A methodology for the determination of the radionuclide contents and activity of samples

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Abstract

γ-Spectrometry has been used in the Department of Radiological Protection and Nuclear Safety of ITN (the Nuclear and Technological Institute, in Lisbon, Portugal) to identify and quantify radionuclides in samples from radiological monitoring of radioactive waste discharges. Real samples from radiological monitoring are characterized by a range of different geometries and heterogeneous matrices, therefore, requiring a diversified range of calibration sources to correctly calibrate the measuring system. Moreover, there are cases when due to the geometry or type of matrix, none of the system calibrations is adequate to correctly measure the sample representing a major problem in the accurate determination of samples' radionuclide concentration. Two detection systems were used, one consisting of a $3'' \times 3''$ NaI(Tl) detector and the other consisting of a HPGe5030. Both detection systems are complete by the associated shielding and calibration sources. The Monte Carlo method was used in support of a methodology to assess the radionuclide contents and activity of the real samples previously mentioned. The efficiency curves measured experimentally from three calibration sources were compared with the corresponding computational ones, obtained by Monte Carlo simulation in order to validate the method and providing the crucial tool needed to simulate any samples' geometry and matrices. The method is then used to analyze unknown samples with different matrix materials. Results from the measurements performed and their comparison with the computational results obtained are presented. Improvements in the understanding of the behavior of the experimental setup, namely the efficiency versus matrix material and geometry are explained.
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1. Introduction

One of the missions of the Department of Radiological Protection and Nuclear Safety of ITN (the Nuclear and Technological Institute, in Lisbon, Portugal) consists on monitoring the radiological hazard associated with radioactive effluents from nuclear medicine discharges, with radwaste repository sites, with NORM's releases into the environment and with contaminated scrap metal, among others. Real samples from the radiological monitoring are characterized by a range of different geometries and heterogeneous matrices, therefore, requiring multiple calibration sources to correctly calibrate the measuring system.

There are cases when due to the geometry or type of matrix, none of the calibrations sources is adequate to correctly calibrate the system. All this represents a major problem to the accurate determination of samples' radionuclide concentration mainly for the radionuclides with less energetic decay schemes.

Two detections systems were used. One consisting of a $3'' \times 3''$ NaI(Tl) detector and associated shielding. The other consisting of a HPGe5030 detector and associated shielding. Calibration sources consisting of a resin with homogeneous distribution of radionuclides, stored in a plastic container were used.

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The detailed geometry of both experimental setups (detectors, calibration sources and shielding systems) has been implemented and a full simulation of both systems was performed using the state of the art Monte Carlo simulation program MCNPX [1].

The efficiency curves measured experimentally from three calibration sources were compared with the corresponding computational ones, obtained by Monte Carlo simulation in order to validate the simulated detectors. The validation Monte Carlo code provides the crucial tool needed to simulate any samples' geometry and matrices providing the correlation between matrices composition and detection efficiency.

2. MCNPX reproduction of the experimental spectrum

The detailed geometry and material composition of the shielding and detection systems has been implemented in MCNPX.

The exact source description was considered. The different radionuclides present in the matrix and the corresponding emission lines are listed in Table 1 [2].

Table 1
Emission lines and probability present on the calibration sources

Isotope	Energy (keV)	Prob.	Isotope	Energy (keV)	Prob.
^{210}Pb	46.5	0.1177	^{85}Sr	514	0.0663
^{241}Am	59.5	0.0824	^{137}Cs	662	0.0704
^{109}Cd	88	0.0841	^{88}Y	898	0.0916
^{57}Co	122	0.0369	^{60}Co	1173	0.1323
^{139}Ce	166	0.0344	^{60}Co	1332	0.1323
^{51}Cr	320	0.0146	^{88}Y	1836	0.097
^{113}Sn	392	0.04	–	–	–

The energy resolution of the detectors was obtained from the fit of the experimental data for the different γ-energies and was considered in the Monte Carlo simulation.

3. Fitting methodology

In order to obtain an efficiency curve from a spectrum, it is necessary to assess the counts under each peak and above background. The background in the measurement facility can be due to ambient activity present in the environment and to the interaction of radiation with the surrounding materials.

