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Advanced Scintillator Crystal for Various Radiation Detection

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Abstract

Recently halide scintillator materials, such as cerium doped $LaBr_3$ or $LaCl_3$ crystals have proven to be novel inorganic scintillator for gamma-ray detection and spectroscopy. Both materials exhibit exceptional scintillation properties such as high light output, fast response, and excellent energy resolution. However, these materials are highly hygroscopic, hard to grow due to thermal stressing then end up cracking and suffer from the self-activity. A search for high resolution, high light yields, and non-hygroscopic materials are highly desirable to advance the scintillator technology. This paper presents the crystal growth of a new scintillator material, lanthanum doped cerium bromide ($CeBr_3$:3%$LaBr_3$), and its optical characteristics. In comparison with the high performance Ce-doped $LaBr_3$ or $LaCl_3$, $CeBr_3$:3%$LaBr_3$ crystals are less hygroscopic, have higher energy resolution (R~ 8% at 662 keV) and higher light yield (67,000 Ph/MeV±3000). Preliminary results on the growth of $CeBr_3$:3%La crystals are presented along with comparison of its luminescence properties with other well-known inorganic scintillators.

Introduction

Varity applications of scintillator crystals for the purpose of nuclear radiation detection for medical applications lead science and crystal growth researchers to search for ideal scintillator for the above applications.

A radiation detector is a device in which incident radiation produces a measurable effect. The case where the effect is a production of light pulses is called a scintillator detector; a scintillator is a substance that glows when hit by high-energy particles or photons. Scintillators can be used not only to count nuclear radiation (like Geiger-Müller counters), but also to measure the energy of the radiation. Scintillators are the most widely used detectors for spectroscopy of energetic photons (x-rays and gamma rays). These detectors are commonly used in nuclear and high-energy physics research, medical imaging, diffraction, non-destructive testing, nuclear treaty verification and safeguards, and geological exploration. Their capability to detect a wide assortment of radiations (including charged particles and neutrons), great variety in size and constitution of scintillators make them as the best choice in different applications.

Performance of scintillation detector depends first of all on the properties of the scintillation material. The ideal material should posses but not be limited to the following critical properties (Laqua et.al, 1995; Anon, 2005; Wiliam, 1994)

- It should convert the energy of particles or gamma quanta into detectable light with high scintillation efficiency.
- This conversion should be linear—the light yield should be proportional to deposited energy over as wide a range as possible.
- The medium should be transparent to the wavelength of its own emission for good light collection.
- The decay time of induced luminescence should be short so that fast signal pulses can be generated.

- The material should be of good optical quality and subject to manufacture in sizes large enough to be interest as a practical detector.
- High-energy resolution.

There is lack of a perfect material that meets all these criteria, and the choice of a particular scintillator is always a compromise among these and other factors. And as a result of this there is a continued interest in search for new scintillators with enhanced performance. When evaluating scintillators for its applicability in gamma ray spectroscopy and dedicated detector system high energy resolution, good linearity, high light yield, fast response, high stopping efficiency, low cost, and minimal after glow are considered to be the most important requirement. (Knoll, 1999; Dorenbos, 1995; Knoll, 2000).

During the 20 years after the discovery of NaI(Tl+) by Hofstadter in 1948 there has been significant effort to research and develop other scintillators for detection of high energy photons (x rays and gamma rays). Besides NaI(Tl+), classical scintillators like CdWO4 in 1948, CsI(Tl+) in 1950(William 1995), CsI(Na+) in 1965, CaF2(Eu2+) in 1966 were discovered in this period. In 1973 Weber and Monchamp, reported on the luminescence properties of Bi4Ge3O12 (BGO). This material is of particular interest because of its high density of 7.13 g/cm-3, which provides a good stopping power for high-energy gamma rays (Hofstadter, 1948; Dorenbos et.al, 1995; Kroger, 1948; Hofstadter, 1950) .

