The role of gamma-ray spectrometry and Monte Carlo simulation in the characterisation of meteorites

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Abstract

Gamma-ray spectrometry has proved to be a powerful tool in meteorite identification due to the presence of certain mid- and long-lived cosmogenic radionuclides in such samples. In particular, this technique can be made totally non-destructive by measuring each sample as-it-is and calculating the full-energy-peak efficiency through Monte Carlo simulation of the full radionuclide decay. In general, this framework can be applied whenever it is needed to characterise non-destructivery the γ-ray emission of a sample with non-standard geometry.

Keywords: meteorite, gamma-ray spectrometry, simulation, non-destructive

1 Introduction

A novel protocol for a completely non-destructive identification and characterisation of meteorites has recently been proposed by Rossini et al.[1]. In this protocol, the sample identification is made by means of γ-ray spectrometry, looking for radionuclides produced by primary cosmic rays having half-life smaller than the age of the atmosphere ($\sim 10^9$ y)[2]. This would indicate that the sample spent much time outside of the Earth atmosphere after its formation and therefore proves its meteoric origin. Such radionuclides are[3]: ^{26}Al ($t_{1/2} = 7.6 \cdot 10^5$ y), ^{60}Co ($t_{1/2} = 5.27$ y), ^{22}Na ($t_{1/2} = 2.6$ y), ^{54}Mn ($t_{1/2} = 312$ d), ^{46}Sc ($t_{1/2} = 84$ d) and ^{48}V ($t_{1/2} = 16$ d).

Carrying out non-destructive measurements is a key point in modern techniques for the characterisation of valuable samples, but they can make the data acquisition and data processing more complicated. In particular, concerning γ-ray spectrometry, the calculation of efficiencies must take into account various factors such as the sample self-absorption, its geometry and inhomogeneities. In particular, the efficiencies have to be computed by a Monte Carlo simulation based on a thorough description of the geometry and composition of the detector and the sample. In this work, simulations are carried out using either Geant4[4] or Arby[5], a toolkit developed within INFN Milano-Bicocca to act as an interface to Geant4. Arby has been originally developed for rare event Physics in the field of neutrinoless double beta decay ($0\nu\beta\beta$), particularly within the CUORE collaboration, but it is now used also for detector simulation in Applied Physics.

The potential of this technique, together with its future perspectives, are here discussed with some practical examples. This protocol of measurement can be applied to various samples coming from different areas of research, whenever it's needed to characterise the γ-ray emission avoiding sample disruption.

2 Gamma-ray spectrometry with HPGe detectors

Gamma-ray spectrometry consists in the measurement of the γ-ray emission of a sample with the best possible energy resolution in order to optimise the peak identification and the signal-to-noise ratio. This can be done using High Purity Germanium (HPGe) detectors. If seeking for a low-activity radionuclide, it is also important to reduce the background by introducing passive shieldings made of Pb and Cu, and to choose the materials for the detector and shielding to be as radio-pure as possible.

Some of the measurements in this work were carried out at the Radioactivity Laboratory of the University of Milano-Bicocca (Italy), whereas some samples had to me measured in a low-background underground facility. STELLA (SubTErranean Low Level Assay) is an extremely low-background γ-ray spectrometry facility[6] located under $\sim$ 3800 mwe (meters of water equivalent) of rock under the Gran Sasso massif, in Italy, as part of Laboratori Nazionali del Gran Sasso (LNGS). The detector used in Milano-Bicocca is a Broad Energy Germanium (BEGe) detectory by ORTEC (at the 1332.52 keV ^{60}Co peak: relative efficiency 50%, energy resolution 1.8 keV). The detector used at STELLA is a p-type coaxial low-background HPGe detector from ORTEC (at the 1332.52 keV ^{60}Co peak: relative efficiency 84%, energy resolution 1.9 keV).

3 Simulation with Arby

Arby[5] is an interface to Geant4 developed within the INFN division of Milano-Bicocca. Despite being originally designed for rare event Physics, it is now also used for Monte Carlo simulation in Applied Physics. It is a versatile pre-compiled toolkit which simplifies the access to Geant4. Arby interprets the geometry from a configuration file and reads the beams or decays to be simulated from command line or from a macro included when launching the simulation. As a consequence, no compilation is needed when the the configuration is changed, which makes Arby a very useful tool when handling simulation with many samples and many detectors.

3.1 Detector geometry

The starting point for the optimisation of the detector geometry in the simulation is the detector datasheet. It describes all the volumes present in the detector and its external shielding and it has been implemented in Arby for all the used detectors. However, computing the efficiencies with this geometry might result in big discrepancies with the true values. The detector may be slightly different from what is declared and its ageing is another factor contributing to this. In fact, for example, the p/n doped parts of the p-i-n junction, which are not active parts, tend to slowly diffuse in the active part, represented by the bulk. All these factors can be simplified by optimising the thickness of some absorbing layers obtaining some equivalent values which take into account all these effects.

