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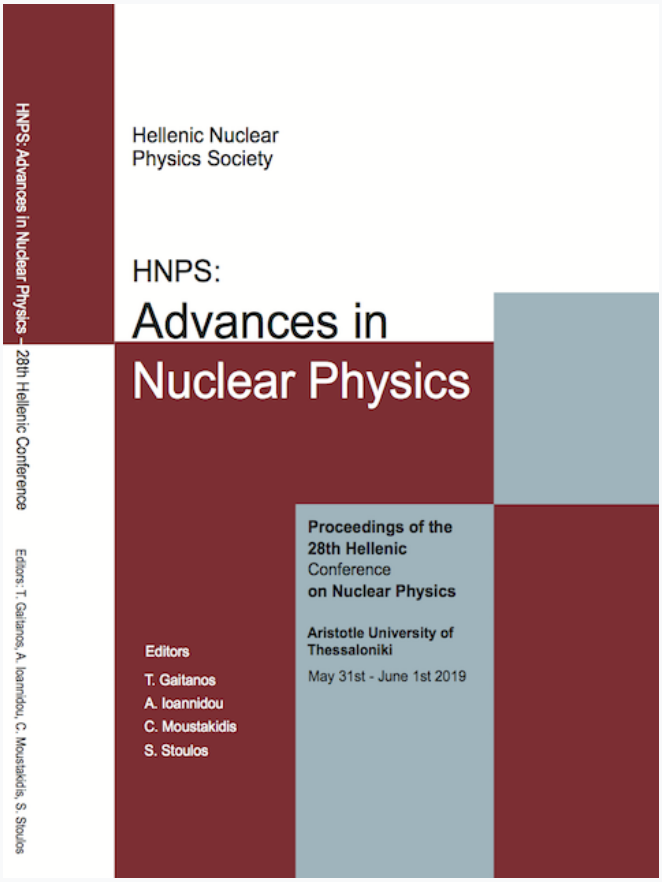

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Development of a semi-empirical calibration method by using a $LaBr_3(Ce)$ scintillation detector for NORM samples analyses

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Abstract In this study, a semi-empirical calibration method for NORM samples measurement by using a $LaBr_3(Ce)$ scintillator was developed based on a combination of experimental gamma spectrometry measurements and MCNP-X simulations. The aim of this work is to provide us with full energy peak efficiency calibration curves in a wide photon energy range which is of particular importance when selected photon energies of ^{234}Th, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{208}Tl and ^{226}Ra are to be measured with accuracy.

Keywords $LaBr_3(Ce)$, efficiency calibration, gamma spectrometry, MCNP-X

INTRODUCTION

The last decade $LaBr_3(Ce)$ scintillation detectors have become commercially available and are very promising due to their high light yield (> 65000 photons/MeV) that results in a better energy resolution compared to NaI(Tl) detectors (< 3% FWHM at ^{137}Cs), their decay time of 35 ns and their material density (5.29 g/cm^3) [1, 2]. Also, there is no need for nitrogen cooling and it is easier to be simulated comparing to HPGe detectors. Thus, $LaBr_3(Ce)$ detectors could be a suitable choice for environmental radiation monitoring [3] and in-situ measurements of NORM [4].

In this study, a semi-empirical calibration method for NORM samples measurement was developed based on a combination of experimental gamma spectrometry measurements and MCNP-X simulations. The aim of this work is to provide us with full energy peak efficiency calibration curves in a wide photon energy range which is of particular importance when selected photon energies of ^{234}Th, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{208}Tl and ^{226}Ra are to be measured with accuracy.

A Canberra scintillation detector $LaBr_3(Ce)$ (Model LABR-1.5x1.5) and four reference multi-nuclide volume sources made of epoxy material of different densities were used. Experimental efficiency calculations were performed with the volume sources adapted on an acetal holder which was positioned in a vertical direction along the detector axis of symmetry. MCNPX simulations were performed in order to evaluate the full energy peak efficiency calibration for this source-detector configuration. The models were validated by using the experimental results.

MATERIALS AND METHODS

EXPERIMENTAL SET-UP

The gamma spectrometry system consists of a Canberra scintillation detector $LaBr_3(Ce)$ (Model LABR-1.5x1.5) with a 1.5x1.5 in. crystal in a hermetically sealed aluminum housing, including a

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photomultiplier tube, an internal magnetic/light shield and a 14-pin connector (Fig.1), a digital signal processing unit (Osprey Digital Tube Base Multi Channel Analyzer), a high voltage supply system (670 V) and lead shielding structure with a thickness of 5 cm. Genie^{TM} 2000 Gamma Analysis software was used for spectrum acquisition and analysis.

