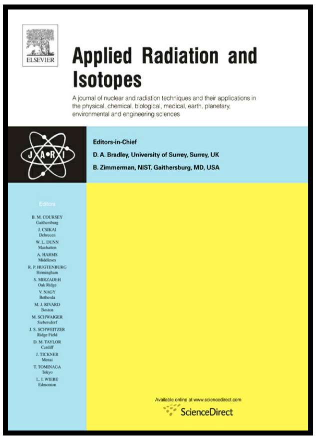

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Efficiency measurement and Monte Carlo simulations of a CeBr$_3$ scintillator

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Abstract

CeBr$_3$ crystals meet many of the demands of high performance scintillators, due to their excellent timing properties, good effective Z and high photon yield. It is important to characterize their efficiency and to verify whether modern Monte Carlo codes are reliable enough to reproduced the observed values. We report here on the measurement of both total and photopeak efficiency of a 1” diameter × 1” height CeBr$_3$ crystal for gamma-ray energies up to 1.4 MeV at several distances, using a variety of low energy gamma rays sources. The measured experimental efficiencies are compared with simulations developed in the framework of PENELOPE and GEANT4.

Keywords: CeBr$_3$, inorganic scintillators, experimental efficiency, Monte Carlo, PENELOPE simulation, GEANT4 simulation

1. Introduction

Cerium halide scintillator crystals and cerium doped lanthanum halide crystals are of strong interest in nuclear spectroscopy, medical imaging and other applications, due to their performance in terms of energy resolution and time response.

Out of these materials, the use of LaBr$_3$(Ce) has generalized in the last years due to its excellent energy resolution of the order of 3% (FWHM) at 662 keV. Its fast rise time and decay constant of the order of 16 ns leads to excellent time resolution, which can be as low as 98 ps for 1-inch in height and 1-inch in diameter cylinder crystals at ^{60}Co energies [1].

A very good alternative to LaBr$_3$(Ce) is the CeBr$_3$ scintillator [2], with very promising intrinsic properties, such as fast rise time and high photon yield, and a competitive market price [3]. Good energy resolution of about 4% at ^{137}Cs and similar time response of CeBr$_3$ to LaBr$_3$(Ce) has been reported [2]. Furthermore it has the strong advantage of not having internal activity, which can be crucial for certain low-counting or low-background applications.

Here we report on the measurement and simulation of the absolute total and photopeak efficiencies of a small CeBr$_3$ cylindrical crystal for standard calibration sources up to 1408 keV γ-rays energies. For this purpose fast Hamamatsu and Photonis photomultipliers are used.

2. Experimental details

For our tests we have used a cylindrical detector with dimensions 1-inch in height and 1-inch in diameter, commercially available from the company Scionix. The crystal is housed in 1.25 mm Al case with a thin Al entrance window of 0.8 mm, and fitted with a glass light guide in order to couple it to the photosensor. The CeBr$_3$ scintillator crystal is covered by a reflector layer of teflon in all his faces except the one equipped with the light guide. The crystal plus reflector system is embedded in a shock-absorbing material and finally by a aluminum housing where the scintillator is encapsulated. Figure 1(a) shows a schematic 2D view of the detector geometry.

The geometrical information, as provided by the manufacturer, was validated by means of a CT image acquired with and ARGUS PET/CT pre-clinical scanner [4] (Figure 1(b)), and it was incorporated in our PENELOPE and GEANT4 simulation procedures (see below).

