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Detection properties and internal activity of newly developed La-containing scintillator crystals

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ABSTRACT

In this work we will present the characterization, in terms of gamma response and internal activity of newly developed crystals that contains Lanthanum in their chemical formula. In particular we tested two $LaBr_3$:Ce,Sr, one CLLBC and two CLLB crystals with different volumes. These crystals just overcome the prototype stage and, even if the production in still not standardized at least for large optics, they have been very recently commercialized in sizes interesting for high-energy gamma-ray spectroscopy, as for application in nuclear physics experiments. In particular, we will report on the study of the decay time, light yield and energy resolution with gamma rays, on the response as a function of the gamma interaction point and on the internal activity due to the presence of Lanthanum.

1. Introduction

Since its introduction in the field of inorganic scintillator crystals in 2001, $LaBr_3$:Ce has undeniably become a reference for gamma spectroscopy applications due to its outstanding detection properties: high light yield, excellent energy resolution, fast decay time constant and a wavelength of emission well matched with standard photomultiplier tubes (PMTs), [1–14].

It was only in the latest years that a renewed effort in the material science community led to the development of some new scintillators that imposed their presence in the field as they can be considered real competitors to $LaBr_3$:Ce. In particular, we can mention $CeBr_3$ that provides detection properties approaching that of $LaBr_3$:Ce with the advantage of having no internal activity, [15–20], SrI_2:Eu that is brighter than $LaBr_3$:Ce but has a very slow decay time constant and suffer of self-absorption, [20–24] and CLYC (Cs_2LiYCl_6:Ce), a Lithium-containing elpasolite crystal, that not only provides an energy resolution better than 4.5% at 662 keV for gamma rays but is also sensitive to neutrons via the n-capture reaction on Lithium, [25–29].

In this new crystals development effort, some interest has been devoted to Lanthanum Bromide-containing scintillators, such as co-doped $LaBr_3$:Ce, CLLB ($Cs_2LiLaBr_6$:Ce) and CLLBC ($Cs_2LiLa(Br_6)90\%(Cl_6)10\%$:Ce) since, in principle, these materials have the potential to approach or even supersede the detection properties of $LaBr_3$:Ce.

Aliovalent co-doping of $LaBr_3$:Ce has been considered since few years, [30–32]. Anyway, despite the fact that the co-doping with Ca+ or Sr+ has showed a considerable improvement in the $LaBr_3$:Ce light production and an enhancement in the alpha/gamma discrimination, the availability of such crystals was still limited in quantity and size and so a large exploitation of co-doped $LaBr_3$:Ce was not started yet. As a consequence, the papers available in literature concerning the detection properties of this material, are mostly signed by the crystal producers.

The same argument is valid for CLLB and CLLBC. While the CLYC has been extensively studied, with a consequent proliferation of papers focused on CLYC detection properties and possible fields of application, the other scintillators in the elpasolite crystals family still remain less explored alternatives. In particular, CLLB showed a less effective n/γ Pulse Shape Discrimination (PSD) with respect to CLYC but it is expected to provide an improved gamma-ray energy resolution and CLLBC, due to the presence of ^{6}Li and ^{35}Cl, is sensitive to both thermal and fast neutrons, as well as to gamma rays.

In this communication we will discuss the detection properties and the internal activity of two $LaBr_3$:Ce,Sr and two CLLB crystals, recently commercialized by Saint Gobain [33], and one CLLBC scintillator, recently commercialized by Radiation Monitor Devices, Inc [34]. In this work we estimated the crystals decay time, light yield, energy resolution and internal background. In particular we are interested in the possibility to use these crystals for high-energy gamma ray spectroscopy experiments, for which large volume and homogeneity in the light yield are very crucial features.

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Table 1
The tested scintillators.

Crystal	Dimensions inches^3	Producer	Home base
CLLB (CLLB1)	⌀ 1"x1"	Saint Gobain	IPNO
CLLB (CLLB2)	⌀ 2"x2"	Saint Gobain	IPNO
CLLBC	⌀ 1"x1"	RMD	INFN-MI
$LaBr_3$:Ce,Sr	⌀ 1.5"x1.5"	Saint Gobain	IPNO
$LaBr_3$:Ce,Sr	⌀ 1.5"x1.5"	Saint Gobain	INFN-MI*
$LaBr_3$:Ce	⌀ 1"x1"	Saint Gobain	IPNO

*The $LaBr_3$:Ce,Sr is owned by Saint Gobain and temporary lent to the "INFN - Sezione di Milano" for testing.

