Scintillation Properties of high-resolution $La(Br_xCl_{1-x})_3$:Ce and high-sensitivity $CeBr_3$

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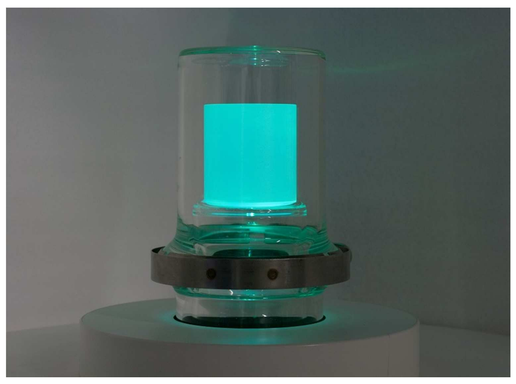

Figure 1: 3” x 3” $CeBr_3$ Crystal.

Materials

There is growing interest in halide-lanthanide scintillators for gamma-ray spectroscopy applications. These Ce^{3+} activated scintillators are distinguished by very high energy resolution and fast detection of gamma rays. The halide-lanthanide scintillators are particularly attractive to bridge the gap between the simple-to-use but relatively low-energy-resolution NaI:Tl scintillators and the more complex, high-energy-resolution cryogenically-cooled germanium detectors.

$CeBr_3$ is unique in respect to intrinsic activity as it is free of naturally radioactive ^{138}La and its ^{227}Ac contamination is significantly lower than in $LaBr_3$:Ce. Due to negligible intrinsic activity, $CeBr_3$ holds significant merit in ultra-high sensitivity detection when:

- dealing with low-intensity gamma-ray emissions (e.g. environmental monitoring),
- in the homeland security field when detection time is critical,
- in space applications such as the BepiColombo ESA/JAXA mission to Mercury [1].

At present, $CeBr_3$ crystals are routinely grown in various boule sizes as large as 4¼” in diameter by Hellma Materials GmbH [2]. Scionix Holland BV manufactures high-quality $CeBr_3$ scintillation detectors. A new material $La(Br_xCl_{1-x})_3$:Ce – denoted as LBC – is under development, that contains a mixture of two halides, lanthanum bromide and lanthanum chloride, activated by Ce^{3+} [3].

This poster covers $CeBr_3$ and LBC in several aspects relevant to gamma-ray spectrometry, such as light yield, energy resolution, intrinsic activity, decay time, non-proportionality of the response, and refractive index measurements.

	$CeBr_3$	$La(Br_xCl_{1-x})_3$:Ce (LBC)	For comparision NaI(Tl)
Light Yield (ph/MeV)	**50.000**	**60.000**	40.000
Energy resolution @ 662 keV (%)	**3.8 – 4.0**	**2.8 – 3.1**	7
Decay Time (ns)	**18-20**	**35**	230
Emission Wavelength (nm)	**380**	**350-380**	415
Background (cps/cc)	**<0.004**	**<1.5**	
Refractive Index	**2.2**		1.85
Density (g/cm^3)	**5.2**	**4.9**	3.36

Table 1: Scintillation and material parameters for $CeBr_3$ and LBC.

Energy Resolution

Energy (keV)	Resolution (FWHM) $CeBr_3$	Resolution (FWHM) LBC	For comparision NaI(Tl)
81	**11.0%**	**8.7%**	10%
122	**8.0%**	**6.4%**	8.5%
356	**4.9%**	**3.7%**	8%
662	**3.8%**	**2.8%**	7%
1332	**2.8%**	**2.2%**	5%

Table 2: Energy resolution versus energy for $CeBr_3$ and LBC.

Sample Spectra

Fig. 2 shows the response of $CeBr_3$ and NaI(Tl) to ^{60}Co where the well-known lines at 1173 and 1332 keV are seen. The resolution of the two peaks is much better for $CeBr_3$ and the efficiency is also higher in comparison to a NaI(Tl) detector of same size.

