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VALIDATION OF EFFICIENCY TRANSFER FOR MARINELLI GEOMETRIES

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ABSTRACT :

In the framework of environmental measurements by gamma-ray spectrometry, some laboratories need to characterize samples in geometries for which a calibration is not directly available. A possibility is to use an efficiency transfer code, e.g. ETNA. However, validation for large volume sources, such as Marinelli geometries, is needed. With this aim in mind, ETNA is compared, initially to a Monte Carlo simulation (PENELOPE) and subsequently to experimental data obtained with a high-purity germanium detector (HPGe).

1. INTRODUCTION

In the framework of low level activity measurements, detection limits as low as possible are required. This is achieved using dedicated “low-level” spectrometers and also using large volume samples. For low density materials, big volume sources like Marinelli geometries can provide the best detection limits. Unfortunately, this type of geometry increases problems of coincidence summing effect and matrix effects. Therefore, the establishment of the efficiency calibration curve is particularly complex. A good compromise for laboratories using volume sources, but without the possibility to establish an efficiency curve, is to calculate efficiency transfer (ET) factors to derive calibration from a reference geometry to another one, taking into account the sample matrix effect with the knowledge of its mass attenuation coefficient.

The Laboratoire National Henri Becquerel (LNHB) developed a tool for the calculation of the Efficiency Transfer for Nuclide Activity measurements (ETNA). The ETNA software offers a practical and convenient solution to some problems encountered in measurement laboratories (Piton *et al.*, 2000, Lépy *et al.*, 2004). Particularly, it can be used to calculate the efficiency of

the detector under measurement conditions different from those of calibration (ET factors). Databases are included, making it possible to record characteristics of different measurement geometries and update data on materials (attenuation coefficients). The goal of this work is further validation of ETNA and its limits of use. ETNA has already been validated for some specific cases, point sources or volume sources far from the detector window. Here, the work is focused on measurement conditions appropriate for environmental samples, with samples close to the detector, with large volume, including Marinelli geometries. Two approaches are used in the present work:

- i. calculation: based on the previous intercomparison exercise (Vidmar *et al.*, 2010) where four general Monte Carlo codes and five dedicated packages for efficiency determination in gamma-ray spectrometry were compared using simple case studies;
- ii. experiment: measurements performed with an actual high-purity germanium (HPGe) detector used at the LNHB and standard solutions in different geometries.

With this aim in mind, and using the previous work testing efficiency transfer codes, we compare the results of a Monte Carlo simulation, ETNA calculation and experimental values on a real detector with Marinelli geometries. It must be noted that, in order to avoid the coincidence summing effects, only mono-energetic radionuclides are used for the experimental validation.

The Monte Carlo code chosen is PENELOPE2008 which simulates coupled electron and photon transport in arbitrary materials. The version used includes several generic programs that allow easy implementation. The PENMAIN routine is used to build a simulating model based on the geometry subroutine package PENGEOM, which performs particle tracking in material systems consisting of homogeneous regions (bodies) limited by quadric surfaces. The

output file (energy deposition spectrum) provides the distribution of absorbed energy in the detector. This is obtained as a histogram representing the probability distribution function (per eV and per initial particle). The full energy peak (FEP) efficiency is obtained by multiplying the probability distribution function corresponding to the energy of interest (full-energy deposition) by the bin energy width. In this work, to determine ET factors using the Monte Carlo code, two simulations must be run, one for each geometrical condition. The ET factor is the ratio of the FEP efficiency for the measurement geometry by the one for the reference condition.

2. CALCULATION VALIDATION

2.1 Cylindrical geometries

The first step of the study is part of the exercise led by Vidmar *et al* (2010) whose goal was to compare ET calculations for some simple case studies, using two types of detector and four different geometries. All parameters for the different samples were given to ensure that all participants had exactly the same parameters in each case. Regarding the detectors, the authors used the same approach and provided all parameters for two detectors, one n-type and one p-type; as an example, geometrical parameters for the p-type detector are given in Table 1. The task for each laboratory was to calculate ET factors in the 20-2000 keV energy range. The codes featuring in this comparison fell into two categories: specialized codes written specifically for efficiency calculations in gamma-ray spectrometry and general Monte-Carlo simulation tools adapted to the task at hand. From the first group, the codes GESPECOR (Sima and Arnold, 2002), ETNA (Piton et al, 2000), DETEFF (Cornejo Diaz and Jurado Vargas, 2008), ANGLE (Jovanovic et al., 1997) and EFFTRAN (Vidmar, 2005) were tested. The representatives of the second group were GEANT 3.21 (Brun et al., 1987), MCNP (Briesmeister, 2000; X-5 Monte Carlo Team, 2003), PENELOPE (Salvat et al., 2003,