In the last few years there has been an effort to develop new scintillator materials characterized by a high light output, a fast decay time, high detection efficiency, a good energy resolution and a low non-proportionality. This renewed interest in scintillators was also initiated by the demand for high density and fast scintillating crystals for application in medical diagnostics, high energy physics and high count rate experiments (Menefee et.al, 1950; Webber and Monchamo, 1973). This research, starting around 1980, is still actively being pursued and has resulted in several new scintillators amongst which are mentioned in subsequent paragraphs. Moszynski et al(2004) studies of pure CsI crystals showed a high light output of 124000 $\pm$ 12000 ph/MeV, energy resolution of 4.3 $\pm$ 0.1% for 662 keV gamma rays from a 137Cs source and a good (but unspecified) proportionality of the light yield versus energy of gamma rays for the best sample at liquid nitrogen temperature. Moszynski et al.(2004) also performed a study on pure NaI crystals which showed a light output of 44000 $\pm$ 4000 ph/MeV, energy resolution of 5.9 $\pm$ 0.2% for 661.6 keV gamma rays from a 137Cs source and a good (but unspecified) proportionality of light yield versus energy of gamma rays at liquid nitrogen. Their observations suggested that non-proportionality response of Na(Tl) is the result of the energy transfer to thallium luminescence centers (Nestor and Huang, 1975; Shah et. al, 2004; Moszynski, et. al, 2004).

BaF2 has been a scintillator of choice in some application primarily due to its fast decay components (τ = 0.6 ns), which enables timing resolution comparable to fast plastic scintillators. BaF2 is non-hygroscopic, has a high effective atomic number (Zeff $\approx$ 56), and high specific gravity (ρ = 4.89 g/cm3). Typical energy resolution has been reported to be 7-8% at 662keV and its light yield about 21% relative to that of NaI(Tl) (Moszynski, et. al, 2004).

Shah et al,(2004) reported on the scintillation properties of LuI3:Ce. The emission spectrum was found to peak at $\approx$ 474 nm, the light output of LuI3:Ce was measured to be $\approx$ 50000 photons/MeV at room temperature and energy resolution at 662 keV was measured to $\approx$10%. With regards to non-proportionality Shah et al(2004) study shows that LuI3:Ce is a fairly proportional scintillator. LuI3:Ce shows a 10% non-proportionality in light yield over an energy range of 14 keV to 662 keV. (Balcerzyk et. al, 2002).

For NaI(Tl) and most other alkali halides, the scintillation response peaks between 10 and 15 keV and decreases as the photon energy. Finally, the scintillation response levels at higher energy Contrary to alkali halide, oxide bases scintillating materials show an increasing scintillation response with increasing photon energy, which levels at higher energies (Ishibashi et. al, 1989; Jones, 1962).

Most crystals studied for scintillation properties have displayed a few shortcomings in either their scintillation properties or their implementation as scintillation detectors. For some scintillators like NaI, the utilization of its expected good properties has been limited by difficulties in a

cooling down of the crystals independently of a photodetector (Jones, 1962).

Attention has been given to Ce3+ -doped material, the characterization of their optical and scintillation properties, and the elucidation of scintillation mechanism. LaBr3 crystals have hexagonal (UCl3 type) structure with P63/m space group and their density is 5.29 g/cm3 (Aitken et. al, 1967; Hawrami et. al, 2008; Hawrami et. al, 2006).

The compound melts congruently at 783 oC and therefore its crystals can be grown using melt-based methods such as Bridgman and Czochralski. This is fortunate because melt based processes are being studied for of large volume crystals. Shah et al (2004) have successfully grown crystals of LaBr3 with size up to ~2.3 cm3 with the Bridgman process and have also shown that the energy resolution of the 662 keV photopeak recorded with LaBr3:Ce scintillator has been measured to be 3.2%.