The ambient background spectrum was obtained from blank measurements and then subtracted from the counting spectrum obtained for each calibration source (Fig. 1).

The contribution from the background arising from the interactions of the γ-rays in the detector and shielding materials was taken into account in the fitting algorithm. A fitting function consisting of a linear combination of a Gaussian (for each peak) and a polynomial (for the background counts) was adjusted to both the experimental and computational spectra. The fitting process was implemented using the software "Root" [3].

Several fitting methodologies have been studied.

Better results were obtained fitting a small range of energy around each peak. With this method, the number of degrees of freedom associated with the fitting function is small and the background variations can be accounted with a polynomial function of order 1 or 2.

In a first step, individual functions (polynomial and gaussian) were adjusted in predetermined energy ranges. The fitting parameters of these adjusted functions were then used as the starting parameters for another fitting process with a linear combination of a polynomial with a Gaussian. The resulting function is then fitted to the same energy range.
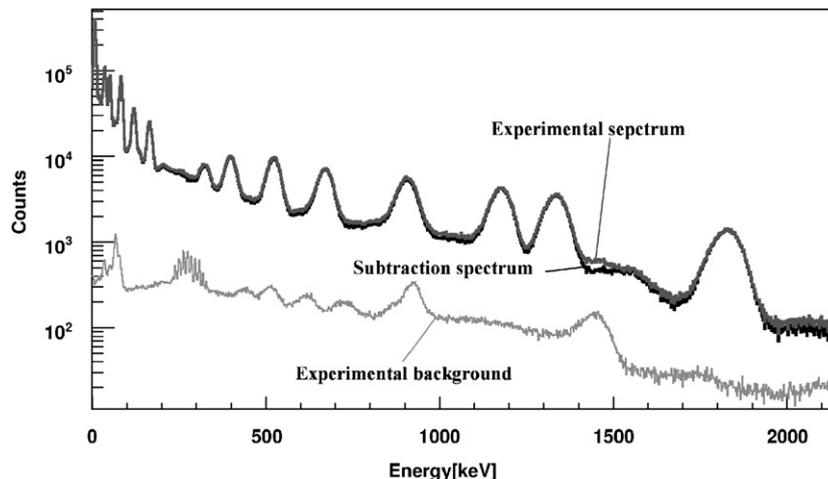

Fig. 1. Subtraction of the ambient background present in the measuring facility. Counting spectra (red), natural background (green) and the counting spectra without the natural background (blue) NaI(Tl) detector.

4. Efficiency calculation

In order to obtain the total efficiency (1), the counts in each peak must be extracted. This is done by analytical integration of the corresponding Gaussian fitted function with the parameters obtained from the last fitting step. This integral value is then divided by the number of γ's emitted from the source with that specific energy.

$$\varepsilon_{\text{total}}(E) = \varepsilon_{\text{detector}}(E)\varepsilon_{\text{geometric}}$$
$$= \frac{N_{\text{particles detected}}(E)}{N_{\text{particles emitted}}(E)}. \tag{1}$$

5. Results

As can be seen in Fig. 2, simulation and experimental spectra show a reasonable agreement despite the small energy shift present on the experimental spectra. This energy shift is presumably due to non-linearity effects in the experimental setup. Figs. 3–5 show the relative differences of the efficiency spectra obtained from the fitting procedure for the experimental and simulated spectra for all geometries using the NaI detector.

The displayed error bars account for the statistical error obtained from the fitting procedure and systematic error.

Fig. 6 show the relative differences of the efficiency spectrum obtained from the fitting procedure for the experimental and simulated spectra for the 162 ml Jar using the HPGe5030 detector.

As can be seen, a reasonable agreement exists between the computational and experimental results.