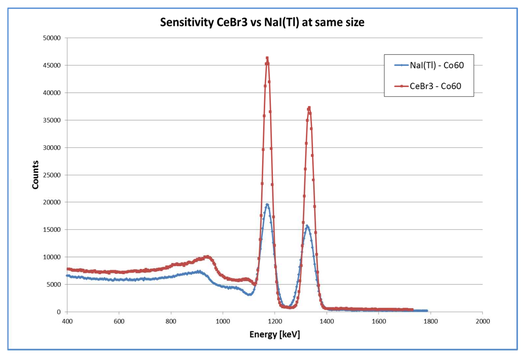

Figure 2: Comparison of Co-60 spectra for $CeBr_3$ (red) and NaI(Tl) (blue), each 2”x2” in size.

At 1332 keV $CeBr_3$ gives 2.8% resolution, while LBC yields 2.2%, as shown in Fig.3.

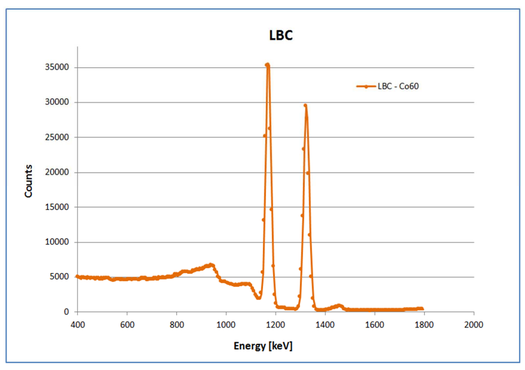

Figure 3: Co-60 Spectrum for 1”x1” LBC.

Detection Efficiency

Both $CeBr_3$ and LBC absorb gamma radiation better than NaI(Tl) primarily due to higher density. In the energy range from 100 keV to 3 MeV, efficiencies exceed those of NaI(Tl) [4].

Energy (keV)	Ratio Peak Counts $CeBr_3$ to NaI(Tl)	Ratio Peak Counts LBC to NaI(Tl)	For comparision NaI(Tl)
662	**130%**	**128%**	100%

Table 3: Relative efficiency of $CeBr_3$ and LBC in comparison to NaI(Tl).

Non-Proportionality of the Response

For low gamma energies (x-rays) the response of the scintillator is non-proportional. According to well studied charge transport behavior in scintillation processes, the energy resolution of a scintillator is expected to be impacted by the non-proportionality (nPR). The deviation of nPR from 100% is about 1.8 times larger for $CeBr_3$ larger than for LBC. This is reflected in the different energy resolutions (3.8% for $CeBr_3$ vs. 2.8% for LBC).

Energy (keV)	nPR $CeBr_3$	nPR LBC
32.2	**89.2%**	**95.3%**
59.5	**92.5%**	**96.0%**
81	**94.4%**	**96.6%**
122	**95.8%**	**97.5%**
356	**99.2%**	**99.4%**
662	**100%**	**100%**

Table 4: Non-Proportionality of the response (nPR) for $CeBr_3$ and LBC. Peaks for ^{241}Am, ^{57}Co, and ^{133}Ba are first calibrated to ^{137}Cs. After this calibration nPR is obtained as the ratio of the measured energy divided by the real energy.

Refractive Index

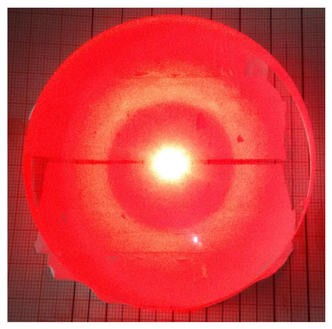

Figure 4: Pfund ring from HeNe laser in $CeBr_3$.

Wavelength (nm)	Refractive Index $CeBr_3$
380	**2.24 ± 0.03 (extrapolated)**
405	**2.21 ± 0.03**
532	**2.17 ± 0.03**
633	**2.12 ± 0.03**

Table 5: Refractive Index of $CeBr_3$ versus wavelength.

