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Luminescence Properties of $La_2Be_2O_5$ Single Crystal Scintillator

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We synthesized an undoped $La_2Be_2O_5$ single crystal by the floating zone method and investigated its scintillation properties. The synthesized sample was confirmed to have a single-phase structure of $La_2Be_2O_5$ as shown by the powder X-ray diffraction pattern. The diffuse transmittance spectrum of the synthesized sample had transmittance at 80% in the range of 250–850 nm. The scintillation spectrum had a broad emission band from 280 to 600 nm, which was derived from unidentified lattice defects. In the scintillation decay profile, the obtained decay time constants were 1.1, 6.2, and 386 ns. From pulse height spectra, the light yield of the $La_2Be_2O_5$ single crystal was 1000 photons/MeV. In afterglow profiles, the afterglow level was 42.9 ppm.

1. Introduction

A scintillator is a type of optical material that can immediately transform ionizing radiation with high photon energy into ultraviolet-visible light with low photon energy. Its primary application is a radiation detector, which is composed of a scintillator and a photodetector. A scintillator has been widely used in various fields including nuclear medicine,[1,2] security,[3,4] astrophysics,[5] and resource exploration.[6] The required properties of a scintillator depend on its applications. However, high chemical stability, large effective atomic number (Z_{eff}), high density (ρ), fast decay time constant, high light yield (*LY*), and low afterglow level (*AL*) are generally required for a scintillator. Since a scintillator with all these required properties has not been found yet, various new scintillators have been developed.[7–13] Such development of new scintillators has been conducted in various forms: glass,[14–17] transparent ceramics,[18–20] single crystals,[21–25] organic–inorganic composites,[26,27] and plastics.[28,29]

In this study, we focused on the $La_2Be_2O_5$ single crystal. $La_2Be_2O_5$ has been studied as the host material of a scintillator in the field of radiation measurement since it has a high chemical/radiation resistance, a relatively large Z_{eff} (52.9), and a high ρ (6.1).[30,31] For example, it has been reported that the Ce-doped $La_2Be_2O_5$ single crystal has a broad emission band centered at 450 nm[30] and that the *LY* of the Ce-doped $La_2Be_2O_5$ is 58% compared with that of $Bi_4Ge_3O_{12}$.[32]

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On the other hand, the scintillation properties of the undoped $La_2Be_2O_5$ single crystal have not been studied. In a previous study of the photoluminescence (PL) properties of undoped $La_2Be_2O_5$, the emission wavelength was observed in the range of 280–600 nm due to unidentified lattice defects.(33) This emission wavelength is compatible with the wavelength sensitivity of the general photodetector. Therefore, undoped $La_2Be_2O_5$ could also be a candidate for a scintillator. In addition, it is important to understand the scintillation properties of an undoped material. One reason for this is that the energy migration efficiency and interaction probability between ionizing radiation and materials in the scintillation process basically depend on the host material. For the above reason, we synthesized the $La_2Be_2O_5$ single crystal by the floating zone (FZ) method and investigated its scintillation properties.

