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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 LaBr₃: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.

Keywords: Scintillator crystals, Elpasolite, Internal activity, LaBr3:Ce

1. Introduction

Since its introduction in the field of inorganic scintillator
crystals in 2001, LaBr₃: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 9 new scintillators that imposed their presence in the field as they 10 can be considered real competitors to LaBr3:Ce. In particu-11 lar, we can mention CeBr₃ that provides detection properties 12 approaching that of LaBr₃:Ce with the advantage of having 13 no internal activity, [15] - [20], SrI₂:Eu that is brighter than 14 LaBr₃:Ce but has a very slow decay time constant and suffer 15 of self-absorption, [20] - [24] and CLYC (Cs₂LiYCl₆:Ce), a 16 Lithium-containing elpasolite crystal, that not only provides an 17 energy resolution better than 4.5% at 662 keV for gamma rays 18 but is also sensitive to neutrons via the n-capture reaction on 19 Lithium, [25] - [29]. 20

In this new crystals development effort, some interest has been devoted to Lanthanum Bromide-containing scintillators, such as co-doped LaBr₃:Ce, CLLB (Cs₂LiLaBr₆:Ce) and CLLBC (Cs₂LiLa(Br₆)90%(Cl₆)10%:Ce) since, in principle, these materials have the potential to approach or even supersede the detection properties of LaBr₃:Ce.

Aliovalent co-doping of LaBr₃:Ce has been considered since few years, [30] - [32]. Anyway, despite the fact that the codoping with Ca+ or Sr+ has showed a considerable improvement in the LaBr₃: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 LaBr₃: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 ⁶Li and ³⁵Cl, 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 LaBr₃: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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2. Equipments and methods

In this work we compared the detection properties and the 58 internal activity of six cylindrical-shaped Lanthanum-containing 59 crystals. In particular we characterized two CLLB crystals with 60 dimension of $\oslash 1$ "×1" and $\oslash 2$ "×2" (identified hereby as CLLB1 61 and CLLB2, respectively), one CLLBC crystal with dimensions 62 of $\oslash 1$ "×1", two Strontium co-doped LaBr₃:Ce with dimension 63 of $\oslash 1.5" \times 1.5"$ and one standard LaBr₃:Ce crystal with dimen-64 sion of $\oslash 1" \times 1"$, for comparison. With the exception of the 65 CLLBC, which was procured from RMD, all the other tested 66 crystals have been supplied from Saint Gobain and, at the time 67 of the purchase, the CLLB2 and the co-doped LaBr3:Ce were 68 the biggest commercially available optics for these scintillator 69 materials. The characteristics of the tested crystals are summa-70 rized in table 1. 71

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 77 out at the "Institut de Physique Nucléaire d'Orsay" (IPNO) and 78 at the Gamma Spectroscopy Laboratory of the "INFN - Sezione 79 di Milano". In both laboratories, the scintillation light was read-80 out coupling the crystals with a high quantum efficiency, low 81 gain-PMT from Hamamastu, the R6231-100-SEL-MOD, and 82 a common scintillators preparation procedure was applied for 83 the tests. In Orsay the PMT anodic signals have been collected 84 with a 14-bit CAEN digitizer (DT5730), while in Milan a stan-85 dard spectroscopic chain composed by a preamplifier, a spec-86 troscopic amplifier (TENNELEC TC244) and a multichannel 87 analyzer (ORTEC ASPEC MCA 926) was used for the signal 88 collection. 89

90 3. Results and discussion

91 3.1. Decay time and integration time

At IPNO, in order to evaluate the CLLB and LaBr₃: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 ¹³⁷Cs 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.

Table 1: The tested scintillators.