6. Conclusions

The measured efficiency curves obtained using the three calibration sources were compared with the
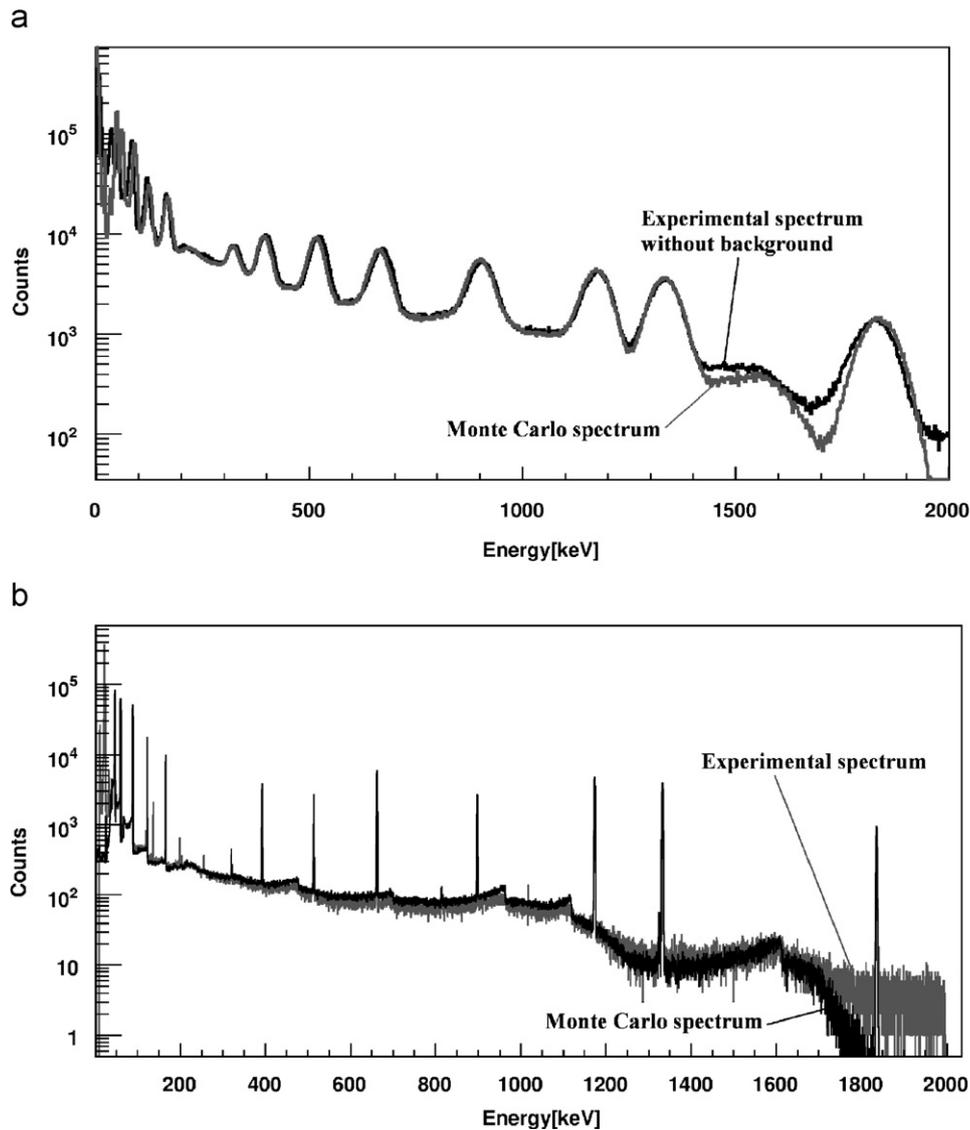