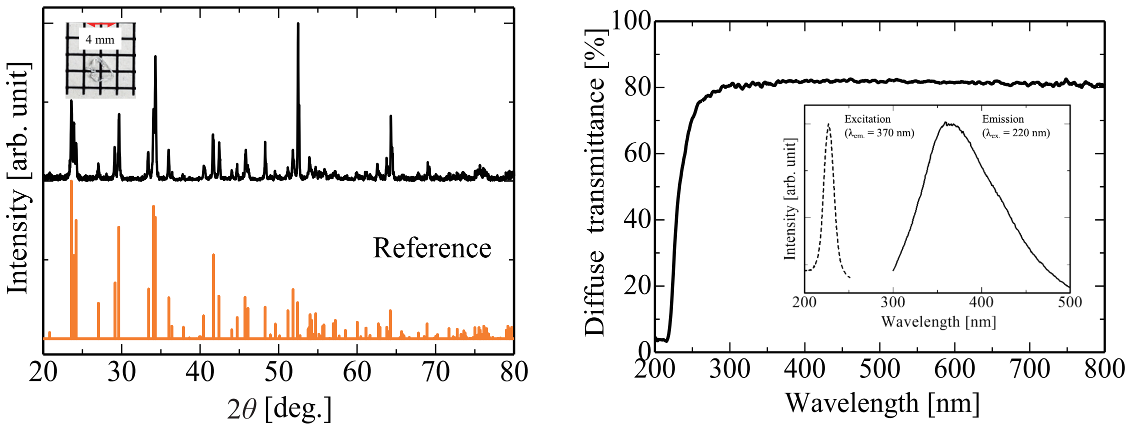
2. Experimental Method

The $La_2Be_2O_5$ single crystal was synthesized by the FZ method. La_2O_3 (99.99%) and BeO (99.99%), which were raw materials, were mixed to form into a cylindrical rod by applying hydrostatic pressure. The formed rod was sintered at 1200 ℃ for 12 h in air and then crystal growth was conducted in an FZ furnace (Canon, Machinery FZD0192). The growth parameters used were a pull-down speed of 3 mm/h and a rotation speed of 30 rpm. The obtained crystalline rod was cut to obtain the evaluation sample, and a polishing machine (Buehler, MetaServe 250) was used for polishing the surface. To confirm the crystal phase, the powder X-ray diffraction (PXRD) pattern was obtained with a diffractometer (MiniFlex600, Rigaku). The diffuse transmittance spectrum was evaluated using a spectrophotometer (SolidSpec-3700, Shimadzu). The PL excitation and emission spectra were evaluated using a spectrofluorometer (JASCO, FP-8600). The resolutions of the spectrofluorometer on the sides of excitation and emission spectra were 10 and 20 nm, respectively.

Scintillation spectra were evaluated using our original setup.(34) As the radiation source, an X-ray generator (Spellman, XRB80N100/CB) with an X-ray tube was used, and the bias voltage and tube current were 80 kV and 1.2 mA, respectively. Scintillation decay time and afterglow profiles were evaluated using an afterglow characterization system,(35) and pulse height spectra under γ-rays from ^{137}Cs were evaluated with our previous setup.(27,36)

3. Results and Discussion

Figure 1(a) shows the picture and PXRD patterns of the $La_2Be_2O_5$ single crystal. The synthesized sample appeared colorless and transparent, and the sample size was approximately $4 \times 4 \times 1.0$ mm^3. The PXRD pattern of the synthesized sample corresponded to the reference pattern ($La_2Be_2O_5$: International Centre for Diffraction Data No. 76–1652). Therefore, it was considered that the synthesized sample had a single-phase structure of $La_2Be_2O_5$. Figure 1(b) shows the diffuse transmittance spectrum and PL excitation and emission spectra of the $La_2Be_2O_5$ single crystal. The diffuse transmittance of the synthesized sample was approximately 80% in the range of 250–800 nm. In addition, there was clear absorption in the range of 220–250 nm, which would be due to the optical band gap of $La_2Be_2O_5$.(37) In the PL excitation spectrum,



(a) (b)

Fig. 1. (Color online) (a) Picture and PXRD patterns of $La_2Be_2O_5$ single crystal. The reference pattern is International Centre for Diffraction Data No. 76-1652. (b) Diffuse transmittance spectrum and PL excitation and emission spectra of $La_2Be_2O_5$ single crystal.

the excitation band was observed at approximately 230 nm, which corresponded to the absorption wavelength observed in the diffuse transmittance spectrum. Under excitation at 230 nm, a broad emission band was observed in the range of 300–500 nm. The broad emission band was reported in previous research to be attributed to unidentified lattice defects.(38)

Figure 2(a) shows the scintillation spectrum of the $La_2Be_2O_5$ single crystal. The synthesized sample had a broad emission band located in the range of 300–600 nm. The broad emission band would be derived from unidentified lattice defects.(33,38) In addition, at least two different defects would contribute to the emission since the scintillation spectrum had two emission peaks at 350 and 390 nm. Figure 2(b) shows the scintillation decay time profile of the $La_2Be_2O_5$ single crystal. The scintillation decay curve was well approximated by a sum of three exponential functions, and the obtained decay time constants were 11, 62, and 386 ns. The fastest component came from an instrumental response function (IRF). In addition, the others would be assumed to be the value related to the emission band, which was observed in the scintillation spectrum. Note that the intensities were 1793 for the first component, 578 for the second component, and 82 for the third component.

Figure 3(a) shows the pulse height spectra of the $La_2Be_2O_5$ single crystal under γ-rays from ^{137}Cs (662 keV). In the measurement, the sharping time was set to 2 μs. As a reference, $(Lu,Y)_2SiO_5$ (LYSO) with an *LY* of 22000 photons/MeV was used. The synthesized sample had a clear photoabsorption peak at 364 ch, and its *LY* was calculated to be 1000 photons/MeV after considering a peak position with LYSO and the quantum efficiency of the photomultiplier tube. Figure 3(b) shows the afterglow profile of the $La_2Be_2O_5$ single crystal. *AL* was defined by the signal intensity at 20 ms after X-ray irradiation with the pulse width of 2 ms. The *AL* of the synthesized sample was 42.9 ppm, which was lower than that of the Tl-doped CsI (~300 ppm) evaluated using the same measurement system.(39)

