Evaluation of production samples of the scintillators $LaBr_3$:Ce and $LaCl_3$:Ce

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Abstract

We report on the evaluation of the performance of two recently developed scintillator materials, $LaCl_3$:Ce and $LaBr_3$:Ce, at the task of gamma ray spectroscopy. Their performance is compared to a standard scintillator used for gamma ray spectroscopy — a 25 mm diameter 25 mm tall cylinder of NaI:Tl. We measure the pulse height, energy resolution, and full-energy efficiency of production $LaBr_3$:Ce and $LaCl_3$:Ce scintillation crystals of different sizes and geometries for a variety of gamma-ray energies. Using production rather than specially selected crystals will establish whether immediate large-scale use is feasible. The crystal is excited by gamma rays from one of six isotopic sources (^{125}I, ^{241}Am, ^{57}Co, ^{22}Na, ^{137}Cs, and ^{60}Co) placed 15 cm away from the scintillator. Our measurements show that both $LaCl_3$ and $LaBr_3$ outperform NaI:Tl in almost all cases. They outperform NaI:Tl at all energies for the photopeak fraction and counting rate measurements, and for energy resolution at higher energies (above 200 keV for $LaCl_3$ and 75 keV for $LaBr_3$). The performance of production crystals is excellent and these scintillators should be considered for immediate use in systems where stopping power and energy resolution are crucial.

Keywords: scintillation crystals; rare-earth trihalides; $LaCl_3$:Ce; $LaBr_3$:Ce; gamma spectroscopy

1. Introduction

The use of scintillators in gamma ray spectroscopy is very wide spread. The important properties for scintillation crystals are high light output, high stopping power, fast decay time, good linearity and low cost. The most commonly used scintillator is NaI:Tl, which has a peak emission of 415 nm, a light output of 38,000 photons/MeV a density of 3.7 g/cc, and a decay time of 230 ns. NaI:Tl is also widely used in gamma cameras for medical imaging. Recently, there is a growing interest in employing a large volume of scintillator material such as NaI:Tl in the detection and identification of radioisotopes through their emission lines for homeland security. Different radioisotopes can have fairly close emission energies. To distinguish between the different isotopes will require the scintillation material to have excellent energy resolution. The energy resolution is typically 5–6% FWHM for 662 keV gamma rays with NaI:Tl. Recently, new cerium doped lanthanum halide scintillators, $LaCl_3$:Ce [1] and $LaBr_3$:Ce [2], have been discovered. These new scintillators have about 3% FWHM energy resolution for 662 keV gamma rays, and have a more linear response with energy than NaI:Tl [3,4]. However, most of these measurements were performed using small research crystals. Recently, Balcerzyk *et al.* [5] have compared the performances of a production sample of $LaCl_3$:Ce and NaI:Tl. In this paper, we report on the performances of the production crystals of $LaCl_3$:Ce and $LaBr_3$:Ce.

2. Materials

The scintillation properties of $LaCl_3$:Ce and $LaBr_3$:Ce are described in [3] and [4] respectively. $LaCl_3$:Ce has a density of 3.9 g/cc, a light output of 50,000 photons/MeV, and a decay time of 20 ns. Its peak emission wavelength is 350 nm. On the other hand, $LaBr_3$:Ce has a density of 5.3 g/cc, a light output of 60,000 photons/MeV, and a decay time of 25 ns. Its peak emission wavelength is 370 nm. We acquired production crystals of $LaCl_3$;Ce and $LaBr_3$:Ce of various sizes from Saint Gobain, Inc. (France) and Radiation Monitoring Devices, Inc. (United States). In addition, we also acquired a standard reference NaI:Tl crystal (25 mm diameter 25 mm tall cylinder) from Saint Gobain, Inc. to compare the performances. All these crystals are hygroscopic and therefore come encapsulated by the supplier in an aluminum case with a front glass window. Table 1 shows the 14 individual scintillator crystals that were acquired and measured.