2008) and EGS (Nelson et al., 1985). LNHB participated in this exercise using both approaches: the ETNA code as dedicated software and PENELOPE for Monte Carlo simulation. The required efficiency transfer factors were derived for the reference geometry (a large polyethylene cylindrical container filled with aqueous solution, at 1 mm from the detector window) and three measurement geometries:

- (i) “Point”: point source at 2 cm from the detector window,
- (ii) “Soil”: cylindrical container filled with quartz, at 1 mm from the detector window,
- (iii) “Filter”: thin cylindrical container filled with cellulose, at 1 mm from the detector (i.e. a single air filter).

The form and dimensions were kept simple to ensure that the geometrical conditions would not affect the comparison results.

Table 2 gives the relative deviation, R , between LNHB results calculated with PENELOPE and the mean results of the intercomparison across the entire energy range, for both detectors types (A= p-type, B=n-type), where:

$$R = \frac{(FP - FM)}{FM} \times 100$$

FP is the ET factor computed with PENELOPE, and FM is the mean value of the ET factor calculated by the participants. For each case, a very good agreement is achieved with the Monte Carlo approach, with relative differences similar to the standard deviation of the results of the exercise.

Similarly, Table 3 gives the relative deviation between the values calculated with ETNA and the mean results of the intercomparison. Good agreement is seen, even if the relative deviations are slightly higher than those of the PENELOPE case. However, the maximum

relative deviation is still only 2.5% at low energy with detector B. Comparison of the results between PENELOPE and ETNA are given in Table 4.

It can be assumed that both our PENELOPE simulation and ETNA calculations are validated for these cylindrical geometries. These results form the basis on which to continue this study; since PENELOPE led to slightly better values, in the following, it will be considered the reference.

2.2 Marinelli geometries

In order to pursue this work, a further case study was considered, with two Marinelli geometries, “450D2” and “SG3000cut”, using detector A as described in the previous study. The Marinelli sources are presented in Figure 1 with the material and thickness used. In all cases, the geometries use a plastic container filled with water. The Marinelli containers are positioned at 1 mm from the detector window. The particular aim of this part of the work is to establish the ET factor between the reference sample of the exercise (cylinder with water) and Marinelli geometries.

The ET factors obtained by Monte Carlo simulation for these new geometries are presented in Table 5. The criterion for the PENELOPE simulation was to get a statistical uncertainty less than 1 % in order to establish the reference values. These are compared with ET factors calculated by ETNA. The relative deviations between PENELOPE and ETNA are also given in the last two columns.

2.3 Discussion

Table 5 shows the results for the two Marinelli geometries, 450D2 and SG3000cut. Both geometries are similar in terms of the detector, with the source around the crystal, the

only differences between them are the container dimensions, as seen in Figure 2. The thickness of the 450D2 is only 1 cm along the side of the crystal, whereas the SG3000cut is 5 cm. Results obtained with ETNA are in good agreement with PENELOPE calculation. All relative deviations are below 6% across the entire energy range. However, we also performed comparison for a “SG3000” geometry, which is a standard container for 3000 cm³ volume source, for which the container extends below the crystal bottom; in this case, we observed higher relative deviations that cannot be only explained by the attenuation coefficients difference between ETNA and PENELOPE. This highlighted problem is currently being studied.

This first part of the study confirms that ETNA can calculate efficiency transfer factor for volume geometries, including the case of Marinelli containers, provided that the container bottom is at the level, or above, the base of the detecting crystal. This is established by comparison with Monte Carlo simulation, and remains a validation of the calculation. Now, experimental validation is required to validate the use of the code in practical cases. This was performed using the 450D2 geometry.

3. EXPERIMENTAL VALIDATION

As a next step, only the case of the 450D2 Marinelli geometry is considered, for which the ETNA calculation is validated by the previous results. This step consists of an ETNA calculation and PENELOPE simulation for a real detector in use at LNHB, allowing comparison with experimental data.

