Available online at www.sciencedirect.com
SCIENCE DIRECT®
Applied Radiation and Isotopes 60 (2004) 683–687
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Combination of nonlinear function and mixing method for fitting HPGe efficiency curve in the 59–2754 kev energy range

M.S. Dias[a,*], V. Cardoso[a], V.R. Vanin[b], M.F. Koskinas[a]

[a] *Instituto de Pesquisas Energeticas e Nucleares (IPEN-CNEN/SP), Centro do Reator de Pesquisas, CP 11049, Pinheiros CEP, São Paulo, SP 05422-970, Brazil*

[b] *Laboratório do Acelerador Linear, Instituto de Física da Universidade de São Paulo, CP 66318, CEP 05315-970, São Paulo, SP, Brazil*

Received 11 September 2003; received in revised form 22 October 2003; accepted 23 October 2003

Abstract

A nonlinear function in combination with the method of mixing activity-calibrated and uncalibrated gamma-ray sources is applied for fitting the experimental peak efficiency of HPGe spectrometers in the 59–2754 keV energy range. In addition, a step function was developed for fitting the gamma-ray background under the peak. Calibrated ^{241}Am, ^{133}Ba,^{137}Cs, ^{152}Eu, ^{60}Co and ^{88}Y sources, as well as one ^{24}Na source (treated as uncalibrated) were used for obtaining the experimental peak efficiency curve. The results were compared to the conventional linear polynomial fitting. From the fitted parameters, the ^{24}Na activity was determined and compared with the result obtained from absolute measurements in a $4\pi\beta$–γ coincidence system.

Keywords: Gamma-ray; Fitting; HPGE; Efficiency; Marquardt

1. Introduction

Gamma-ray spectrometry by means of HPGe detectors is widely used for applications due to their excellent energy resolution. In nuclear metrology, this technique has been used for the activity determination of radioactive sources when suitable standards are not available, as well as for the determination of gamma-ray emission probability per decay (Fonseca et al., 1998; Simões et al., 2001). In these cases, an efficiency curve as a function of the gamma-ray energy becomes necessary, and the efficiency values for the desired energies are obtained by interpolation. This subject is discussed by Seymour et al. (1988).

The absolute efficiency can be calculated as the ratio between the detected and emitted gamma-rays

$$\varepsilon(E) = \frac{N}{A\Delta t I_\gamma} \cdot f, \tag{1}$$

where N is the net counts of the full energy absorption peak corresponding to the gamma-ray of energy E and emission probability per decay I_γ.Δt is the measuring time and A is the source disintegration rate. Factor f takes into account corrections due to dead time, detection geometry, decay, summing effects and self absorption.

The relative efficiency is given by

$$\varepsilon_r(E) = \alpha \frac{N}{I_\gamma} \cdot f, \tag{2}$$

where α is an arbitrary constant.

The efficiency curve in the 59–2754 keV range shows three 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

*Corresponding author. Fax: +55-11-816-9188.
E-mail address: msdias@net.ipen.br (M.S. Dias).

0969-8043/$ - see front matter © 2003 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2003.10.011

almost linearly in log-scale up to around 1200 keV. For higher energies the curve continues to decrease but following a different pattern due to pair production interactions. Therefore three regions can be distinguished: at lower, intermediate and higher energies.

An alternative procedure is described by Janssen (1990), which makes use of low order spline functions for fitting the efficiency curve in the 2–3000 keV. In this case, five or more regions are chosen, but this requires a high number (>40) of experimental points. This procedure has been successfully used by Daza et al. (2001) for measuring environmental samples. Hayashi et al. (2000) applied a third-order polynomial function for fitting the relative gamma-ray efficiency curve in the 430–2750 keV energy region.

Previous papers have dealt with nonlinear fitting in the 50–1400 keV energy range, Venturini and Vanin (1993), and the use of the mixing technique, Tramontano and Vanin (1999). The present work proposes a nonlinear function divided into three regions and covering the 59–2754 keV energy interval. This function was combined with the mixing technique, in order to include uncalibrated sources in the fitting procedure. Several calibrated and one uncalibrated have been used.

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 present paper proposes an alternative step function for fitting the background region under the peak. This function is composed by a second degree polynomial coupled with a sigmoid function, in order to yield a smooth step under the peak.

For nonlinear calculations, the algorithm suggested by Marquardt (1963) was used. 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 (1998), Geraldo and Smith (1990) and Mannhart (1981).

