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A fitting algorithm based on simulated annealing techniques for efficiency calibration of HPGe detectors using different mathematical functions

S. Hurtado [a,*], M. García-León [b], R. García-Tenorio [c]

[a] Servicio de Radioisótopos, Centro de Investigacion, Tecnologia e Innovacion (CITIUS), Universidad de Sevilla, Avda. Reina Mercedes s/n, 41012 Sevilla, Spain
[b] Departamento de Física Atómica, Molecular y Nuclear, Facultad de Física, Universidad de Sevilla, Aptd. 1065, 41080 Sevilla, Spain
[c] Departamento de Física Aplicada II, E.T.S.A. Universidad de Sevilla, Avda, Reina Mercedes 2, 41012 Sevilla, Spain

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ABSTRACT

In this work several mathematical functions are compared in order to perform the full-energy peak efficiency calibration of HPGe detectors using a 126 cm^3 HPGe coaxial detector and gamma-ray energies ranging from 36 to 1460 keV. Statistical tests and Monte Carlo simulations were used to study the performance of the fitting curve equations. Furthermore the fitting procedure of these complex functional forms to experimental data is a non-linear multi-parameter minimization problem. In gamma-ray spectrometry usually non-linear least-squares fitting algorithms (Levenberg–Marquardt method) provide a fast convergence while minimizing χ^2_R, however, sometimes reaching only local minima. In order to overcome that shortcoming a hybrid algorithm based on simulated annealing (HSA) techniques is proposed. Additionally a new function is suggested that models the efficiency curve of germanium detectors in gamma-ray spectrometry.

1. Introduction

Determination of the full-energy peak (FEP) efficiency of HPGe detectors is essential for quantitative measurements of radionuclides in gamma-ray spectrometry. The functional dependence between efficiency and energy is not well established [1] so fitting curve equations are usually fitted to the experimental efficiency data points for FEP calibration of the detector. Several fitting curve equations have been proposed [1] with linear and non-linear dependence on different number of parameters. In this work we propose a new analytical function that achieves a good performance interpolating and extrapolating from energy range given by experimental data.

Alternately various authors have used semi-empirical functions [2–5] to describe the FEP efficiency further over the energy range of experimental efficiency data. Cross sections (photoelectric, Compton and gamma conversion) and geometrical dimensions of the detector are common parameters included in semi-empirical functions. However, most of semi-empirical formulae were intended for planar germanium detectors or involved too many parameters incompatible with generally few available experimental data points. Recently the semi-empirical method has been improved including extended sources, extending the energy range to 10 keV, and involving only a few parameters [6]. An analogous explanation could be applied to the use of spline technique fitting FEP efficiency curves [7,8].

In this work the fitting results were obtained with a hybrid simulated annealing strategy and compared with those results obtained with classical techniques, considering their advantages and disadvantages.

2. Experimental data

The experimental measurements were performed with a Canberra n-type ReGe (Reverse electrode Germanium) detector, with relative photopeak efficiency of 30% at 1332 keV. The endcap of the detector was made of aluminium and its thickness was 1.5 mm. A lead shield (15 cm thick regular lead) and an inner copper layer (5 mm) surround the detector against environmental radiation. The preamplified signals from the detector were sent to a Canberra Spectroscopy Amplifier model 2020 and a Canberra ADC model 8701 connected to an *Accuspec*™ card. Gamma-ray peaks were analysed with *Genie*2000™ using a Gaussian peak with a step background fit.

Calibration sources were obtained using standard solutions that contain: ^{210}Pb (105.8 ± 0.4 Bq/g) and ^{241}Am (329 ± 1 Bq/g) supplied by CIEMAT (Madrid, Spain); and ^{137}Cs (53.7 ± 0.8 kBq/g), ^{133}Ba (29.74 ± 0.44 kBq/g) and ^{152}Eu (54.6 ± 1.1 kBq/g) supplied by DAMRI (France). Point calibration sources were obtained by

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

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Table 1
Experimental FEP efficiencies ε_{exp} for point source geometry