3.1 Experimental setup

The detector under study is a 100 cm³ n-type HPGe detector, which is an Ortec GMX-15-70-S model. For the higher energy ⁶⁰Co gamma line at 1.33 MeV, the detector has a relative efficiency of 15 % and an energy resolution of 1.8 keV. All the detector parameters, dimensions and materials, are given in Figure 3. The efficiency calibration of the detector is obtained using standard solutions and is accurately established for point sources at 10 cm and for the “SG500” cylindrical volume source of 500 cm³ at 8.33 cm from the detector window. Moreover, two 450D2 Marinelli standards were prepared: the radionuclides chosen were ¹³⁹Ce and ¹³⁷Cs, emitting photons with 166 and 662 keV, respectively. These two radionuclides allowed to check for possible problems at low energies.

3.2 Monte Carlo simulation for point source

As in the previous part of the study, the criterion for the PENELOPE simulation was to obtain a statistical uncertainty less than 1 %. First of all, in order to be sure that the PENELOPE simulation is correct, even if the dimensions were obtained with an X-ray analysis, a first simulation was made for a point source at 10 cm from the top of the detector. The results showed a relative deviation between PENELOPE simulation and the experimental calibration of approximately +10%. This result suggests a problem with the dimensions of the crystal used in the simulation because a constant relative deviation as a function of energy is synonymous with a difference of solid angle. As the external crystal dimensions were checked by X-ray analysis, the only solution is the dimensions of the dead layer of the crystal side, the front dead layer being validated by comparison with the experimental calibration in the low-energy range. Knowing a similar problem on another detector in LNHB, a further simulation was made with a crystal diameter of 46.6 mm instead of 48.6 mm, i.e. with a dead layer thickness of 1 mm. The simulation with this new diameter gave relative deviations below 1.5 % for both energies, which confirmed the dead layer problem. This diameter was adopted for the rest of this study.

3.3 Efficiency transfer for volume sources

With this optimized parameter for the crystal active diameter, the Monte Carlo simulation was performed for volume geometries for which the experimental efficiency is established. The SG500 was simulated by a cylinder with a diameter of 9.41 cm, lateral at the side of 1.6 mm and bottom thickness of 1 mm. The relative deviation with the experimental results are -4 % for ^{139}Ce and +3.3 % for ^{137}Cs . This result confirms the simulation of the germanium detector and allowed the determination of the efficiency transfer between this reference geometry and the Marinelli 450D2 geometry to be carried out.

Table 6 shows the efficiency transfer between the SG500 at 8.33 cm (as reference) and the Marinelli geometry 450D2 on top of the detector. There is a good agreement with experimental values at 662 keV, the relative deviations being -3.9 % for PENELOPE and +1.3 % for ETNA. At 166 keV, the relative deviations increase to +2.4 % and +5.1 %, respectively.

4. CONCLUSION

The present validation work, including Monte Carlo simulations and comparisons with experimental data, demonstrates that ETNA can be used for large geometries including Marinelli containers and that the efficiency transfer can be obtained with 2-3% uncertainty for energies higher than 100 keV. This is achieved for a large energy range in only one calculation, so that using ETNA is much faster than Monte Carlo methods to obtain reliable results for a large energy range. The main objective of this work is reached, and this result confirms the interest of such software when a laboratory does not have standard solutions for

different geometries. The next step of this study will be apply for Marinelli geometries whose bottom extends below the crystal in order to validate that ETNA could be used in these cases.

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Table 1

Detector parameters. All dimensions are given in millimeters (mm). The housing diameter is in all cases the same as the window diameter.

Parameter	
Crystal type	P
Crystal material	Ge
Crystal diameter (including the side of the dead layer)	60
Crystal length (including the top dead layer)	60
Dead layer thickness (top and side)	1
Hole diameter	10
Hole depth	40
Window diameter	80
Window thickness	1
Window material	Al
Crystal-to-window distance	5
Housing length	80
Housing thickness	1
Housing material	Al

Table 2

Relative deviation (%) between ET calculated by PENELOPE and the mean results of the intercomparison.

