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Reliability of two calculation codes for efficiency calibrations of HPGe detectors

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Abstract

This paper reports the reliability of efficiency calibrations for γ -ray detectors using the calculation codes ANGLE and LabSOCS. For experimental verification, three HPGe detectors under various laboratory geometry configurations were used for this study. An overall comparison between experimental and calculated efficiency calibration curves is presented and comments on the various error sources affecting the final results are given. The deviations are generally below 10%, which could be acceptable for many applications. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Among the methods of radioactivity measurement, γ -ray spectrometry is one of the most widely used. It is a non-destructive method and in general does not require sample preparation. High-resolution γ -ray spectrometry based on germanium detectors has been routinely applied in various fields of research and industry. It allows qualitative and quantitative analysis of radioactive material.

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 and sample shapes). Standard samples are in general costly or even unavailable because of the complexity of some configurations. Also the standard samples would need to be renewed, for instance due to the short half-life of some radionuclides. There is also a difficulty in establishing the efficiency calibration using standard sources because the density and composition of

the standard sources must be known in order to consider the self-absorption especially in the case of volume samples. Furthermore the density and composition of the sample to be analysed must be known, this is not always possible. To overcome these drawbacks, some calculation codes for efficiency calibration are nowadays commercially available. Lepy and all the participants to the EUROMET action 428 (2000) performed an interesting work in this topic presenting how one can transfer a Ge detector efficiency calibration from point source geometry to other geometries using some calculation codes. This paper reports the results obtained using the commercial calculation codes ANGLE and LabSOCS. These calculation codes allow to predict efficiency calibration curves for various Ge γ -ray detectors. Calculated efficiency curves, in the γ -ray energy range 59–1836 keV, were compared with experimental ones using standard samples in more general measurement configurations.

2. Summary of ANGLE and LabSOCS calculation codes

ANGLE is a PC friendly user code for calculation of efficiency calibration curves for semi-conductor detectors. The paper of Jovanovic et al. (1997) and references

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therein describe the concept behind it. This code was obtained from the University of Montenegro, Faculty of Sciences. The main features of this code of interest for a γ -ray spectrometry user can be outlined as follows. The ANGLE version used in this work (version 1.01) is executable under DOS/WINDOWS platforms and the code source is written in the PASCAL data processing language. ANGLE manages true and closed-end coaxial HPGe, open and closed-end Ge(Li), planar LEPD and “well-type” detectors for point, cylindrical and Marinelli sources positioned coaxially. The main inputs for ANGLE execution are the technical specifications of the detector of interest (type, dimensions of the crystal...). Although these data are rarely available in the usual documentation of the detector, the manufacturer can easily supply them. Inputs include also the measurement configuration (geometry, source type, container...) and the reference efficiency calibration curve. Very careful attention has to be paid for the latter input. In fact, the reference efficiency curve is made experimentally using a standard point source in a coaxial position (e.g. at 10 cm from the crystal). For a given γ -ray detector, only one reference efficiency calibration curve is needed to carry out ANGLE calculations. In principle, ANGLE can be used for all detectors, provided that the technical specifications are available in order to be inserted in the input file. The calculation time for a complex configuration does not exceed a minute using PCs with CPU greater than 200 MHz.

LabSOCS version 3.0 (Laboratory SOURCEless Calibration Software) is a Monte Carlo based code, whose main features are described by Bronson and Wang (1996) and Bronson et al. (1998). This code, based on a REXX platform, is integrated in the *Genie 2000* γ -ray spectrometry system of Canberra, which runs on WINDOWS based PCs. Before using the code, the considered germanium detector must undergo a so-called “Characterisation”, which is carried out by the LabSOCS manufacturer. This “Characterisation” is a procedure to obtain the response of the detector for sources located inside a 500 m radius sphere, centred in the detector, and over the photon energy range 45 keV–7 MeV. Although this procedure requires weeks, it has the advantage that in principle all detector geometry configurations can be used. Other input data are only those concerning the physical parameters of the measurement configuration such as source and container characteristics and source-detector displacement. LabSOCS already includes sample geometry templates of common commercially available containers, but its great advantage is that in principle all the features of any cylindrically symmetric object can be modelled. Each LabSOCS template allows a wide variety of parameters, which can be specified as necessary. Moreover, the efficiency calculation can be performed for all detector-

