CeBr₃ Scintillator Development for Possible Use in Space Missions

W. Drozdowski, P. Dorenbos, A. J. J. Bos, G. Bizarri, A. Owens, and F. G. A. Quarati

Abstract—CeBr₃ crystals have been studied to assess their utility as potential gamma ray spectrometers for future ESA planetary missions. Pulse height spectra, scintillation time profiles, X-ray excited emission spectra, and photoluminescence spectra have been recorded as a function of temperature between 78 and 600 K. In addition, the influence of exposing CeBr₃ to various doses of gamma rays from a strong ⁶⁰Co source on its scintillation performance has been investigated.

Index Terms—CeBr₃, energy resolution, LaBr₃:Ce, radiation damage, scintillation yield, scintillator, self-absorption.

I. INTRODUCTION

N PREVIOUS papers [1], [2] we presented main results on our "Gamma Ray Scintillator Development" program, focused on studying the feasibility of using lanthanum bromide scintillators as gamma ray detectors for the European Space Agencies (ESA) BepiColombo Cornerstone mission to the planet Mercury [3]–[5]. We identified LaBr₃:5%Ce as a good choice for such a task due to its high light output, excellent energy resolution, low value for absorption and scattering losses, and very good proton radiation hardness. Only the resistance of this material to high dose gamma radiation was to some extent disappointing: a gamma dose of 1 kGy decreased the photoelectron yield of a $\sim 1 \text{ cm}^3$ sample by 8% and increased its energy resolution at 662 keV from 3.0 to 3.8%. Although this drawback did not undermine the utility of LaBr₃:5%Ce for the BepiColombo mission, it motivated us to extend the area of interest to other scintillators, which could be useful for future space applications.

While investigating proton resistance and scintillation properties of lanthanum bromide as a function of cerium concentration [1], we also studied cerium bromide (referred to as LaBr₃:100%Ce). Like LaBr₃:5%Ce, CeBr₃ proved to be very resistant to protons of energies and fluences up to 100 MeV and 10^{12} protons/cm². For small irregular samples of CeBr₃, placed on a photomultiplier tube (PMT) window without any coupling grease, we observed photoelectron yields

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up to 17100 phe/MeV and energy resolutions close to 4.0 % at 662 keV. Although in the same study similar scrap samples of LaBr₃:5%Ce displayed yields up to 25100 phe/MeV together with an excellent resolution of 2.5%, we kept in mind that the growth technology of CeBr₃ had not been optimized and there might still be some room for improvement; note that Shah *et al.* reported a resolution of 3.6% for <1 cm³ CeBr₃ [6]. Moreover, CeBr₃ has a slightly higher density (5.18 vs. 5.07 g/cm³) and probably a lower intrinsic count rate than LaBr₃:5%Ce, the latter being one of the most important criteria for ESA. Therefore CeBr₃ is an interesting candidate for further development directed at space applications.

In this paper we present the results of pulse height measurements performed on small irregular samples of CeBr₃ and a cylindrical $\otimes 0.5'' \times 0.5''$ sample with both flat surfaces polished. Based on comparison of corresponding pulse height spectra we estimate the dependence of photoelectron yield and energy resolution of CeBr₃ on crystal size. We show radio- and photoluminescence spectra, as well as scintillation time profiles, recorded at various temperatures between 78 and 600 K. We discuss the effect of self-absorption in CeBr₃ and its influence on scintillation decay times. We also examine the yield as a function of temperature. Finally, we demonstrate the effect of 1–100 kGy gamma irradiation on yield, resolution and proportionality of CeBr₃. With respect to gamma radiation hardness the current CeBr₃ samples turn out to be better than the LaBr₃:5%Ce samples studied previously [2].

II. MATERIALS AND EXPERIMENT

The crystals of CeBr₃ and LaBr₃:5%Ce were grown by Saint-Gobain Cristaux & Détecteurs (SGC&D). Single samples were cleaved out from larger volumes of scrap material. In addition, a cylindrical $0.5'' \times 0.5''$ CeBr₃ sample with polished flat surfaces was prepared by SGC&D.