Crystal	Dimensions	Producer	Home base
	inches ³		
CLLB (CLLB1)	⊘ 1"x1"	Saint Gobain	IPNO
CLLB (CLLB2)	⊘ 2"x2"	Saint Gobain	IPNO
CLLBC	⊘ 1"x1"	RMD	INFN-MI
LaBr3:Ce,Sr	⊘ 1.5"x1.5"	Saint Gobain	IPNO
LaBr3:Ce,Sr	⊘ 1.5"x1.5"	Saint Gobain	INFN-MI*
LaBr ₃ :Ce	⊘ 1"x1"	Saint Gobain	IPNO

*The LaBr₃:Ce,Sr is owned by Saint Gobain and temporary lent to the "INFN - Sezione di Milano" for testing. ⁵For the CLLB, the data distribution has been fitted with a double exponential decay curve, as:

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

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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 LaBr₃: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.



Figure 1: 662-keV ¹³⁷Cs peak position as a function of the digitizer gate length for the CLLB1 (top) and the co-doped LaBr₃:Ce (bottom). The error bars are within the size of the markers. The red lines are the fitting functions.

Fig. 2 shows the 662-keV FWHM-energy resolution mea-99 sured as a function of the signal integration time, for the two 100 tested crystals. For the co-doped LaBr₃:Ce we can observe that 101 the energy resolution is better than 2.5% for a gate width longer 102 than 350 ns and up to 800 ns, so we decided to use a gate of 103 at least 500 ns when integrating the digitized signals produced 104 with this crystal, for the gamma spectroscopy measurements. 105 For the CLLB, the energy resolution is slightly bigger than 4%, 106 for a gate width between 600 ns and $2 \mu s$, i.e., when mostly the 107 fast component is collected. Increasing the gate width up to 108 $8\,\mu$ s, the energy resolution decreases up to values bigger than 109 5%; in this case, the slow decay time component starts to con-110 tribute, but background noise and, possibly, pile-up events, con-111 tribute as well with a consequent degradation of the energy res-112 olution. 113

3.2. Light yield

The gamma ray response, for the CLLB, the LaBr₃:Ce,Sr and the LaBr₃:Ce crystals under study, has been measured, at 116



Figure 2: 662-keV FWHM-energy resolution as a function of the digitizer gate length for the CLLB1 (top) and the co-doped LaBr₃:Ce (bottom). The error bars are within the size of the markers.

the IPNO, in the energy range between 60 keV to 1.7 MeV using standard gamma-ray emitting sources (²²Na, ⁶⁰Co, ¹⁵²Eu, 118 ¹³³Ba, ²⁰⁷Bi and ¹³⁷Cs). The PMT was operated at 800 V (cor-119 responding to a gain of 7.2 10^4) while collecting the light of the 120 two CLLB crystals and at 750 V (corresponding to a gain of 121 4.7 10⁴) for the LaBr₃:Ce and the LaBr₃:Ce,Sr. For this set of 122 measurements we used the CAEN digitizer with a gate length of¹⁴⁷ 123 4 μ s, to acquire the charge spectra for the CLLB1 and CLLB2 124 crystals and a gate of 500 ns for the LaBr₃:Ce and LaBr₃:Ce,Sr. 125 For each acquired spectrum, we performed a gaussian fit on the 126 main emission peaks to evaluate the position and the full width 127 at half maximum (FWHM). While the statistical uncertainties 128 129 associated to the light yield measurements are estimated to be smaller than 0.5%, the systematic variances associated to the 130 measures reproducibility have been estimated to be of the order 131 of $\pm 2\%$. 132

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 LaBr₃ is considerably brighter than the standard one.

This effect is well visible in the comparison of ¹⁵²Eu spectra acquired with the two crystals, shown in fig. 4. The higher light yield of the LaBr₃: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 better energy resolution leading to a better separation of close peaks (at 1085.9 keV and 1112.1 keV for example).