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Table 1

Production scintillator crystals that were acquired and measured

Material	Geometry (Diameter × Height)	Number of Crystals	Supplier
NaI:Tl	25 mm × 25 mm	1 each	St. Gobain
$LaCl_3$	13 mm × 13 mm	3 each	St. Gobain
$LaCl_3$	25 mm × 25 mm	3 each	St. Gobain
$LaCl_3$	38 mm × 38 mm	1 each	St. Gobain
$LaBr_3$	13 mm × 13 mm	3 each	St. Gobain
$LaBr_3$	25 mm × 25 mm	1 each	St. Gobain
$LaBr_3$	10 mm × 28 mm	1 each	RMD
$LaBr_3$	10 mm × 30 mm	1 each	RMD

3. Methods

The scintillator crystal being studied was coupled with Viscasil 600k silicone optical coupling fluid to a Hamamatsu R-1306 photomultiplier tube operated at –1200 V or a Photonis XP2060B with a VD200K base operated at –1000 V. The photomultiplier tube output was amplified by an Ortec 113 preamplifier followed by an Ortec 672 shaper amplifier set to a 1 μs rise and fall time. This amplified output was digitized and histogrammed with an Ortec TRUMP PCI-8k multi-channel analyzer and the data analyzed with the Ortec MAESTRO-32 software. The crystal was excited by gamma rays from one of six isotopic sources (^{125}I [27.5keV], ^{241}Am [60 keV], ^{57}Co [122 keV], ^{22}Na [511, 1275 keV], ^{137}Cs [662 keV], and ^{60}Co [1173, 1330 keV]) placed 15 cm away from the scintillator.

3.1. Light output

A pulse height spectrum is acquired for each crystal with each excitation source. The photopeak is identified and the MAESTRO software used to fit the photopeak region to a Gaussian distribution plus background. The fit value for the center of this distribution is recorded for each scintillator. The (relative) light output is defined as the center value for this scintillator and source divided by the center value for the NaI:Tl reference crystal excited with the same source. The primary use of this measure is to estimate the amplitude of the signal generated in each scintillator.

3.2. Energy resolution

Using the data and subsequent photopeak fit obtained in the light output measurement, the energy resolution for each crystal and excitation source is defined as the photopeak full width at half maximum (FWHM) divided by the center. For convenience, the relative energy resolution (compared to NaI:Tl) is also computed by dividing the energy resolution for this scintillator and source by the energy resolution for the NaI:Tl reference crystal excited with the same source. The primary use of this measure is to estimate the ability of each scintillator to reject background events.

3.3. Photopeak fraction

Using the data and subsequent photopeak fit obtained in the energy resolution measurement, the photopeak fraction for each crystal and excitation source is defined as the number of events within the photopeak region (defined as being within plus or minus one FWHM value of the photopeak center) divided by the total number of events in the spectrum. The photopeak fraction is not calculated for ^{22}Na, which has two emission gamma rays that are far apart in energy making it difficult to estimate the background contribution for each photopeak. On the other hand, we calculated the photopeak fraction for the higher energy emission of ^{60}Co (1330 keV) by assuming that the photopeak and background events of its two emission gamma rays (1173 keV and 1330 keV) are approximately the same because both gamma ray energies are fairly close together and have the same emission probability. For convenience, the relative photopeak fraction (compared to NaI:Tl) is also computed by dividing the photopeak fraction for this scintillator and source by the photopeak fraction for the NaI:Tl reference crystal excited with the same source. The primary uses of this measure are to estimate the efficiency of each scintillator for detecting gamma rays of a specific energy, and to help estimate signal to background levels.

3.4. Counting rate

Using the data and subsequent photopeak fit obtained in the photopeak fraction measurement, the counting rate is defined as the number of events within the spectrum divided by the live time of the system. The relative counting rate (compared to NaI:Tl) for each crystal and excitation source is defined as its counting rate divided by the counting rate in the NaI:Tl spectrum. For convenience, the relative photopeak rate (compared to NaI:Tl) is also computed by multiplying the relative photopeak fraction for this scintillator and source by the relative counting rate. The primary use of this measure is to estimate the efficiency of each scintillator for detecting gamma rays of a specific energy.