2. Equipments and methods

In this work we compared the detection properties and the internal activity of six cylindrical-shaped Lanthanum-containing crystals. In particular we characterized two CLLB crystals with dimension of ⌀1"×1" and ⌀2"×2" (identified hereby as CLLB1 and CLLB2, respectively), one CLLBC crystal with dimensions of ⌀1"×1", two Strontium co-doped $LaBr_3$:Ce with dimension of ⌀1.5"×1.5" and one standard $LaBr_3$:Ce crystal with dimension of ⌀1"×1", for comparison. With the exception of the CLLBC, which was procured from RMD, all the other tested crystals have been supplied from Saint Gobain and, at the time of the purchase, the CLLB2 and the co-doped $LaBr_3$:Ce were the biggest commercially available optics for these scintillator materials. The characteristics of the tested crystals are summarized in Table 1.

The scintillators are encapsulated in an 0.5 mm-thick Aluminum housing to protect them from moisture, the space between the crystal and the housing is filled with diffusive material and the scintillation light is collected from a 5 mm-thick window.

The measurements presented in this work have been carried out at the "Institut de Physique Nucléaire d'Orsay" (IPNO) and at the Gamma Spectroscopy Laboratory of the "INFN - Sezione di Milano". In both laboratories, the scintillation light was read-out coupling the crystals with a high quantum efficiency, low gain-PMT from Hamamastu, the R6231-100-SEL-MOD, and a common scintillators preparation procedure was applied for the tests. In Orsay the PMT anodic signals have been collected with a 14-bit CAEN digitizer (DT5730), while in Milan a standard spectroscopic chain composed by a preamplifier, a spectroscopic amplifier (TENNELEC TC244) and a multichannel analyzer (ORTEC ASPEC MCA 926) was used for the signal collection.

3. Results and discussion

3.1. Decay time and integration time

At IPNO, in order to evaluate the CLLB and $LaBr_3$:Ce,Sr characteristic decay time constants, and thus the best suited integration time for the gamma-ray spectroscopic measurements, we irradiated the detectors with a ^{137}Cs source and we evaluated the variation of the 662 keV peak position, in terms of QDC channels, as a function of the digitizer gate length, Fig. 1. For the CLLB, the data distribution has been fitted with a double exponential decay curve, as:

$$y = A_0 + A_1(1 - e^{(-x/\tau_1)}) + A_2(1 - e^{(-x/\tau_2)}) \quad (1)$$

In addition to the fast component of 154.39 ± 0.04 ns, with a relative intensity of 63%, we observed a slow component of 1096.8 ± 0.3 ns, with a relative intensity of 37%. For the $LaBr_3$:Ce,Sr, instead, no slow component was observed; the data distribution is fitted with a single exponential decay distribution, for which we estimated a decay time constant equal to 30.80 ± 0.01 ns.

Fig. 2 shows the 662-keV FWHM-energy resolution measured as a function of the signal integration time, for the two tested crystals. For the co-doped $LaBr_3$:Ce we can observe that the energy resolution is better than 2.5% for a gate width longer than 350 ns and up to 800 ns, so we decided to use a gate of at least 500 ns when integrating the digitized signals produced with this crystal, for the gamma spectroscopy measurements. For the CLLB, the energy resolution is slightly bigger than 4%, for a gate width between 600 ns and 2 μs, i.e., when mostly the fast component is collected. Increasing the gate width up to 8 μs, the energy resolution decreases up to values bigger than 5%; in this case, the slow decay time component starts to contribute, but background noise and, possibly, pile-up events, contribute as well with a consequent degradation of the energy resolution.