¹⁴⁵ If we normalize the light yield to that of LaBr₃:Ce, used ¹⁴⁶ as reference, then the ratio of the light yields for the different



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



Figure 4: ^{152}Eu spectra acquired with the LaBr_3:Ce (in red) and with the LaBr_3:Ce,Sr (in blue).

crystals can be summarized as follow:

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It also interesting to compare the two CLLB samples: for 148 the smaller optic we collect 25 % more light than for the bigger 149 one. This can be partially explained by a loss of light at the 150 level of the PMT/crystal interface for the CLLB2 crystal. As a 151 matter of fact, this crystal has a surface of 2 inches in diame-152 ter thus matching the entrance window of the PMT, on which 153 a smaller surface photocathode (\oslash 46 mm) is deposited; as a 154 consequence, a small fraction of the scintillation light has less 155 chance of being collected. To this effect, lower optical prop-156 erties of the CLLB2 with respect to the CLLB1 may also con-157 tribute. 158

Performing a linear fit on the data distributions of fig.3 we 159 could estimate that the deviation from linearity is less than 1% 160 for all the tested crystals, in the investigated energy range. 161

3.3. The energy resolution and response uniformity

The FWHM-energy resolution was measured in the Gamma 163 Spectroscopy Laboratory in Milan, for all the tested crystals. 164 For this study we irradiated the scintillators in the energy range between 276 keV and 1.33 MeV and we acquired the spectra 166

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with a standard spectroscopic chain, selecting a shaping time₁₈₇ $_{167}$ of 2 μ s for the CLLBs and for the CLLBC and 0.5 μ s for the LaBr₃:Ce and the LaBr₃:Ce,Sr in order to optimize the energy resolution measurements.

The ¹³⁷Cs spectra, acquired with the tested crystals, are reported in fig. 5. For the two co-doped LaBr₃: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.



Figure 5: ¹³⁷Cs spectra acquired with the crystals under study: in black the LaBr₃:Ce, in pink the LaBr₃:Ce,Sr, in green the CLLB1, in red the CLLB2 and in sky-blue the CLLBC. Being the spectra acquired with the two LaBr₃:Ce,Sr really similar, we just reported one, to help the readiness of the figure.

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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.



Figure 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.

While the energy resolution measured with the CLLBC really approaches that of LaBr₃: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 ¹³⁷Cs beam along the X, Y and Z axes and we studied 188 the variation of the centroid, the FWHM and the area of the 662 189 keV peak as a function of the incident radiation position. The 190 ¹³⁷Cs source, providing an activity of 400MBq, is collimated 191 with a 8-cm thick heavy metal collimator, equipped with an 192 exit hole of 1 mm in diameter; this set-up is placed on a support 193 that allows rigid translations on two directions via a microme-194 ter screw. The scintillators are then, in turn, placed on a second 195 platform, at a fixed position, in front of the source at a distance 196 of about 2 cm from the exit hole. A non-collimated ⁶⁰Co source 197 is placed nearby and its two gamma peaks are used as calibra-198 tion reference. 199

In fig. 7 we present the variation of the 662 peak position 200 for the scan that we performed on the X and Y axes along the 201 CLLB1, CLLB2, CLLBC and LaBr3:Ce,Sr round surfaces, re-202 spectively. We identified the origin of the axes as the center 203 of the crystals front face and we performed a 2-mm step scan 204 for the elpasolite scintillators and a 4-mm step scan for the co-205 doped LaBr₃:Ce. Imposing the energy value of 662 keV for the 206 centroid of the ¹³⁷Cs peak acquired in the center of the crystal 207 face, we then scaled accordingly the peaks acquired in differ-208 ent positions. While we observed a variation of the peak posi-209 tion that does not exceed 3 keV along the X and Y axis for the 210 CLLB1 and the LaBr₃:Ce,Sr, for the other two tested crystals 211 the variation results more pronounced, mostly at the edges, at 212 the level of 7 keV and 9 keV for the CLLBC and the CLLB2, 213 respectively. 214



Figure 7: ¹³⁷Cs 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 LaBr₃:Ce,Sr (purple round). The origin of the axes corresponds to the center of the crystal surface.