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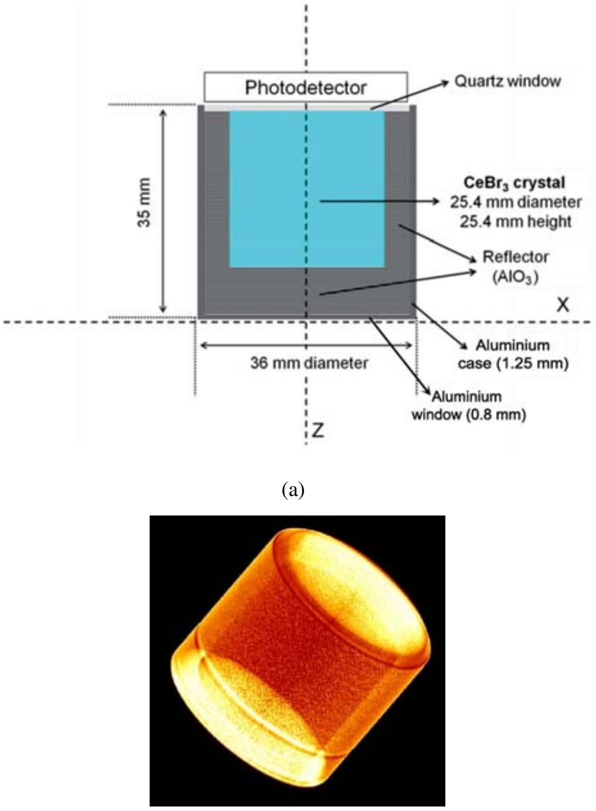

(a)

(b)

Figure 1: (a) Schematic 2D view of the detector geometry (b) 3D rendered CT image of the CeBr$_3$ detector

As photosensors we have used two different fast 8-stage photomultiplier tubes (PMTs), a XP20D0 by Photonis and a R9779 by Hamamatsu [5, 6]. Those photomultipliers are optimized for fast time response and are used to relatively high HV. Due to the very high light output from the CeBr$_3$ crystals, of the order of 45,000 photons/MeV [7], very good energy resolution is provided, but on the other hand this leads to high currents in the photomultiplier and non linear effects. The CeBr$_3$ crystal was coupled to the PMTs using silicon grease. The energy measurements were performed by using the signals from the last dynode of the PMTs.

Absolute photopeak efficiencies were measured using the calibrated sources of ^{137}Cs, ^{60}Co, ^{133}Ba and ^{152}Eu. The CeBr$_3$ detector was held in a supporting structure. The measurements were performed at three different distances from the source to the detector end-cap, namely 50, 150 and 250 mm, with an estimated error of ± 2 mm.

For the determination of the absolute efficiencies room background subtraction has been performed. The measurement live time for the background spectra was the same than for the sources. After subtraction the spectra were analysed and the full-energy peak areas were obtained by fitting to Gaussian functions for the peaks and linear (or step functions) for the background underneath.

In order to measure the total detection efficiency it is crucial to accurately record spectra down to low energies. Precise calibrations and care about non-lineary is also needed. The detectors were calibrated by using standard calibrated sources: ^{137}Cs (X-rays and γ-ray at 661.7 keV), ^{133}Ba (X-rays, 81.0, 276.4, 302.8, 356.0 and 383.8 keV), ^{22}Na (511, 1274.5 keV), ^{152}Eu (X-rays, 121.8, 244.7, 344.3, 778.9, 964.1, 1085.9, 1112.1 and 1408.0 keV) and ^{60}Co (1173.2, 1332.5 keV). Two typical spectra are shown in Figure 2 for the ^{60}Co (top) and ^{22}Na (bottom) sources.

