba, ^{134}Cs and ^{241}Am in a master solution for each counting geometry. Table 1 shows the calibration standard activities and the literature data from Lorenz (1983) for the gamma-ray transitions used.

The counting live time was 3000 s and the counting rate was about 390 cps for the 100 mL geometry and 560 cps for the 850 mL counting geometry. Greater counting time would not improve the precision of the calculation efficiency which is limited by the activity uncertainty.

Curve Fitting

To describe the efficiency curve the following function is proposed:

$$\ln(\epsilon) = \begin{cases} a_0 + a_1 \cdot \ln(E/E_0) + a_2 \cdot \ln^2(E/E_0), & E \leqslant E_0 \\ b_0 + b_1 \cdot \ln(E/E_0) + b_2 \cdot \ln^2(E/E_0), & E > E_0 \end{cases}$$

where: ϵ, total energy absorption peak efficiency at the energy E (keV); E_0, energy (keV) that corresponds to the joining point; $\{a_i, b_i\}$, parameters to be determined together with E_0.

The smooth continuity of the function at the joining point makes $a_0 = b_0$ and $a_1 = b_1$. Thus the proposed function can be written as follows:

$$\ln(\epsilon) = b_0 + b_1 \cdot \ln(E/E_0) + \beta \cdot \ln^2(E/E_0),$$

$$\beta = \begin{cases} a_2, & E \leqslant E_0 \\ b_2, & E > E_0 \end{cases} \quad (2)$$

The vector of parameters $\boldsymbol{\Phi} = \{b_0, b_1, b_2, a_2, E_0\}$ can be estimated by the Least Squares Method. Identifying $Y = \ln(\epsilon)$ and $x = \ln(E/E_0)$, equation (2) can be written as

$$Y(x, \boldsymbol{\Phi}) = b_0 + b_1 \cdot x + \beta \cdot x^2. \quad (3)$$

To use the LSM to estimate parameters of non-linear functions such as (2) and (3) one has to expand the function around an estimate $\boldsymbol{\phi}'$. For an interval $\Delta\boldsymbol{\phi}$ small enough, $Y(x, \boldsymbol{\phi})$ can be developed in Taylor series and only the first order terms need to be kept. If y_i is the experimental value corresponding to x_i, then [see, for instance, Eadie *et al.* (1971), or Kendall and Stuart (1979)]:

$$y_i - Y_i \cong y_i - Y(x_i, \boldsymbol{\phi}') - \sum_k X_{ik} \cdot \Delta\phi_k,$$

where: X_{ik}, $\partial Y(x_i, \boldsymbol{\phi}')/\partial\phi_k'|_{\boldsymbol{\phi}}$; $\Delta\phi_k$, $\Phi_k - \phi_k'$.

Defining $Y_i' = Y_i - Y(x_i, \boldsymbol{\phi}')$ and $\mathbb{V}$ = experimental data covariance matrix with

$$V_{ij} = \mathrm{cov}(\ln \epsilon_i, \ln \epsilon_j),$$

the weighted sum of the squares of the residues is given by:

Table 1. Energy (E) and intensity (I_γ) for the gamma-ray transitions used, and activities, kBq, in the 100 mL ($A_{100\,mL}$) and in the 850 mL ($A_{850\,mL}$) geometry standards. Decay data were taken from Lorenz (1983)