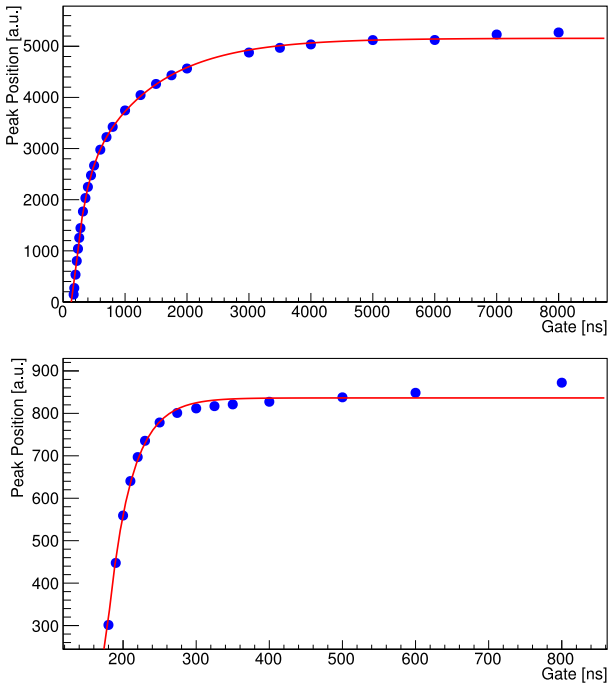

Fig. 1. 662-keV ^{137}Cs peak position as a function of the digitizer gate length for the CLLB1 (top) and the co-doped $LaBr_3$:Ce (bottom). The error bars are within the size of the markers. The red lines are the fitting functions.

3.2. Light yield

The gamma ray response, for the CLLB, the $LaBr_3$:Ce,Sr and the $LaBr_3$:Ce crystals under study, has been measured, at the IPNO, in the energy range between 60 keV – 1.7 MeV using standard gamma-ray emitting sources (^{22}Na, ^{60}Co, ^{152}Eu, ^{133}Ba, ^{207}Bi and ^{137}Cs). The PMT was operated at 800 V (corresponding to a gain of 7.2 10^4) while collecting the light of the two CLLB crystals and at 750 V (corresponding to a gain of 4.7 10^4) for the $LaBr_3$:Ce and the $LaBr_3$:Ce,Sr. For this set of measurements we used the CAEN digitizer with a gate length of 4 μs, to acquire the charge spectra for the CLLB1 and CLLB2 crystals and a gate of 500 ns for the $LaBr_3$:Ce and $LaBr_3$:Ce,Sr. For each acquired spectrum, we performed a gaussian fit on the main emission peaks to evaluate the position and the full width at half maximum (FWHM). While the statistical uncertainties associated to the light yield measurements are estimated to be smaller than 0.5%, the systematic variances associated to the measures reproducibility have been estimated to be of the order of ±2%.

The light produced, expressed in terms of number of photoelectrons (phe), is presented in Fig. 3 as a function of the irradiation energy.

It is interesting to observe that the co-doped $LaBr_3$ is considerably brighter than the standard one.

This effect is well visible in the comparison of ^{152}Eu spectra acquired with the two crystals, shown in Fig. 4. The higher light yield of the $LaBr_3$:Ce,Sr not only results in a higher number of collected photoelectrons at the PMT photocathode, for a given energy, but as well in a

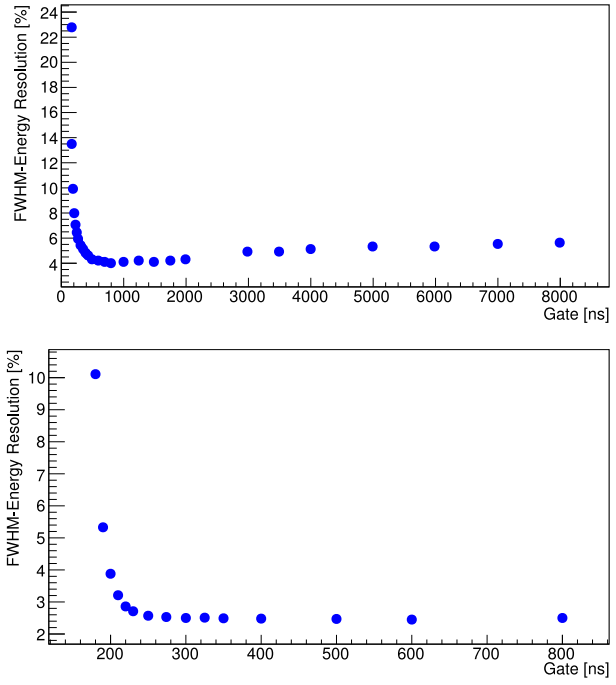

Fig. 2. 662-keV FWHM-energy resolution as a function of the digitizer gate length for the CLLB1 (top) and the co-doped $LaBr_3$:Ce (bottom). The error bars are within the size of the markers.