Fig. 2. Comparison between the experimental spectrum and the spectrum obtained by simulation for the calibration Jar on both detector systems (a) NaI(Tl) detector, (b) HPGe Detector.
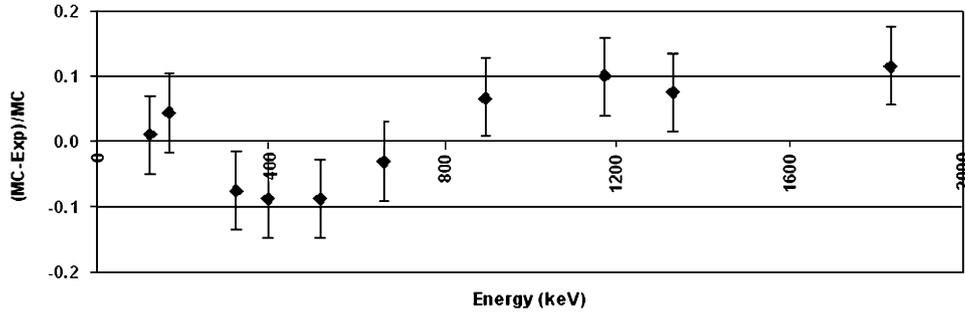
Fig. 3. Relative differences between Simulated and Experimental efficiency obtained for the 162 ml Jar (with 6% of relative error) with the NaI(Tl) 3′ × 3′ detector.
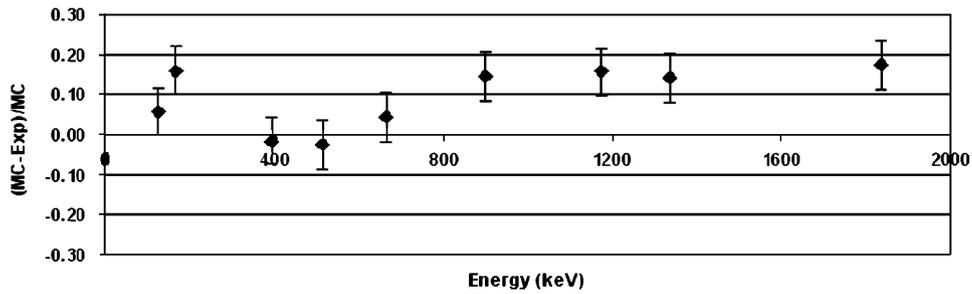

Fig. 4. Relative differences between Simulated and Experimental efficiency obtained for the 180 ml Petri (with 6% of relative error) with the NaI(Tl) 3′ × 3′ detector.
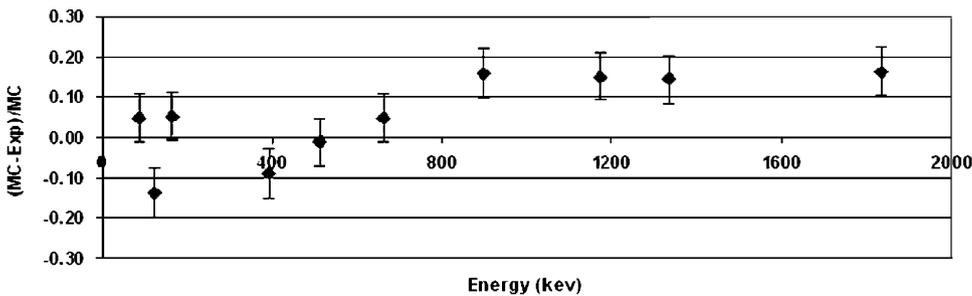

Fig. 5. Relative differences between Simulated and Experimental efficiency obtained for the 11 Marinelli (with 6% of relative error). with the NaI(Tl) 3′ × 3′ detector.
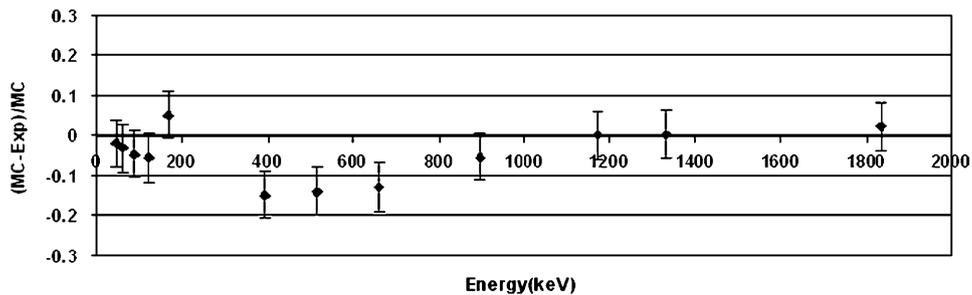