Room temperature pulse height spectra were collected under 662 keV gamma excitation from a ¹³⁷Cs source. For proportionality studies we also employed a ²⁴¹Am, a ²²Na, and a variable-energy X-ray source. The samples were coupled to the quartz window of a PMT with Viscasil grease (General Electric, 60000 cSt). To improve the light collection efficiency, several layers of dried teflon tape were used in a configuration forming a reflective "umbrella" covering the crystal. The output signal from a Hamamatsu R1791 photomultiplier tube, supplied with high voltage of 600 V, was processed by a custom-made preamplifier, an Ortec 672 spectroscopy amplifier, an Ortec AD114 analog-to-digital converter, and a multichannel analyzer. PMT gain saturation due to high peak currents was avoided by using a voltage divider described in [7]. From the channel position

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Fig. 1. A^{137} Cs pulse height spectrum of the $\oslash 0.5'' \times 0.5''$ CeBr₃ crystal. The data symbols come from the experiment; the solid curve is a Gaussian fit.

of the 662 keV photopeak in the pulse height spectra and the mean of the single photoelectron response peak, the scintillation yield expressed as the number of photoelectrons from the PMT photocatode detected per unit of energy deposited in the crystal was obtained. To provide a compatibility with previous measurements on LaBr₃:5%Ce, the shaping time was set at 3μ s.

Temperature-dependent pulse height spectra and scintillation time profiles were recorded with a setup described in detail by Bizarri *et al.* [8]. The crystals were kept in clean vacuum inside a Janis cryostat and excited by a 137 Cs source. The scintillation decay measurement was based on the delayed coincidence single photon counting method [9].

A typical setup consisting of a Philips PW2253 X-ray tube with a Cu anode, operated at 60 kV and 20 mA, an Acton Research Corporation VM504 monochromator, a Hamamatsu R943-02 photomultiplier, a Janis cryostat, and a LakeShore 331 programmable temperature controler, was used to record X-ray excited emission spectra at various temperatures ranging from 78 to 600 K. For optical excitation a FL-1039 xenon lamp combined with a Jobin Yvon Gemini 180 monochromator was employed. Both radio-and photoluminescence spectra were corrected for the spectral sensitivity of the detection part.

A Gammacell 220 Excel irradiator with a 60 Co source inside was used to expose the crystals to gamma-rays. The dose rate was about 3.8 kGy/h. The samples were transported to the irradiator in air-tight aluminum boxes. The doses from 1 to 100 kGy were delivered by varying the irradiation time.

III. RESULTS AND DISCUSSION

A. Photoelectron Yield and Energy Resolution

To determine photoelectron yield and energy resolution of CeBr₃, we measured pulse height spectra of four scrap samples and a cylindrical $0.5'' \times 0.5''$ crystal. The small samples had parallelepiped-like shapes, with the largest faces of $0.5-1 \text{ cm}^2$ areas. In each measurement, one of these faces was coupled to the PMT window. An example of a pulse height spectrum is

TABLE I PHOTOELECTRON YIELD Y AND ENERGY RESOLUTION R of CEBr₃AS FUNCTIONS OF CRYSTAL HEIGHT h



Fig. 2. Photoelectron yield of $CeBr_3$ samples as a function of height. The data symbols are the values from Table I; the solid curve is a fit based on (1).

shown in Fig. 1, whereas the values of photoelectron yield and energy resolution derived from all recorded spectra are collected in Table I. Note that the observed yields are generally higher than reported in [1] for LaBr₃:100%Ce due to the use of the optical coupling grease.

The 0.6 mm high sample of CeBr₃ displays a photoelectron yield of 20300 phe/MeV, which is about 25% less than measured for LaBr₃:5%Ce samples of similar size, under identical experimental conditions. The 3.9% energy resolution of CeBr₃ at 662 keV, although very good itself, is also inferior to 2.5% observed for LaBr₃:5%Ce. Larger crystals of CeBr₃ show lower yields and increased resolutions. For the $0.5'' \times 0.5''$ sample we get 16000 phe/MeV and 5.2%, respectively.

The decrease of light output with increasing crystal height is a known effect, already characterized quantitatively for some oxide scintillators [10], [11], and recently also for LaBr₃:5%Ce [1]. A simple two-ray ("2R") model proposed by Wojtowicz *et al.* [10] introduces a loss parameter μ , describing the scintillation light loss inside the material caused by optical absorption and photon scattering. The square data symbols in Fig. 2 correspond to the photoelectron yields in Table I. The solid curve results from fitting the "2R" equation

$$Y(h) = Y_0 \frac{1 - e^{-2\mu h}}{2\mu h}$$
(1)

and provides the following values of fit parameters: $\mu = 0.203 \pm 0.017 \text{ cm}^{-1}$, and $Y_0 = 20320 \pm 170 \text{ phe/MeV}$. Hence in CeBr₃ the scintillation light loss due to self-absorption and scattering is larger, compared to LaBr₃:5%Ce [1], by a factor of 3. This



Fig. 3. Average photoelectron yields of CeBr₃ and LaBr₃:5%Ce as functions of excitation energy. Standard deviations are shown.