4. Results

4.1. Light output

Fig. 1 to 4 show the energy spectra of the gamma rays from a ^{137}Cs source, as measured with the reference NaI:Tl crystal, the 25 mm diameter $LaCl_3$:Ce crystal, the 25 mm diameter $LaBr_3$:Ce crystal, and the 10 × 28 mm $LaBr_3$:Ce crystal respectively. The light output measurements were inconclusive. The data were all acquired with the R1306 photomultiplier tube, but we subsequently discovered that the relatively high light outputs of the lanthanum halides caused significant nonlinearity when using this photomultiplier tube. Subsequent tests with the Photonis XP2060B showed that nonlinearity was not an issue with that photomultiplier tube, and also that while the nonlinearity affected the light output measurement, it did not affect any of the other measurements.

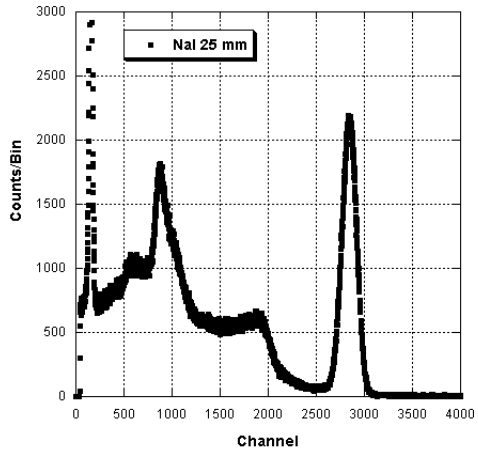

Fig. 1. Energy spectrum of gamma rays from a ^{137}Cs source, as measured with the reference NaI:Tl crystal.

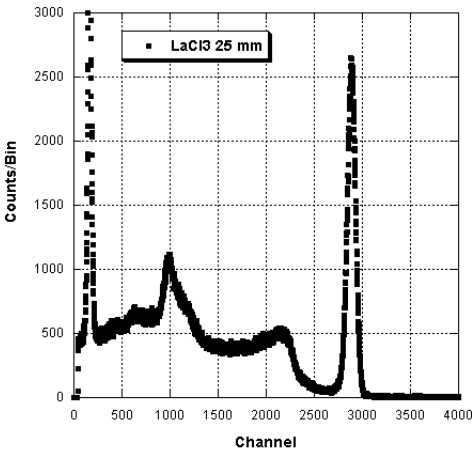

Fig. 2. Energy spectrum of gamma rays from a ^{137}Cs source, as measured with the 25 mm diameter $LaCl_3$:Ce crystal.

4.2. Energy resolution

Fig. 5 shows the energy resolution for each crystal and excitation source as well as the relative energy resolution (the energy resolution divided by the NaI:Tl energy resolution). Table 2 lists the energy resolution in full-width at half-maximum percent. A standard deviation is given for those geometries where we have acquired more than one production sample of the same material and geometry. The data show that $LaCl_3$:Ce outperforms NaI:Tl for energies above 200 keV, while $LaBr_3$:Ce outperforms NaI:Tl for energies above 75 keV. The small standard deviation indicates that the production samples are good quality and uniform. In addition, the energy resolution does not depend strongly on the crystal size or supplier (although one of the $LaBr_3$:Ce crystals provided by RMD has slightly inferior performance).