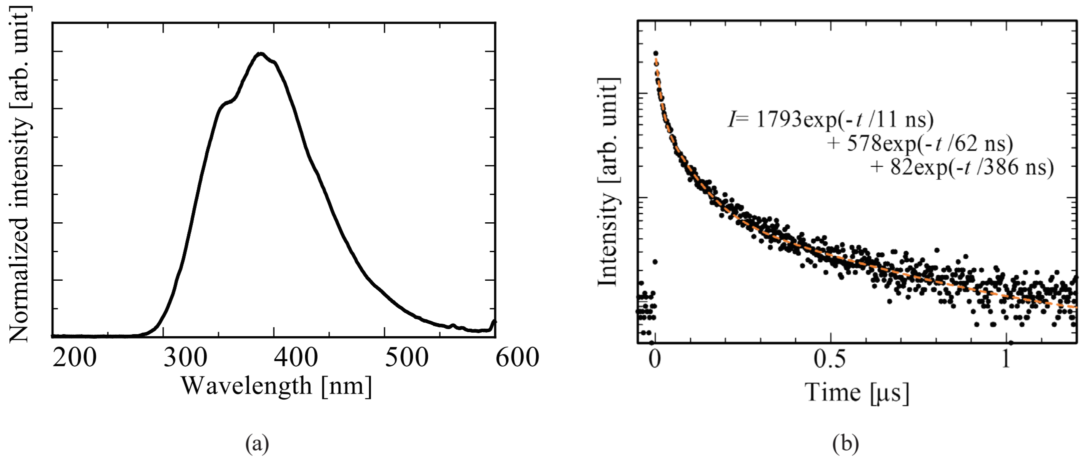


Fig. 2. (Color online) (a) Scintillation spectrum and (b) scintillation decay time profile of $La_2Be_2O_5$ single crystal.

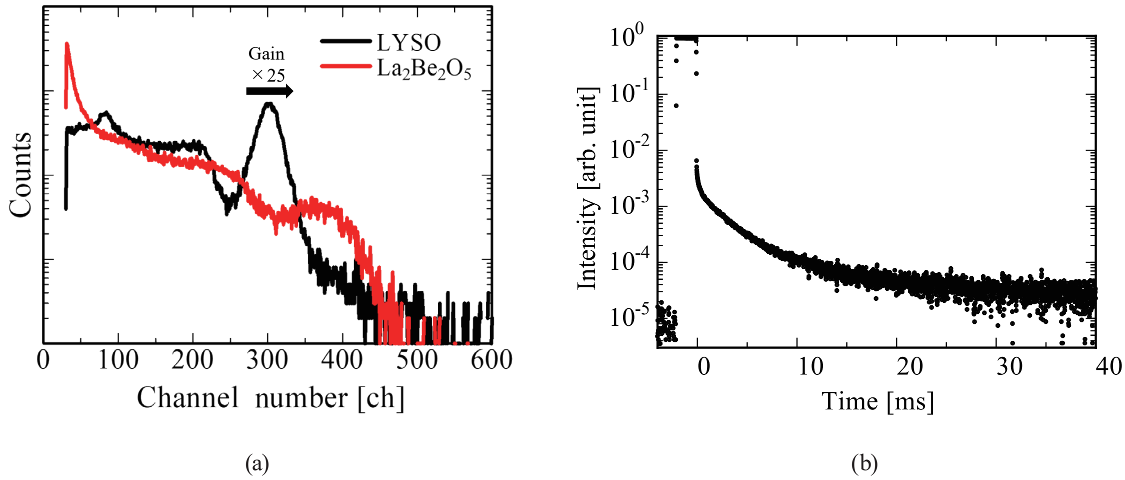


Fig. 3. (Color online) (a) Pulse height spectra of LYSO and $La_2Be_2O_5$ single crystal under γ-rays from ^{137}Cs. (b) Afterglow profile of $La_2Be_2O_5$ single crystal.

4. Conclusion

In this study, we synthesized a $La_2Be_2O_5$ single crystal to evaluate its scintillation properties. From the PXRD pattern, the synthesized sample had a single-phase structure of $La_2Be_2O_5$. In the scintillation spectrum, the $La_2Be_2O_5$ single crystal had a broad emission band in the range of 280–600 nm, which was due to unidentified lattice defects. Pulse height spectra revealed that the *LY* of the $La_2Be_2O_5$ single crystal was 1000 photons/MeV. The *AL* was 42.9 ppm, which was lower than that of Tl-doped CsI. Judging from these scintillation properties, the $La_2Be_2O_5$ single crystal would hold potential as a new scintillator for X-ray and γ-ray detection. However, the *LY* of the $La_2Be_2O_5$ single crystal is clearly lower than that of commercial scintillators such as $Bi_4Ge_3O_{12}$, LYSO, and $CdWO_4$. Therefore, it is essential to improve *LY*, and a solution may be to control the lattice defect derived from the emission center by doping impurity ions such as alkali metal or alkali earth metal.

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