Hawrami et al.(2006), studies on the influence of anion on the spectroscopy and scintillation mechanism in pure and Ce3+ -doped K2LaX5 and LaX3 (X = Cl, Br, I) observed decreasing scintillation rates of K2LaX5:Ce3+ along the halide series from Cl to I. Hawrami et al (2008)., recently discovered, Europium doped halide scintillator, and SrI2 in particular is a promising scintillator for gamma ray spectroscopy exhibiting attractive scintillation properties such as highest reported energy resolution(2.5%) amongst all scintillation materials, very high light output (of ~120,000 photons/MeV) and fast principal decay constant (of 1.2 ms).

Medical Applications

The field of medical imaging is in rapid evolution and is based on five different modalities: X-rays radiology, emission tomography, ultrasonic tomography, magnetic resonance imaging (MRI), and electrophysiology with electro-and magneto encephalography (EEG and MEG). The first medical image is arguably the x-ray image that Röntgen took of his wife's hand in 1895. While Röntgen used photographic film to convert the x-rays into a form observable by the human eye, within one year powdered phosphor materials such as CaWO4 replaced photographic film as the x-ray conversion material and have been an integral part of medical imaging devices ever since. More recently, direct optical techniques such as bioluminescence and infrared transmission are also emerging as powerful imaging tools for non-too deep organs. Only X-rays radiology and emission tomography are using scintillators and are described here in more details (Hofstadter, 1948) The energies of X-rays and gamma rays used in medical diagnostics, in particular medical imaging, are summarized in Table 1. In X-ray imaging in almost all cases radiation is generated by means of X-ray tubes. The maximum energy is determined by the tube voltage, which in general is p150 KVp.

Table 1. Imaging modalities and radiation energies

The lower-energy part of the continuous X-ray spectrum is cut off by means of filters as it will not go through the body anyway and would only contribute to the radiation dose. The information of interest, i.e. the image resulting from projections displaying differences in absorption, is mainly obtained from approximately the top half of the energy range. In nuclear medicine, radiation is generated by radiopharmaceuticals introduced into the body. On average, the gamma-ray energies are higher than in X-ray imaging. In principle, I do not want any absorption at all. Information is obtained from emission, i.e. projection of the distribution of the radio nuclides. Apart from mammography, in all cases energies are so high that inorganic scintillators are required for efficient, large-area radiation detection. The gamma camera, employing NaI:Tl as scintillation material, is a well-known example. The variety of imaging techniques and the ever-growing wish for better images and faster data acquisition imply a diversity of detector requirements that continue to change. Often the specifications of available detectors, especially scintillators, do not meet these requirements. Consequently, many groups are working on inorganic-scintillator R&D. I will limit the discussion to a few cases for which much scintillator research and detector development are going on, i.e. fluoroscopy, X-ray Computed Tomography (CT) and Positron

Imaging modalities and radiation energies

Imaging modality		Energies
X-ray imaging	Mammography	25 kVp, ~18 keV
	Radiography, chest	150 kVp
	Fluoroscopy	150 kVp
	X-ray CT	150 kVp
Nuclear medicine	Scintigraphy	~80/140 keV
	SPECT	60–511 keV
	PET	511 keV

CT, computed tomography.
SPECT, single photon emission computed tomography.
PET, positron emission tomography.

Emission Tomography (PET). In fact, virtually all medical imaging modalities that require the detection of energetic photons (x- and γ- rays) use scintillator for their detection. These modalities include planar x-ray imaging, x-ray computed tomography (x-ray CT), SPECT (single photon emission computed tomography) and PET (positron emission tomography). See Figure 1.

