



Scintillation properties of Zr co-doped Ce:(Gd, La)₂Si₂O₇ grown by the Czochralski process



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HIGHLIGHTS

- Czochralski growth of Ce:(Gd,La)₂Si₂O₇ single crystals.
- Co-doping effect of a stable tetravalent ion in Ce:(Gd,La)₂Si₂O₇ system.
- Photon nonproportional response of Zr co-doped Ce:(Gd,La)₂Si₂O₇.

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ABSTRACT

(Gd_{0.75}Ce_{0.015}La_{0.235})₂Si₂O₇ (Ce:La-GPS) single crystals co-doped with 0, 100, 200, 500 and 1000 ppm Zr were grown by the Czochralski process, and their scintillation properties were investigated. We investigated the co-doping effect of a stable tetravalent ion in Ce:La-GPS for the first time. The scintillation decay times in the faster component were shortened with increasing the Zr concentration. While the non-co-doped sample showed ~63 ns decay time, the Zr 100, 200, 500 and 1000 ppm co-doped samples showed ~61, ~59, ~57, ~54 ns, respectively. Additionally, light output, photon nonproportional response (PNR) and other optical properties were investigated.

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1. Introduction

Silicate type scintillators such as Ce:Gd₂SiO₅ (Ce:GSO) (Takagi and Fukuzawa, 1983) and Ce:(Lu,Y)₂SiO₅ (Ce:LYSO) (Cooke et al., 2000) have been applied in various fields: medical imaging, oil well logging and astronomy because of their high light output, decay time etc. Ce:GSO shows a good light output of ~12,500 photons/MeV and fast scintillation decay time of ~56 ns (Melcher et al., 1996). Moreover, some researchers have reported new promising scintillation materials, pyrosilicate-type scintillators

such as Ce:Gd₂Si₂O₇ (Ce:GPS) (Kawamura et al., 2006) and Ce:Lu₂Si₂O₇ (Ce:LPS) (Pidol et al., 2004) recently. They show more than twice as high light output when compared with Ce:GSO (Kawamura et al., 2007). Here, melt growth of the heavy Ce doped (~10%) GPS can be realized, and a large-size crystal with a diameter of over 1 inch is expected to be grown, which is not the case for the pure GPS. However, the heavily Ce doped GPS, shows a light yield decrease due to concentration quenching and self-absorption. Recently discovered Ce:(Gd,La)₂Si₂O₇ (Ce:La-GPS) overcomes these problems by substituting major part of Ce³⁺ for La³⁺, which has almost same ionic radius with that of Ce³⁺ (Suzuki et al., 2012). Scintillation properties of Ce:La-GPS grown by floating-zone (FZ) method have been investigated in previous research (Suzuki et al., 2012) (Kurosawa et al., 2013); 9%-La substituted crystal showed good light output (~36,000 photons/MeV) and good energy

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resolution (~5.0%, 662 keV, FWHM). Furthermore, photon non-proportional response (PNR) of the FZ-grown Ce:La-GPS was evaluated with a Si-avalanche photodiode (Si-APD) (Kurosawa et al., 2013), and our previous work reported the PNR of Cz grown Mg co-doped Ce:La-GPS single crystal with a photomultiplier tube (PMT) (Murakami et al., submitted for publication). Although the FZ-grown sample had a small size, we have succeeded in growing larger size of Ce:La-GPS scintillators by the Czochralski (Cz) process up to ~1 inch diameter. Here, we confirmed that if at least 25–50% of La is substituted for Gd site, then the GPS single crystals can be grown by the Cz process (Yoshikawa et al., 2015).

Shimura et al. reported the co-doping effect of several metal copopants on the properties of Ce:GSO: the light output for 100–300 ppm of Zr co-doping was improved by ~20% for conventional sample (Ce:GSO) (Shimura et al., 2006). Although the mechanism of the improvement has not been revealed clearly, Shimura et al. have pointed out the possibility that originally present Ce⁴⁺ which causes degradation of light yield might be reduced in the GSO crystal by the charge compensation between Ce⁴⁺ and Zr⁴⁺ which replaces the Gd³⁺ site. Thus, in this work, we report the Zr co-doping effect on Ce:La-GPS single crystal for several Zr concentrations.

2. Material and method

(Gd_{0.75}Ce_{0.015}La_{0.235})₂Si₂O₇ single crystals co-doped with 0, 100, 200, 500 and 1000 ppm Zr, were grown by the Cz process. At least 99.99% purity of Gd₂O₃, La₂O₃, SiO₂, CeO₂ and ZrO₂ powders were used as starting materials. The mixture of powders were melted in an iridium crucible with a diameter of 50 mm in argon and 2%-oxygen mixed atmosphere. Bulk crystals with 1 inch diameter were pulled up using the single crystal seed with the composition of (Gd_{0.75}Ce_{0.015}La_{0.235})₂Si₂O₇. Here, the pulling rate and the rotation rate were 0.5 mm/h and 10 rpm, respectively.

The grown crystals were cut and mirror-polished. Parts of the grown crystals were crushed into powders for X-ray diffraction (XRD) analysis, which was performed within the 2θ range of 10–60° using a diffractometer (Rigaku, RINT 2000). Here, the X-ray source was Cu-Kα line with an acceleration voltage of 40 kV and beam current of 40 mA. Subsequently, transmittance and reflectance spectra were measured using spectrometers: V-550 (JASCO) and UV-2550 (SHIMADZU), respectively. The samples dimensions were 5 mm × 5 mm × 0.2 mm. Photoluminescence (PL) emission and excitation spectra were measured with an absolute PL quantum yields measurement system (Hamamatsu Photonics C9920-02G).