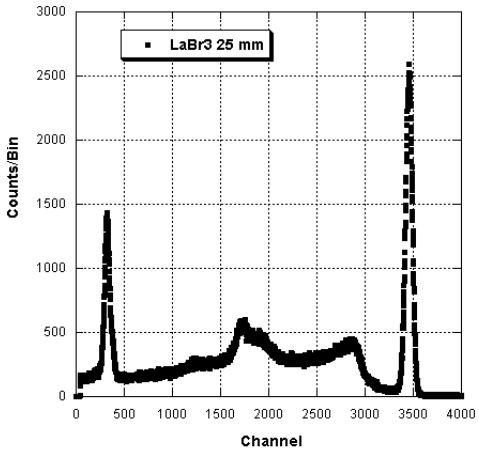

Fig. 3. Energy spectrum of gamma rays from a ^{137}Cs source, as measured with the 25 mm diameter $LaBr_3$:Ce crystal.

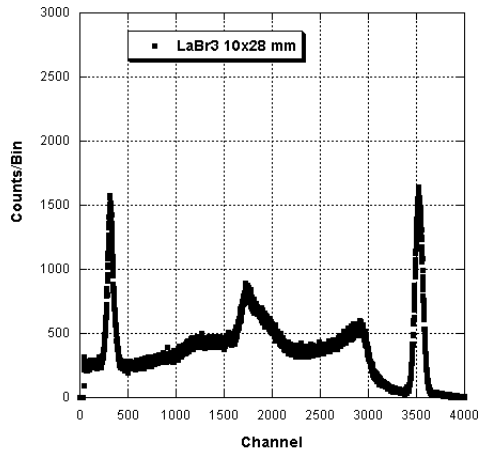

Fig. 4. Energy spectrum of gamma rays from a ^{137}Cs source, as measured with the 10 × 28 mm $LaBr_3$:Ce crystal.

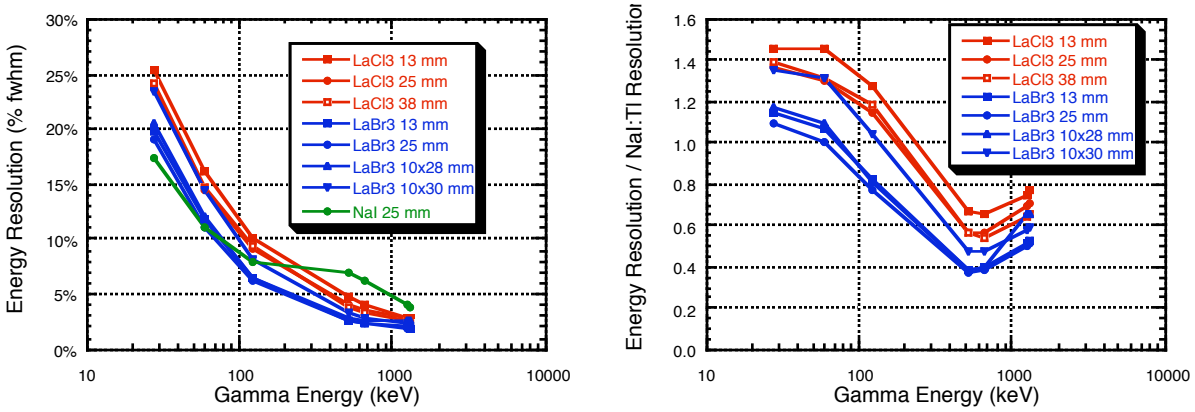

Fig. 5. Energy resolution in percent FWHM (left) and relative energy resolution compared to NaI:Tl (right), as measured for each crystal and excitation source.

Table 2

Energy resolution in percent FWHM as measured for each crystal and excitation source