In principle for the ideal scintillator, the first important requirement for a scintillator to be used in medical imaging devices is the stopping power for the given energy ranges of X- and γ-rays to be considered and, more precisely, the conversion efficiency. Clearly, materials with high Z and high density are favored but the position of the K-edge is also important For low-energy X-ray imaging (below 63 KeV), the attenuation coefficient of yttrium, cesium, and iodine are quite high and crystals such as YAP and CsI are good candidates to be used for each application. However, the ideal scintillator does not exist, and so each modality must compromise, choosing from available materials the most suitable combination of properties. Scintillating materials are used in a variety of medical imaging devices. Traditionally, inorganic scintillators have played an important role in the detection and visualization of radiation. (William W. Moses, Stephen, 1995).

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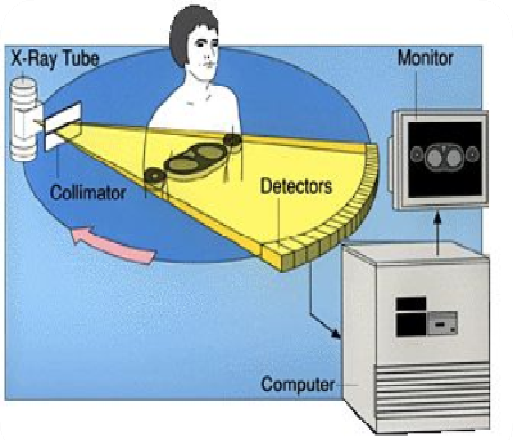
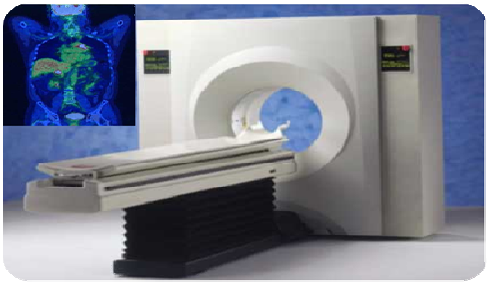
Fig.1. Sample image obtained using a combination of PET and CT imaging technology.





Characterization: Pulse Height Measurements

Pulse height measurements are generally performed to obtain information on the light yield, energy resolution and decay time of the scintillating material under ionizing radiation. A typical set-up for recording pulse height spectra is shown schematically in Figure 2. The crystal is optically coupled to the window of the PMT with silicon-based grease. If there is air between the scintillator and PMT, the light beam indicated to the scintillator surface at angle $\alpha \geq \arcsin(1/nSc)$ will not be able to go out from the scintillator due to the total internal reflection (nSc is the refractive index of the scintillator) To collect as much light as possible, it is wrapped in several layers of 0.1mm thick UV reflecting Teflon tape. Gamma rays and/or X-rays

emanating from a radioactive source excite the crystal. In most case the 662 KeV of a 137Cs source is used

A Hamamatsu R1791 PMT (P) detects the scintillation light. The output signal is integrated with a homemade pre-amplifier of the charge-sensitive type (A1), having an RC-time of 50 μs. Next, the signal is amplified once again and filtered using Gaussian shaping by an Ortec 572/672 spectroscopic amplifier (A2). Shaping times of 0.5 to 10 μs are available. Here, shaping time is defined as the time during which the output of the preamplifier is being integrated and shaped into a Gaussian pulse. Finally, the analogue signal is converted to a digital pulse by a conventional ADC unit analogue to digital converter (ADC). That way, the signal can be stored and processed by a computerized data handling system (PC). A Hamamatsu R6427 photomultiplier tube (PMT) was used for recording pulse height spectra with 662 keV gamma rays from a ^{137}Cs source. For better light collection, crystals were wrapped with Teflon tape on all sides except the face in contact with the PMT. The crystals were coupled with the PMT window with index matching silicone grease.