2. Area under the full absorption peak

To describe the background under the full absorption peak, the following function is proposed:

$$B(x) = a_0 + a_1 x + a_2 x^2 + a_3 \left(\frac{1}{1 + e^z} \right), \tag{3}$$

where $z = (x - x_p)/S$ and $S = FWHM/2\sqrt{\ln 2}$. x_p is the peak centroid channel and x is the channel in the background region.

This function corresponds to a second degree polynomial associated with a sigmoid step function. In this first approach, the values of x_p and $FWHM$ were estimated previously and then treated as constants. Future versions of the code will consider these variables as free parameters to be included in the fitting procedure. All other parameters were calculated by covariance matrix methodology. The overall uncertainty in the peak area is the composition of the statistical uncertainty in the gross counts and the background uncertainty calculated by this fitting.

3. Efficiency curve fitting

3.1. Linear function

The use of a linear function associated with the mixing technique is described by Tramontano and Vanin (1999). In the present work, the best fitting for the 59–2754 keV energy range was obtained by means of the following function:

$$\begin{aligned} \ln \varepsilon(E) = {} & C_{a_0} + U_{a_0} + a_1 \ln(E/E_b) + a_2 \ln^2(E/E_b) \\ & + a_3 \ln^3(E/E_b) + a_4 \ln^4(E/E_b) \\ & + a_5 \ln^5(E/E_b), \end{aligned} \tag{4}$$

where ε is the peak efficiency for gamma-ray energy E and E_b is a reference energy selected to bring the range of log values around zero. C_{a_0} and U_{a_0} are parameters related to the activity-calibrated and uncalibrated sources, respectively.

The relative variances of the experimental efficiencies are given by

$$\begin{aligned} \left(\frac{\sigma(c_\varepsilon)}{c_\varepsilon} \right)^2 = {} & \left(\frac{\sigma_A}{A} \right)^2 + \left(\frac{\sigma_N}{N} \right)^2 + \left(\frac{\sigma_{I\gamma}}{I_\gamma} \right)^2 \\ & + \left(\frac{\sigma_{f_\tau}}{f_\tau} \right)^2 + \left(\frac{\sigma_{f_\lambda}}{f_\lambda} \right)^2 + \left(\frac{\sigma_{f_s}}{f_s} \right)^2, \end{aligned} \tag{5}$$

$$\left(\frac{\sigma(u_\varepsilon)}{u_\varepsilon} \right)^2 = \left(\frac{\sigma_N}{N} \right)^2 + \left(\frac{\sigma_{I\gamma}}{I_\gamma} \right)^2 + \left(\frac{\sigma_{f_\tau}}{f_\tau} \right)^2 + \left(\frac{\sigma_{f_\lambda}}{f_\lambda} \right)^2 + \left(\frac{\sigma_{f_s}}{f_s} \right)^2, \tag{6}$$

where N is the net area under the full absorption peak for the selected gamma-ray energy, A the source activity, I_γ the gamma-ray emission probability per decay, f_τ the dead time correction factor, obtained by means of a reference pulser peak at the end of the gamma-ray spectrum, f_λ the nuclear decay correction and f_s the cascade summing correction factor, obtained by means of a Monte Carlo code, as described by Dias et al. (2002).

Eqs. (5) and (6) correspond to the calibrated and uncalibrated sources, respectively.

The fitted efficiency relative variances are calculated by

$$\left(\frac{\sigma_\varepsilon(E)}{\varepsilon(E)}\right)^2 = \vec{F} V_{\vec{a}} \vec{F}^t, \tag{7}$$

where

$$\vec{F} = \left[0 \quad 1 \quad \ln\left(\frac{E}{E_b}\right) \quad \ln^2\left(\frac{E}{E_b}\right) \quad \ln^3\left(\frac{E}{E_b}\right) \quad \ln^4\left(\frac{E}{E_b}\right) \quad \ln^5\left(\frac{E}{E_b}\right)\right] \tag{8}$$

and V_u is the covariance matrix of U_ε.