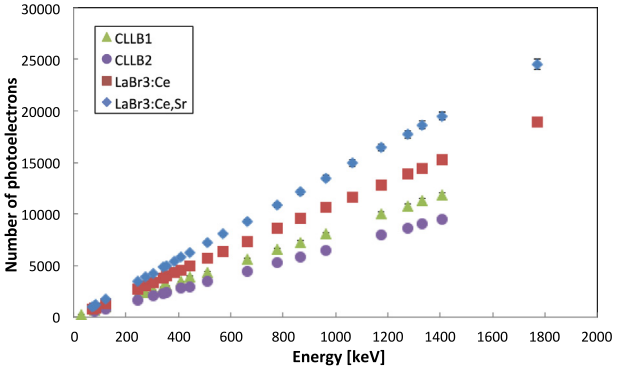

Fig. 3. Number of phe measured as a function of the irradiation energy. The error bars are within the marker size.

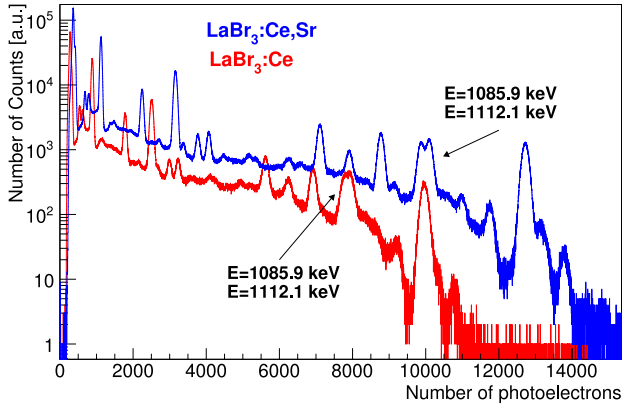

Fig. 4. ^{152}Eu spectra acquired with the $LaBr_3$:Ce (in red) and with the $LaBr_3$:Ce,Sr (in blue) . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

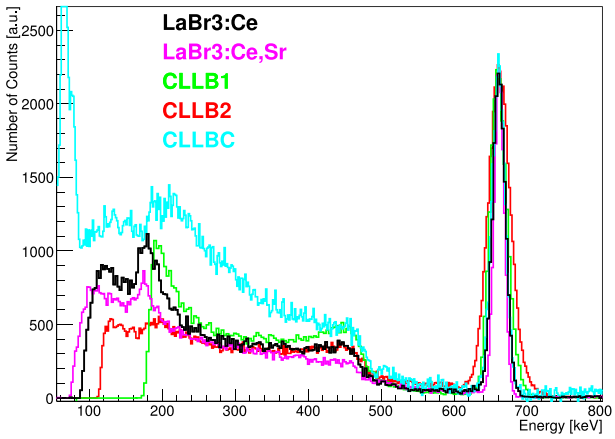

Fig. 5. ^{137}Cs spectra acquired with the crystals under study: in black the $LaBr_3$:Ce, in pink the $LaBr_3$:Ce,Sr, in green the CLLB1, in red the CLLB2 and in sky-blue the CLLBC. Being the spectra acquired with the two $LaBr_3$:Ce,Sr really similar, we just reported one, to help the readiness of the figure . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

better energy resolution leading to a better separation of close peaks (at 1085.9 keV and 1112.1 keV for example).

If we normalize the light yield to that of $LaBr_3$:Ce, used as reference, then the ratio of the light yields for the different crystals can be summarized as follow:

$LaBr_3$:Ce	$LaBr_3$:Ce,Sr	CLLB1	CLLB2
1	1.29	0.78	0.63

It also interesting to compare the two CLLB samples: for the smaller optic we collect 25% more light than for the bigger one. This can be partially explained by a loss of light at the level of the PMT/crystal interface for the CLLB2 crystal. As a matter of fact, this crystal has a surface of 2 inches in diameter thus matching the entrance window of the PMT, on which a smaller surface photocathode (⌀ 46 mm) is deposited; as a consequence, a small fraction of the scintillation light has less chance of being collected. To this effect, lower optical properties of the CLLB2 with respect to the CLLB1 may also contribute.

Performing a linear fit on the data distributions of Fig. 3 we could estimate that the deviation from linearity is less than 1% for all the tested crystals, in the investigated energy range.

3.3. The energy resolution and response uniformity

The FWHM-energy resolution was measured in the Gamma Spectroscopy Laboratory in Milan, for all the tested crystals. For this study we irradiated the scintillators in the energy range between 276 keV and 1.33 MeV and we acquired the spectra with a standard spectroscopic chain, selecting a shaping time of 2 μs for the CLLBs and for the CLLBC and 0.5 μs for the $LaBr_3$:Ce and the $LaBr_3$:Ce,Sr in order to optimize the energy resolution measurements.