The experimental data were obtained from four multiple gamma-ray large volume certified sources of different epoxy material density (0.5, 0.9, 1.5 and 2.0 g/cm^3), an active volume of 260 ml and covering of polyethylene material. The main features of the sources are shown in Table 1.

Experimental efficiency calculations were performed for ^{210}Pb, ^{241}Am, ^{137}Cs and ^{60}Co with the volume sources adapted on an acetal holder (Fig.1) which was positioned in a vertical direction along the detector axis of symmetry. The acquisition time was 4 h and the dead time less than 2%. All measurements were performed at 21 ° C.

MCNP-X simulations (Fig.1) were performed in an energy range of 46.54 – 1461 keV including the energies used in the experimental calculations for the four multiple gamma-ray large volume sources of different epoxy material density.

All compounds and pure materials were defined in the MC input file. These materials were aluminum with density of 2.7 g/cm^3, polyethylene with density of 0.91 g/cm^3, acetal with density of 1.41 g/cm^3, $LaBr_3$(Ce) crystal with density of 5.08 g/cm^3 and epoxy with a variety of densities (0.5, 0.9, 1.5 and 2.0 g/cm^3), depending on the volume source which is taken into account. The aluminum housing of the detector crystal was 0.5 mm and the concentration per weight of Cerium (Ce) was 5%, according to the Canberra manufacturer.

Fig. 1. *The $LaBr_3$(Ce) detector, the acetal holder and the MCNP-X simulation of the gamma volume source (from left to right)*

Table. 1. *Characteristics of the four multiple gamma-ray large volume sources*

Multi Volume Source Density (g/cm^3)	Isotopes	T½ (d)	Reference Activity (kBq)	Date of Calibration
0.5 ±0.1	^{210}Pb, ^{241}Am, ^{137}Cs, ^{60}Co	8140, 157753, 11001, 1924	11.3±11.4% 1.13±3.6%, 2.52±2.9%, 3.15±2.9%	1/5/2008
0.9 ±0.1	^{210}Pb, ^{241}Am, ^{137}Cs, ^{60}Co	8140, 157753, 11001, 1924	12.3±11.4% 1.23±3.6%, 2.6±3.6%, 3.25±2.9%	1/3/2007
1.5 ±0.1	^{210}Pb, ^{241}Am, ^{137}Cs, ^{60}Co	8140, 157753, 11001, 1924	12.6±11.4% 1.26±3.6%, 2.66±3.6%, 3.32±2.9%	1/3/2007
2.0 ±0.1	^{210}Pb, ^{241}Am, ^{137}Cs, ^{60}Co	8140, 157753, 11001, 1924	11.4±11.4% 1.14±3.6%, 2.54±3.6%,, 3.17±2.9%	1/5/2008

POTENTIAL SELECTION OF NORM NUCLIDES FOR ACTIVITY DETERMINATION

The spectrum of gamma rays in NORM samples comes mainly from the natural emitters of ^{40}K (1460.82 keV), products of ^{238}U decay such as ^{226}Ra (186.21 keV), ^{214}Pb (295.22, 351.93 keV), ^{214}Bi (609.31, 1120.29 keV) and products of ^{232}Th decay such as ^{228}Ac (338.32, 911.2, 969.97 keV), ^{212}Pb (238.63 keV) and ^{208}Tl (583.19 keV) [5].

The main factors that affect the NORM sample analysis by using the $LaBr_3(Ce)$ detector are: peak energy interferences, peak energy abundance, $LaBr_3(Ce)$ peak efficiency and FWHM, $LaBr_3(Ce)$ intrinsic background (Fig.2) and possible radioactive equilibrium between progenies and daughter nuclides in natural radioactive series.

The potential selection of nuclides and their corresponding energies for activity determination in NORM samples are shown in Table 2.

Table. 2. *Characteristics of selected nuclides for NORM analysis*

Norm Series	Nuclide	Peak Energies (keV)	Abundance (%)
^{238}U	^{234}Th	63.28	4.8
	^{234m}Pa	1001.03	1.021
	^{214}Pb	295.22, 351.93	18.41, 35.6
	^{214}Bi	609.31, 1120.29	45.49, 14.91
^{238}U (and ^{235}U)	^{226}Ra (and ^{235}U)	186.21	3.55 (and 57.2)
^{232}Th	^{228}Ac	911.2, 968.97	25.8, 15.8
	^{208}Tl	583.19	85