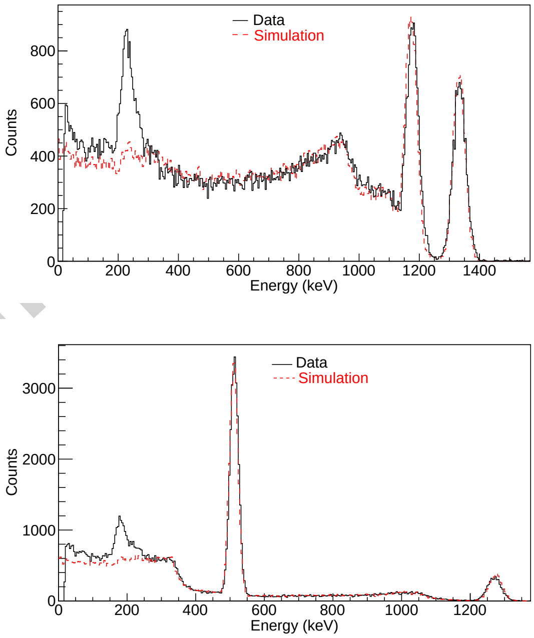

Figure 2: ^{60}Co spectrum (top panel) and ^{22}Na spectrum (bottom panel). In black the background subtracted energy spectra for sources measured with the 1-inch in height 1-inch diameter CeBr$_3$ crystal at 5-cm distance. The red dashed lines show the simulated spectra using the GEANT4 toolkit. The simulations use the nominal activity of the sources and do not include any normalization factor. The good separation for the 1173- and 1332-keV peaks in the ^{60}Co spectrum gives an idea of the energy resolution of the crystal.

The energy resolution has been obtained from our measurements. Both photomultiplier tubes yield similar results. The best combination for a good energy resolution was found for the XP20D0 photomultiplier coupled to the CeBr$_3$ crystal. Figure 3 shows the energy resolution for different γ-ray energies for both $CeBr_3$+XP20D0 and $CeBr_3$+R9779 detector configurations.

We have used an energy resolution function

$$R = a + \frac{b}{\sqrt{E}}\,, \tag{1}$$

where a and b coefficients have been determined from a fit to the experimental relative resolutions R as a function of energy E. This model can be easily implemented into the Monte Carlo simulations. The energy resolution was found to be 5.0% at 662 keV, which is higher than the quoted value of 4% given by the manufacturer, and than the reported value in [7] due to the use of fast photomultipliers.

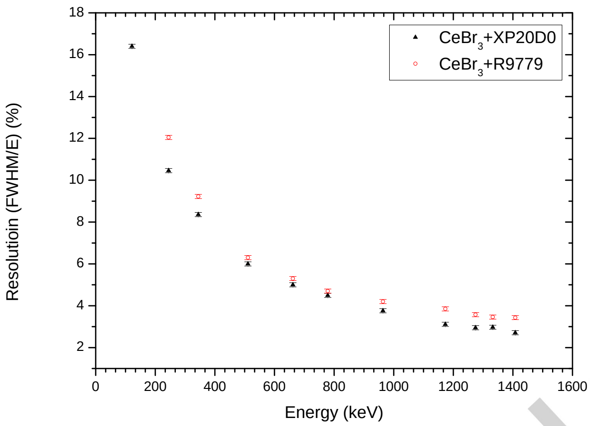

Figure 3: Energy resolution for different γ-ray energies from the standard calibrated sources listed in the text. The results for the $CeBr_3$ crystal coupled to the R9779 PMT are shown by empty circles, while for the XP20D0 PMT are represented by filled triangles.

3. Monte Carlo simulations

In order to evaluate the performance of $CeBr_3$ detectors in different scenarios, it is useful to have a complete and accurate model of the emission and detection of the radiation. Monte Carlo simulations are commonly used for this task.

There exist several Monte Carlo codes able to simulate the transport of radiation through matter, e.g. GEANT4 [8, 9], MCNP [10], EGS5 [11] and PENELOPE [12, 13] among others. We have chosen both PENELOPE (2008 version, with the standard simulation parameters, as recommended in [13]) and GEANT4 (version 10) to simulate the $CeBr_3$ photopeak and total efficiencies, together with the energy spectra. Since the geometry of the system is a key factor for the simulations we have included a detailed definition following the description given in Section 2. It contains the space coordinates of the crystal and the housing, the shock-absorbing material (included as thick polyethylene layer), and the aluminum casing of the crystal where the incident γ-rays may lose part of their energy.