3.2. Nonlinear function

The selected nonlinear function is an extension of the model proposed by Venturini and Vanin (1993) for a wider energy range. This function is divided into three regions, corresponding to low, intermediate and high gamma-ray energies, respectively

$$\ln(\varepsilon) = \begin{cases} a_0 + a_1 \ln(E/a_4) + a_2 \ln^2(E/a_4), & E \leqslant a_4, \\ b_0 + b_1 \ln(E/a_4) + b_2 \ln^2(E/a_4), & a_4 < E \leqslant a_5, \\ c_0 + c_1 \ln(E/a_5) + c_2 \ln^2(E/a_5), & a_5 < E, \end{cases} \tag{9}$$

where ε is the peak efficiency for gamma-ray energy E; a_4 and a_5 correspond to energy values at the two junction points (between low and intermediate energies and between intermediate and high energies).

The present approach can be considered as a simplification of the method developed by Janssen (1990) and is suitable when the number of available experimental points is smaller than 40. Moreover, the behavior of the efficiency curve in the energy region from 60 to 3000 keV is smooth and does not require a large number of junction points. In the present paper, these junction points are obtained as fitting parameters and not chosen arbitrarily.

The continuity condition for function (9) and its derivative imply that

$$a_0 = b_0 \quad \text{and} \quad c_0 = b_0 + b_1 \ln(a_5/a_4) + b_2 \ln(a_5/a_4)^2$$
$$a_1 = b_2 \quad \text{and} \quad c_1 = b_1 + 2b_2 \ln(a_5/a_4).$$

Including the mixing parameters C_{a_0} and U_{a_0}, the final expression becomes

$$\ln(\varepsilon) = C_{a_0} + U_{a_0} + b_1 \ln(E/a_4) + \beta \ln^2(E/a_4) + \vartheta \ln^2(E/a_5), \tag{10}$$

where

if $E \leqslant a_4$, then $\beta = a_2$ and $\vartheta = 0$,

if $a_4 < E \leqslant a_5$, then $\beta = b_2$ and $\vartheta = 0$,

if $a_5 < E$, then $\beta = 0$ and $\vartheta = c_2$.

The least square method adopted for estimating the parameters for the nonlinear function is described by Venturini and Vanin (1993).

4. Experimental

A HPGe detector Model EGC-20 Intertechnique was used, with 20.6 cm^2 sensitive area, 99.7 cm^3 active volume and 20% relative efficiency. The amplifier time constant was set to 2 μs and the spectra were stored in a 4096 multichannel analyzer. The source–detector distance was set to 19.3 cm. The radioactive sources used were IAEA standards of ^{241}Am, ^{133}Ba, ^{152}Eu, ^{137}Cs and ^{60}Co. In addition, a ^{88}Y standard source, calibrated in our laboratory by means of $4\pi\beta$–γ coincidence system, was used.

One ^{24}Na calibrated source was included in the fitting procedure in order to apply the mixing technique and extend the energy range up to 2754 keV. This source was treated as uncalibrated in the fitting procedure in order to extract its activity and compare with the reference activity. This reference value has been obtained by calibration in a $4\pi\beta$–γ coincidence system. For this purpose a ^{24}Na solution was produced by means of ^{23}Na(n,γ)^{24}Na reaction at the IEA-R1 research reactor from IPEN, São Paulo. The source was prepared by depositing an aliquot on 10 μg cm^{-2} thick Collodion film. The gamma-ray window was set at the 1368.63 keV gamma-ray total absorption peak.

5. Results and discussion

Table 1 shows the experimental results for the peak efficiency. The values for ^{24}Na uncalibrated source were normalized to 1000 at 1368.60 keV gamma-ray energy. The last columns give the uncertainty in the applied correction factors.

Table 2 shows the fitting parameters for the linear function. This table includes all the correlation factors and covariances among different parameters. The reduced chi-square was 1.19. Table 3 shows the corresponding results for the nonlinear function. The reduced chi-square was 1.22, close to the one obtained with the linear function.

Fig. 1 shows the behavior of residuals obtained with the nonlinear function (Eq. (9)). It can be seen that there is no systematic trend of the points around the fitted line, indicating no appreciable bias.