source displacements, not only for coaxial sample placement. The execution time did not exceed a minute for the geometries presented in this study. More complicated configurations usually do not require more than a few minutes. The final LabSOCS output is an efficiency calibration curve which is directly stored among the calibration files of the spectrum analysis software and can be used just like those generated by traditional calibration techniques. The user must enter uncertainties on calculated efficiency calibration curves. The relative standard uncertainties suggested by the manufacturer for standard laboratory conditions are of the order of 7% at low energies (50–100 keV), 6% at medium energies (100–400 keV) and 4% at high energies (400–7000 keV).

Both ANGLE and LabSOCS take into account the self-absorption in the considered samples. The composition, density and the shape of the considered samples are among the inputs of these codes.

It is important to note that the detector and geometry configurations presented by ANGLE are pre-defined and fixed, while LabSOCS allows reproducing the actual technical specifications. The same considerations are valid also for the modelling of the sample geometries: for example both Marinelli and Cylinder have sloping sides, which can be reproduced perfectly with LabSOCS and only approximately with ANGLE.

3. Materials and methods

The work was carried out using 3 (indicated with A, B and C) closed-end coaxial γ -ray detectors (p-type) 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 summarised in Table 1. The three detectors were considered for ANGLE calculation, while detector C was also used for LabSOCS comparison being the only one characterised by the manufacturer.

Seven radioactive sources, which are considered as standards, were used to establish the experimental calibration curves to be compared with those calculated by ANGLE and LabSOCS. The characteristics of these standards are given in Table 2. The ^{152}Eu point sources and vial are certified standards. Marinelli 1, Marinelli 2 and Cylinder are prepared in the authors' laboratory from certified multi-radionuclide standards. All the calibrations were made with the standards placed along the axis of the detectors. The three reference calibration curves, which are required as an input for ANGLE calculations (one for each detector), were made with Multi- γ point source at distances 117, 135 and 145 mm from the end caps of detectors A, B and C respectively. For the experimental calibration curves, which have

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

Detector	Relative efficiency (%)	Crystal length-diameter (mm)	Energy resolution (FWHM)	
			At 122 keV	At 1330 keV
A	30	65.2–55.1	679 eV	1.67 keV
B	20	43.5–56.8	921 eV	1.66 keV
C	43.5	60.5–60.5	867 eV	1.78 keV

Table 2
Characteristics of the seven radioactive standards used in this study (four are certified and others are prepared in the authors' laboratory)

Standard	Activity	State and chemical composition	Dimensions	Standard certification	Measurement distance from the detectors
Multi- γ point source	See Table 3	Solid	Point	ISOTOPE, Germany	105 cm from det C
^{152}Eu point source 1	45.6 kBq on 29/11/99	Solid	Point	CERCA, France	141 mm for det A
^{152}Eu point source 2	377 kBq on 27/07/98	Solid	Point	DAMRI, France	330 mm from det B
^{152}Eu vial	203.5 kBq on 10/95	12 ml liquid (density 1.25 g/cm ³) ZnCl ₂ solution	Inside radius 0.9 cm	ENEA, Italy	300 mm from det B
Marinelli 1	See Table 4	1 l liquid (density 1.0 g/cm ³) water	Inside radius 6.8 cm	Prepared from a certified CERCA standard	Contact with det A
			Cavity radius 3.9 cm Cavity depth 6.9 cm		Contact with det C
Marinelli 2	See Table 4	1 l liquid (density 1.4 g/cm ³) ZnCl ₂ solution	Inside radius 6.8 cm	Prepared from a certified ENEA standard	Contact with det A
			Cavity radius 3.9 cm Cavity depth 6.9 cm		Contact with det C
Cylinder	See Table 4	400 ml liquid (density 1.4 g/cm ³) ZnCl ₂ solution	Inside radius 5.5 cm	Prepared from a certified ENEA standard	Contact with det A
					Contact with det C

been compared with those calculated by ANGLE and LabSOCS, these distances are included in Table 2.