Fig. 4. Room temperature X-ray excited emission spectra of $CeBr_3$ and $LaBr_3{:}5\%Ce.$

feature makes it more difficult to maintain high light output of CeBr₃ with increasing crystal size.

Fig. 3 presents average photoelectron yields of CeBr₃ and LaBr₃:5%Ce, measured as functions of excitation energy on several irregular (a few cm³) samples of each material. CeBr₃ appears to be a less proportional scintillator than LaBr₃:5%Ce, which contributes to its inferior energy resolution [12].

B. Radio- and Photoluminescence

Room temperature X-ray excited emission spectra of CeBr₃ and LaBr₃:5%Ce are shown in Fig. 4. Compared to LaBr₃:5%Ce, the Ce³⁺5d - 4f luminescence in CeBr₃, peaking at 390 nm, is clearly shifted to longer wavelengths, and its doublet structure due to spin-orbit splitting of the $4f^2F_{5/2}$ and $^2F_{7/2}$ levels, is less marked. We note that the magnitude of the shift is larger than reported by Shah *et al.* [6], who observed a maximum at 370 nm.



Fig. 5. X-ray excited emission spectra of CeBr₃ at various temperatures.



Fig. 6. Photoluminescence excitation and emission spectra of $\rm CeBr_3$ at 300 and 360 K.

Fig. 5 provides an overview of CeBr₃ radioluminescence spectra, displayed on energy scale as a function of temperature. Between 300 and 600 K the spectra are formed by the 5d - 4femission of Ce³⁺ ions. The high-energy side shifts to lower wavenumbers with temperature, whereas the low-energy side remains unchanged. Such a tendency suggests the presence of a self-absorption mechanism, originating in overlapping of the 4f - 5d absorption with the 5d - 4f emission. As a consequence of thermal broadening of the former, the width of the latter decreases with temperature. Below 300 K an anticorrelation between the emission bandwidth and temperature is maintained. In addition, an extra band of unknown nature arises on the low-energy side of the cerium band, around 420 nm. Its relative contribution to radioluminescence of CeBr₃ increases towards low temperature, getting close to 10% at 78 K.

TABLE II Values of Scintillation Decay Time Constants τ_1 , τ_2 With Contributions, Reabsorption Ratio ξ , and SWL-TO-LWL Ratio I_{SWL}/I_{LWL} in CeBr₃ as Functions of Temperature T

<i>T</i> (K)	$ au_1$ (ns)	calculated ξ	I_{SWL}/I_{LWL}		$- \tau_2(ns)$
			calculated	measured	- t ₂ (13)
78	17.4 (97%)	0.0354	1.79	0.349	717 (3%)
100	17.3 (97%)	0.0267	1.81	0.371	529 (3%)
150	17.2 (97%)	0.0179	1.82	0.224	503 (3%)
200	17.4 (97%)	0.0354	1.79	0.134	461 (3%)
300	18.7 (98%)	0.140	1.60	0.0763	432 (2%)
400	20.3 (99%)	0.250	1.39	0.0483	427 (1%)
500	22.2 (99%)	0.360	1.19	0.0942	418 (1%)
600	24.6 (99%)	0.475	0.974	0.179	253 (1%)



Fig. 7. Scintillation time profiles of CeBr₃ at 78 and 600 K.

Fig. 6 presents excitation and optically excited emission spectra of CeBr₃, recorded at 300 and 360 K. The excitation spectra, monitoring the 390 nm emission, consist of a distinct excitonic peak at 225 nm, observed previously also in LaBr3:Ce [13], and two $Ce^{3+}4f - 5d$ bands with a hardly resolvable structure due to the 100% cerium concentration. At 300 K an additional band of unknown origin appears around 330 nm. This band can be permanently reduced upon heating the crystal once to at least 360 K. Moreover, such a treatment reveals that there is an overlap between the cerium absorption and emission bands. The emission spectra in Fig. 6, excited at 280 nm, are similar in shape to the X-ray excited spectrum in Fig. 4. The photoluminescence spectrum measured at 360 K, however, shows an increased contribution of the short-wavelength side, compared to 300 K. In this way it resembles more the spectrum published by Shah et al. [6].