Radionuclide	Energy (keV)	Intensity (%)	ε_{exp}
X-ray ^{137}Cs	36.3	1.30(5)	0.57(2)
^{210}Pb	46.56	4.25	0.617(11)
^{133}Ba	53.15	2.19(2)	0.62(3)
^{241}Am	59.53	35.9(4)	0.632(8)
^{133}Ba	80.997	34.06(27)	0.637(14)
^{152}Eu	121.78	28.58(6)	0.583(11)
^{133}Ba	276.4	7.16(2)	0.339(8)
^{152}Eu	344.281	26.5(4)	0.267(7)
^{133}Ba	356.01	62.05(19)	0.264(5)
^{137}Cs	661.65	85.1(2)	0.146(2)
^{152}Eu	778.92	12.94(2)	0.126(3)
^{152}Eu	964.05	14.60(2)	0.104(3)
^{152}Eu	1112.09	13.64(2)	0.094(3)
^{152}Eu	1408.02	21.01(2)	0.077(2)
^{40}K	1460.8	11.0	0.076(2)

pipetting and drying a volume of standard solutions over aluminium planchets. The ^{40}K sources were obtained by packing 10 g of KCl from Merck.

The measurement geometry was a point-like source measured at 16 cm source-to-detector window distance to avoid coincidence-summing effect. The experimental efficiency points for reference geometry are shown in Table 1.

3. Simulated annealing algorithm

Fitting methods minimize an objective function (reduced chi-square χ^2_R) in order to find the optimal values of parameters that reach the global minimum. Mathematical algorithms for fitting FEP efficiency data are usually based on least-squares or Levenberg–Marquardt (LM) method [5,9–11] depending on linear or non-linear functional parameters, and are described in detail in Refs. [12,13]. However, LM algorithm is a gradient-based method that involves the calculation of derivatives and has the disadvantage of converging towards local minima when the initial parameter estimates are poor [1,5].

An alternative minimization technique, the Simplex method, has already been applied for peak fitting in alpha-ray spectrometry [14]. This paper describes an algorithm based on simulated annealing (SA) method. SA derives its name from an analogy to the cooling of heated metals. As a metal cools, the atoms fluctuate between relatively higher and lower energy levels. If the temperature is dropped slowly enough, the atoms will all reach their ground state. However, if the temperature is dropped too quickly, the system will get trapped in a less-than-optimum configuration. If the energy function of this physical system is instead replaced by an objective function χ^2_R then its progression towards the global minimum is analogous to the physical progression towards the ground state. In the SA algorithm random steps [15] in the parameter space are performed (using a control parameter T). A step is always accepted if the objective function is lowered, and it is sometimes accepted with a certain, decreasing probability, if an uphill step is taken. This scheme allows the algorithm of escaping from local minima.

The hybrid simulated annealing (HSA) algorithm shown in this work (see Fig. 1) is based specifically on a variation of the direct search method called Simplex [16] adding the SA technique to select the parameters of the function. Finally the HSA algorithm links the output parameters of the SA method to an LM algorithm in order to fast converge to global minima and to obtain uncertainties of the optimal parameters.