The scan along the Z axe is shown in fig. 8, in this case the origin of the axe is identified at the crystal/PMT interface 216 and we imposed the energy value of 662 keV for the centroid 217 of the ¹³⁷Cs peak acquired in the center of the crystal length. 218 The response of the LaBr₃:Ce,Sr and the CLLBC is quite stable 219 along the scintillator axe, with a deviation below 8 keV for the 220 CLLBC response when the gamma rays are detected in the rear 221 face of the crystal. For the CLLB crystals instead, we observed 222 a strong response anisotropy, with the ¹³⁷Cs peak placed at 223 higher ADC channels when the interaction between the gamma 224 ray and the scintillator occurs closer to the crystal/PMT inter-225 face. We can estimate the contribution of this anisotropy to be 226 at the level of 17 keV (2.6%) and 32 keV (4.8%) for the CLLB1 227 and CLLB2, respectively. 228



Figure 8: ¹³⁷Cs 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 LaBr₃:Ce,Sr (purple round). The position Z=0 corresponds to the PMT entrance window. 253

Fig. 9 presents the variation of the energy resolution, at 662 229 keV, as a function of the interaction point along Z, for the elpa-230 solite crystals and for the co-doped LaBr₃:Ce. For the CLLB1, 231 we observe no evident correlation between the interaction point 232 and the measured energy resolution. For the collimated source 233 we achieve an energy resolution between 3.1 and 3.5% at 662 234 keV, thus strongly competitive with other similar high-energy 235 resolution scintillator, such as LaBr3:Ce or CeBr3. For the 236 CLLB2 crystal, instead, moving the irradiation point far from 237 the PMT entrance window, the energy resolution changes from 238 7% to 4.3%. The fact that we cannot achieve, for the CLLB2, 239 an energy resolution approaching that of LaBr₃:Ce, even with 240 a collimated source, might be explained by the 9 keV drift that 241 was observed along the X and the Y axes. 242

243 3.4. Internal activity

To study the internal activity of the crystals, we placed them, 244 in turn, in a lead box and we acquired the self-produced signals 245 for more than 48h. With the exception of the LaBr₃:Ce,Sr lent 246 by Saint-Gobain to the "INFN - Sezione di Milano", that was 247 placed in a 10-cm-thick lead box, the internal activity of all the 248 other crystals has been studied using a 5-cm-thick lead box. The 249 estimated counting rates, for the different crystals, for energies 250 higher than 100 keV, are reported in table 2, together with the 251 α contribution to the total counting rate. 252



Figure 9: Energy resolution of the ¹³⁷Cs peak as a function of the interaction point along the Z axis for the CLLB1 (red square), CLLB2 (blue diamond), CLLBC (green triangle) and LaBr₃:Ce,Sr (purple round). The position Z=0 corresponds to the PMT entrance window.

Table 2: Self activity counting rate.

Crystal	Couting Rate	lpha contribution	α range
	[cts/s/cm ²]	[%]	[GEE MeV]
LaBr ₃ :Ce	0.47	2.79	1.7 - 2.7
LaBr3:Ce,Sr IPNO	0.81	15.0	2.2 - 3.4
LaBr3: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 of the LaBr₃:Ce,Sr INFN-MI that was placed in a 10-cm-thick lead box.

The acquired spectra are presented in fig. 10. For each spectrum, we can identify the peak at \sim 1470 keV, due to the emis-254 sion of the γ -ray at 1436 keV from the ¹³⁸La in coincidence 255 with the K_{α} x-ray of ¹³⁸Ba, and the series of α -peaks due to the 256 ²²⁷Ac contamination. These alpha peaks are placed in differ-257 ent regions of the gamma equivalent energy (GEE) range, for 258 the different investigated scintillator materials, thus indicating 259 a different quenching factor for charged particles for the tested 260 crystals. In particular, the alpha peaks are placed between 1.7 261 and 2.7 MeV-GEE for the LaBr3:Ce, between 2.2 and 3.4 MeV-262 GEE for the co-doped LaBr₃:Ce and between 3.0 and 4.6 MeV-263 GEE for the CLLBC and the CLLB crystals, in agreement with 264 previous works ([35] - [38]). 265

