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Scintillation Properties of Y-stabilized ZrO_2 Transparent Ceramic

Yusuke Endo,* Kensei Ichiba, Satoshi Honjo, Daisuke Nakauchi, Takumi Kato, Noriaki Kawaguchi, and Takayuki Yanagida

Nara Institute of Science and Technology, Takayama, Ikoma, Nara 630-0192, Japan

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We evaluated the photoluminescence (PL) and scintillation properties of Y-stabilized ZrO_2 (YSZ) transparent ceramic (TC) compared with those of a YSZ single crystal (SC). Each material form shows the same emission origin. Afterglow levels of the SC and TC forms are 240 and 330 ppm, respectively, which are comparable to that of Tl:CsI. Considering that TC exhibits transmittance exceeding 60% in the NIR region, YSZ TC can be expected to be applied to scintillators emitting NIR photons by doping with NIR emission centers.

1. Introduction

Scintillators can immediately convert high-energy ionizing radiation into numerous low-energy photons.[1–4] The ionizing radiation can be measured by converting scintillation photons into electrical signals using photodetectors[5–7] such as Si photodiodes,[8–10] photomultiplier tubes,[11–13] and InGaAs PIN photodiodes.[14–17] These measurement systems are used in a wide range of fields, including medical imaging,[18–20] well-logging,[21–23] nuclear physics,[24–26] and security.[27–29] The prioritization of scintillation properties varies depending on the application, and no scintillators have met all the necessary criteria. For these reasons, a wide variety of scintillators have been developed with diverse compositions and material forms, including single crystals (SCs),[30–40] ceramics,[41–49] and glasses.[50–62]

The present study is focused on Y-stabilized ZrO_2 (YSZ) transparent ceramic (TC) as a scintillator host. YSZ has an effective atomic number (Z_{eff} = ~37) and a density (ρ = ~5.7 g/cm^3) surpassing those of Ce:$Y_3Al_5O_{12}$ (Z_{eff} = 32 and ρ = 4.6 g/cm^3). YSZ is applied to a variety of fields such as dentistry[63] and jewelry.[64] Although the scintillation properties of YSZ SC have been reported,[65,66] those of TC remain unexplored. Moreover, ceramics can be produced at a lower cost than SC. If YSZ TC with SC-level performance can be fabricated below the melting point of YSZ (~2700 °C), it would provide an advantage for the development of new scintillators. In this study, we evaluate the photoluminescence (PL) and scintillation properties of YSZ TC compared with those of SC to examine its potential as a scintillator.

*Corresponding author: e-mail: endo.yusuke.ey5@ms.naist.jp
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2. Experimental Setups

YSZ SC and TC were prepared using YSZ nanopowder (Tosoh, TZ-10YS) containing 10 mol% Y_2O_3 and 90 mol% ZrO_2. YSZ SC was prepared by following the procedure below. The powder was formed into a cylindrical rod by applying hydrostatic pressure; then, the obtained rod was sintered at 1400 ℃ for 8 h. The sintered rod was melted in a floating zone (FZ) furnace (Crystal Systems, FZ-T-12000-X-VPO-PC-YH). The crystal growth conditions were as follows: the pull-down speed was 100 mm/h and the rotation speeds of the upper and lower shafts were 20 and 0 rpm, respectively. After crystal growth, the relatively crack-free part was sectioned selectively and polished to make an evaluation sample. In the procedure to prepare YSZ TC, the powder was sealed in a graphite die and two punches in the spark plasma sintering (SPS) equipment (Sinterland, LABOX-300), and was subsequently sintered under the conditions shown in Fig. 1. After that, the obtained ceramic was annealed at 700 ℃ for 4 h[67] and then polished to prepare an evaluation sample.

X-ray diffraction (XRD) patterns were observed using an X-ray diffractometer (Rigaku, MiniFlex 600). Diffuse transmittance spectra were measured using a spectrophotometer (Shimadzu, Solidspec-3700). PL emission spectra and quantum yields were measured with Quantaurus-QY (Hamamatsu Photonics, C11347). PL decay curves were evaluated with Quantaurus-τ (Hamamatsu Photonics, C11367), and decay time constant (τ) values were determined by approximating the curves with exponential functions. X-ray-induced scintillation spectra, decay curves, and afterglow levels (ALs) were measured using our original experimental setups.[68,69]

3. Results and Discussion

Figure 2 shows the appearance, XRD patterns, and diffuse transmittance spectra of the YSZ SC and TC samples. The SC appears colorless and transparent, and the TC appears brownish and

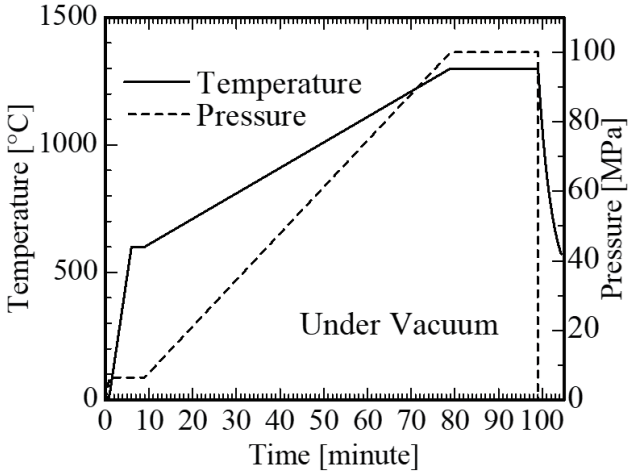

Fig. 1. SPS conditions.