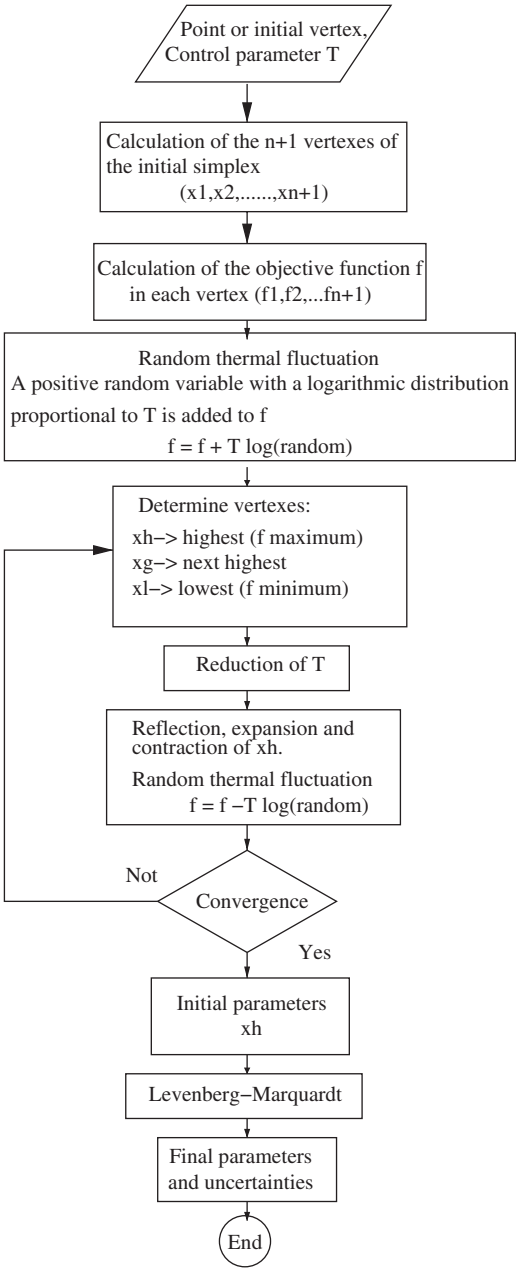

Fig. 1. Hybrid simulated annealing algorithm (HSA).

4. Fitting curve equations

In gamma-ray spectrometry the most common function [17] used and presented in analysis software is the polynomial logarithm (see Table 2), sometimes the energy range is divided into a low-energy (<200 keV) and a high-energy part for fitting them separately [1]. McNelles and Campbell [18] attempted to fit the FEP data of their 25 cm^3 coaxial detector using a non-linear function. They obtained an average deviation of 1% in the energy range of 122–1333 keV. Later on, Gray [19] stressed the advantageous features of linear function. The average deviation obtained in the energy range of 80–1850 keV was 1.8%. Jäckel [20] maintained the functional form of the low-energy part of the efficiency curve where the curvature is higher and obtained a deviation of 2% from 60 to 1333 keV. Finally, Sánchez-Reyes [21] extended the energy range from 63 to 3054 keV using a 5-parameter function.

Table 2
Fitting curve equations used for fitting experimental FEP efficiency values and their original energy range as shown in their corresponding reference

Polynomial logarithm	Sánchez-Reyes
$\ln \varepsilon = \sum_i a_i (\ln(E))^i$ $i = 0-5$ or 0–6 81–3254 keV	$\ln \varepsilon = a_1 - (a_2 + a_3 \exp(-a_4 E)) \exp(-a_5 E) \ln E$ 63–3054 keV
McNelles and Campbell	Jäckel
$\varepsilon = a_1 E^{-a_2} + \sum_{i=2}^{4} a_{2i-1} \exp(-a_{2i} E)$ 122–1333 keV	$\ln \varepsilon + \text{constant} = (a_1 + a_2 \ln(E) + a_3 (\ln(E))^2) \cdot \arctan[\exp(a_4 + a_5 \ln(E) + a_6 (\ln(E))^2)]$ 60–1333 keV
This paper	Gray
$\varepsilon = a_1(\exp(-a_2 E^{a_3}) + \exp(-a_4 E^{a_5}))(1 - \exp(a_6 E^{a_7}))$ 36–1500 keV	$\varepsilon = 1/E \cdot \sum_i a_i (\ln E)^{i-1}$ $i = 1-8 (\neq 5, 7)$ 80–1850 keV

Additionally, a new function is proposed in this work in order to describe the low-energy part of the efficiency curve, but without semi-empirical framework to avoid the need for many FEP data points. The terms included in that function were selected to qualitatively reproduce the physical meaning of semi-empirical functions, and extending the energy range below 30 keV in order to be useful in modern gamma-ray spectrometry linked to n-type germanium detectors. The term $\exp(-a_2 E^{a_3}) + \exp(-a_4 E^{a_5})$ reflects the transmission of photons in the materials between the source and the detector. The term $1 - \exp(a_6 E^{a_7})$ accounts for the absorption of the photons in the detector. In principle this function could be extended to p-type detectors but a checking would be advisable.

5. Results

First of all, we have carried out a comparison between the LM and the hybrid simulated annealing (HSA) algorithm. A polynomial logarithm of sixth order and the function proposed in this paper have been used in order to perform the comparison of fitting algorithms. The results (see Table 3) consisted of the obtained χ^2_R for each function with the same initial values for their parameters (a_i), and the total number of iterations of each function (n) performed to reach the previous χ^2_R value. The LM algorithm performs the fitting procedure faster than SA algorithm for the polynomial logarithm due to the simple linear dependence on the parameters but both algorithms reach the same final value of χ^2_R. However, if a complex function is fitted, as the one proposed in this paper, the selection of the initial values of the parameters becomes important. The LM method may get anchored to a local minimum but the SA algorithm reaches the global minimum independently of the initial values of the parameters exhibiting in this manner the power of this algorithm.

