$CeBr_3$ Scintillators for Gamma-Ray Spectroscopy

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***Abstract*—In this paper, we report on a new scintillator, cerium bromide ($CeBr_3$), for gamma-ray spectroscopy. Crystals of this scintillator have been grown using Bridgman process. In this material Ce^{3+} is an intrinsic constituent as well as a luminescence center for the scintillation process. Samples of $CeBr_3$ showed high light output (~ 68000 photons/MeV) and fast decay constant (~ 17 ns). Furthermore, they exhibited excellent energy resolution for gamma-ray detection. For example, energy resolution of $< 4\%$ full width at half maximum (FWHM) has been achieved using this scintillator for 662 keV photons (^{137}Cs source) at room temperature. High timing resolution (< 200 ps—FWHM) has been recorded with $CeBr_3$-photomultiplier (PMT) and BaF_2-PMT detectors operating in coincidence using 511 keV positron annihilation gamma-ray pairs.**

***Index Terms*—Ce^{3+}, gamma-ray spectroscopy, radiation detectors, scintillators.**

I. Introduction

SCINTILLATION spectrometers are widely used in detection and spectroscopy of energetic photons (X-rays and γ-rays) at room temperature. These detectors are commonly used in nuclear and particle physics research, medical imaging, diffraction, nondestructive testing, nuclear treaty verification and safeguards, nuclear nonproliferation monitoring, and geological exploration [1], [2].

Important requirements for the scintillation crystals used in these applications include high light output, high stopping efficiency, fast response, good proportionality, low cost and availability in large volume. These requirements cannot be met by any of the commercially available scintillators. As a result, there is continued interest in search for new scintillators with enhanced performance [3]–[6].

In this paper, properties of a new scintillator, cerium bromide ($CeBr_3$), are discussed. In this material, Ce^{3+} is an intrinsic constituent as well as a luminescence center for the scintillation process. The γ-ray stopping efficiency of $CeBr_3$ is significantly higher than that of NaI:Tl, the most common scintillation detector. For example, the mean attenuation length for 511 keV photons is 2.15 cm and 2.92 cm for $CeBr_3$ and NaI:Tl respectively. In our investigation, small crystals of $CeBr_3$ have been grown using the Bridgman process and their scintillation properties have been characterized. High light output, good proportionality, fast response and excellent energy and timing resolution have been measured for small $CeBr_3$ crystals. Based on the results, $CeBr_3$ is very promising for γ-ray spectroscopy. Its properties are very similar to those of another recently discovered scintillator, cerium doped lanthanum bromide ($LaBr_3$:Ce) [7].

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II. Crystal Growth

$CeBr_3$ has hexagonal crystal structure and its density is 5.2 g/cm^3. The compound melts congruently at 722°C and therefore its crystals can be grown using melt-based methods such as Bridgman and Czochralski. This is fortunate because these melt-based processes are well suited for growth of large volume crystals [8]. In our research, we have used Bridgman method for growing $CeBr_3$ crystals because this technique is easy to implement and it can provide good indication of the feasibility of producing high quality crystals of $CeBr_3$ from the melt. Ultra-dry $CeBr_3$ was used with 99.99% purity. A two zone Bridgman furnace was used with temperature in the hotter zone above the melting point of $CeBr_3$ (722°C) and that of the cooler zone less than 722°C. $CeBr_3$ crystals ($< 1\ cm^3$) were grown in quartz ampoules using the Bridgman method. Small sections were cut from the solid ingots and polished using nonaqueous slurries (due to hygroscopic nature of $CeBr_3$) prepared by mixing mineral oil with Al_2O_3 grit. Some crystals were then packaged to prevent long exposure to moisture.

III. Scintillation Properties

Scintillation properties of small Bridgman grown $CeBr_3$ crystals ($\leq 0.3\ cm^3$) have been characterized. This investigation involved measurement of the light output, the emission spectrum, and the fluorescent decay time of the crystals. Energy and timing resolution of $CeBr_3$ crystals and their proportionality of response were also measured.