Source	E (keV)		I_γ (%)		$A_{100\,mL}$	$A_{850\,mL}$
^{241}Am	59.537	(1)	35.82	(12)	0.928 (14)	1.871 (28)
^{57}Co	122.06135	(30)	85.68	(13)	0.639 (7)	1.290 (14)
	136.4743	(5)	10.67	(13)		
^{133}Ba	80.997	(10)	36.94	(61)	0.989 (12)	1.991 (25)
	276.398	(2)	7.12	(7)		
	302.853	(1)	18.3	(2)		
	356.017	(2)	62.1	(7)		
	383.851	(3)	8.92	(9)		
^{134}Cs	604.69	(2)	97.63	(3)	0.874 (17)	1.760 (35)
	795.84	(1)	85.52	(3)		
^{60}Co	1173.238	(4)	99.89	(2)	0.972 (19)	1.909 (38)
	1332.502	(5)	99.9816	(15)		

$$S \cong (\mathbf{y}' - \mathbb{X}\cdot\Delta\boldsymbol{\phi})'\mathbb{V}^{-1}(\mathbf{y}' - \mathbb{X}\cdot\Delta\boldsymbol{\phi}) \quad (4)$$

To minimize S it is necessary to solve the set of equations $\partial S/\partial\,\Delta\phi_k = 0$. The solution is:

$$\Delta\boldsymbol{\phi} = (\mathbb{X}'\cdot\mathbb{V}^{-1}\cdot\mathbb{X})^{-1}\cdot\mathbb{X}'\cdot\mathbb{V}^{-1}\cdot\mathbf{y}',$$

with

$$\boldsymbol{\Phi} = \boldsymbol{\phi}' + \Delta\boldsymbol{\phi}. \quad (5)$$

The determination of $\boldsymbol{\Phi}$ is an interactive process. Starting from $\boldsymbol{\phi}'$ which defines $\mathbf{y}'$, one needs to reduce $\Delta\boldsymbol{\phi}$. Equation (5) is then used to find the vector $\boldsymbol{\Phi}$ that will make S minimum.

At convergence, the covariances between the Φ_k are given by $\mathrm{cov}(\Phi_i, \Phi_j) = [(\mathbb{X}'\cdot\mathbb{V}^{-1}\cdot\mathbb{X})^{-1}]_{ij}$ and the variance of each Φ_j is given by the $i = j$ term.

Since the fractional error of the gamma-ray energies used are less than 10^{-5}, the experimental data fit was done by supposing exact independent variable values. Meanwhile, the covariance between the experimental data were accounted for. These covariances come from the use of more than one gamma-ray transition from the same radionuclide. The efficiency values corresponding to such gamma-ray transitions are correlated by the sample activity errors. The correlations betwen the gamma-ray intensities should be also accounted for. Since these correlations are not found in the current literature, the intensity values were considered to be statistically independent. Each efficiency variance includes an additional error which comes from the standard preparation and is equal to 2% (fractional). From equation (1) and from the above considerations, it follows that:

$$\mathrm{var}(\ln\epsilon) = \mathrm{var}(C)/C^2 + \mathrm{var}(A_0)/A_0^2 + \mathrm{var}(I_\gamma)/I_\gamma^2 + (2\%)^2 \quad (6)$$

and for each pair of efficiencies (ϵ_i, ϵ_j), calculated from the same standard activity A_0, we have:

$$\mathrm{cov}(\ln\epsilon_i, \ln\epsilon_j) = \mathrm{var}(A_0)/A_0^2 + (2\%)^2.$$

The summing effect was evaluated for ^{60}Co, ^{133}Ba and ^{134}Cs, using the nuclear decay data from Anderson *et al.* (1986) and Sergeenkov *et al.* (1981, 1986). Table 2 shows the measured summing effect correction factors for the 100 mL and for the 850 mL geometries.

Table 3 shows the estimated vector of parameters $\boldsymbol{\Phi}$ for both counting geometries. The χ^2 is given by equation (4) and corresponds to 7 degrees of freedom.