In the following the results of the comparison between the functions using HSA algorithm for fitting purposes are shown. The statistical parameters used in this comparison were the objective function χ^2_R, and the average relative deviation RD (%). In Figs. 2–6 the fits performed using the functions are shown together with the standardized residuals. The experimental data have been also shown together with their uncertainties (as one sigma). The final values for the statistical parameters are shown in Table 4.

We notice that all functions give similar values of χ^2_R but a few ones (polynomial of fifth order, Sánchez-Reyes, and Gray) provide a standard deviation higher than 1%. The remaining of the functions (polynomial of sixth order, McNelles and Campbell, Jäckel and the function presented in this paper) give a standard deviation comprised between 0.6% and 0.8%. However, McNelles and Campbell function has a strange behaviour out of experimental energy range generating non-realistic oscillations for energies above 1500 keV. Therefore the remaining three functions (polynomial of sixth order, Jäckel and the function presented in this paper) provide lower standard deviations and follow a correct behaviour across all energy range.

Table 3
Comparison between LM and HSA algorithms using polynomial logarithm of sixth order and the function proposed in this work

a_i	χ^2_R(LM)	n	χ^2_R(HSA)	n
Polynomial 6 order				
1.0×10^{-1}	0.20	16	0.20	34051
1.0×10^{-2}	0.20	16	0.20	37030
1.0×10^{-3}	0.20	16	0.20	38053
1.0×10^{-4}	0.20	16	0.20	28037
1.0×10^{-5}	0.20	16	0.20	25035
This work				
1.0×10^{-1}	3479	2	0.23	90122
1.0×10^{-2}	3478	3	0.23	64088
1.0×10^{-3}	3478	2	0.23	69093
1.0×10^{-4}	3478	2	0.23	28052
1.0×10^{-5}	3478	2	0.23	31040

The objective function is shown for LM χ^2_R(LM) and HSA algorithm χ^2_R(HSA), just as the number of iterations (n) for different initial values of parameters (a_i).

Next we have used Monte Carlo simulation to develop a deeper comparison between the three functions. The GEANT4 code [22] and an optimization of the detector dimensions provides us a tool to obtain FEP efficiencies. The calculation of FEP efficiencies at 30, 2000 and 3000 keV allows us to test the capability of each function to extrapolate outside the experimental energy range. In Table 5 the FEP efficiencies calculated using MC method and extrapolating each function together with their associated relative deviations with regard to the MC FEP efficiency values. As it can be seen the lower average relative deviation at the three energies is provided for our function (2.09%) followed by the polynomial of sixth order (4.9%) and the Jäckel function (12.1%).

In order to allow the use of the proposed function in future works the values of the parameters for the proposed function are the following: $a_1 = 3.16$, $a_2 = 0.049$, $a_3 = 0.729$, $a_4 = 0.753$, $a_5 = 0.219$, $a_6 = -0.011$ and $a_7 = 0.92$.

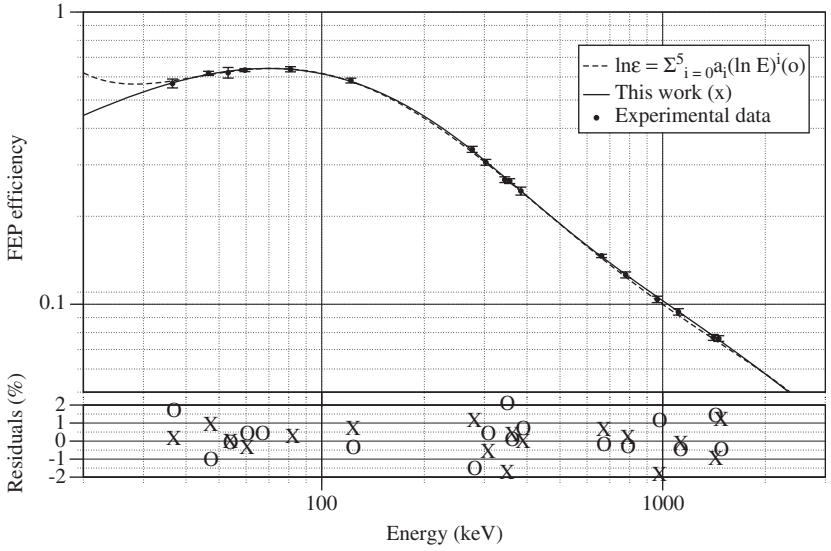

Fig. 2. Fitting experimental FEP efficiencies using polynomial logarithm of sixth order and the proposed function.

