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Lanthanum halide scintillators: Properties and applications

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Abstract

BrilLanCe®-350 and BrilLanCe®-380, Saint-Gobain Crystals' trade-names for $LaCl_3$:Ce and $LaBr_3$:Ce are being brought to market under exclusive license to Delft and Bern Universities. We are reporting the properties of crystals produced with commercially viable processes and find they match others' observations. These scintillators are bright (60,000 photons/MeV for $LaBr_3$:Ce) and have very linear response, a combination that leads to very good energy resolution (<3% at 662 keV and about 6% at 122 keV for $LaBr_3$:Ce). The materials also have fast scintillation decay times (<30 ns) which supports counting applications at very high rate. These fast light output properties also lead to very fast timing (<300 ps coincidence resolving time on 30 mm long pieces of $LaBr_3$:Ce). These excellent properties are retained at high temperature with only moderate light loss (<10%) at 175 °C in both materials. The detectors carry natural background in La^{138} and Ac^{227}, the latter having been substantially reduced in recent processing. BrilLanCe®-350 is now available in detectors up to 51 mm diameter while 38 mm diameter is available for BrilLanCe®-380. Larger sizes are expected.

Keywords: Fast timing; $LaCl_3$; $LaBr_3$; Scintillators; Energy resolution; X-rays

1. Introduction

In 2000 and 2001 two families of anhydrous halide scintillators were reported and extensively studied [1–6] Saint-Gobain Crystals is bringing two of them to market, $LaCl_3$:Ce and $LaBr_3$:Ce with trade names BrilLanCe®-350 and BrilLanCe®-380 thanks to an exclusive license to the inventors, Delft and Bern Universities.

The early reports on small crystals of $LaBr_3$:Ce were very attractive: light yield of 61,000 photons/MeV, energy resolution of 3% (FWHM) at 662 keV, density of 5.3 g/cm^3 combined with a decay time of 35 ns and no intense slow component all superior to other widely used scintillators like NaI:Tl^+. The comparative properties are summarized in Table 1.

One purpose of this article is to report that these attractive properties are retained in larger crystals produced with commercially scalable processes. There do not appear to be deleterious effects from self-absorption or inhomogeneities at least to 75 mm crystal sizes.

Exceptional energy resolution naturally leads to application of those new crystals in γ-ray spectroscopy [7], a use we show can be extended to at least 175 °C a point of interest for application to geophysical well logging and measurement while drilling. Some very promising results have also been reported on timing for PET applications [8]. These properties coupled with the availability of big pieces in BrilLanCe®-350 could be of interest for physics applications.

The fast decay time aids high rate detection. We have optimized BrilLanCe®-350 detectors for low-energy X-rays and find the energy resolution at 5.9 keV is close to that with NaI:Tl^+, but much higher counting rates can be reached.

Making these crystals commercially available has required addressing several issues:

- $LaBr_3$:Ce is more hygroscopic than NaI:Tl^+ .
- The new crystals have a hexagonal crystal structure with considerable anisotropy in properties like thermal expansion between the *c* and *a* directions. This also renders them birefringent.

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Table 1
Comparison of properties of BrilLanCe®-350, BrilLanCe®-380 and NaI:Tl$^+$

	NaI:Tl$^+$	$LaCl_3$:Ce BrilLanCe®-350	$LaBr_3$:Ce BrilLanCe®-380
Density (g/cm^3)	3.67	3.79	5.29
Decay time (ns)	240	28	26
Light out (ph/MeV)	39 000	46 000	63 000
E_{res} at 662 keV	7%	<4%	<3%
Wavelength (nm)	415	350	380

- $LaBr_3$:Ce has a relatively weak (1 0 0) cleavage plane, which makes crystal growth tricky.

2. Doping, emission spectra, index of refraction

All measurements herein are for $LaCl_3$:10%Ce and $LaBr_3$:5%Ce, unless noted otherwise.

We find $LaCl_3$:10%Ce has an emission spectrum peaking at 350 nm, whence the designation BrilLanCe®-350, and likewise $LaBr_3$:5%Ce peaks at 380 nm from which the designation BrilLanCe®-380 is derived. These emission spectra allow use of standard faceplate photomultipliers (PMTs) without need of fused silica windows.