Gamma Ray Energy (keV)	$LaCl_3$ 13 mm	$LaCl_3$ 25 mm	$LaCl_3$ 38 mm	$LaBr_3$ 13 mm	$LaBr_3$ 25 mm	$LaBr_3$ 10 x 28 mm	$LaBr_3$ 10 x 30 mm	NaI:Tl 25 mm
27.5	25.43 ± 0.14	23.64 ± 0.30	24.27	19.92 ± 0.28	19.1	20.5	23.47	17.45
60	16.26 ± 0.30	14.54 ± 0.24	14.66	11.87 ± 0.29	11.18	12.11	14.56	11.14
122	10.23 ± 0.12	9.11 ± 0.19	9.47	6.55 ± 0.13	6.15	6.44	8.26	7.96
511	4.71 ± 0.06	4.04 ± 0.06	3.92	2.72 ± 0.07	2.61	2.76	3.31	7.05
662	4.00 ± 0.05	3.47 ± 0.14	3.30	2.47 ± 0.10	2.35	2.49	2.89	6.14
1275	2.96 ± 0.05	2.76 ± 0.13	2.54	2.04 ± 0.05	1.99	2.58	2.33	3.98
1330	2.92 ± 0.01	2.71 ± 0.19	2.47	1.99 ± 0.05	1.95	2.51	2.24	3.79

4.3. Photopeak fraction

Fig. 6 shows the photopeak fraction for each crystal and excitation source as well as the relative photopeak fraction (compared to the 25 mm NaI:Tl crystal). Table 3 lists the photopeak fraction. The data show that both $LaCl_3$:Ce and $LaBr_3$:Ce outperform NaI:Tl at all energies, except at high excitation energies for the small size crystals. This is expected as the probability of re-absorbing Compton scatter is lower in the smaller crystals. However, for similar crystal geometry (25 mm) $LaCl_3$:Ce only outperforms NaI:Tl for energies below 100 keV.

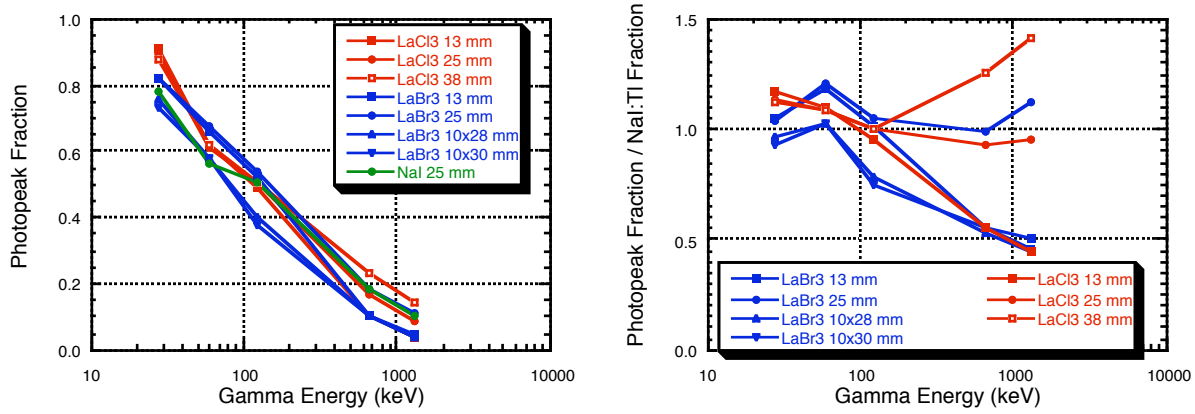

Fig. 6. Photopeak fraction (left) and relative photopeak fraction compared to NaI:Tl (right), as measured for each crystal and excitation source

Table 3

Photopeak fraction as measured for each crystal and excitation source

Gamma Ray Energy (keV)	$LaCl_3$ 13 mm	$LaCl_3$ 25 mm	$LaCl_3$ 38 mm	$LaBr_3$ 13 mm	$LaBr_3$ 25 mm	$LaBr_3$ 10 x 28 mm	$LaBr_3$ 10 x 30 mm	NaI:Tl 25 mm
27.5	0.914 ± 0.001	0.894 ± 0.004	0.879	0.825 ± 0.006	0.817	0.761	0.729	0.784
60	0.623 ± 0.003	0.614 ± 0.005	0.615	0.664 ± 0.010	0.681	0.580	0.580	0.565
122	0.485 ± 0.002	0.509 ± 0.004	0.512	0.517 ± 0.003	0.538	0.403	0.384	0.512
662	0.100 ± 0.001	0.168 ± 0.002	0.226	0.099 ± 0.001	0.178	0.095	0.100	0.180
1330	0.044 ± 0.001	0.092 ± 0.001	0.138	0.050 ± 0.001	0.108	0.042	0.045	0.097