3.1. PENELOPE simulations

PENELOPE is a Monte Carlo code written in the FORTRAN programming language for the simulation of the transport in matter of electrons, positrons and photons with energies from a few hundred eV to 1 GeV. It is more specialized than GEANT4, but it suits well this work needs, since it is fast and robust, and it has been extensively used for many applications. For PENELOPE simulations, we used the *penmain* program for PENELOPE [13]. This program allows the user to define the main parameters of the simulation such as geometry, materials, source model, number of showers simulated and time of simulation in a simplified manner. We have chosen a point-like source, emitting photons of the energies corresponding to the isotopes ^{22}Na, ^{137}Cs, ^{60}Co, ^{133}Ba, and ^{152}Eu. For this energy range below 1.5 MeV, the dominant interaction processes are the photoelectric absorption, coherent (Rayleigh) scattering and incoherent (Compton) scattering and (very unlikely) pair production (only for energies above 1022 keV) [14]. The emission of electrons or positrons in the disintegration of these isotopes was not taken into account, due to the fact that we assumed all these particles will be absorbed in the aluminum cover of the detector and do not reach the crystal.

The source was placed at a distance of 50, 150 and 250 mm to the center of the end cap, following the experimental setup explained in the previous section. A large number of disintegrations was simulated, in order to ensure a number of at least 10^5 interactions in the detector for each simulation case.

The energy spectra of $CeBr_3$ detector for the chosen isotopes have been generated assuming the energy resolution model discussed above (equation 1). The total energy deposited in the scintillator was also simulated. The extraction of efficiencies for sources emitting mono-energetic γ-rays is rather simple and can be obtained from the total spectrum and the photopeak areas. The uncertainties associated with the efficiencies have been obtained from the statistics of detected events, and were less than 1% in all cases. Only statistical errors have been considered.

3.2. GEANT4 simulations

GEANT4 is a simulation code written in the C++ programming language that allows the construction of different geometrical setups, the interaction between particles and the tracking of particles in media. Concerning the physics, different electromagnetic packages are available. In our case the standard physics package as well as the Penelope and the Livermore (for low energy physics) packages have been used. GEANT4 has libraries with the definition of the standard nuclear sources, i.e. including the emission of β-particles from the β-decays with experimental branching ratios and associated γ-ray intensities. We have considered these point-like sources placed at the given experimental distances. It is worth mentioning that in these simulations the point source was placed in the middle of a 1 mm thick of mylar cylinder capsule reproducing the the experimental use of an encapsulated source. The resolution of the detector have been implemented in the code following a the energy resolution model based on equation 1 fitted to the experimental values.

Figure 2 shows in red two examples of the spectra obtained from the simulations of a ^{60}Co (top) and a ^{22}Na (bottom) sources. Good agreement is found when comparing with the experimental spectra (in black), except for the region of the backscattering peak at around 200 keV. This is because in the simulations we did not include other materials around the detector such as holding structures and in particular other scintillator detectors, present at the time of the measurement. The use of the encapsulated source is justified to guarantee that the e$^+$ from the β^+ decay of the ^{22}Na nuclei annihilate with e$^-$ from the medium, which allows us to properly reproduce the intensity of the 511 keV peak in the ^{22}Na spectrum (see bottom panel of figure 2).

Once we checked that we were able to reproduce experimental spectra, and in order to optimize the run time and minimize the statistical uncertainty, mono-energetic γ-"particles" with incoming energy corresponding to those from γ-rays of the standard sources have been simulated directly, similarly to what was done in PENELOPE. The γ-events were shot isotropically with an aperture of 30° from distances of 50, 150 and 250 mm between the source and the front-end of the detector (point-like source). The deposited energy in the steps occurring in the CeBr$_3$ crystal volume is added up event by event and saved at the end of every event. The number of γ-events simulated for every energy and distance have been optimized in order to have an statistical error below 2%.

The absolute photopeak efficiency is calculated as the ratio between the detected γ-rays under the photopeak and the emitted γ-rays corrected by the solid angular given by the angular aperture. For the total efficiency we consider all γ-rays which have deposited energy in the crystal, instead of those in the photopeak. The efficiency differences between the three physics packages used are always less than 5% except for the case of 40.97 keV γ-ray energy, where differences between Livermore/Penelope and Standard libraries of 10% are observed.