The ^{137}Cs spectra, acquired with the tested crystals, are reported in Fig. 5. For the two co-doped $LaBr_3$:Ce we measured a FWHM of 15.9 ± 0.1 keV and of 17.1 ± 0.1 keV, for the peak at 662 keV, corresponding to an energy resolution of 2.5% and 2.6% for the IPNO and the "INFN-MI", respectively.

For the elpasolite crystals we measured an energy resolution of 21.7 keV, 27.5 keV and 35.7 keV at 662 keV, for the CLLBC, the CLLB1 and the CLLB2 respectively.

The energy resolution, measured as a function of the irradiation gamma energy, is presented in Fig. 6.

While the energy resolution measured with the CLLBC really approaches that of $LaBr_3$:Ce, for the CLLB1 and CLLB2 the measured values are larger than expected. In order to investigate the observed degradation for the CLLB scintillators, we studied the detectors response as a function of the interaction point along the axes for the crystals.

For this test, we scanned the detectors with a highly collimated ^{137}Cs beam along the X, Y and Z axes and we studied the variation of the centroid, the FWHM and the area of the 662 keV peak as a function of the incident radiation position. The ^{137}Cs source, providing an activity of 400 MBq, is collimated with a 8-cm

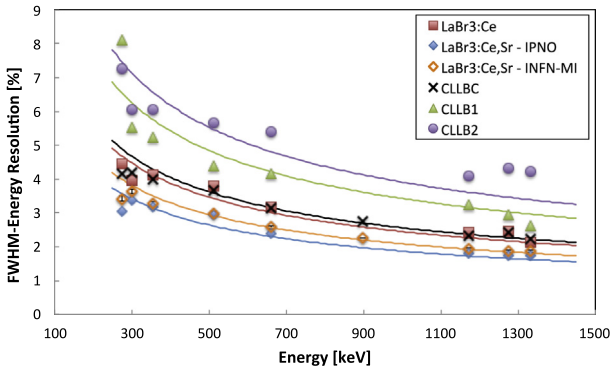

Fig. 6. FWHM-energy resolution, as a function of the irradiation energy. The continuous lines represent the trend $R \propto 1/\sqrt{E}$. The error bars are within the markers size.

thick heavy metal collimator, equipped with an exit hole of 1 mm in diameter; this set-up is placed on a support that allows rigid translations on two directions via a micrometer screw. The scintillators are then, in turn, placed on a second platform, at a fixed position, in front of the source at a distance of about 2 cm from the exit hole. A non-collimated ^{60}Co source is placed nearby and its two gamma peaks are used as calibration reference.

In Fig. 7 we present the variation of the 662 peak position for the scan that we performed on the X and Y axes along the CLLB1, CLLB2, CLLBC and $LaBr_3$:Ce,Sr round surfaces, respectively. We identified the origin of the axes as the center of the crystals front face and we performed a 2-mm step scan for the elpasolite scintillators and a 4-mm step scan for the co-doped $LaBr_3$:Ce. Imposing the energy value of 662 keV for the centroid of the ^{137}Cs peak acquired in the center of the crystal face, we then scaled accordingly the peaks acquired in different positions. While we observed a variation of the peak position that does not exceed 3 keV along the X and Y axis for the CLLB1 and the $LaBr_3$:Ce,Sr, for the other two tested crystals the variation results more pronounced, mostly at the edges, at the level of 7 keV and 9 keV for the CLLBC and the CLLB2, respectively.

The scan along the Z axis is shown in Fig. 8, in this case the origin of the axis is identified at the crystal/PMT interface and we imposed the energy value of 662 keV for the centroid of the ^{137}Cs peak acquired in the center of the crystal length. The response of the $LaBr_3$:Ce,Sr and the CLLBC is quite stable along the scintillator axis, with a deviation below 8 keV for the CLLBC response when the gamma rays are detected in the rear face of the crystal. For the CLLB crystals instead, we observed a strong response anisotropy, with the ^{137}Cs peak placed at higher ADC channels when the interaction between the gamma ray and the scintillator occurs closer to the crystal/PMT interface. We can estimate the contribution of this anisotropy to be at the level of 17 keV (2.6%) and 32 keV (4.8%) for the CLLB1 and CLLB2, respectively.