Energy/keV	Point A	Soil A	Filter A	Point B	Soil B	Filter B
20				-1,6	-1,2	-1,0
45	-1,0	-0,1	-0,8	-1,7	0,3	-1,3
60	-0,7	0,5	-0,6	-0,9	0,9	-0,8
80	-1,2	0,3	-1,1	-0,9	0,4	-0,8
120	-0,2	0,3	-0,8	-0,8	0,4	-0,9
200	0,3	0,2	-0,7	0,7	0,5	-0,6
500	0,4	0,3	-0,3	-0,1	-0,3	-0,6
1000	-0,2	0,8	-1,0	0,4	0,8	-0,2
2000	0,5	-0,2	-0,6	-0,1	0,2	-0,6

Table 3

Relative deviation (%) between ET calculated by ETNA and the mean results of the intercomparison.

Energy/keV	Point A	Soil A	Filter A	Point B	Soil B	Filter B
20				1,5	2,5	2,4
45	-0,4	2,3	0,0	0,1	2,1	0,6
60	0,0	0,2	0,5	0,2	0,3	0,7
80	-0,1	0,1	0,5	0,0	0,1	0,6
120	-0,3	1,3	1,3	-0,2	1,1	1,0
200	-1,1	-0,1	1,0	-1,0	-0,6	1,4
500	-1,3	-0,5	1,0	-1,5	-0,7	1,3
1000	-1,0	-0,6	0,9	-1,5	-0,8	1,3
2000	-1,1	-0,8	0,9	-1,3	-0,8	1,2

Energy/keV	Point A	Soil A	Filter A	Point B	Soil B	Filter B
20				3,1	3,7	3,5
45	0,6	2,3	0,8	1,8	1,8	1,9
60	0,8	-0,3	1,1	1,1	-0,6	1,6
80	1,1	-0,2	1,6	1,0	-0,3	1,5
120	-0,1	1,0	2,1	0,6	0,8	2,0
200	-1,4	-0,3	1,7	-1,7	-1,1	2,0
500	-1,7	-0,8	1,3	-1,4	-0,4	2,0
1000	-0,9	-1,4	1,9	-1,9	-1,6	1,5
2000	-1,6	-0,6	1,5	-1,2	-1,0	1,8

Table 5

Efficiency Transfer (E.T.) factors calculated by ETNA for Marinelli 450D2 and SG3000cut.

Energy/keV	E.T. Penelope simulation		E.T. Etna computation		Relative deviation (%)	
	450D2	SG3000cut	450D2	SG3000cut	450D2	SG3000cut
45	0,5005	0,2202	0,4891	0,2170	-2,3	-1,4
60	0,5217	0,2292	0,5170	0,2291	-0,9	-0,1
80	0,5533	0,2425	0,5476	0,2426	-1,0	0,0
120	0,6033	0,2728	0,5927	0,2657	-1,8	-2,6
200	0,6470	0,2956	0,6301	0,2903	-2,7	-1,8
500	0,6849	0,3362	0,6589	0,3207	-4,0	-4,6
1000	0,6968	0,3597	0,6721	0,3398	-3,7	5,5
2000	0,7057	0,3725	0,6833	0,3568	-3,3	-4,2

Table 6

Efficiency Transfer factors with PENELOPE and ETNA and the relative deviation compared to experimental data for ^{139}Ce and ^{137}Cs .

ENERGY (keV)	E.T. Experimental	E.T. PENELOPE	Relative Deviation (%)	E.T. ETNA	Relative Deviation (%)
165.86	11.6	11.9	2.4	12.2	5.1
661.66	10.3	9.8	-3.9	10.5	1.3