4.4. Counting rate

Fig. 7 shows the relative counting rate (compared to the 25 mm NaI:Tl crystal) and the relative photopeak rate for each crystal and excitation source. Table 4 lists the count rate in counts per second. The data indicate that as expected, the volume of the detector is of primary importance. They also indicate that for detectors with similar volume, both $LaCl_3$:Ce and $LaBr_3$:Ce outperform NaI:Tl at all energies.

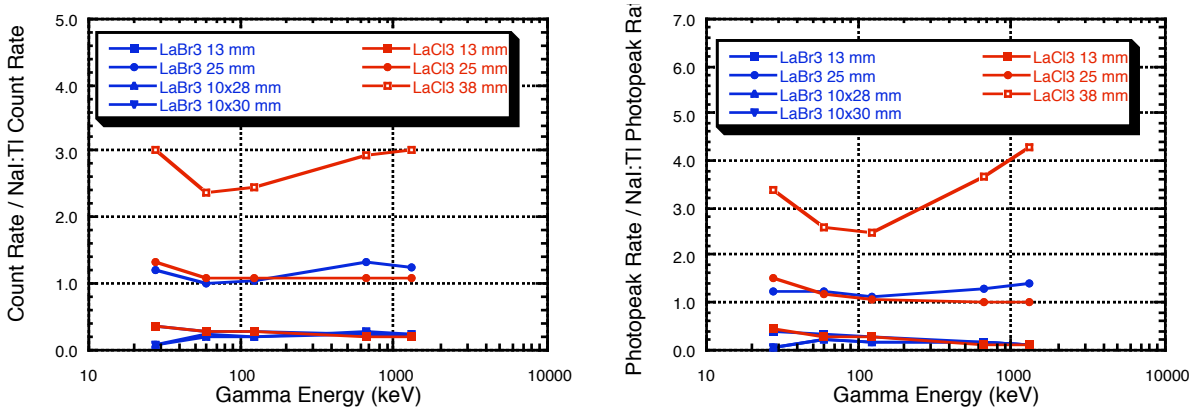

Fig. 7. Relative count rate (left) and relative photopeak count rate (right) compared to NaI:Tl, as measured for each crystal and excitation source.

Table 4

Count rate in counts per second as measured for each crystal and excitation source

Gamma Ray Energy (keV)	$LaCl_3$ 13 mm	$LaCl_3$ 25 mm	$LaCl_3$ 38 mm	$LaBr_3$ 13 mm	$LaBr_3$ 25 mm	$LaBr_3$ 10 x 28 mm	$LaBr_3$ 10 x 30 mm	NaI:Tl 25 mm
27.5	1312 ± 18	4742 ± 63	10843	1285 ± 89	4311	214	226	3582
60	245 ± 2	992 ± 5	2219	238 ± 1	933	187	208	926
122	3421 ± 13	13939 ± 19	32208	3411 ± 26	13775	2364	2466	13107
662	1166 ± 16	6852 ± 53	18481	1474 ± 17	8311	1543	1615	6322
1330	481 ± 4	2947 ± 6	8152	565 ± 4	3419	637	679	2720

5. Summary

Our measurements show that both $LaCl_3$:Ce and $LaBr_3$:Ce outperform NaI:Tl in almost all cases. They outperform NaI:Tl at all energies for the photopeak fraction and counting rate tests, and for energy resolution at higher energies (above 200 keV for $LaCl_3$ and 75 keV for $LaBr_3$). The performance of production crystals is excellent and these scintillators should be considered for immediate use in systems where stopping power and energy resolution are crucial.

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