Fig. 11 shows the direct comparison of the background 266 spectra of the co-doped LaBr₃:Ce crystal (in red the crystal 267 owned by IPNO and in purple the crystal lent by Saint Gob-268 ain to the "INFN - Sezione di Milano") and the standard one (in 269 blue), normalized to account for volume and acquisition time 270 differences. In the LaBr₃:Ce,Sr spectrum, the superior energy 271 resolution of this scintillator led to a clear separation between 272 the peak at \sim 1440 keV, due to the 1436 keV ¹³⁸La γ -ray in co-273 incidence with the L and M x-rays of ¹³⁸Ba, and the neighbor 274 one at 1470 keV. Furthermore we can observe that not only the 275 charged particle quenching factor, but also the level of the in-276 ternal activity is considerably different between the two scintil-277 lator materials with, in particular, the alpha contribution being 278 considerably higher for the LaBr₃:Ce,Sr. The same comparison 279



Figure 10: The internal background for the six tested crystals, from the top: LaBr₃:Ce (in blue), LaBr₃: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 hours.

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 ²²⁷Ac contamination might be due to a better purification of the row materials during the crystal production process.

287 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 LaBr3:Ce showed similar excel-293 lent detection properties, superseding standard LaBr3:Ce. We 294 measured, for both samples a FWHM-energy resolution better 295 than 2.6% at 662 keV but we observed for these scintillators 296 the highest counting rate within the tested crystals. In particu-297 lar, the alpha contribution to the total counting rate results par-298 ticularly high, at the level of 14-15%, indicating a still high 299 Actinium-227 contamination in the material. 300

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 LaBr₃:Ce, the measured internal activity re-



Figure 11: The internal background spectra for LaBr₃:Ce,Sr (in red) LaBr₃:Ce (in blue), normalized to account for volume and time acquisition differences.



Figure 12: The internal background of CLLB1 (in green) and CLLB2 (in black), normalized to account for volume and time acquisition differences.

sults considerably smaller. It is interesting to remark that for the two scintillators the total counting rates are really similar 308 but the α contribution is much stronger for the CLLB1 than for 309 the CLLB2. As the \oslash 2" \times 2" scintillator was delivered few 310 months later than the smaller sample, we can speculate that in 311 the period between the production of the two crystals a more 312 efficient purification process of the raw materials was devel-313 oped. Anyway as the sources of raw materials might vary over 314 time and the purification process is not trivial, to confirm our 315 hypothesis more CLLB crystals should be tested. 316

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 LaBr₃:Ce and the higher α contribution provides scope for improvement in this sense if a more sophisticated purification procedure can be achieved. 322