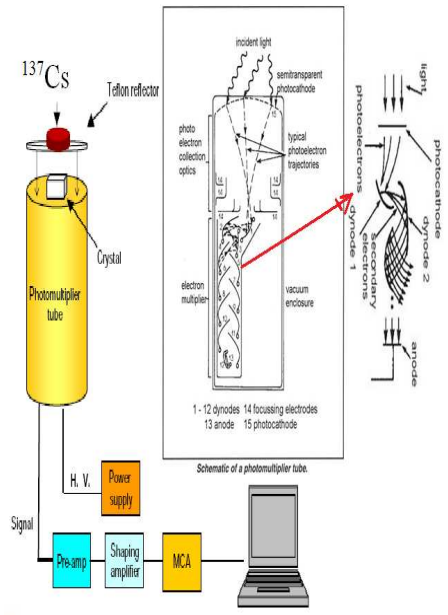


Figure 2: A schematic diagram of the experimental set up of scintillation system

Results and Discussion

In this section, crystal growth of a new scintillator material, lanthanum doped cerium bromide ($CeBr_3$**:**3%La), and its Scintillation characteristics are reported. In comparison with the high performance Ce-doped $LaBr_3$ or $LaCl_3$, $CeBr_3$:3%La crystals are less hygroscopic. Fig.3. shows the spectrums of $CeBr_3$:3%La under ^{137}Cs γ-rays excitation at 662 *KeV*. Energy resolution obtained was about 8 %. By comparing with Lanthanum Bromide ($LaBr_3$), the light outputs were estimated to be about 67000 *Ph/MeV*±3000. The inset shows a photograph of the cut and polished sample (5x4x2.2mm) crystals which were used for the pulse height measurements. Preliminary results on the growth of $CeBr_3$:3%La crystals are presented along with comparison of its luminescence properties with other well-known inorganic scintillators. The samples were observed to be less hygroscopic when compared to $LaBr_3$. The polished surfaces were found to be slightly foggy after some time, possibly due to reaction with air.

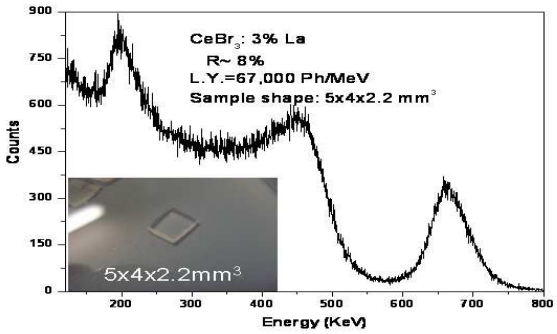


Figure 3: Energy Spectrum of 662 keV γ-rays from a ^{137}Cs source measured at room temperature with of $CeBr_5$:3%La crystal

Light yield and energy resolutions are summarized in table 2, along with properties of other inorganic scintillators that are routinely being used.

Table: 2 Comparison of Characteristics of selected single crystal scintillators

Properties	K_2CeBr_5	$CeBr_3$:La	CsI(Tl	NaI(Tl)	BGO
Density (g/cm^3)	N/A	N/A	4.51	3.67	7.13
Light Output (ph/MeV)	~50,000	~67,000±3000	52000	39000	9000
ΔE/E(FWHM) @ 662 keV	6.3%	~ 8%	10%	7%	>10 %
Peak (nm)	~ 400	~ 400	550	415	480

The inset in Figure 3 shows a photograph of the cut and polished crystal which was used for the pulse height measurements. The samples were observed to be less hygroscopic when compared to $LaBr_3$. The polished surfaces were found to be slightly foggy after some time, possibly due to reaction with air.

Conclusions

Ce-based compound scintillator crystals, $CeBr_3$:3%La have been successfully grown by vertical Bridgman method. Crystals are less hygroscopic than the $LaBr_3$ and crystal growth is relatively easy compared to $LaBr_3$ .Furthermore, they have higher energy resolution (R~ 8%) and high light yield (67,000 Ph/MeV±3000) in response to gamma rays than other well known inorganic scintillators. $CeBr_3$:3%La crystals have the desired properties for meeting the need for advanced scintillator-based gamma-ray spectrometers for nuclear non-proliferation testing, medical imaging, environmental remediation, and oil exploration.

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Appendix: At the end of the paper

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