The ratio between the efficiencies obtained with the calibrated and uncalibrated curves gives the activity of the uncalibrated source. For the case of linear fit these efficiencies were calculated at reference energy E_b. In the case of nonlinear fit this ratio was calculated at 800 keV, which is near the middle point of the second region. This

Table 1
Experimental efficiency and partial relative uncertainties involved (in percent)

Energy (keV)	Efficiency $\times 10^{-3}$	σ_N (%)	σ_A (%)	σ_{I_γ} (%)	σ_τ (%)	σ_λ (%)	σ_{fs} (%)
59.54	3.8615	0.13	1.10	1.11	0.29	0.00	0.26
81.00	4.0025	0.09	1.20	0.82	0.31	0.50	0.15
121.78	3.6627	0.08	1.60	0.46	0.31	0.22	0.08
244.70	2.2886	0.22	1.60	0.53	0.31	0.22	0.12
276.40	2.0482	0.27	1.20	0.42	0.31	0.50	0.08
302.85	1.8783	0.17	1.20	0.33	0.31	0.50	0.08
344.28	1.6574	0.12	1.60	0.41	0.31	0.22	0.04
356.02	1.6033	0.09	1.20	0.22	0.31	0.50	0.08
383.85	1.4873	0.26	1.20	0.32	0.31	0.50	0.08
411.13	1.3883	0.59	1.60	0.45	0.31	0.22	0.12
443.96	1.2851	0.49	1.60	0.45	0.31	0.22	0.08
661.66	0.8664	0.14	1.00	0.23	0.38	0.27	0.00
778.90	0.7429	0.28	1.60	0.46	0.31	0.22	0.06
867.39	0.6730	0.59	1.60	0.59	0.31	0.22	0.15
898.04	0.6526	0.73	1.50	0.30	0.30	0.13	0.06
964.05	0.6127	0.28	1.60	0.41	0.31	0.22	0.12
1112.10	0.5409	0.31	1.60	0.44	0.31	0.22	0.10
1173.20	0.5165	0.25	0.50	0.02	0.31	0.07	0.04
1332.50	0.4631	0.26	0.50	0.01	0.31	0.07	0.04
1368.60	1000.0[a]	0.30	[b]	0.00	0.32	0.02	0.02
1408.00	0.4417	0.26	1.60	0.43	0.31	0.22	0.11
1836.10	0.3486	0.84	1.50	0.03	0.30	0.13	0.06
2754.00	498.00[a]	0.42	[b]	0.00	0.32	0.02	0.04

[a] The values for ^{24}Na were normalized to 1000 at 1368.60 keV.
[b] Uncalibrated source (error equal to zero).

Table 2
Estimated parameters for the linear function (Eq. (4))

Parameter	Value	U_{a_0}	C_{a_0}	a_1	a_2	a_3	a_4	a_5
U_{a_0}	0.0162 (84)	**0.000070**	0.000000	−0.000013	−0.000057	−0.000062	−0.000026	−0.000004
C_{a_0}	−7.6887 (48)	0.012	**0.000023**	0.000017	−0.000006	−0.000021	−0.000009	−0.000001
a_1	−0.859 (11)	−0.141	0.313	**0.000129**	0.000018	−0.000137	−0.000086	−0.000014
a_2	−0.031 (11)	−0.642	−0.127	0.150	**0.000113**	0.000110	0.000038	0.000004
a_3	−0.176 (18)	−0.405	−0.235	−0.654	0.560	**0.000341**	0.000184	0.000029
a_4	−0.095 (10)	−0.294	−0.187	−0.734	0.342	0.962	**0.000108**	0.000018
a_5	−0.0117 (17)	−0.241	−0.140	−0.726	0.230	0.906	0.986	**0.000003**
χ^2/v	1.19							

The values in the parentheses are the standard deviation in the last digits. Covariances between parameters are shown in the upper triangle (including the main diagonal). Correlations are shown in the lower triangle. The last row gives the reduced chi-square.

Table 3
Estimated parameters for the nonlinear function (Eq. (9))

Parameter	Value	U_{a_0}	C_{b_0}	b_1	a_2	b_2	c_2	a_4	a_5
U_{a_0}	1.491(41)	**0.001678**	0.001617	0.000435	−0.000634	−0.000408	−0.000469	−0.471193	−0.489759
C_{b_0}	−6.212(41)	0.972	**0.001649**	0.000432	−0.000650	−0.000406	−0.000351	−0.476151	−0.490303
b_1	−1.045(15)	0.687	0.688	**0.000239**	−0.000068	−0.000195	−0.000231	−0.131877	−0.172814
a_2	−0.398(19)	−0.816	−0.001	−0.234	**0.000360**	0.000087	0.000026	0.184668	0.158440
b_2	0.064(13)	−0.766	−0.769	−0.970	0.353	**0.000169**	0.000184	0.120217	0.152412
c_2	−0.186(26)	−0.441	−0.333	−0.576	0.052	0.546	**0.000672**	0.109286	0.151518
a_4	284(12)	−0.971	−0.990	−0.721	0.822	0.781	0.356	**140.35500**	144.89100
a_5	1290(13)	−0.937	−0.947	−0.877	0.655	0.920	0.458	0.9589215	**162.66300**
χ^2/v	1.22								

The values in the parentheses are the standard deviation in the last digits. Covariances between parameters are shown in the upper triangle (including the main diagonal). Correlations are shown in the lower triangle. The last row gives the reduced chi-square.