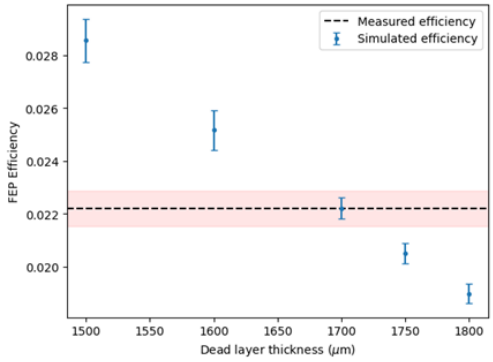

Fig. 1 Variation of the Full-Energy-Peak (FEP) Efficiency for the 60 keV peak from a calibrated ^{241}Am source as a function of the thickness of the frontal n-type-doped germanium layer. An equivalent layer thickness of 1700 µm is the value which best reproduces the experimental result (dashed line).

Figure 1 shows an example of optimisation of the thickness of the frontal n-type layer in the Milano-Bicocca BEGe detector to match the measured efficiency (dashed line). Figure 2 shows the difference between the simulated efficiencies with the nominal n-type dead layer thickness (declared in the datasheet) and the optimised value for the same detector. This optimisation has been carried out using radioactive sources

containing certified amounts of ^{241}Am (3.45 kBq), ^{137}Cs (2.72 kBq) and ^{60}Co (3.21 kBq), supplied by Eckert&Ziegler. Although the difference shown in this case is quite prominent, the typical effect is small, but still not negligible.

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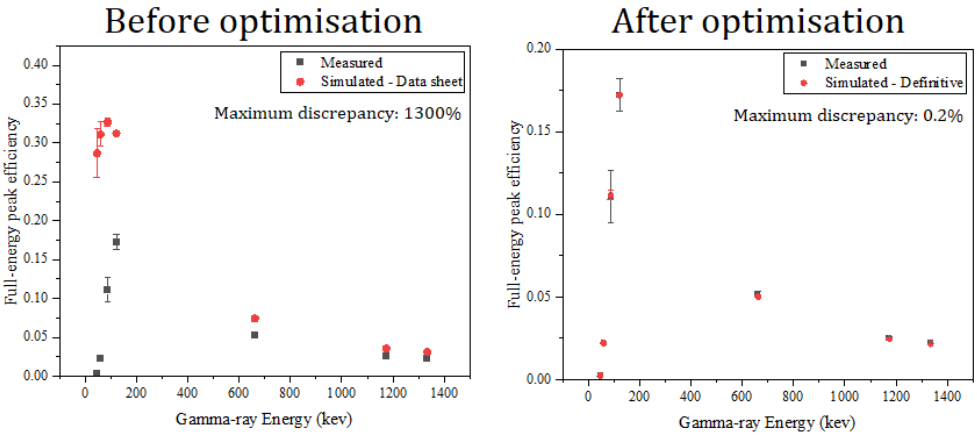

Fig. 2 The optimisation of the thickness of various dead layers in the detector geometry can return significant improvement in the estimation of efficiencies: the case of the Milano-Bicocca BEGe detector is here shown, where this effect is particularly effective. This is a key factor in the reliability of the specific activity calculated for each measured radionuclide.

3.2 Sample geometry

When the characterisation of the detector results in a trusted model for the detector geometry, it is eventually possible to simulate the processes occurring in the experimental setup and extract the efficiencies. However, while performing γ-ray spectrometry on whole samples with complex geometry, this is not straightforward.

In fact, the geometry of the sample plays a crucial role in the systematic uncertainties on the simulated efficiency and therefore on the specific activity estimated for each radionuclide. This is clearly pointed out in a previous work[7], where the measurement and analysis procedure is also disclosed. In particular, a first approach could be to simplify the geometry to a solid[7], but better accordance is obtained by considering the sample as a composition of different solids[1]. Table 1 presents the results from these two approaches in the estimation of the ^{26}Al specific activity in the samples in Figure 3. The approximated geometry for sample R001 in the two approaches is shown in Figure 4. It follows that:

- the relative uncertainty, estimated through the calculation of the uncertainty budget as shown in this Section, is smaller when the simulation geometry describes more thoroughly the sample. Indeed, this value takes into account the uncertainty on the

sample size, which is much bigger if the description of the sample does not quite match its actual shape;
- the values obtained in the two estimations are not consistent with each other, as a bad sample description introduces a systematic uncertainty, which also shows that the uncertainty on the rough estimation had been under-assessed in [7].

Table 1 Difference between the activities estimated with a rough description of the sample geometry using simple geometric shapes[7] and an accurate description using many solids to better approximate the object[1]. These results are obtained from the characterisation of both samples R001 & R002 at STELLA.

	^{26}Al activity (Bq/kg)	Relative uncertainty
Rough est. [7]	0.62 ± 0.07	11.3 %
Good est. [1]	0.92 ± 0.06	6.5 %
t-Student test	3.25σ	$> 1.96\sigma$ values not consistent with each other

This difference is mainly due to the uncertainty on the sample size. In fact, Table 2 shows the data used for the estimation of the uncertainty budget on the value of the efficiency, in the simplified geometry[7].