Fig. 6. Relative differences between Simulated and Experimental efficiency obtained for the 162 ml Jar (with 6% of relative error) with the HPGe5030 detector.

corresponding computational ones, obtained by Monte Carlo simulation. Results can be further improved by fine tuning the fitting methodology and detector para-meters. A detailed study of the coincidences and back-grounds must be performed in order to improve the obtained efficiency results.
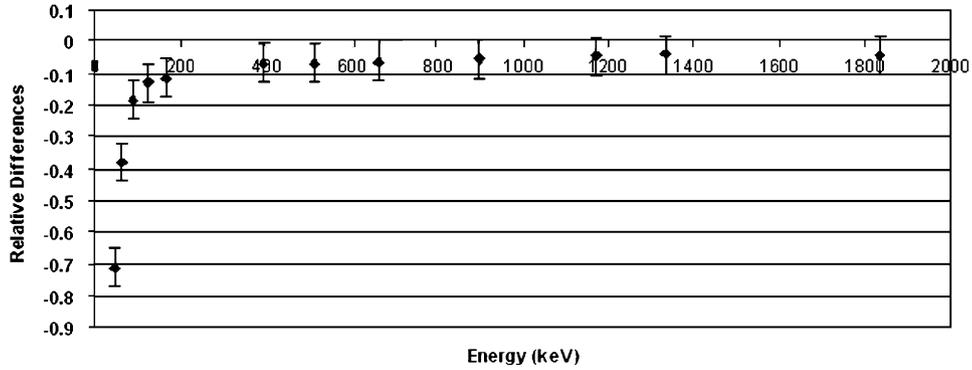
Fig. 7. Relative differences between a calibration source efficiency curve and a simulated efficiency curve for a real sample of soil obtained for the HPGe5030 detector.
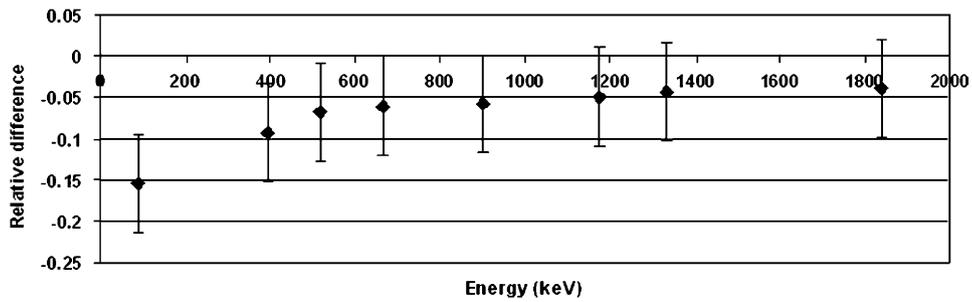

Fig. 8. Relative differences between a calibration source efficiency curve and a simulated efficiency curve for a real sample of soil obtained for the NaI(Tl) detector.

Table 2
Soil compositions used during simulations with density 1.6 g/cm^3

Isotope	Atomic fraction	Isotope	Atomic fraction	Isotope	Atomic fraction
C	0.02	K	0.019	Na	0.009
O	0.511	Ca	0.007	Ti	0.003
Al	0.046	Fe	0.013		
Si	0.37	Mg	0.002		

With the methodology described above, we are already capable of point and predict differences due to matrix composition and shapes in the radionuclide content of samples. Figs. 7 and 8 show how the determination of the content of radionuclides in a soil sample (Table 2), especially for the ones with less energetic decay schemes, can be affected by an incorrect calibration of the measuring system. The possibility of assess an efficiency curve for any geometry and matrix without a calibration source will allow to optimize and extend the use of the present measurement systems. Nevertheless, the results must still improve so it will become possible to have an accurate correlation between the existent calibration matrices and any other matrices of interest.

References

[1] Los Alamos National Laboratory, MCNPX version 2.5f.
[2] Analytics, Certificate of Calibration, Standard Source.
[3] Organisation européenne pour la recherche nucléaire, Root version 5.04/00.