4. Results and discussion

The absolute total detection efficiency has been determined for the case of the 662 keV energy from the ^{137}Cs source. Figure 4 shows the experimental spectrum at 5 cm source-detector distance. In order to measure the experimental absolute efficiency at 662 keV, the counts under the 30 keV peak (from the characteristic X-rays from the ^{137}Ba daughter) and the backscattered peak were fit, and the total counts were subtracted out from the total area under the background-subtracted ^{137}Cs spectrum. For the simulation, the GEANT4 toolkit was employed. Since we are using mono-energy γ-rays the total spectrum was just integrated. The experimental and simulation results are summarized in Table 1, showing a very good overall agreement. For completion, the photopeak efficiency is obtained by fitting the 661.6 keV photopeak and finding the total area under the peak. The results together with the GEANT4 simulations values are also provided in Table 1.

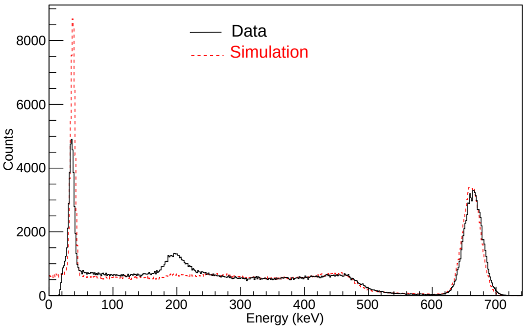

Figure 4: Background-subtracted ^{137}Cs spectrum for a 5-cm distance from the source to the 1-inch CeBr$_3$ crystal. To obtain the absolute efficiency only the contribution of the 662 keV γ-ray has been considered. For comparison a simulated spectrum using the GEANT4 toolkit is shown in dashed lines.

Given the good agreement between the experimental data and simulation for the total absolute efficiency at 662 keV, we report here values of 0.46% and 0.43%

Table 1: Total and photopeak absolute efficiencies for the $CeBr_3$ crystal at 662 keV for 5, 15 and 25 cm distances. Together with the experimentalvalues shown in column two and four, the efficiencies obtained from GEANT4 simulations are shown in columns three and five.

Distance (cm)	$\varepsilon^{exp}_{total}$ (%)	$\varepsilon^{GEANT4}_{total}$ (%)	$\varepsilon^{exp}_{photopeak}$ (%)	$\varepsilon^{GEANT4}_{photopeak}$ (%)
5	0.569±0.057	0.562	0.154±0.015	0.166
15	0.099±0.009	0.091	0.027±0.002	0.027
25	0.042±0.004	0.037	0.011±0.001	0.011

Table 2: Summary of experimental (second column) and simulated (third and fourth columns) absolute photopeak efficiencies for 15 cm distance between the detector and the standard sources for the given energies. These data are plotted in red in Figure 5.

E (keV)	$\varepsilon^{exp}_{photopeak}$ $(\cdot 10^{-3})$	$\varepsilon^{GEANT4}_{photopeak}$ $(\cdot 10^{-3})$	$\varepsilon^{Penelope}_{photopeak}$ $(\cdot 10^{-3})$
31.8	1.01(9)	1.16	0.76
81.0	1.41(12)	1.34	1.47
121.8	1.34(10)	1.32	1.46
302.8	0.77(7)	0.76	0.80
511.0	0.43(3)	0.38	0.39
778.9	0.21(2)	0.22	0.23
1173.2	0.135(11)	0.13	0.14
1274.5	0.123(10)	0.12	0.13
1332.5	0.118(10)	0.11	0.12

for the absolute total efficiency for 1173 and 1332 keV energies of the ^{60}Co source at 5 cm from the detector, based on our GEANT4 simulations.