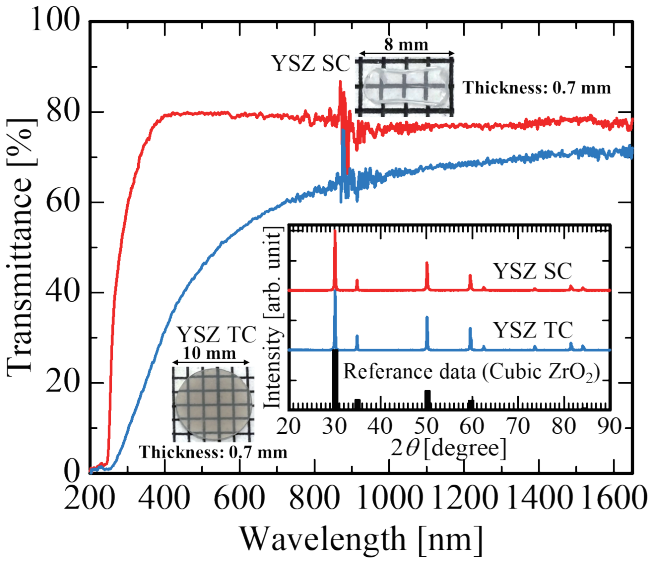

Fig. 2. (Color online) Appearance, XRD patterns, and diffuse transmittance spectra of samples.

transparent. The brownish coloration is due to oxygen vacancies generated during the SPS process.[67] The XRD patterns of the samples are consistent with the reference pattern of cubic ZrO_2,[70] and no extra peaks were detected. Therefore, the samples have a single phase of cubic ZrO_2, which belongs to the Fmm space group of the fluorite structure.[71,72] SC exhibits transparency with 80% transmittance from 400 to 1650 nm, while TC exhibits transparency and the highest transmittance at approximately 1600 nm.

Figure 3 shows PL emission spectra and decay curves with the fitted lines. Except for the instrumental response function (IRF), PL τ values of YSZ SC and YSZ TC are 11 and 14 ns, respectively. Similar spectral shapes and τ values are noted in previous reports on YSZ;[65,66] thus, the emission origin would be an oxygen vacancy. The difference in spectral shape between the SC and TC samples would be attributed to the concentration of oxygen vacancies. Quantum yields of YSZ SC and YSZ TC are 1.4 and 0.6% under excitation at 280 nm and monitoring at 350–680 nm, respectively.

Figure 4 shows the scintillation spectra and decay curves with the fitted lines. The spectra show broad emission bands with peaks at 450 nm. The difference in spectral shape between PL and X-ray excitation would be attributed to the variation in the number and distribution of excited states generated by a single wavelength and X-ray excitation. Except for the IRF, the τ values of both YSZ samples are 27 ns. Similar spectral shapes and τ values are described in previous reports on YSZ.[65,66] Considering the previous reports and the results of PL, the emission would be due to an oxygen vacancy. ALs were evaluated by comparing the emission intensity during 2 ms of X-ray irradiation with that measured 20 ms after the irradiation. The ALs of YSZ SC and YSZ TC samples are 240 and 330 ppm, respectively. These values are comparable to that of Tl:CsI (~300 ppm[69]), a commercial scintillator. The reason why YSZ SC shows a higher AL than YSZ TC would be because there are fewer defects in YSZ SC than in YSZ TC.

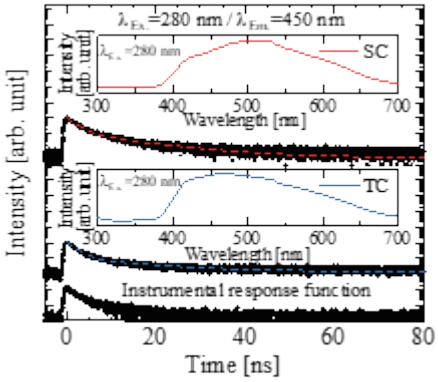

Fig. 3. (Color online) PL emission spectra and decay curves with fitted lines of samples.

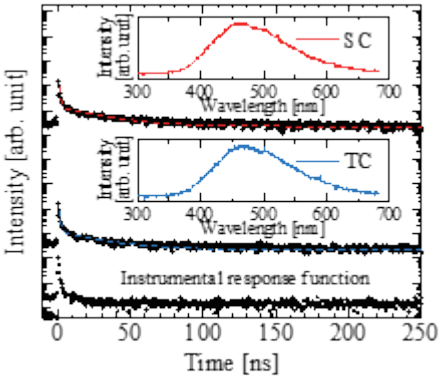

Fig. 4. (Color online) Scintillation emission spectra and decay curves with fitted lines of samples.

4. Conclusions

In the present study, we evaluated the PL and scintillation properties of YSZ TC compared with those of YSZ SC. The emission of the samples is due to an oxygen vacancy. Regarding scintillator performance, the same scintillation τ values suggest that both forms can exhibit comparable performance in photon-counting applications. On the other hand, the AL of YSZ SC is lower than that of YSZ TC: therefore, the SC form can exhibit higher performance in integrated-type applications. The TC form shows transmittance exceeding 60% in the NIR region; it can be expected to be applied to scintillators emitting NIR photons by doping with NIR emission centers. Therefore, the scintillation properties of YSZ TC doped with NIR emission centers will be evaluated in future work.

Acknowledgments

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