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References 328

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- 329 [1] E.V.D. van Loef, et al., "High-energy-resolution scintillator: Ce3+ activated LaBr₃", Appl. Phys. Lett. Vol. 79 Is. 10, pp. 1573-1575 (2001) 330
- C.W.E. van Eijk et al., "Energy resolution of some new inorganic-331 [2] scintillator gamma-ray detectors" Rad. Meas. Vol. 33 Is. 5, pp. 521-525 332 (2001)333
- [3] E.V.D. van Loef et al., "Scintillation properties of LaBr₃:Ce3+ crystals: 334 fast, efficient and high-energy-resolution scintillators", Nucl. Inst. and 335 Meth. A, Vol. 486 Is. 1-2, pp. 254-258 (2002) 336
- 337 [4] W.W. Moses, "Current trends in scintillator detectors and materials", Nucl. Inst. and Meth. A, Vol. 487 Is. 1-2, pp. 123-128 (2002) 338
 - K.S. Shah et al., "LaBr3:Ce scintillators for gamma ray spectroscopy", [5] IEEE Trans. Nucl. Sci., Vol. 50 Is. 6, pp. 2410-2413 (2003)
 - K.S. Shah et al., "High energy resolution scintillation spectrometers", [6] IEEE Trans. Nucl. Sci., Vol. 51 Is. 5, pp. 2395-2399 (2004)
 - J. Glodo et al., "Effects of Ce concentration on scintillation properties of [7] LaBr3:Ce", IEEE Trans. Nucl. Sci., Vol. 52 Is. 5, pp. 1805-1808 (2005)
 - G. Bizzarri et al., "Scintillation properties of circle divide 1x1 inch(3) [8] LaBr3:5%Ce3+ crystal", IEEE Trans. Nucl. Sci., Vol. 53 Is. 2, pp. 615-619 (2006)
 - [9] A. Iltis et al., "Lanthanum halide scintillators: Properties and applications", Nucl. Inst. and Meth. A, Vol. 563, pp. 359-363 (2006)
 - [10] M. Moszynskia et al., "New Photonis XP20D0 photomultiplier for fast timing in nuclear medicine", Nucl. Inst. and Meth. A, Vol. 567 Is. 1, pp. 31-35 (2006)
- [11] F. Camera et al., "Investigation of the self activity and high energy 353 gamma-rays response of a 1" x 1" and 3" x 3" LaBr3:Ce scintillators' 354 2007 IEEE-NSS Conf. Rec., Vol.1-11, pp. 1386 355
- [12] F. Quarati et al., "X-ray and gamma-ray response of a 2" x 2" LaBr3:Ce 356 scintillation detector", Nucl. Inst. and Meth. A, Vol. 574 Is. 1, pp. 115-120 357 358 (2007)
- [13] P.R. Menge et al., "Performance of large lanthanum bromide scintilla-359 360 tors", Nucl. Inst. and Meth. A, Vol. 579 Is. 1, pp. 6-10 (2007)
- 361 [14] R. Nicolini et al., "Investigation of the properties of a 1" x 1" LaBr3:Ce scintillator", Nucl. Inst. and Meth. A, Vol. 582 Is. 2, pp. 554-561 (2007) 362
- [15] K.S. Shah et al., "CeBr3 Scintillators for Gamma-Ray Spectroscopy", 363 IEEE Trans. Nucl. Sci., Vol. 52 Is. 6, pp. 3157-3159 (2005) 364
- [16] R. Sejin et al., "Luminescence and scintillation properties of a CeBr₃ sin-365 gle crystal", IEEE Trans. Nucl. Sci., Vol. 55 Is. 3, pp. 1221-1224 (2008) 366
- [17] P. Guss et al., "CeBr3 as a room-temperature, high-resolution gamma-ray 367 detector", Nucl. Inst. and Meth. A, Vol. 608 Is. 2, pp. 297-304 (2009) 368
- [18] L.M. Fraile et al., "Fast timing study of a CeBr₃ crystal: Time resolution 369 below 120 ps at Co-60 energies", Nucl. Inst. and Meth. A, Vol. 701, pp.
- 370 235-242 (2013) 371
- [19] F. Quarati et al., "Scintillation and detection characteristics of high-372 sensitivity CeBr3 gamma-ray spectrometers", Nucl. Inst. and Meth. A, 373 Vol. 729, pp. 596-604 (2013) 374
- [20] A. Giaz et al., "Prelinimary investigation of scintillator materials prop-375 erties: SrI2:Eu, CeBr3 and GYGAG:Ce for gamma rays up to 9 MeV", 376 Nucl. Inst. and Meth. A, Vol. 804, pp. 210-220 (2015) 377
- [21] N.J. Cherepy et al., "Strontium and barium iodide high light yield scintil-378 lators", App. Phys. Lett., Vol. 92 Is. 8, 083508 (2008) 379
- N.J. Cherepy et al., "Scintillators with potential to supersede Lanthanum 380 [22] Bromide", IEEE Trans. Nucl. Sci., Vol. 56 Is. 3, pp. 873-880 (2009) 381
- 382 [23] B.W. Sturm et al., "Evaluation of Large Volume SrI2(Eu) Scintillator Detectors", 2010 IEEE-NSS Conf. Rec., pp. 1607-1611 383
- M.S. Alekhin et al., "Scitillation Properties of and Self Absorption in 384 [24] SrI₂:Eu2+", IEEE Trans. Nucl. Sci., Vol. 58 Is. 5, pp. 2519-2527 (2011) 385
- [25] J. Glodo et al., "Scintillation properties of 1 inch Cs(2)LiYCl(6): Ce crys-386 tals", IEEE Trans. Nucl. Sci., Vol. 55 Is. 3, pp. 1206-1209 (2008) 387
- 388 [26] J. Glodo et al., "Cs2LiYCl6:Ce Scintillator for Nuclear Monitoring Applications", IEEE Trans. Nucl. Sci., Vol. 56 Is. 3, pp. 1257-1261 (2009) 389