A. Light Output and Energy Resolution

The light output of $CeBr_3$ samples was measured by comparing their response to 662 keV γ-rays (from ^{137}Cs source) with the response of a calibrated BGO scintillator to the same isotope (see Fig. 1). This measurement involved optical coupling of $CeBr_3$ sample to a photomultiplier tube (with multi-alkali S-20 photocathode), irradiating the scintillator with 662 keV photons and recording the resulting pulse height spectrum. In order to maximize light collection, the $CeBr_3$ crystal was wrapped in reflective white Teflon tape on all faces (except the one coupled to PMT). An index matching silicone fluid was also used at the PMT-scintillator interface. A pulse height spectrum was recorded with a $CeBr_3$ crystal. This experiment was then repeated with a BGO scintillator. Comparison of the 662 keV

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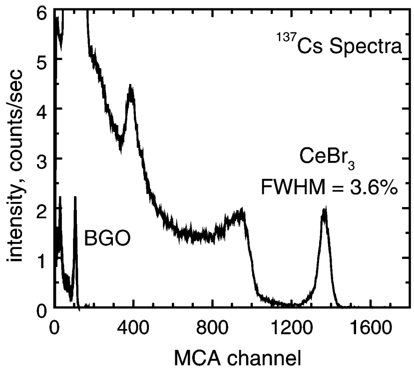

Fig. 1. $^{137}\mathrm{Cs}$ spectra collected with $\mathrm{CeBr_3}$ and BGO crystals coupled to PMT. The light output of $\mathrm{CeBr_3}$ has been estimated to be $\sim 68\,000$ photons/MeV. The energy resolution of 662 keV peak for $\mathrm{CeBr_3}$ is 3.6% (FWHM) at room temperature.

photopeak position obtained with $\mathrm{CeBr_3}$ to that with BGO provided estimation of light output for the $\mathrm{CeBr_3}$ crystal. Fig. 1 shows pulse height spectra for both $\mathrm{CeBr_3}$ and BGO under $^{137}\mathrm{Cs}$ irradiation and amplifier shaping time of 4.0 μsec. This shaping time is long enough to allow full light collection from both scintillators. The PMT bias and amplifier-gain were the same for both spectra. Based on the recorded photopeak positions for $\mathrm{CeBr_3}$ and BGO, and by taking into account the photocathode quantum efficiency for BGO and $\mathrm{CeBr_3}$, we estimated light output of $\mathrm{CeBr_3}$ crystal to be about 68 000 photons/MeV. This light output is amongst the highest values measured for inorganic scintillators [1], [9].

The energy resolution of the 662 keV photopeak recorded with $\mathrm{CeBr_3}$ scintillator has been measured to be $\sim 3.6\%$ [full width at half maximum (FWHM)] at room temperature as shown in Fig. 1. This is substantially better than the energy resolution of 6 to 7% (FWHM) at 662 keV obtained with established scintillators such as NaI:Tl and CsI:Tl. The energy resolution of $\mathrm{CeBr_3}$ crystals at 662 keV approaches that of room temperature semiconductor detectors such as CdTe and CdZnTe (2 to 3% FWHM at 662 keV).

B. Emission Spectrum

We have measured the emission spectrum of the $\mathrm{CeBr_3}$ scintillators. The $\mathrm{CeBr_3}$ samples were excited with radiation from a Philips X-ray tube having a Cu target, with power settings of 30 kVp and 15 mA. The scintillation light was passed through a McPherson monochromator and detected by a Hamamatsu R2059 photomultiplier tube with a quartz window. An emission spectrum for a $\mathrm{CeBr_3}$ sample is shown in Fig. 2. The peak emission wavelength for the $\mathrm{CeBr_3}$ sample is at about 371 nm. This emission is due to $5\mathrm{d} \rightarrow 4\mathrm{f}$ transition of Ce^{3+}. The peak emission wavelength of 371 nm for $\mathrm{CeBr_3}$ is attractive for gamma ray spectroscopy since it matches well with the spectral response of photomultiplier tubes as well as a new generation of silicon photodiodes.

C. Decay Time

Decay time spectrum of a $\mathrm{CeBr_3}$ crystal has been measured using the delayed coincidence method [10]. Fig. 3 shows the decay time spectrum recorded for a $\mathrm{CeBr_3}$ sample along with a fit (a single component described by an exponential rise and decay time constants plus a background constant) to the data.