Fig. 9 presents the variation of the energy resolution, at 662 keV, as a function of the interaction point along Z, for the elpasolite crystals and for the co-doped $LaBr_3$:Ce. For the CLLB1, we observe no evident correlation between the interaction point and the measured energy resolution. For the collimated source we achieve an energy resolution between 3.1 and 3.5% at 662 keV, thus strongly competitive with other similar high-energy resolution scintillator, such as $LaBr_3$:Ce or $CeBr_3$. For the CLLB2 crystal, instead, moving the irradiation point far from the PMT entrance window, the energy resolution changes from 7% to 4.3%. The fact that we cannot achieve, for the CLLB2, an energy resolution approaching that of $LaBr_3$:Ce, even with a collimated source, might be explained by the 9 keV drift that was observed along the X and the Y axes.

3.4. Internal activity

To study the internal activity of the crystals, we placed them, in turn, in a lead box and we acquired the self-produced signals for more than 48 h. With the exception of the $LaBr_3$:Ce,Sr lent by Saint-Gobain to the "INFN - Sezione di Milano", that was placed in a 10-cm-thick lead box, the internal activity of all the other crystals has been studied using a 5-cm-thick lead box. The estimated counting rates, for the different crystals, for energies higher than 100 keV, are reported in Table 2, together with the α contribution to the total counting rate.

The acquired spectra are presented in Fig. 10. For each spectrum, we can identify the peak at ~1470 keV, due to the emission of the γ-ray at 1436 keV

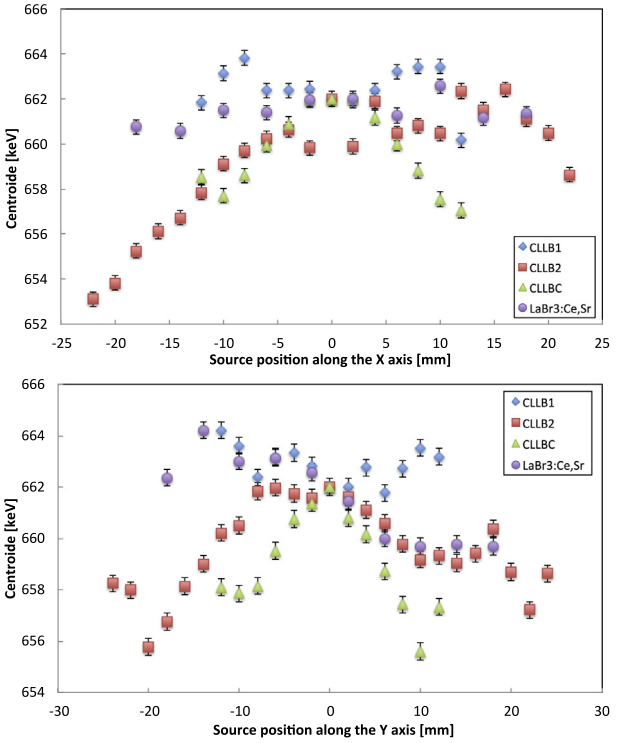

Fig. 7. ^{137}Cs peak position as a function of the interaction point along the X (top) and Y axis (bottom), for the CLLB1 (red square), CLLB2 (blue diamond), CLLBC (green triangle) and $LaBr_3$:Ce,Sr (purple round). The origin of the axes corresponds to the center of the crystal surface.

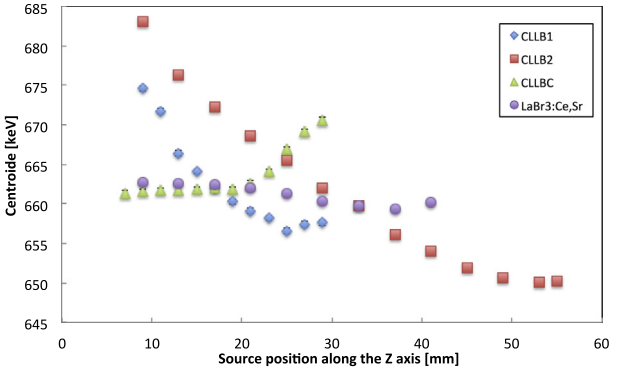

Fig. 8. ^{137}Cs peak position as a function of the interaction point along the Z axis for the CLLB1 (red square), CLLB2 (blue diamond), CLLBC (green triangle) and $LaBr_3$:Ce,Sr (purple round) . The position Z=0 corresponds to the PMT entrance window.