- 390 32{27] J. Glodo et al., "Selected Properties of Cs2LiYCl6, Cs2LiLaCl)6, and Cs₂LiLaYBr₆ Scintillators", IEEE Trans. Nucl. Sci., Vol. 58 Is. 1, pp. 333-338 (2011)
- H2020-INFRAIA-2014-2015 Grant. Agreement 654002 ENSAR2^[28] D.W.Lee et al., "Pulse-shape analysis of Cs₂LiYCl₆:Ce scintillator for neutron and gamma-ray discrimination", Nucl. Inst. and Meth. A, Vol. 664 Is.1, pp. 1-5 (2012)
 - [29] A. Giaz et al., "The CLYC-6 and CLYC-7 response to gamma-rays, fast and thermal neutrons", Nucl. Inst. and Meth. A, Vol. 810, pp. 132-139 (2016)
 - [30] M.S. Alekhin et al.,"Improvement of gamma-ray energy resolution of LaBr₃:Ce3+ scintillation detectors by Sr2+ and Ca2+ co-doping". App. Phys. Lett., Vol. 102 Is. 16, 161915 (2013)
 - [31] M.S. Alekhin et al., "Improvement of LaBr3:5%Ce scintillation properties by Li+, Na+, Mg2+, Ca2+, Sr2+, and Ba2+ co-doping", Journ. App. Phys., Vol. 113 Is. 22, 224904 (2013)
 - [32] A. Benedetto et al., "The effect of LaBr3:Ce single crystal aliovalent codoping on its mechanical strength", Nucl. Inst. and Meth. A, Vol. 784, pp. 17-22 (2015)
 - [33] https://www.crystals.saint-gobain.com/products/crystal-scintillationmaterials
 - [34] https://www.dynasil.com/company/rmd
 - [35] F. Camera et al., "Investigation of the self activity and high energy γ -rays response of a 1" x 1" and 3" x 3" LaBr3:Ce scintillators", 2007 IEEE-NSS Conf. Rec., N24-186, pp.1386-1388
 - [36] A.F. Iyudin et al., "Peculiarities of Intrinsic Background in LaBr3:Ce and CeBr3 Scintillating Crystals", Inst. and Exp. Techn., Vol. 52, Is. 6, pp.774-781 (2009)
 - [37] R.S. Woolf et al., "Characterization of the internal background for thermal and fast neutron detection with CLLB", Nucl. Inst. and Meth. A, Vol. 838, pp. 147-153 (2016)
 - [38] K. Yang et al., "Enhanced α - γ discrimination in co-doped LaBr₃:Ce", IEEE Trans. Nucl. Sci., Vol. 63 Is. 1, pp. 416-421 (2016)

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