All the observations concerning radio- and photoluminescence spectra indicate that we are dealing with three different processes. An apparent overlap of cerium absorption and emission bands is responsible for self-absorption in CeBr₃ and thermally-dependent changes of the Ce³⁺5d - 4f bandwidth. Below room temperature the shape of the radioluminescence spectra is affected by a band around 420 nm. Finally, the 330 nm absorption band produces a distortion of the short-wavelength side of the $Ce^{3+}5d-4f$ emission. The influence of these processes on light output of CeBr₃ will be discussed later on.

C. Scintillation Kinetics

Scintillation time profiles of CeBr₃ were recorded at various temperatures between 78 and 600 K and fitted with double-exponential decay curves. Two extreme cases are compared in Fig. 7. At liquid nitrogen temperature ca. 97% of the scintillation light is emitted in a fast component with a time constant of 17.4 ns. Besides, a slow component of 717 ns is present. At 600 K the fast decay time constant is prolonged to 24.6 ns, whereas the slow decay time constant is shortened to 253 ns. Systematic changes of both decay times with temperature are summarized in Table II; the errors are below 2% for τ_1 and 4% for τ_2 . Note that in spite of small uncertainties obtained from fitting, the values for the slow component may be less accurate, as the measured profiles were distorted by afterpulses.

To explain the thermal prolongation of the fast decay time constant in CeBr₃, we focus our attention again on the process of self-absorption. In literature its influence on luminescence decay times is often referred to as "radiation trapping" and several examples with corresponding mathematical models can be found [14]–[16]. In our approach we combine and extend these models, introducing the following assumptions:

- a) due to spin-orbit splitting of the two 4f levels the Ce³⁺5d 4f emission consists of two components: short-wavelength luminescence (SWL) and long-wavelength luminescence (LWL), terminating at ${}^{2}F_{5/2}$ and ${}^{2}F_{7/2}$, respectively;
- b) if there was no self-absorption, α of all photons would be emitted as LWL and (1α) as SWL, both with a decay time constant of τ_0 ;
- c) in the presence of self-absorption ξ of SWL photons are absorbed by Ce³⁺ ions, while $(1-\xi)$ of SWL photons and all LWL photons are not absorbed and leave the crystal;
- all the absorbed SWL photons are re-emitted as SWL or LWL; their further distribution is subject to b) and c).

Using a similar method to the one described in [14], we derive a formula expressing the observed decay time τ , prolonged by radiation trapping, as a function of reabsorption probability ξ

$$\tau = \frac{\tau_0}{1 + (\alpha - 1)\xi}.$$
 (2)



Fig. 8. A significant difference between the values of SWL-to-LWL ratios, based on scintillation time profiles and X-ray excited emission spectra of CeBr₃.

Although (2) implies that $\tau > \tau_0$, it specifies no explicit relation between the decay time and temperature. This relation, however, is hidden inside the parameter ξ , which can be found experimentally. Adapting the approach of Wojtowicz *et al.* [16] to our case, we get a link between the reabsorption probability ξ and relative intensities I_{SWL} , I_{LWL} of the SWL and LWL bands

$$\xi = 1 + \frac{\alpha}{\alpha - 1} \frac{I_{\rm SWL}}{I_{\rm LWL}}.$$
(3)

For further analysis we take a value of $\alpha = 0.35$ from a liquid helium temperature emission spectrum of LaBr₃:0.5%Ce [13], for which absence of self-absorption is assumed. The Ce^{3+} radiative lifetime in CeBr₃ we fix at $\tau_0 = 17$ ns. Employing (2) and using the measured fast decay constants τ_1 of CeBr₃, for each temperature we calculate the value of reabsorption probability ξ and, in turn, with (3) the value of SWL-to-LWL ratio $I_{\rm SWL}/I_{\rm LWL}$. Alternatively, the $I_{\rm SWL}/I_{\rm LWL}$ values can be derived from Gaussian fits to the radioluminescence spectra of CeBr₃. As shown in Table II and Fig. 8, the SWL-to-LWL ratios measured by the X-ray excited emission spectra are definitely too low, compared to those evaluated from scintillation decays. We suspect that beside an "ordinary" cerium self-absorption, responsible for thermal prolongation of the fast decay time, an efficient "extraordinary" absorption exists in CeBr₃. SWL photons absorbed in the latter process are never re-emitted, hence they do not affect the scintillation time profiles. Therefore we do not observe any further prolongation of the decay time. Nevertheless, the SWL band is strongly suppressed, which may contribute to lower yield of CeBr₃. The "extraordinary" absorption may be related to the 330 nm excitation band in Fig. 6.