In order to minimise the statistical counting uncertainties, a γ -ray spectrum which is an average of five γ -ray spectra, was used to establish the experimental calibration curve for the given detector, which was then compared with that calculated by ANGLE and Lab-

SOCS. The γ -ray spectrometry electronic instrumentation used in this work is a Canberra chain monitored with *Genie 2000* software. The acquisitions of the γ -ray spectra were carried out with a sufficient counting statistics for the γ -lines of interest (standard uncertainties over the peak areas below 2%). All the γ -spectra were acquired with an average dead time below 2%

(maximum value: 10% for ^{152}Eu), in order to minimise the standard uncertainty due to corrections for the dead time. The acquisition time range for each spectrum was 2–6 h. The γ -ray energy range considered in this study is 59–1836 keV for all the detectors (see Tables 3 and 4).

The coincidence-summing effect, which is not negligible, especially in close geometries (Marinelli, Cylinder) or in high efficiency detectors (Detector C), has been taken into account in this study. In fact, the determined experimental efficiency curves obtained with Marinelli 1, Marinelli 2 and Cylinder have been corrected for the summing effect of γ -lines of ^{88}Y , ^{60}Co and ^{152}Eu (Debertin and Schotzig, 1979; Semkow et al., 1990; De Felice et al., 2000). As an example, this correction is up

to 8% for ^{88}Y in the case of detector C in Cylinder configuration.

4. Results and discussion

The relative deviations between the theoretical (ϵ_{theo}) and the experimental (ϵ_{exp}) efficiency calibration values are calculated as $(\epsilon_{\text{exp}} - \epsilon_{\text{theo}})/\epsilon_{\text{exp}}$. They are presented in % in Figs. 1–3. The average of the absolute values of these deviations, calculated in the low-energy range and in the high-energy range, respectively 59–700 keV and 700–1836 keV, are shown in Tables 5–7.

Table 5 and Fig. 1 are dedicated to the results obtained using ANGLE with detector A. Table 6 and Fig. 2 present the results obtained using ANGLE with detector B. Table 7 and Fig. 3 summarise the results obtained using both ANGLE and LabSOCS with detector C.

Concerning ANGLE, the results agree within 5% for all the studied configurations (detector, geometry, standard) except for detector C in the high-energy range in both Marinelli 1 and Marinelli 2 configurations. In these cases the deviations are within 11%.

Concerning LabSOCS, in the low-energy range, the deviations are <3.3% for all the studied configurations, while in the high-energy range the deviations are <7.2%.

It is important to remember that the experimental efficiency curves, which are compared to those calculated, are affected by various uncertainties. For instance, there is a standard uncertainty of up to 5% on the activity of the prepared standards Marinelli 1, Marinelli 2 and

Table 3

Activity of Multi- γ point source used for the determination of the reference calibration curve required for ANGLE calculations

Radionuclide	γ -ray energy (keV)	Activity (Bq) January 15, 2001
^{241}Am	59.54	5609.20
^{109}Cd	88.03	59829.00
^{57}Co	122.06	2265.51
$^{123}\text{Te-m}$	159.00	2415.73
^{51}Cr	320.08	57054.00
^{113}Sn	391.70	10792.90
^{85}Sr	514.01	14507.70
^{137}Cs	661.66	8868.90
^{88}Y	898.04	21778.20
^{60}Co	1836.06	10955.70
	1173.24	
	1332.50	

Table 4

Activities of Marinelli 1, Marinelli 2 and Cylinder standards used in this study

Radionuclide	γ -ray energy (keV)	Activity in Bq of Marinelli 1 standard on October, 25th 1999 (4.5% standard uncertainty)	Activity in Bq of Marinelli 2 standard on October, 1st 1999 (3% standard uncertainty)	Activity in Bq of the Cylinder standard on October, 1st 1999 (3% standard uncertainty)
^{241}Am	59.54	976.76	2868.62	1196.50
^{109}Cd	88.03	4692.40	3003.25	1252.67
^{57}Co	122.06	235.20	186.62	77.84
	136.47			
^{139}Ce	165.86	240.79	191.95	80.07
^{51}Cr	320.08	9540.24	2664.67	1111.45
^{113}Sn	391.70	1340.61	511.87	213.51
^{85}Sr	514.01	955.55	603.85	251.90
^{137}Cs	661.66	1432.05	587.86	245.19
^{88}Y	898.04	1453.90	726.49	303.02
	1836.06			
^{60}Co	1173.24	1420.37	819.80	341.94
	1332.50			