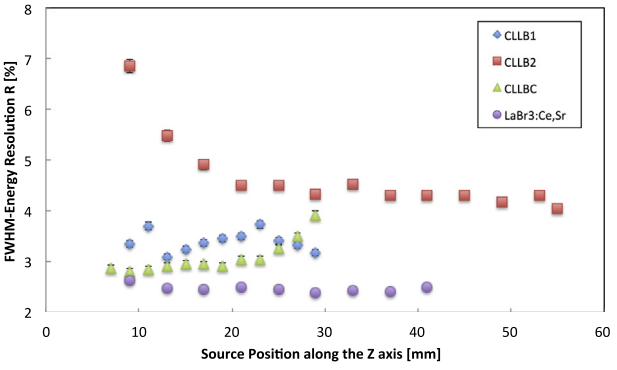

Fig. 9. Energy resolution of the ^{137}Cs peak as a function of the interaction point along the Z axis for the CLLB1 (red square), CLLB2 (blue diamond), CLLBC (green triangle) and $LaBr_3$:Ce,Sr (purple round). The position Z=0 corresponds to the PMT entrance window.

Table 2
Self activity counting rate.

Crystal	Counting rate [cts/s/cm^2]	α contribution [%]	α range [GEE MeV]
$LaBr_3$:Ce	0.47	2.79	1.7–2.7
$LaBr_3$:Ce,Sr IPNO	0.81	15.0	2.2–3.4
$LaBr_3$:Ce,Sr INFN-MI*	0.76	13.7	2.2–3.4
CLLB1	0.28	7.1	3.0–4.6
CLLB2	0.30	1.9	3.0–4.6
CLLBC	0.54	7.4	3.0–4.6

*The activity was measured placing all the crystals in a 5-cm-thick lead box with the exception of the $LaBr_3$:Ce,Sr INFN-MI that was placed in a 10-cm-thick lead box.

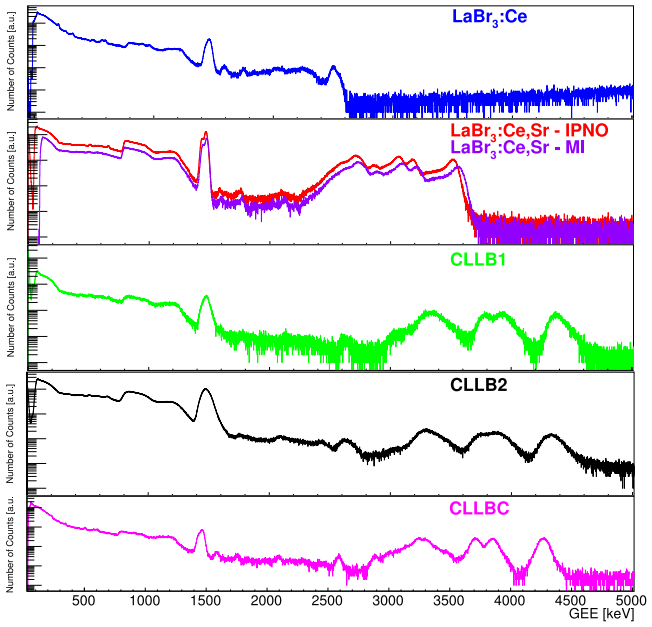

Fig. 10. The internal background for the six tested crystals, from the top: $LaBr_3$:Ce (in blue), $LaBr_3$:Ce,Sr (in red and purple), CLLB1(in green) and CLLB2 (in black), CLLBC (in pink). The spectra have been acquired placing the detectors in a lead box and acquiring self-triggered signals for more than 48 h . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

from the ^{138}La in coincidence with the K_α X-ray of ^{138}Ba, and the series of α-peaks due to the ^{227}Ac contamination. These alpha peaks are placed in different regions of the gamma equivalent energy (GEE) range, for the different investigated scintillator materials, thus indicating a different quenching factor for charged particles for the tested crystals. In particular, the alpha peaks are placed between 1.7 and 2.7 MeV-GEE for the $LaBr_3$:Ce, between 2.2 and 3.4 MeV-GEE for the co-doped $LaBr_3$:Ce and between 3.0 and 4.6 MeV-GEE for the CLLBC and the CLLB crystals, in agreement with previous works [35–38].