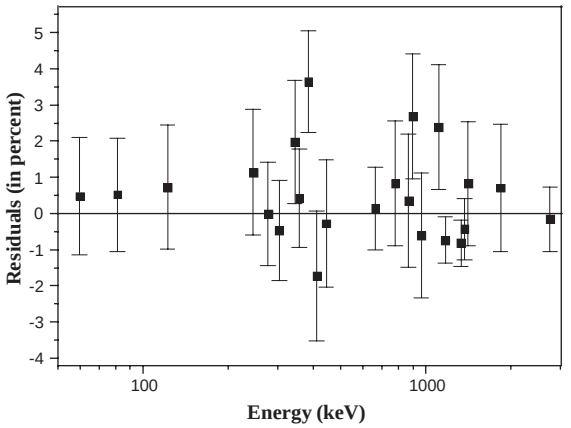

Fig. 1. Percent residuals between experimental and fitted efficiencies as a function of the gamma-ray energy. The error bars correspond to the standard deviation in the experimental value (in percent).

value has been chosen in order to minimize the overall uncertainty. The results obtained for the two performed fits were: 2219 (21) and 2215 (18) Bq, corresponding to linear and nonlinear fits, respectively. These two values agree within their uncertainties. The reference activity measured with the $4\pi\beta$–γ coincidence system resulted—2215 (11) Bq in very good agreement with both fitted values.

As a conclusion, a nonlinear fit dividing the 59–2754 keV energy interval into three regions can give satisfactory results. The accuracy in the interpolation is comparable to the linear fit using a fifth degree polynomial. Since the nonlinear function is a composition of simple second degree polynomials, it is expected that the interpolation should be more reliable than a linear function with several parameters, specially in regions where there are a few experimental points. In these regions a high degree polynomial may show artificial oscillations in the efficiency curve.

References

Daza, M.J., Quintana, B., García-Talavera, M., Fernández, F., 2001. Efficiency calibration of a HPGe detector in the (46.54–2000) keV energy range for the measurement of environmental samples. Nucl. Instrum. Methods A 470, 520.

Dias, M.S., Takeda, M.N., Koskinas, M.F., 2002. Cascade summing corrections for HPGe spectrometers by the Monte Carlo Method. Appl. Radiat. Isot. 56, 105.

Fonseca, K.A., Koskinas, M.F., Dias, M.S., 1998. Measurement of the gamma-ray probability per decay of ^{126}I. Appl. Radiat. Isot. 49 (9–11), 1373.

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.

Hayashi, N., Miyahara, H., Mori, C., 2000. Highly precise measurement of the relative gamma-ray detection efficiency curve. J. Nucl. Sci. Technol. NS-37 (2), 139.

Janssen, H., 1990. Spline techniques for fitting efficiency curves in gamma-ray spectrometry. Nucl. Instrum. Methods A 286, 398.

Mannhart, W., 1981. A small guide to generating covariances of experimental data. PTB-FMRB-84.

Marquardt, D.W., 1963. An algorithm for least-squares estimation of nonlinear parameters. J. Soc. Appl. Math. 11, 431.

Seymour, R.S., Andreacom, M.S., Pierce, J., 1988. Evaluation of energy efficiency fitting functions for HPGe detectors. J Radi. Nucl. Chem. 123 (2), 529.

Simões, D., Koskinas, M.F., Dias,, M.S., 2001. Measurement of the gamma-ray probability per decay of ^{42}K. Appl. Radiat. Isot. 54, 443.

Tramontano, R., Vanin, V.R., 1999. Mixing activity-calibrated and uncalibrated gamma-ray sources in efficiency calibration. Appl. Radiat. Isot. 51, 323.

Venturini, L., Vanin, V.R., 1993. HPGe detector efficiency calibration for extended sources in the 50–1400 keV energy range. Appl. Radiat. Isot. 44 (7), 999.

Winkler, G., 1998. On the role of covariances for uncertainty estimates in radioactivity measurements. Appl. Radiat. Isot. 49 (9–11), 1153.