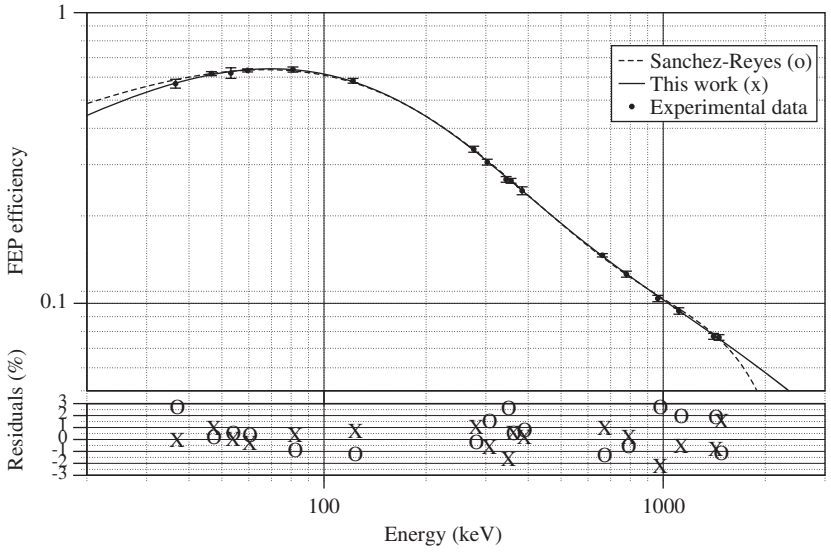

Fig. 3. Fitting experimental FEP efficiencies using Sánchez-Reyes function and the proposed function.

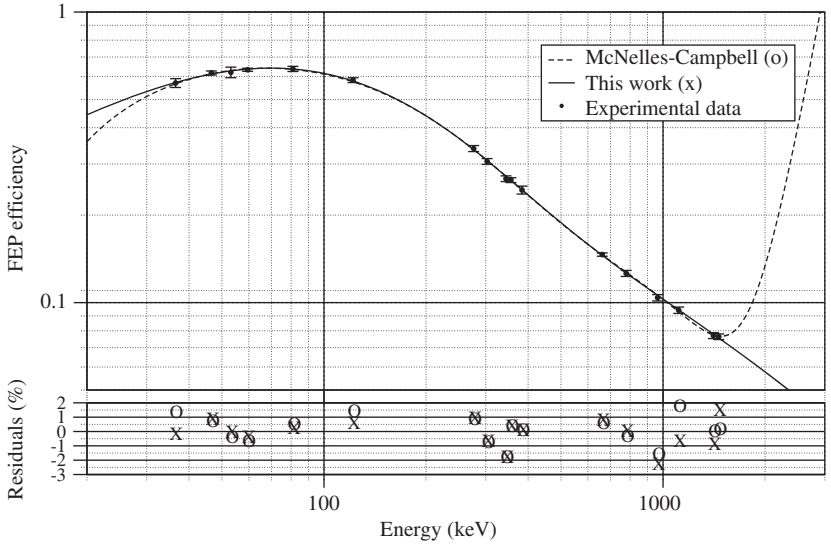

Fig. 4. Fitting experimental FEP efficiencies using McNelles and Campbell function and the proposed function.