Fig. 11 shows the direct comparison of the background spectra of the co-doped $LaBr_3$:Ce crystal (in red the crystal owned by IPNO and in purple the crystal lent by Saint Gobain to the "INFN - Sezione di Milano") and the standard one (in blue), normalized to account for volume and acquisition time differences. In the $LaBr_3$:Ce,Sr spectrum, the superior energy resolution of this scintillator led to a clear separation between the peak at ~1440 keV, due to the 1436 keV ^{138}La γ-ray in coincidence with the L and M X-rays of ^{138}Ba, and the neighbor one at 1470 keV. Furthermore we can observe that not only the charged particle quenching factor, but also the level of the internal activity is considerably different between the two scintillator materials with, in particular, the alpha contribution being considerably higher for the $LaBr_3$:Ce,Sr. The same comparison has been performed to the background spectra of the two CLLB crystals, again normalized in terms of volume and acquisition time, Fig. 12. In this case the gamma and beta activity, due to the presence of the radioactive isotope of the Lanthanum, is higher for the bigger crystal while it is present a much lower ^{227}Ac contamination might be due to a better purification of the row materials during the crystal production process.

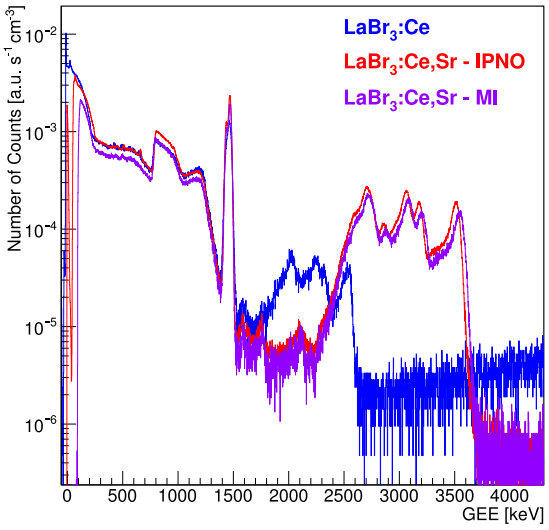

Fig. 11. The internal background spectra for $LaBr_3$:Ce,Sr (in red) $LaBr_3$:Ce (in blue), normalized to account for volume and time acquisition differences . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

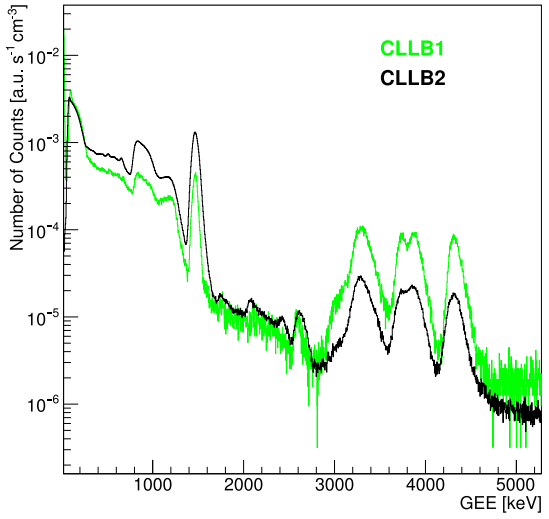

Fig. 12. The internal background of CLLB1 (in green) and CLLB2 (in black), normalized to account for volume and time acquisition differences . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Conclusions

In this work we studied the detection properties and the internal activity of three new developed La-containing scintillator crystals and, in particular, for two of the studied materials we tested the largest volume optics that were available on the market at the time of the purchase.

The two tested Co-doped $LaBr_3$:Ce showed similar excellent detection properties, superseding standard $LaBr_3$:Ce. We measured, for both samples a FWHM-energy resolution better than 2.6% at 662 keV but we observed for these scintillators the highest counting rate within the tested crystals. In particular, the alpha contribution to the total counting rate results particularly high, at the level of 14% – 15%, indicating a still high Actinium-227 contamination in the material.

For the CLLB scintillators we observed a very strong light yield anisotropy along the crystals longitudinal axes, especially for the bigger optic, with a consequent degradation of the measured energy resolution. Anyway, while for this scintillator material the observed detection properties are still not approaching those of the $LaBr_3$:Ce, the measured internal activity results considerably smaller. It is interesting to remark that for the two scintillators the total counting

rates are really similar but the α contribution is much stronger for the CLLB1 than for the CLLB2. As the $\oslash$ 2" × 2" scintillator was delivered few months later than the smaller sample, we can speculate that in the period between the production of the two crystals a more efficient purification process of the raw materials was developed. Anyway as the sources of raw materials might vary over time and the purification process is not trivial, to confirm our hypothesis more CLLB crystals should be tested.

The CLLBC scintillator tested in this work completely satisfied the high expectation we had concerning the use of this material for gamma detection. The total internal activity is close to that of $LaBr_3$:Ce and the higher α contribution provides scope for improvement in this sense if a more sophisticated purification procedure can be achieved.

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