Table 2. Summing effect correction factors

Energy (keV)	100 mL	850 mL
81	1.067	1.046
276	1.038	1.018
302	1.017	1.008
356	1.017	1.008
383	0.993	0.999
604	1.084	1.058
795	1.078	1.057
1173	1.058	1.044
1332	1.058	1.044

Table 3. Parameter estimates and non-reduced χ^2

Φ	100 mL	850 mL
b_0	−3.1586 (355)	−3.7350 (306)
b_1	−0.917 (95)	−0.884 (108)
b_2	0.0098 (373)	0.0109 (457)
a_2	−1.849 (185)	−1.822 (152)
E_0	170.8 (148)	183.1 (153)
χ^2	6.4	5.6

Table 4 shows the experimental and the fitted efficiency data for the 100 mL and 850 mL geometries. The standard deviations are given by equation (6). The experimental data were corrected for the summing-effect.

Table 5 shows the parameter covariance matrix. In this table the terms in the lower triangle (main diagonal excluded) were replaced by the corresponding correlated values that are given by:

$$\rho(\Phi_i, \Phi_j) = \mathrm{cov}(\Phi_i, \Phi_j)/(\sigma_{\Phi_i}\cdot\sigma_{\Phi_j})$$

where σ stands for the standard deviation.

The calculation of the efficiency standard deviation should take the parameter correlations into account since efficiency estimates for different energies can be very correlated. A typical case corresponds to the radionuclide activity calculated as the weighted average of individual values from different gamma transition measurements. In such case the weighted average activity is given by the LSM as:

$$\bar{A}_0 = (\mathbb{Z}'\mathbb{W}^{-1}\mathbb{Z})^{-1}\mathbb{Z}'\,\mathbb{W}^{-1}\boldsymbol{A}_0$$

with variance $(\mathbb{Z}'\mathbb{W}^{-1}\mathbb{Z})^{-1}$ where $\mathbb{Z} = \mathbf{1}$ = unity vector $\mathbb{W}$, $\boldsymbol{A}_0$ = individual activity vector, $\mathbb{W}$ = individual activity covariance matrix,

$$\mathbb{W} = \mathbb{F}(\mathbb{X}'\mathbb{V}^{-1}\mathbb{X})^{-1}\mathbb{F}' \quad \text{with} \quad F_{ij} = \epsilon_i\partial\,\ln\epsilon_i/\partial\Phi_j.$$

Assessing the Bias by Simulation

To verify any bias concerning the use of a non-linear function, simulations of the parameters and of the fitted efficiency values were performed. A thousand sets $\{y_s\}$ were fitted to function (2) with the y_{s_i} given by:

$$y_{s_i} = y_{\mathrm{fit}_i} + \sum_j A_{ij}\cdot\alpha_j \quad \begin{cases} i = 1, \ldots, 12 \\ j = 1, \ldots, i \end{cases}$$

Table 4. Experimental and fitted efficiency data for both counting geometries. These values are to be multiplied by 10^{-3}

Energy (keV)	100 mL		850 mL	
	exp.	fit	exp.	fit
59.537	14.11 (58)	14.31 (58)	6.42 (29)	6.46 (28)
80.997	31.71 (159)	30.00 (99)	14.72 (93)	14.56 (46)
122.061	46.92 (127)	46.92 (122)	25.43 (63)	25.32 (62)
136.474	45.79 (159)	47.55 (133)	25.80 (76)	26.45 (68)
276.398	28.76 (143)	27.39 (85)	16.90 (81)	16.62 (54)
302.853	26.16 (124)	25.21 (73)	15.60 (74)	15.34 (46)
356.017	22.21 (106)	21.78 (58)	13.49 (64)	13.33 (37)
383.851	21.00 (104)	20.35 (54)	12.84 (63)	12.48 (34)
604.69	13.34 (40)	13.54 (37)	8.31 (27)	8.43 (24)
795.84	10.47 (32)	10.60 (28)	6.42 (21)	6.67 (18)
1173.238	7.48 (23)	7.53 (20)	4.87 (14)	4.80 (13)
1332.502	6.79 (21)	6.73 (20)	4.36 (16)	4.31 (14)

Table 5. Correlation (in italic) and covariance matrix for the parameter vector Φ calculated for the 850 mL geometry