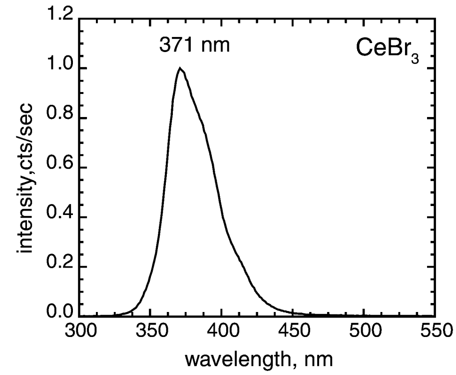

Fig. 2. Emission spectrum of a $\mathrm{CeBr_3}$ scintillator upon exposure to X-rays.

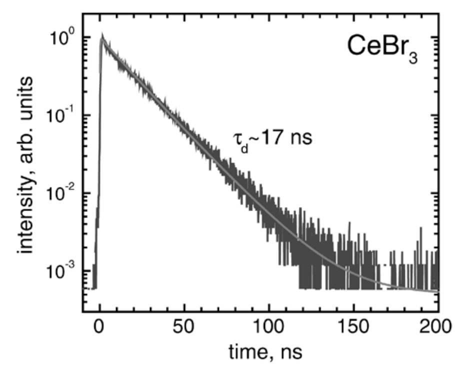

Fig. 3. A time profile of $\mathrm{CeBr_3}$ scintillation (jagged line). Solid line shows an exponential fit with a decay time constant of 17 ns.

The time constant of the measured decay is 17 ns and covers all of the integrated light output of the sample. The rise time of the scintillation has been estimated to be about 0.1 ns. The *initial photon intensity*, a figure of merit for timing applications, has been estimated to be $\sim 4000\ \mathrm{photons/(ns \cdot MeV)}$ for $\mathrm{CeBr_3}$. It is higher compared to all common inorganic scintillators, including $\mathrm{BaF_2}$ that at 2,300 $\mathrm{photons/(ns \cdot MeV)}$ is a benchmark for timing applications. The fast decay and particularly short rise time constants suggest that excitation process of Ce ions in this material is very fast. As studies of $\mathrm{CeF_3}$ [11], [12] have shown there are few ways that those ions could be excited. Those include: Direct ionization, impact ionization, and finally sequential capture of electron-hole pairs.

D. Coincidence Timing Resolution

Coincidence timing resolution of $\mathrm{CeBr_3}$ crystals has been measured. This experiment involved irradiating a $\mathrm{BaF_2}$ and a $\mathrm{CeBr_3}$ scintillator, each coupled to a fast PMT (Hamamatsu H5321) with 511 keV positron annihilation γ-ray pairs (emitted by a $^{22}\mathrm{Na}$ source). The $\mathrm{BaF_2}$-PMT detector formed a "start" channel in the timing circuit, while the $\mathrm{CeBr_3}$-PMT detector formed the "stop" channel. The signal from each detector was processed using two channels of a Tennelec TC-454 CFD. The time difference between the start and stop signals was digitized with a Tennelec TC-862 TAC and a 16-bit ADC, resulting in a TDC with 7.5 ps per bin resolution. Data were accumulated until the coincidence timing distribution had approximately 10 000 counts in the maximum bin. Fig. 4 shows a coincidence timing resolution plot acquired in this manner with $\mathrm{CeBr_3}$ and $\mathrm{BaF_2}$ crystals and the timing resolution was measured to be < 200 ps (FWHM). Using the same setup, the timing resolution

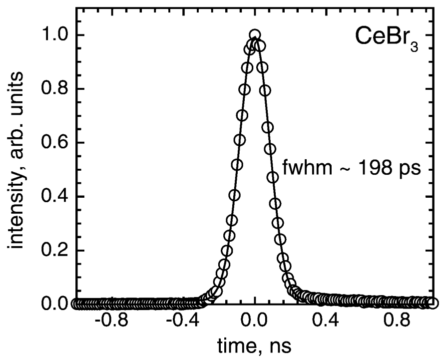

Fig. 4. Timing resolution for $\mathrm{CeBr_3}$ and $\mathrm{BaF_2}$ crystals in coincidence. For two $\mathrm{BaF_2}$ crystals in coincidence, the timing resolution with the same setup was 210 ps (FWHM).