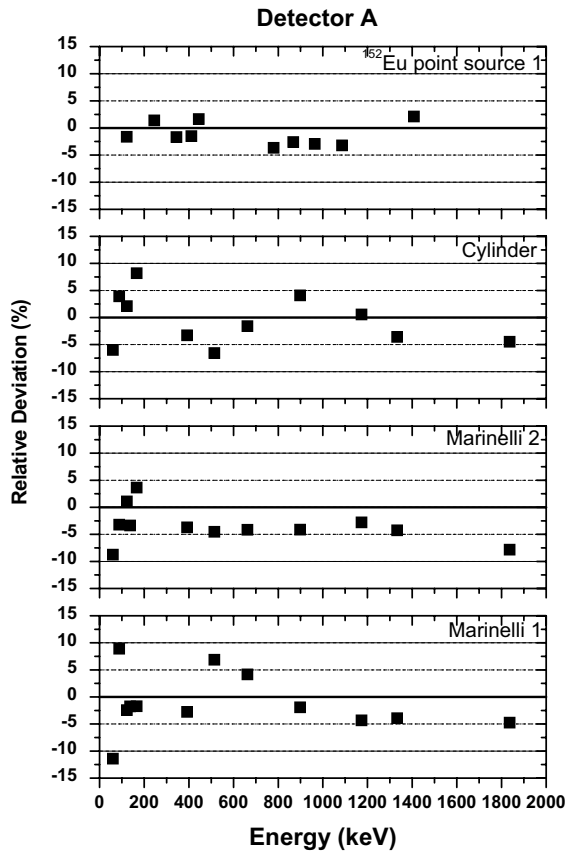


Fig. 1. Results obtained with detector A. Relative deviations between the ANGLE calculated and the experimental efficiency calibration values for ^{152}Eu point source 1, Marinelli 1, Marinelli 2 and Cylinder standards.

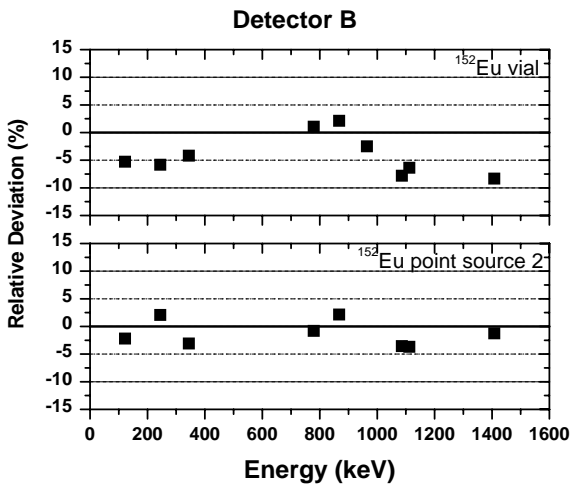


Fig. 2. Results obtained with detector B. Relative deviations between the ANGLE calculated and the experimental efficiency calibration values for ^{152}Eu point source 2 and ^{152}Eu vial standards.

