Optical and Scintillation Properties of $\text{Sr}_3\text{BGa}_3\text{Si}_2\text{O}_{14}$ ($B = \text{Nb}, \text{Ta}$) Single Crystals

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Abstract

Optical and scintillation properties of $\text{Sr}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$ [SNGS] and $\text{Sr}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$ [STGS] single crystals with the langasite-type crystal structure were investigated as a novel scintillator materials. In the transmittance spectra of the SNGS and STGS polished specimens, absorption peaks around 380 and 505 nm were observed and the absorptions are considered to be attributable to the excess oxygen in the crystals. An emission peak around 420 nm was observed in the X-ray radioluminescence spectrum of the SNGS crystal. On the other hand, there was an emission peak around 335 nm in the X-ray radioluminescence spectrum of the STGS crystal.

Keywords

$\text{Sr}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$, $\text{Sr}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$, scintillator, single crystal, langasite-type

Introduction

Langasite-type materials with the $A_3BC_3D_2O_{14}$ chemical composition ($A = La^{3+}, Ca^{2+}, Sr^{2+}, Ba^{2+}, B = Nb^{5+}, Ta^{5+}, Ga^{3+}, C = Al^{3+}, Ga^{3+}, Fe^{3+}, In^{3+}$, and $D = Ga^{3+}, Si^{4+}$) have been investigated as a material for laser devices and a piezoelectric crystal for sensor devices which can be used at high temperature and oscillators (H. Takeda, et.al., 2005; J. Bohm, et.al., 1999; K. Shimamura, et.al., 1996). The langasite-type materials have several advantages as a functional element for some applications. The single crystal can be grown with relative ease by various conventional melt-growth methods as represented by the Czochralski (Cz) method (H. Kawamata, et.al., 1998), Vertical Bridgmann (VB) method (T. Taishi, et.al., 2007) and micro-pulling-down (μ -PD) method (Y. Yokota et.al., 2011, 2012, 2013), and their large bulk single crystals with several inches diameter have been already developed for mass production (S. Uda, et.al., 2005). In addition, the langasite-type single crystals have strong chemical, impact and temperature resistant properties, and the elements can be used under various conditions.

In our previous reports, we clarified the optical and scintillation properties of the $Ca_3NbGa_3Si_2O_{14}$ [CNGS] and $Ca_3TaGa_3Si_2O_{14}$ [CTGS] which were one of the langasite-type materials with the ordered structure (S. Kurosawa, et.al., 2014; Y. Futami, et.al., 2012). The CNGS and CTGS crystals indicated emission around 300~500 nm under UV, α -ray and X-ray excitation. On the other hand, there are other langasite-type materials which are composed of $A = Sr, B = Nb$ or $Ta, C = Ga$ and $D = Si$, $Sr_3NbGa_3Si_2O_{14}$ [SNGS] and $Sr_3TaGa_3Si_2O_{14}$ [STGS], and their single crystals have been grown by the Cz methods as a piezoelectric material (I.H. Jung, et.al, 2001; Z. Wang, et.al., 2004). Their crystal growths by a micro-pulling-down (μ -PD) method, phase identifications, crystallinities, and piezoelectric properties have been investigated in our previous reports (Y. Yokota, et.al., 2011, 2012). The density and effective atomic number are larger than that of the CNGS and CTGS due to the difference of the A site ion (Ca^{2+} and Sr^{2+}). However, there is no report about the investigations about the SNGS and STGS as a scintillator material. The our report (S. Kurosawa, et.al., 2014) revealed that the Ta-containing scintillator single crystals as the CTGS could demonstrate well-measurable scintillation response. Therefore, it is worthwhile to develop novel Ta-containing crystals with a larger effective atomic number.

On these backgrounds, in this study, the SNGS and STGS single crystals were grown by the μ -PD method, and their optical and scintillation properties were measured to evaluate them as a novel scintillator materials with larger density and effective atomic number than the CNGS and CTGS.

Experimental

$Sr_3NbGa_3Si_2O_{14}$ and $Sr_3TaGa_3Si_2O_{14}$ single crystals were grown by the μ -PD method in air. The detail of the crystal growth was described in the previous report (Y. Yokota et.al., 2011, 2012).

As-grown SNGS and STGS single crystals with approximately $\phi 2$ mm diameter and several centimeters length were obtained by the crystal growth. The as-grown crystals were cut and polished for the measurements of optical and scintillation properties.

Transmittance spectra of the SNGS and STGS polished specimens were measured in the wavelength range from 200 to 800 nm using a spectrophotometer (JASCO V530). X-ray radioluminescence spectra of the polished specimen under X-ray irradiation from an X-ray tube (40 kV, 40 mA) were measured by the spectrofluorometer (EDINBURGH INSTRUMENTS FLS920) and the charge coupled device [CCD] with the optical cable, respectively. Pulse-height spectra under γ -ray or α -ray irradiation were investigated using the polished specimens exposed to a ^{137}Cs or ^{241}Am radiation source with a photomultiplier [PMT] (HAMAMATSU R7600U-200). Specimens were covered by Teflon tape except for one plane and the plane was attached to the light entrance window of the PMT with optical grease. Signals from the PMT were converted to digital signals by a multi-channel analyzer (AMPTEK CO. Pocket MCA 8000A). In the pulse-height spectrum measurement, $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ [BGO] single crystal with 8,000 ph/MeV light yield was used as a standard scintillator.