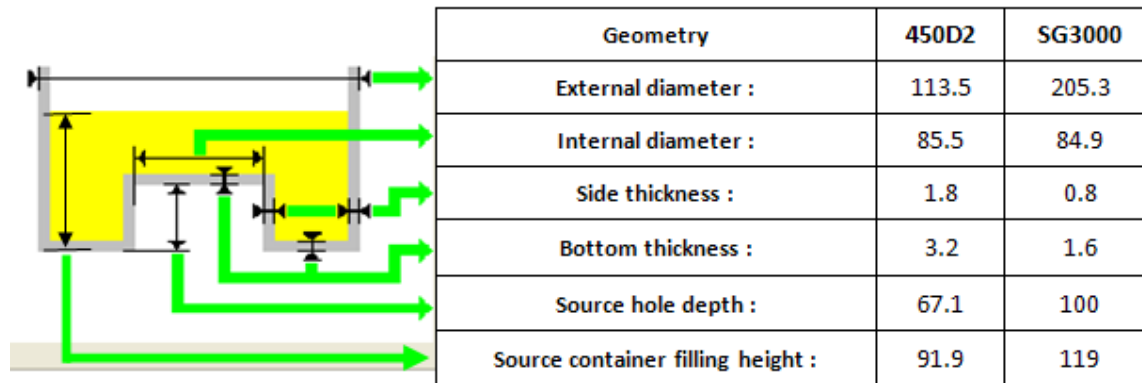
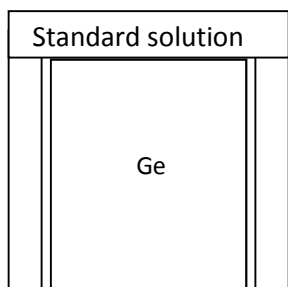
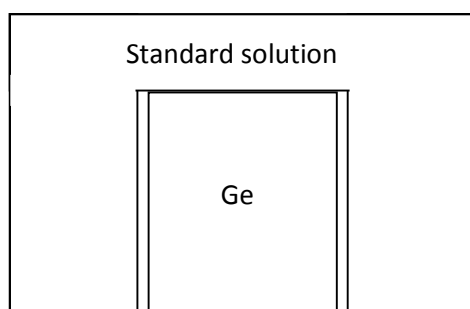


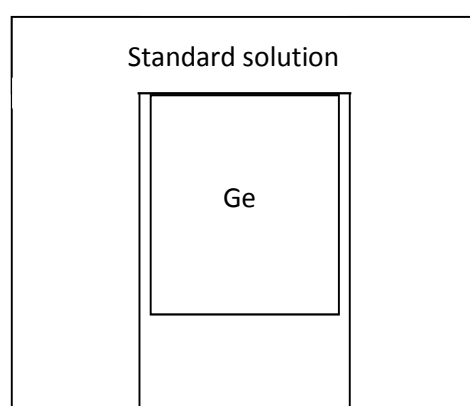
Figure 1. Dimensions (in mm) for the Marinelli geometries.



Marinelli 450D2

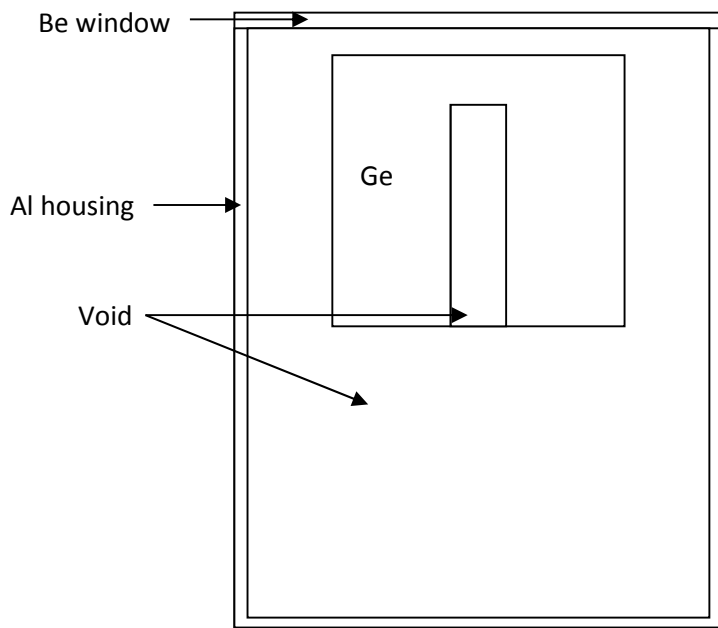


Marinelli SG3000cut



Marinelli SG3000

Figure 2. Marinelli positions on the same HPGe detector



Detector parameters :

- Crystal material: Ge
- Crystal diameter: 48.6
- Crystal length: 55.2
- Dead layer thickness: 0.0003
- Hole diameter: 9.5
- Hole depth: 47.2
- Window diameter: 70
- Window thickness: 0.5
- Window material: Be
- Crystal to window distance: 4.4
- Housing length: 125
- Housing thickness: 1.8
- Housing material: Al

Figure 3. Detector parameters of the experimental setup. All parameters are given in millimeters (mm).