The absolute photopeak efficiencies were measured for the γ-lines from ^{60}Co, ^{152}Eu, ^{133}Ba and ^{22}Na standard sources. The results are shown in Figure 5, where the points represent the experimental photopeak efficiencies for the three distances of 5, 15, and 25 cm, and summarized in Table 2. In the figure, solid and dashed lines show the results from GEANT4 (using the Penelope physics package) and PENELOPE simulations, respectively. A very good agreement between Monte Carlo simulations and the experimental values is found. This is specially true for the GEANT4 simulations, where the match at 15 and 25 cm is almost perfect.

Concerning PENELOPE, apart from the two lowest energies of 30.61 and 40.97 keV, the simulated efficiencies are consistent with the experimental data but they show systematically higher values than both the experimental data and the GEANT4 simulations. This effect is larger at 5 cm source-detector distance, where a mismatch of the solid angle subtended by the γ-rays would have a bigger effect.

The agreement between experiment and both Monte Carlo simulations is slightly worse at lower energies, specially at 5 cm distance, as can be seen in the inset of Figure 5, where a zoom view of the 0-400 keV energy range is shown in linear scale. This could be due to the simulations not accurately including the effective thickness of the different materials around the crystal, which has a larger effect for the absorption at lower energies and closer distances. The physics packages used in the simulations may have an impact as well.

A comparison of the efficiency of a $LaBr_3(Ce)$ detector with the same dimensions would be meaningful, since it is expected to exhibit analogous properties due to the similar atomic number and mass of lanthanum and cerium elements. In Fig. 4 of Ref. [1] the photopeak efficiencies of a 1-inch $LaBr_3(Ce)$ crystal fitted with a R9779 PMT are shown for several detector-source distances. The values at 15 cm can be directly compared to the measurements in the present work, showing very similar results. Additionally, in Ref. [15] the total and photopeak efficiencies are measured and simulated for an identical $LaBr_3(Ce)$ crystal at 662 keV and ^{60}Co with the sources on the detector surface.

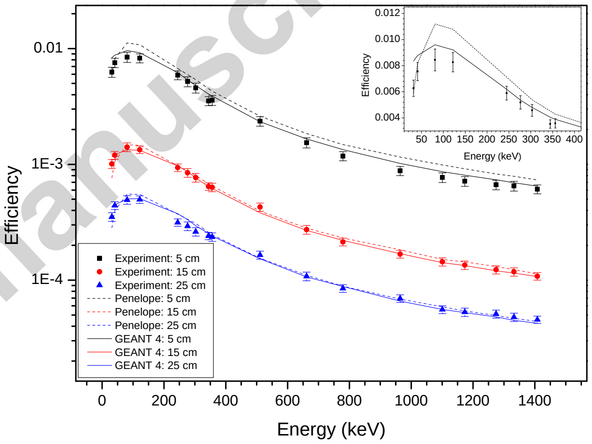

Figure 5: Absolute photopeak efficiency for 5 cm (black), 15 cm (red), and 25 cm (blue). The dots represent the experimental efficiency, the dashed line shows the results from PENELOPE simulations and the solid line from the GEANT4 simulations. The inset shows the low energy region at 5 cm distance.

5. Summary and conclussion

In this work we have evaluated the efficiency of a small cylindrical $CeBr_3$ crystal 1-inch in height and 1-inch in diameter, both experimental and by means of Monte Carlo simulations. We report on the simulations using the GEANT4 and PENELOPE platforms for the abstoule photopeak efficiencies and show that they successfully reproduce the experimental results obtained with certified sources. The simulated absolute total efficiency is also consistent with the measured values.

The validation of efficiency simutalions for simple source geometries will make it possible to broaden calculations to other geometries, such as extended sources and complex detector arrays. Together with the results described in [2] these results make $CeBr_3$ a good candidate for application to fast timing, as an competitor to $LaBr_3(Ce)$ with similar efficiency, energy resolution and and time resolution.

6. Acknowledgements

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