The simulation to obtain the efficiency for the 1809 keV peak has been repeated varying the sample size in the range of each length uncertainy (± 2 mm). This contribution weights for 9.4% in sample R001 and 14.3% in R002.

The same approach has been applied with the sample composition in terms of the four majority elements (Fe, O, Mg, Si): the percentage of the highest-Z element Fe has been varied by its uncertainty (0.6%) and redistributed to the other three majority elements. The composition has been obtain through EDS measurements[1]. The contribution to the uncertainty on the efficiency is 2.5% in sample R001 and 0.9% in R002.

The final uncertainty on the efficiency takes into account of these two contributions in addition to the statystical one. As the activity of ^{26}Al is computed also through the sum-peak of the 1809 keV peak and the 511 keV e^+e^- annihilation peak, its uncertainty takes also into account the uncertainty on the efficiency for the sum-peak. This uncertainty budget estimation proves that the sample size and shape plays an important role in the uncertainty of the calculated efficiencies, thus activities, and explains the unmatching values in Table 1.

However, to this extent, both the estimated values for the activity of ^{26}Al differ from Earth rocks by more than 12 standard deviations, which confirms that these samples are part of a meteorite[1, 8]. This result is obtained also with a rough description of the sample.

An even more precise estimation would be obtained by having an actual voxelised model of the sample, which can be imported in Arby/Geant4 as source geometry. The easiest way to get it is by means of a 3D laser scanner, which is the path that is currently being investigated for the future measurements.

Table 2 Variation of the simulated efficiency for the 1809 keV peak from ^{26}Al as a function of the uncertainties on the sample R001&R002 geometry.

	Sample R001	**Sample R002**
Efficiency - reference value	0.04286	0.04419

Size variation	**Sample R001**	**Sample R002**
Efficiency with lengths +2 mm [1]	0.04414	0.04419
Efficiency with lengths −2 mm [1]	0.04545	0.04835
Difference due to **size** variation	**9.4%**	**14.3%**

Composition variation	**Sample R001**	**Sample R002**
Efficiency with composition: +0.6 % Fe [2]	0.04341	0.04451
Efficiency with composition: −0.6 % Fe [2]	0.04233	0.04411
Difference due to **composition** variation	**2.5%**	**0.9%**

[1] All sample lengths modified by their uncertainty (2 mm).

[2] The amount of Fe (highest-Z) has been modified by its uncertainty (0.6%) and redistributed to the other main components (Si, O. Mg).

4 Discussion and Conclusion

The use of γ-ray spectrometry on whole samples is getting wider and wider. It enables to evaluate the specific activity of γ emitters in samples which are not to be disrupted by the measurements due to their value or cultural interest.

However, this experimental protocol complicates the estimation of efficiencies, which can only be obtained through Monte Carlo simulation in order to take into account of various effects such as the sample self-absorption. The key points in this procedure have been discussed, namely:

- the geometry of the detector has to be optimised with calibrated radioactive sources and Monte Carlo simulation in order to tune the thickness of the layers;
- the geometry of the sample must be as careful as possible in order not to introduce systematic uncertainties in the estimated activities.

In particular, the main future perspective regards the second point, where it is currently being studied the feasibility of using a voxelised version of the sample obtained by 3D laser scanning, to minimise systematic errors given by a bad description of the sample.

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Fig. 3 The bulk samples characterised in the cited previous works[1, 7]: two pieces of stony meteorite from a private collection, named R001 (mass: 12.61(36) g) and R002 (mass: 14.65(14) g).

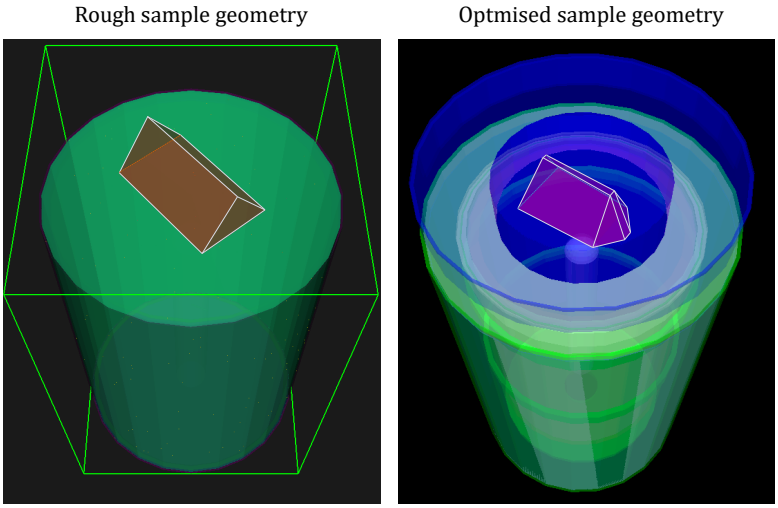

Fig. 4 The simplified geometry for sample R001[7] (left) and the multi-solid description used in the detailed simulation[1].