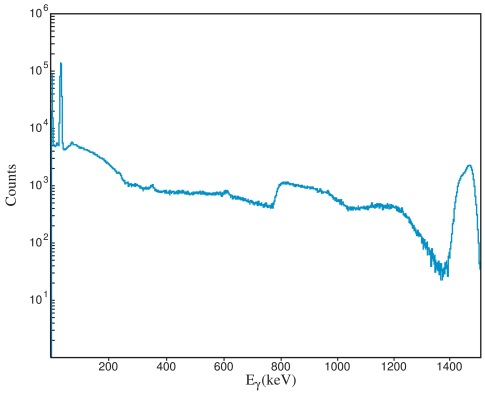

Fig. 2. *Background spectra of $LaBr_3(Ce)$: Internal contamination of ^{138}La and ^{227}Ac*

RESULTS AND DISCUSSION

In Fig. 3 and 4 the four semi-empirical efficiency calibration curves are presented in an energy range 46.54 – 1461 keV including the energies used in the experimental efficiency calculations. Black dots represent the MCNP-X data and red dots the experimental data (^{210}Pb, ^{241}Am, ^{137}Cs and ^{60}Co).

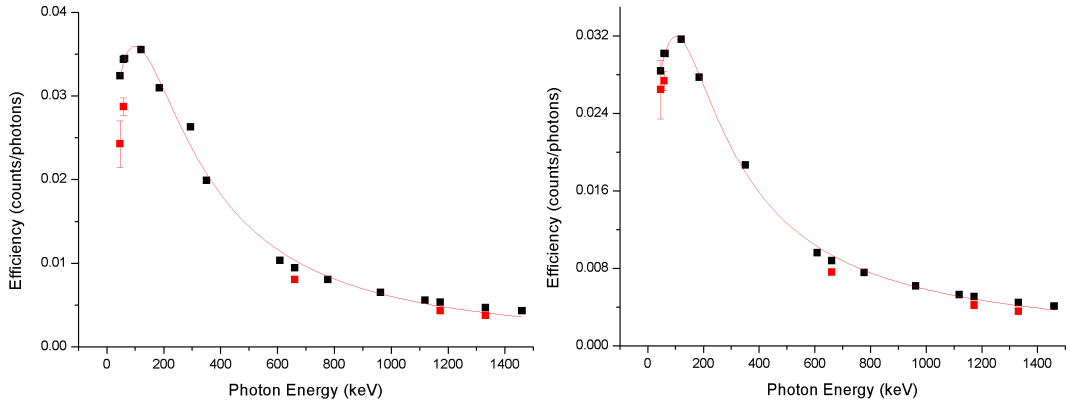

Fig. 3. *Efficiency calibration curves for the 0.5 g/cm^3(left) and 0.9 g/cm^3(right) volume source*

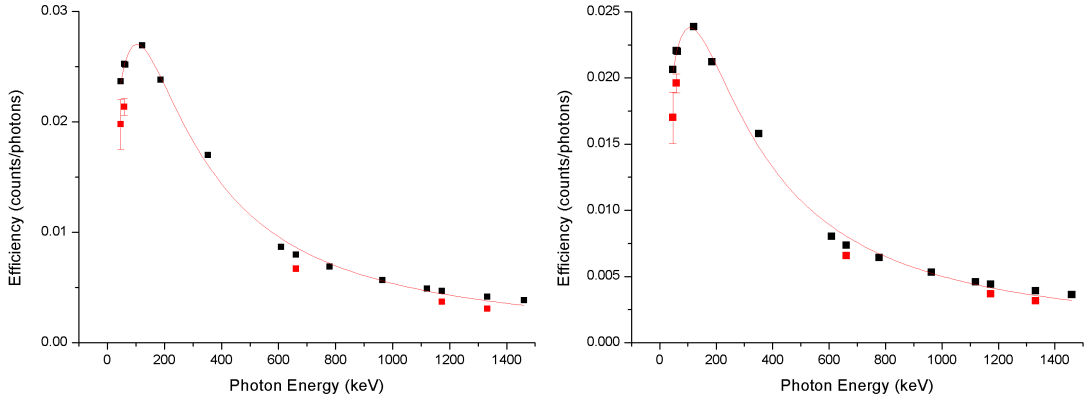

Fig. 4. *Efficiency calibration curves for the 1.5 g/cm^3(left) and 2.0 g/cm^3(right) volume source*

CONCLUSIONS

In this study a preliminary semi-empirical efficiency calibration method by using a $LaBr_3$(Ce) for NORM samples analysis was performed. In the future, the deviations between simulations and experimental data will be eliminated when the accurate dimensions of the $LaBr_3$(Ce) will be determined. The accurate NORM samples analyses will be helpful for validating the in-situ techniques which will be developed for large volume sources radiological characterization.

Acknowledgments

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