We do not yet have a direct measurement of index of refraction for BrilLanCe®-380 ($LaBr_3$:Ce), due to the hygroscopic nature of the crystal. We have estimated the refractive index from the dielectric function calculated with the Wien2K program package (Institute for Materials Chemistry, TU Vienna, Austria) using an all-electron scheme including relativistic effects. A gap correction to fit experimental data was performed in order to account for the typical error on the gap width made by DFT. Calculations were made at the Jean Rouxel Institute of Materials, Nantes, France. The results are shown as a function of wavelength in Fig. 1.

Also of interest is the change in refractive index as a function of Ce content which is summarized in the following two equations for a and c-axes at 380 nm:

$$n_a = 2.040 + [\mathrm{Ce}] \times 0.034$$

$$n_c = 2.074 + [\mathrm{Ce}] \times 0.082.$$

Typically, cylindrical detectors are mounted with the cylinder's axis along the crystallographic c-axis.

3. Performance—spectroscopy

Energy resolution results seen in others' work in smaller crystals are at least preserved in detectors 38 mm in diameter and 38 mm long as seen in Table 2 and Fig. 2 below. The clear separation of the 136 keV line for Co^{57} is noteworthy. Measurements were taken over 2048 channels with 1 μs time constants on a Photonis XP2060B. These are best results, not average ones.

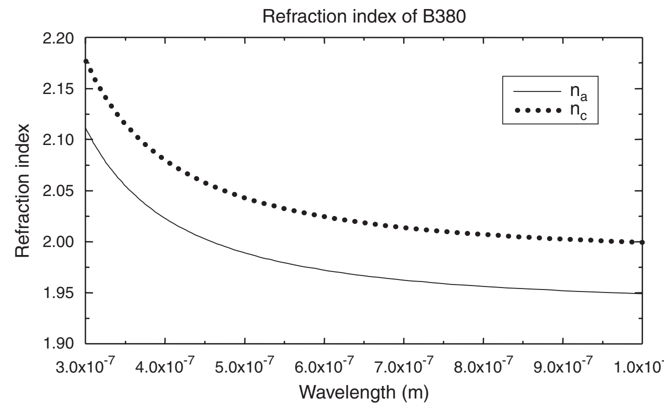

Fig. 1. Calculated optical indexes of pure $LaBr_3$ along c and a-axes as a function of wavelength.

Table 2
Best energy resolution FWHM (%) for 38 × 38 mm BrilLanCe®-350, BrilLanCe®-380 units

Isotope	Energy (keV)	B-350 (%)	B-380 (%)
Cs^{137}	662	3.5	2.6
Co^{57}	122	7.4	5.9
Am^{241}	59.5	11.4	9.7

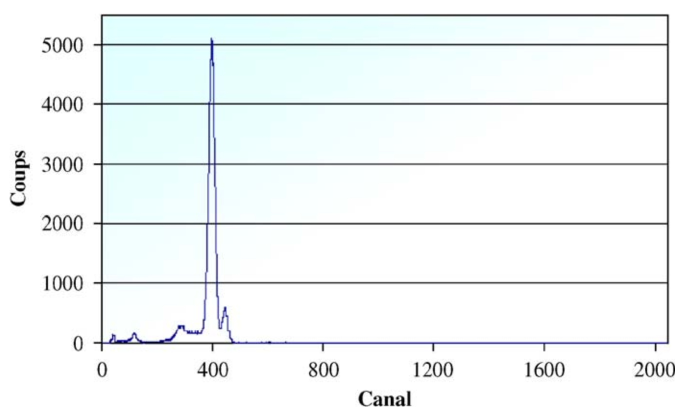

Fig. 2. Pulse height spectrum of a 38 × 38 mm Brillance®-380 detector excited by Co^{57} (122 keV).

These excellent energy resolutions suggest that even in these larger sizes, the materials are free of self absorption and non-uniform response that sometimes plague other crystals due to non-uniformity in growth or the presence of different Ce sites. To check the uniformity question further we measured the performance homogeneity of a 3%-doped BrilLanCe®-350 3″ × 3″ crystal. A collimated Cs^{137} source was directed at the various locations shown in Fig. 3. The signal was detected with a Hamamatsu R1307 PMT on electronics with a 1 μs shaping. The variation of response over the several positions was small.

- Standard deviation: Pulse Height = 0.4%.
- Standard deviation: Energy Resolution = 0.2%.