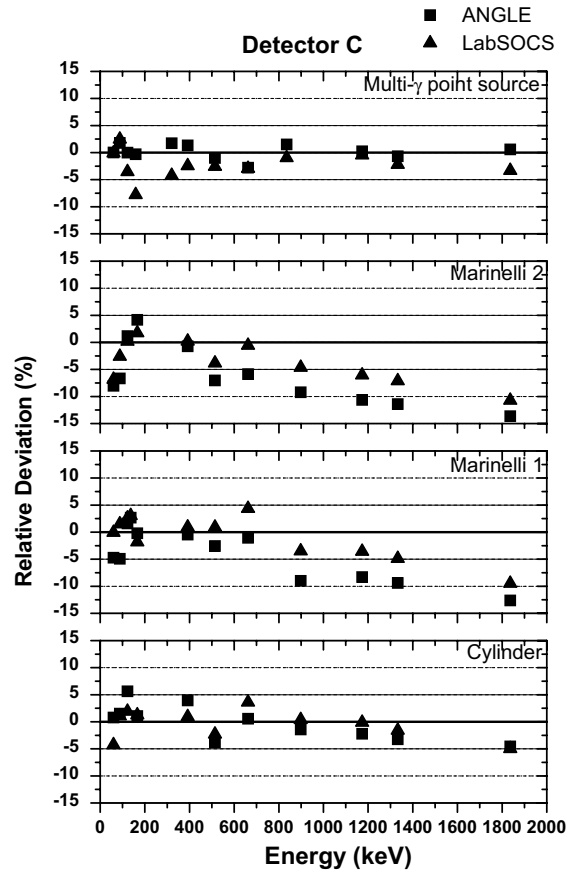


Fig. 3. Results obtained with detector C. Relative deviations between both the ANGLE and the LabSOCS calculated and the experimental efficiency calibration values for Multi- γ point source, Cylinder, Marinelli 1 and Marinelli 2 standards.

Cylinder (up to 3.5% from the certificates; a factor of 1.5 has been arbitrarily included to consider uncertainties due to dilutions and handling of the standards). Also geometry positioning and counting statistics affect the accuracy of the results. A further uncertainty can arise from the method used in this study for the coincidence summing effect correction. A final uncertainty can be attributed to the theoretical models on which the codes are based.

5. Conclusions

Nowadays, due to the wide variety of radioactive samples to be measured, calculation codes for γ -spectrometry efficiency calibration are helpful. The concept of such simulation could also be applied to efficiency calibration for other techniques such as alpha spectrometry or X-ray fluorescence analysis. Also other kinds of semi-conductor γ -ray detectors such as CdTe/

Table 5
Averages of deviations (%) between experimental and calculated efficiency values using ANGLE in the configuration of detector A

Standard	ANGLE results obtained with detector A	
	Average deviation (%) in the low-energy range (59–700 keV)	Average deviation (%) in the high-energy range (700–1836 keV)
Marinelli 1	5.1	3.7
Marinelli 2	4.1	4.7
Cylinder	4.5	3.2
¹⁵² Eu point source 1	1.6	2.9

These averages are determined in the low-energy range (from 59 to 700 keV) and in the high-energy range (from 700 to 1836 keV).

Table 6
Averages of deviations (%) between experimental and calculated efficiency values using ANGLE in the configuration of detector B

Standard	ANGLE results obtained with detector B	
	Average deviation (%) in the low-energy range (100–700 keV)	Average deviation (%) in the high-energy range (700–1500 keV)
¹⁵² Eu point source 2	2.4	2.3
¹⁵² Eu via I	5.1	4.7

These averages are determined in the low-energy range (from 100 to 700 keV) and in the high-energy range (from 700 to 1500 keV).

Table 7
Averages of deviations (%) between experimental and calculated efficiency values using both ANGLE and LabSOCS in the configuration of detector C

Standard	ANGLE results obtained with detector C		LabSOCS results obtained with detector C	
	Average deviation (%) in the low-energy range (59–700 keV)	Average deviation (%) in the high-energy range (700–1836 keV)	Average deviation (%) in the low-energy range (59–700 keV)	Average deviation (%) in the high-energy range (700–1836 keV)
Multi- γ point source	1.1	0.8	3.3	1.7
Marinelli 1	2.3	11.2	1.9	7.2
Marinelli 2	4.2	9.8	2.0	5.4
Cylinder	2.5	2.9	2.2	1.8

These averages are determined in the low-energy range (from 59 to 700 keV) and in the high-energy range (from 700 to 1836 keV).

CdZnTe may be considered, although their energy resolution and efficiency are poor.

The use of ANGLE requires the knowledge of specific detector parameters and an experimentally determined calibration curve, but has the advantage to be extendable to different detectors. Conversely, LabSOCS calculation requires only information about the positioning, dimensions and nature of the source, but its use is limited to a previously “characterised” detector.

The results obtained are promising and further improvement may be expected with the development of the techniques for the summing effect estimation. In all the fields where deviations of the order of 10% are acceptable, especially for those scenarios where the

sample size, geometry and positioning from the detector are changing frequently, ANGLE and LabSOCS can be considered reliable for γ -ray detector efficiency calibrations.

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