D. Thermal Dependence of Yield

Fig. 9 displays the values of photoelectron yield of $CeBr_3$ as a function of temperature, determined using two different experimental techniques: pulse height and radioluminescence measurements. Based on the pulse height spectra we observe a downward trend with temperature, with a 22% loss of yield at



Fig. 9. Yield of $CeBr_3$ as a function of temperature, determined by two different techniques. Both curves are normalized; 1.0 stands for yield at 300 K.

TABLE III PHOTOELECTRON YIELD Y AND ENERGY RESOLUTION R of CeBr₃ Before AND AFTER GAMMA IRRADIATION

Cumulative dose (kGy)	Y(arb. units)	<i>R</i> at 662 keV (%)
0	1.000	3.9 ± 0.1
1	0.984 ± 0.011	4.0 ± 0.2
11	0.964 ± 0.006	4.0 ± 0.2
111	0.922 ± 0.013	4.2 ± 0.2

600 K relative to 100 K. The thermal stability of yield in CeBr₃ is poorer than in LaBr₃:5%Ce, for which such a loss does not exceed 15% [8]. The areas under the X-ray excited emission spectra confirm the decrease of yield with temperature. Moreover, they indicate a much larger scale of this effect, particularly below room temperature. This suggests a contribution of light emitted in very slow components, which could not be recorded in pulse height and decay measurements because of the 3 μ s shaping time. These hypothetical components might be linked to the low-temperature band at 420 nm in the CeBr₃ radioluminescence. To prove this, however, a time-resolved spectroscopy technique should be employed.

E. Gamma Resistance

Studies of gamma radiation hardness of CeBr₃ were performed on three parallelepiped-like scrap samples with volumes up to 0.5 cm². Pulse height spectra were recorded directly before and immediately after each irradiation treatment. Time intervals of 3–4 days between consecutive irradiations were introduced to observe a possible recovery. In these periods both photoelectron yield and energy resolution were monitored continuously, however no significant changes were noticed. Apparently there is no spontaneous recovery in ⁶⁰Co-irradiated CeBr₃ on a time scale of a few days.

Gamma radiation induced deterioration of yield and resolution of CeBr₃ is characterized quantitatively in Table III. In the "Y" column average values of yield with standard deviations between particular samples are given; 1.000 stands for yield before the first irradiation. Since the initial resolutions of the three



Fig. 10. Photoelectron yield and energy resolution of $CeBr_3$ and $LaBr_3:5\%Ce$ as functions of absorbed gamma dose.

samples were not the same due to differences in size and surface quality, data in the "R" column refer to the best one. Nevertheless, for the two other samples the increase of resolution following each gamma dose was also very small, not exceeding a few tenths of percentage point upon 111 kGy.

To assess the gamma radiation hardness of CeBr₃, in Fig. 10 we compare the deterioration of its yield and resolution with that of a $\oslash 15 \text{ mm} \times 5 \text{ mm} \text{ LaBr}_3:5\%$ Ce crystal, reported in [2]. Although both materials show a decreased photoelectron yield and an increased energy resolution after each ⁶⁰Co exposure, the scale of this effect turns out to be smaller in CeBr₃. In particular, in LaBr₃:5%Ce a kind of "damage shock" can be observed, i.e., the most significant change of yield and resolution takes place after the first irradiation. In CeBr₃ such effect does not occur. The yield of CeBr₃ seems to be a nearly linear, slowly decreasing function of delivered dose, whereas the increase of energy resolution is almost negligible. Apparently, for some unknown reason CeBr₃ is more resistant to high dose gamma radiation than LaBr₃:5%Ce.

IV. CONCLUSION

Of the LaBr₃:Ce scintillator family, cerium bromide (LaBr₃:100%Ce) is a noteworthy modern scintillator, characterized by high photoelectron yield above 20000 phe/MeV, excellent energy resolution of 4%, fast scintillation response of 19 ns, and reasonably good proportionality and thermal stability of yield. At present in all these respects CeBr₃ is inferior to LaBr₃:5%Ce, nevertheless this status may be biased by advanced optimization of crystal growth and scintillation performance of the latter. On the other hand, CeBr₃ seems to be more gamma radiation hard than LaBr₃:5%Ce. Furthermore, it has a higher density and is expected to display a lower intrinsic count rate. All these features make CeBr₃ an interesting candidate for space applications. First efforts to improve CeBr₃ should be probably aimed at reducing absorption processes in the crystal in order to increase light output and enable a growth of large crystals with yield and resolution maintained at a good level.

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