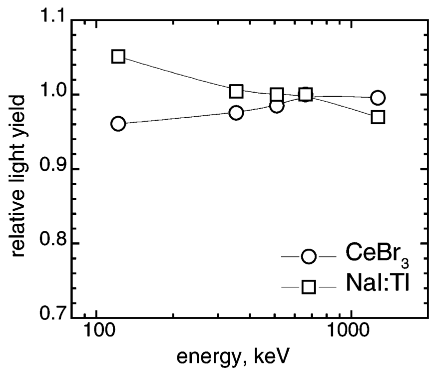

Fig. 5. Proportionality of response plot measured for $\mathrm{CeBr_3}$ and NaI:Tl crystals. Within the measured region the nonproportionality of $\mathrm{CeBr_3}$ and NaI:Tl is 4% and 9%, respectively.

of two $\mathrm{BaF_2}$ crystals in coincidence was measured to be 210 ps (FWHM).

These results confirm that $\mathrm{CeBr_3}$ is well suited for applications requiring fast response, high count-rates, and good timing resolution. Based on its high timing resolution, $\mathrm{CeBr_3}$ scintillators can be expected to provide accurate time-of-flight (TOF) information.

E. Proportionality of Response

We have evaluated the proportionality of response of $\mathrm{CeBr_3}$ scintillators. Non-proportionality (as a function of energy) in light yield can be one of the important reasons for degradation in energy resolution of established scintillators such as NaI:Tl and CsI:Tl [13]. As a result, we have measured light output of $\mathrm{CeBr_3}$ under excitation from isotopes such as $^{57}\mathrm{Co}$ (122 keV γ-rays), $^{22}\mathrm{Na}$ (511 keV and 1274 keV γ-rays) and $^{137}\mathrm{Cs}$ (662 keV γ-rays). A $\mathrm{CeBr_3}$ crystal was wrapped in Teflon tape and coupled to a PMT. Pulse height measurements were performed using standard NIM equipment with the scintillator exposed to different isotopes. Same settings were used for PMT and pulse processing electronics for each isotope. From the measured peak position and the known γ-ray energy for each isotope, the light output (in photons/MeV) at each γ-ray energy was estimated. The data points were then normalized with respect to the light output value at 662 keV energy and the results (shown in Fig. 5) indicate that $\mathrm{CeBr_3}$ is a very proportional scintillator. Over the energy range from 122 to 1274 keV, the nonproportionality in light yield is 4% for $\mathrm{CeBr_3}$, which is better than that for many established scintillators [14]. For comparison we also measured proportionality of response of NaI:Tl crystal that is shown in Fig. 5 as well. In this case the proportionality is about 9%. Higher proportionality of $\mathrm{CeBr_3}$ can be expected to enhance its energy resolution.

Overall, these measurements clearly indicate that $\mathrm{CeBr_3}$ is a promising scintillator. It has high light output, fast response, and shows very high energy and timing resolution. It also shows good proportionality of response over the γ-ray energy range from 122 to 1274 keV.

IV. SUMMARY

In our research, we have discovered a new scintillation material, $\mathrm{CeBr_3}$, for γ-ray detection. Our efforts concentrated on growth of $\mathrm{CeBr_3}$ crystals using Bridgman method, as well as characterization of the scintillation properties of these crystals. Performed measurements indicate that $\mathrm{CeBr_3}$ is a promising scintillator. It has high light output, fast response, and shows good energy and timing resolution. Based on its performance this new scintillation material appears to be very promising for applications such as: Medical imaging (positron emission tomography (PET) and single photon emission computed tomography (SPECT), nuclear and particle physics research, X-ray diffraction, nondestructive evaluation; nuclear treaty verification and nonproliferation monitoring, environmental cleanup, and geological exploration. Preparation of large crystals of $\mathrm{CeBr_3}$ and evaluation of their properties is necessary in order to investigate the full potential of this remarkable scintillator.

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