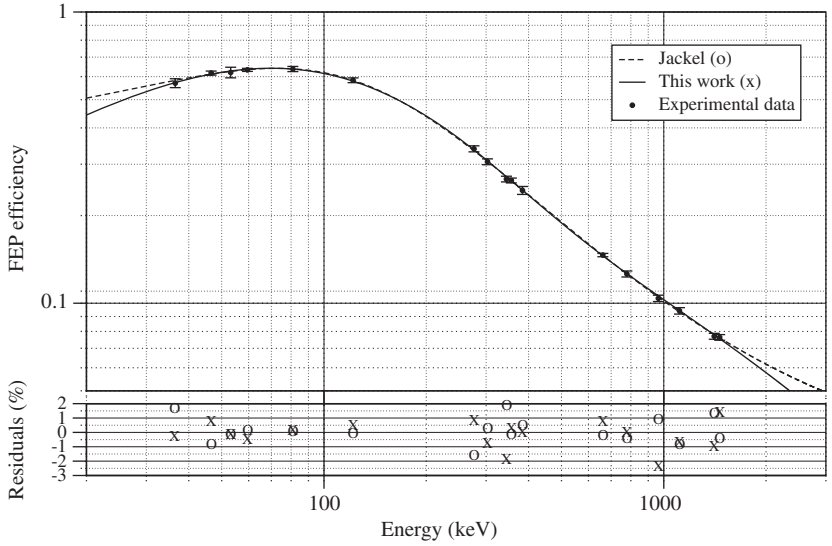

Fig. 5. Fitting experimental FEP efficiencies using Jäckel function and the proposed function.

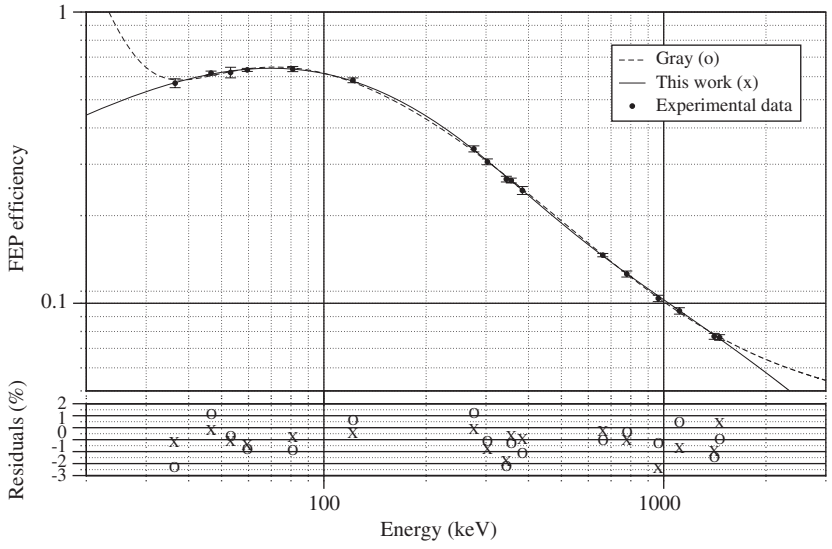

Fig. 6. Fitting experimental FEP efficiencies using Gray function and the proposed function.

Table 4
Reduced chi-square χ^2_R values and average standard deviations RD obtained in the fitting procedures using fitting curve equations shown in Table 2

Function	χ^2_R	RD (%)
Polynomial 5 order	0.51	1.22
Polynomial 6 order	0.20	0.75
Sánchez-Reyes	0.46	1.19
McNelles and Campbell	0.32	0.77
Jäckel	0.18	0.67
Gray	0.48	1.36
This work	0.23	0.65

Table 5
Extrapolated experimental FEP efficiencies values for energies 30, 2000 and 3000 keV using Jäckel function, polynomial of sixth order and the function proposed in this work, and Monte Carlo (MC) FEP efficiency values for the same energies

Function	30 keV	2000 keV	3000 keV
MC	0.516(4)	0.0581(7)	0.0406(8)
Polynomial logarithm	0.57(2)	0.059(1)	0.040(1)
D (%)	10.6	1.87	−2.3
Jäckel	0.56(2)	0.062(2)	0.049(2)
D (%)	8.6	6.2	21.6
This work	0.53(1)	0.058(1)	0.040(1)
D(%)	3.3	−0.68	−2.3

D (%) is the relative deviation between MC and extrapolated FEP efficiency values.

6. Conclusions

The HSA algorithm is powerful and more robust than the classical fitting algorithms (LM) reaching the global minimum of χ^2_R regardless of the functional form used and the initial values of the parameters. All fitting curve equations provide a good behaviour within the experimental energy range but only a few could give a proper FEP efficiency value extrapolating outside that

range. Especially the function proposed in this paper works well in both, interpolating and extrapolating FEP efficiency curve. Additionally Monte Carlo simulation becomes a powerful tool to test curves used in gamma-ray spectrometry.

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