0.00093	0.00192	−0.00083	−0.00293	−0.2875
0.58239	0.01161	−0.00483	−0.01335	−1.5684
−0.59159	*−0.98068*	0.00209	0.00517	0.62416
−0.62952	*−0.81459*	*0.74451*	0.02313	2.20280
−0.61250	*−0.94756*	*0.88960*	*0.94291*	235.98487

where: y_{fit} = fitted data, α = random independent Gaussian numbers with zero mean and unit variance, $\mathbb{A}$ = variance generating matrix for the $\{y_s\}$ related to $\mathbb{V}$ by:

$$V_{ij} = \sum_{k}^{\min[i,j]} A_{ik} A_{jk}.$$

The repeated simulation of a quantity generates a series which is described by the series average (av.), the standard deviation of the series (σ), the standard deviation of the average (σ_{av}), and the standard deviation of the series (σ_σ). A fitting technique can be accepted if it returns on average the simulation initial values within a few σ_{av}, and estimates the variances within a few σ_σ.

Table 6 shows the results of the parameters simulation for the 850 mL geometry. The average estimates seem to be Gaussian distributed. The non-reduced χ^2 average was found to be 6.8. It corresponds to 7 degrees of freedom and seems to be distributed like ordinary χ^2. As can be seen from Tables 3 and 6 the values of σ agree within $<2\ \sigma_\sigma$. However, not all averages agree with their initial values within two σ_{av}. Since the parameters are not quantities of physical interest, the bias of the interpolated efficiencies was evaluted by generating a series of data using the parameter vectors fitted to the sets $\{y_s\}$. Table 7 shows the series averages (ϵ_{av}) and standard deviations of the fitted efficiency simulation. The fitted data are also Gaussian like distributed. The ϵ_{av} and σ from simulation can be compared to their initial values given in Table 4 and it can be seen that they agree within $<3\ \sigma_{av}$. So, it is possible to conclude that the fit presents no appreciable bias.

Conclusion

The fit of the efficiency curve was tested by simulation. No bias was detected, the fitted data are Gaussian like distributed, Gaussian confidence intervals can be assumed, and the χ^2 test can be used. The experimental data were well fitted by the function given by equation (2). Therefore, the proposed function is appropriate to describe the total absorption energy peak efficiency for HPGe detector, in the 50–1400 keV energy range. The lack of data around 200 keV was filled by the smooth continuity condition at the joining point. The matrix formalism allows the experimental data correlations to be accounted for and this is necessary to improve the evaluation of the fitted data standard deviations. The importance of these correlations has already been pointed out [see for example, Mannhart (1981), Gray and Ahamad (1985) and Geraldo and Smith (1990)].

Table 6. Simulation results for the 850 mL geometry

Parameter	av.	σ	$\sigma_{av.}$	σ_σ
b_0	−3.7393	0.0298	0.0009	0.0007
b_1	−0.8881	0.1034	0.0033	0.0023
b_2	0.0146	0.0441	0.0014	0.0010
a_2	−1.839	0.153	0.005	0.003
E_0	183.8	14.8	0.5	0.3

Table 7. Simulated efficiency averages and simulated series standard deviations for the 850 mL geometry. These values are to be multiplied by 10^{-3}

E (keV)	$\epsilon_{av.}$	σ	$\sigma_{av.}$	σ_σ
59.537	6.442	0.289	0.009	0.006
80.997	14.575	0.458	0.014	0.010
122.061	25.314	0.623	0.020	0.014
136.474	26.411	0.679	0.021	0.015
276.398	16.653	0.532	0.017	0.012
302.853	15.364	0.451	0.014	0.010
356.017	13.332	0.365	0.012	0.008
383.851	12.484	0.340	0.011	0.008
604.69	8.423	0.236	0.007	0.005
795.84	6.660	0.176	0.006	0.004
1173.238	4.796	0.130	0.004	0.003
1332.502	4.312	0.135	0.004	0.003

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