Pulse height spectra (for light output and energy resolution) and scintillation decay times were measured at room temperature using a PMT (Hamamatsu Photonics R7600U-200). The measured samples had the dimension of 5 mm × 5 mm × 1 mm. The samples were covered by Teflon™ tape as a reflector and irradiated with a gamma-ray source (¹³⁷Cs, 662 keV). The scintillation photons were detected by the PMT. Then the signal was amplified (ORTEC 113), shaped (ORTEC 572A), and red out with a multi-channel analyzer (AMPTK 8000A). A Ce:Gd₂SiO₅ (Ce:GSO) single crystal whose light yield has been determined with a Si-APD was also measured as a reference. The details of this Si-APD measurement are described in Ref. (Kurosawa et al., 2013) (Murakami et al., 2015, submitted for publication). Scintillation decay times were measured with the PMT and an oscilloscope (Tektronix TDS3052B).

Furthermore, non-proportionality was evaluated with X-ray and gamma-ray sources: ²⁴¹Am (59.5 keV), ⁵⁷Co (14.4, 122 keV), ¹⁵²Eu (40.1, 122, 245, 344 keV), ¹³³Ba (31, 81.0, 276, 303, 356 keV), ¹³⁷Cs (32.2, 662 keV), ²²Na (511 keV), ¹⁰⁹Cd (22.2, 88.0 keV). Thus, this measurement was performed within the energy range of 14.4–662 keV. The measurement systems and the temperature

conditions were the same as those above.

3. Results and discussion

Fig. 1 shows a photograph of Zr 100, 200, 500 and 1000 ppm co-doped Ce:La-GPS single crystals grown by the Cz process. With increasing the Zr amount, bubbles at the crystal tail side appear. The XRD patterns of every grown crystals were corresponded, and they were considered to be monoclinic (Monteverde and Celotti, 1999). The XRD patterns for every grown crystals were corresponded to be the monoclinic structure in the pyrosilicate group (Monteverde and Celotti, 1999).

In Fig. 2 the absorption coefficient spectra are provided for all the sample set. The position of five broad absorption bands within 190–360 nm is consistent with 4f–5d^{1,2,3,4,5} absorption transitions of Ce³⁺ center in La-GPS single crystal host reported in the recent literature (Jary et al., 2014) and sharp absorption lines around 275 nm and 200 nm are due to 4f–4f transitions ⁸S_{7/2}–⁶I_x and ⁸S_{7/2}–⁶G_x, respectively, of Gd³⁺ cation. Slightly different values of residual absorption in transparency region of crystals above 380 nm are most probably due to light scattering processes. Due to too high absorbance values reaching the spectrometer limit the Ce³⁺ absorption band related to 4f–5d² transition around 320 nm becomes visibly saturated.

Fig. 3(a) and (b) show PL emission (ex = 345 nm) and excitation (em = 390 nm) spectra of the Ce,Zr:La-GPS sample set, respectively. Each sample showed typical broad emission doublet of Ce³⁺ with maxima at 370 and 390 nm due to the 5d₁–4f (²F_{7/2}, ²F_{5/2}) transition of Ce³⁺ (Fig. 3(a)). Overall shape of excitation spectra was somewhat different for the Zr co-doped samples in comparison with the non-co-doped one (Fig. 3 (b)) and consistent with absorption spectra in Fig. 2, but separate 4f–5d^{1,2,3} transitions of Ce³⁺ within 270–360 nm are not well resolved in any of the samples (compare with Fig. 2) due to their high absorbance in this spectral region.

Pulse height spectra measured with a ¹³⁷Cs gamma-ray source are shown in Fig. 4. Due to the sample thicknesses were small enough to allow X ray to escape, the escape peak related to the Gd was observed besides the photoelectric peak in each spectrum. From the comparison of MCA channel number of photoabsorption peak, light output degrades with increasing Zr amount (0, 100, 200, 500 and 1000 ppm), and the values were estimated to be 42,000 ± 2,000, ~40,000 ± 2,000, ~39,000 ± 2,000, ~38,000 ± 2,000 and ~34,000 ± 2,000 photons/MeV, respectively. Here the quantum efficiency of photocathode in the PMT for the emission wavelength of GSO (~430 nm) and Ce:La-GPS (~390 nm) were assumed to be 38% and 41%, respectively. Scintillation decay times were estimated by fitting the obtained decay curves with two exponentials as shown in equation (1).

$$I(t) = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + C_0 \quad (1)$$

where A_1 and A_2 denote amplitudes and τ_1 and τ_2 denote fast and slow component decay times, respectively. Moreover, the intensity of each component was calculated as $I_i = A_i \tau_i / \sum (A_j \tau_j)$ ($i, j = 1, 2$). Table 1 lists the obtained values. It was found that the Zr co-doped samples tend to have shorter decay times in the faster component than the non-co-doped one which showed ~63 ns while the Zr 100, 200, 500 and 1000 ppm co-doped samples showed ~61, ~59, ~57, ~54 ns decay times, respectively. The decay times in faster component are still noticeably longer compared to photoluminescence decay times found in (Jary et al., 2014) which are around 30 ns. Thus, even faster component in scintillation decay is already affected (slowed down) by an energy transfer process in the transfer stage of scintillation mechanism and Zr codoping apparently diminishes its effect. At the same time, however, light output becomes degraded with increasing Zr concentration as mentioned above and there is

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