The refractive index was measured via Pfund’s method. A laser beam is incident on a polished $CeBr_3$ disk. The bottom is covered with teflon tape. Backscattered light reaching the upper surface at or above the critical angle is reflected and forms a sharp ring of light on the teflon screen.

Intrinsic Activity

The material used for growing $CeBr_3$ crystals is now available in formerly unknown purity. Accordingly, the intrinsic background in detectors is significantly reduced. Fig. 5 shows the improvements achieved in lowering intrinsic activity of $CeBr_3$. Hellma Materials now offers low background crystals with < 0.004 cps/cc, while typical measured values will even be lower (between 0.002 to 0.003 cps/cc or lower).

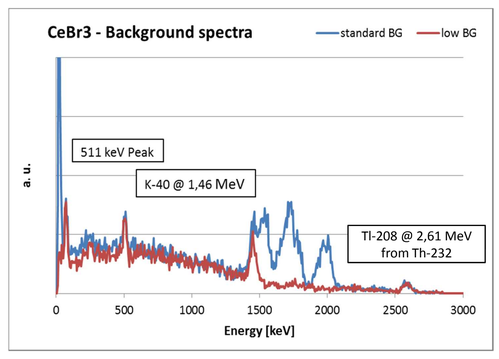

Figure 5: Improvement in intrinsic activity of $CeBr_3$. Low Background $CeBr_3$ (red) in comparison with Standard BG $CeBr_3$ (blue). Most of the peaks in the (red) Low BG curve are unrelated to crystal activity, stemming from cosmic radiation (511 keV peak) or environmental sources (K-40 and Th-232).

The LBC background spectrum is shown in Fig. 6. The count rate in the energy range from 1.6 – 2.7 MeV is presently at the level of 1.1 to 1.4 cps/cc. This does not include background from La-138. Hellma Materials is currently working on improving these Ac-227 related impurities in LBC raw materials.

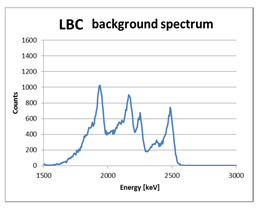

Figure 6: LBC background spectrum.

R&D Objectives

Enhancing $CeBr_3$ Energy Resolution

Present R&D efforts are directed towards improving $CeBr_3$ energy resolution. We have examined several 1”x1” crystals and found the best result as:

- 3.5% energy resolution at 662 keV
- nPR reduced to 0.6 times Standard nPR of $CeBr_3$

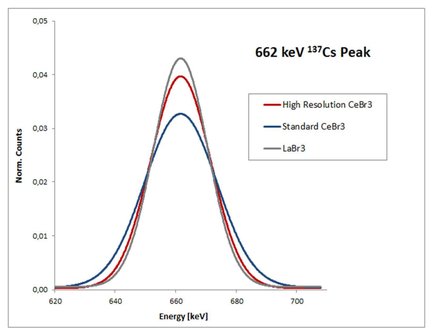

Figure 7: Improvement in energy resolution of $CeBr_3$. 1”x1” $CeBr_3$ crystal with 3.5% resolution at 662 keV (red) in comparison with Standard $CeBr_3$ (blue) and $LaBr_3$ (grey).

Lowering LBC Intrinsic Background

The high background of LBC crystals is of significant concern. Efforts are underway to monitor and improve Ac-227/U-235 related impurities in $LaBr_3$ and $LaCl_3$ raw materials. Hellma Materials aims at achieving background rates < 0.25 cps/cc in the energy range from 1.6 to 2.7 MeV.

For applications tolerant of 1.5 cps/cc background level, LBC crystals are currently available in sizes up to 1.5” x 1.5”.

[1] A. Kozyrev et al., Review of Scientific Instruments 87 (2016) 085112.
[2] F.G.A. Quarati et al., NIM A 729 (2013) 596.
[3] US Patent 7084403 Srivastava et al., General Electric Company.
[4] Poster #60 W. Westmeier, *$CeBr_3$ – a well characterized new scintillator for gamma-ray spectrometry*.

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