Results and discussions

The as-grown SNGS and STGS single crystals were cut perpendicular to the growth direction and cutting planes were polished. The SNGS and STGS polished specimens were shown in Fig.1. The SNGS polished specimen indicated deep pink color while the color depth of the STGS polished specimen was weaker. Both polished specimens indicated high transparency and there were no visible cracks and inclusions in the crystals.

The transmittance spectra of the SNGS and STGS polished specimens were measured as it is illustrated in Fig.2. In the transmittance spectra of the SNGS and STGS specimens, absorption peaks around 380 and 505 nm were observed. The absorption peaks of the SNGS were larger than that of the STGS. The difference in the transmittance spectra between the SNGS and STGS was considered to be due to the oxygen content in the crystals. In the previous reports, the color of the langasite-type crystals and absorptions in the transmittance spectra were attributable to the excess oxygen in the crystals (Y. Yokota, et.al., 2013). The melting point of the STGS ($\sim 1535^\circ\text{C}$) is higher than that of SNGS ($\sim 1432^\circ\text{C}$) and therefore the crystal growth of the STGS single crystal at higher temperature was suggested to decrease the content of the excess oxygen in the crystal compared to the SNGS. In addition, the absorption edges of the SNGS and STGS crystals at room temperature were approximately 280 and 240 nm as it is shown in the inset. The defect absorption bands originated from the excess oxygen are considered to obscure the band-to-band transitions of SNGS and STGS.

Radioluminescence spectrum of the SNGS and STGS specimens under X-ray irradiation were shown in Fig.3. A emission peak around 420 nm was observed in the spectrum of SNGS specimen and the wavelength of the emission peak is almost consistent with that of the CNGS crystal in the previous report (Y. Futami, et.al., 2012). On the other hand, an emission peak at 335 nm was observed in the radioluminescence spectrum of the STGS specimens under X-ray irradiation. The emission wavelength of the STGS was slightly smaller than that of the CTGS, ~340 nm (S. Kurosawa, et.al., 2014). Their emissions by the X-ray irradiation were considered to be originated from the trapped exciton (TE) in the SNGS or STGS crystals, suggesting that the TE was dependent on the excess oxygen which acted as defects in the crystals.

Pulse-height spectra of the SNGS and STGS polished specimens under γ -ray and α -ray irradiation were investigated using the ^{137}Cs and ^{247}Am radiation sources, respectively. Clear photo-peaks were not observed in the pulse-height spectra of the SNGS and STGS specimens under γ -ray irradiation. On the other hand, a broad peak was observed in the pulse-height spectra of the SNGS and STGS crystals under α -ray irradiation (Fig.4). Figure 4(a) is the pulse-height spectra of the SNGS specimen under α -ray irradiation at various shaping times. The broad peak position increased with an increase of the shaping time up to 3 μs while the positions of the peak were almost same in the shaping time region from 3 to 10 μs . In addition, the broad peak was also observed in the pulse-height spectrum of the STGS specimen under α -ray irradiation and the broad peak position was larger than that of the SNGS specimen.

Conclusions

The SNGS and STGS polished specimens which were prepared from the as-grown SNGS and STGS single crystals were used for the measurements of transmittance, X-ray excited radioluminescence and pulse-height spectra under α -ray and γ -ray irradiation. The absorption peaks around 380 and 505 nm originated from the excess oxygen were observed in the transmittance spectra of the SNGS and STGS polished specimens. In the X-ray excited radioluminescence spectra of the SNGS and STGS, emission peaks around 330~420 nm were observed. In addition, a broad peak appeared in the pulse-height spectra of the SNGS and STGS specimen under α -ray irradiation and the position of the peak was changed with the shaping time and the B site ion (Nb^{5+} and Ta^{5+}).

Acknowledgment

This work was partially supported by the New Energy and Industrial Technology

Development Organization (NEDO), Ministry of Education, Culture, Sports, Science and Technology of Japanese government, Grant-in-Aid for Young Scientists, Development of Systems and Technology for Advanced Measurement and Analysis, Japan Science and Technology Agency (JST), Adaptable & Seamless Technology Transfer Program through Target-driven R&D [A-STEP].

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Figure Captions

Figure 1 SNGS and STGS polished single crystals grown by the μ -PD method

Figure 2 Transmittance spectra of the SNGS and STGS polished specimens.

Figure 3 X-ray radioluminescence spectra of the SNGS and STGS polished specimens.

Figure 4 (a) Pulse-height spectra at various shaping time for the SNGS polished specimen under γ -ray irradiation. (b) Pulse-height spectra at 3 μ s shaping time for the SNGS and STGS polished specimen under γ -ray irradiation.

Figure1

Figure2

Figure3

Figure4