Growth of Shape-controlled Ce:Y₃Al₅O₁₂ Scintillator Crystal and their Scintillation Properties

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Abstract

A shape-controlled Ce doped Y₃Al₅O₁₂ (Ce:YAG) single crystal has been grown by a micro-pulling-down method using an Ir crucible with a square die. The as-grown Ce:YAG single crystal has square-shape and flat surfaces with high transparency. The shape-controlled Ce:YAG single crystal showed more than 70% transmittance from 530 to 900 nm without any cutting and polishing. An emission peak around 540 nm was observed in the radioluminescence spectrum under X-ray irradiation. In addition, the light yield and energy resolution under γ -ray irradiation of the as-grown crystal without any cutting and polishing were approximately 23,000 ph/MeV and 16%, respectively.

Keywords: A1. Radiation, A2. Growth from melt, A2. Single crystal growth, B1. Oxides, B3. Scintillators

Introduction

Scintillator materials have been used as radiation detectors for dosimeters, survey meters, positron emission tomography (PET), security devices as well as other applications. Scintillators generate photons when irradiated by α -rays, β -rays, X-rays, γ -rays or neutrons. In radiation detectors for imaging devices such as PET, scintillator crystals are generally used in a typical configuration of rectangular columns to make scintillator array units [1-3]. The rectangular column-shaped scintillator element must have flat plane on the surface and the planes have to be mirror-polished for optical transmission. In general, scintillator single crystals have been grown by the Czochralski and Bridgman methods from the melt. However, these methods can't control the shape of the grown single crystal while they can grow a large bulk single crystal. Therefore, the rectangular column-shaped scintillator elements for some devices have to be prepared from a bulk single crystal after cutting and polishing processes, which increases the manufacturing cost.

We have developed growth of shape-controlled single crystals by a micro-pulling-down (μ -PD) method in order to decrease the manufacturing cost of single crystals [4-9]. The μ -PD method can grow a single crystal by pulling-down the meniscus below the bottom of a crucible which has a die with one or several holes at the bottom. The shape of the meniscus during the crystal growth is limited by the bottom shape of the die, and as a result, a single crystal with the same cross-sectional shape as the bottom shape of the die can be grown. In our previous paper, a square-shaped sapphire single crystal could be grown by the μ -PD method using a Molybdenum crucible with a square-shaped die. The shape-controlled sapphire single crystal showed great crystallinity from initial to end areas [4]. In addition, shape-controlled $\text{Ca}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$ (CNGS) piezoelectric single crystals were grown by the μ -PD method using a platinum crucible and columnar, plate or tube-shaped CNGS single crystals could be obtained through only growth, without cutting and polishing [5-9]. The shape-controlled growth method enables to make the crystal elements with the shape close to an element in a final device.

In this study, we have developed growth of shape-controlled Ce doped $\text{Y}_3\text{Al}_5\text{O}_{12}$ (Ce:YAG) scintillator single crystal to apply the near-net-shape technique using the μ -PD method to a scintillator crystal for a radiation detector. In addition, optical and scintillation properties of the shape-controlled Ce:YAG single crystal without any cutting and polishing were investigated.

Experimental

Shape-controlled Ce:YAG single crystal was grown by the μ -PD method using a Iridium crucible with a square die at the bottom. The bottom of the die is $5 \times 5 \text{ mm}^2$ square and there are 9 holes for capillary-channel of melt supply in the die. The starting materials, $\alpha\text{-Al}_2\text{O}_3$, Y_2O_3 , CeO_2 powders ($> 4\text{N}$ purity), were mixed as nominal composition $(\text{Y}_{1-x}\text{Ce}_x)_3\text{Al}_5\text{O}_{12}$ $x = 0.01$, and the mixed powders was set into the Ir crucible. The crucible was placed in the center of a high-frequency (HF) induction coil and it was surrounded by aluminum insulators. The crucible was heated by the HF coil in N_2 up to the melting point of YAG ($\sim 1950^\circ\text{C}$) and crystal growth was performed using an undoped YAG crystal with c -axis orientation as a seed. Growth rate was $0.02 \sim 0.05 \text{ mm/min}$, and liquid-solid interfaces during the crystal growth were observed by a charge coupled devices (CCD) camera through holes of the after-heater and insulators.

Laue image, pole figure and X-ray rocking curve (XRC) measurements on the surface of the as-grown Ce:YAG single crystal were performed by a back-reflection Laue camera with a CCD camera (Rigaku RASCO-TU) and X-ray diffractometer (Rigaku ATX-G),

respectively. ω scan was performed for the reflection (4 4 4) from a surface plane of the as-grown crystal.

All measurements of optical and scintillation properties were performed using the as-grown Ce:YAG single crystal without any cutting and polishing. The transmittance spectrum of the grown crystal was measured by a spectrophotometer (JASCO V530) from 200 nm to 900 nm. The radioluminescence spectrum of the grown crystal under X-ray irradiation (40 kV, 40 mA) was obtained by the CCD camera and a spectroscopy with an optical cable. Pulse-height spectrum and decay curve of the as-grown Ce:YAG crystal were investigated to estimate the light yield and decay time. The radiation measurements were performed by investigating the response of the Ce:YAG single crystal exposed to a ^{137}Cs radiation source emitting γ -rays with a photomultiplier tube (PMT, HAMAMATSU R7600U-200). The single crystal was covered by Teflon tapes except for one plane which 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 (AMPTK CO. Pocket MCA 8000A) and the decay curve was measured by an oscilloscope (TEKTRONIX TDS3035B). In the pulse-height spectrum measurement, a Ce1% doped Gd_2SiO_5 (Ce:GSO) single crystal with 11,000 ph/MeV light yield was used as a reference to estimate the light yield of the Ce:YAG single crystal. The sample thickness of the Ce:YAG single crystal was 5 mm in the transmittance and light yield measurements.

Results and Discussions

Ce:YAG single crystal with the shape of rectangular column was grown by the μ -PD method and the Ir crucible with the die which had the square-shaped bottom. The liquid-solid interface during crystal growth is shown in Fig.1(a). After the seed crystal was touched to the meniscus on the bottom of the die, the seed crystal was pulled down and the cross-sectional shape of the grown crystal gradually got close to the bottom shape of the die. Finally, the shape-controlled Ce:YAG single crystal could be grown. Figure 1(b) is the as-grown Ce:YAG single crystal without any cutting and polishing. The as-grown Ce:YAG single crystal showed high transparency with yellow color and the shape was a rectangular column with $5 \times 5 \text{ mm}^2$ cross-sectional shape along the growth direction. In addition, the surfaces perpendicular to the growth direction were flat and the clear surfaces are considered to realize the high transparency.

The back-scattering Laue image on the surface of the shape-controlled Ce:YAG single crystal is illustrated in the inset of Fig.2. A clear Laue pattern is observed in the image and the result revealed that the grown Ce:YAG was a single crystal. Figure 2 shows the XRC on the surface of the shape-controlled Ce:YAG single crystal. A clear sharp peak from (111) plane was observed in the XRC measurement and the full-width at half maximum (FWHM) on the XRC was approximately 15 arcsec (0.0042°). The result suggests that the shape-controlled Ce:YAG single crystal has excellent crystallinity.

The transmittance spectrum of the shape-controlled Ce:YAG single crystal without any cutting and polishing was measured (Fig.3(a)). The transmittance was more than 70% in the wavelength range from 530 to 900 nm. Absorption peaks around 215, 340 and 460 nm were observed, originating from the 4f-5d transition of Ce^{3+} ion [10]. Radioluminescence spectra of the shape-controlled Ce:YAG single crystal under X-ray irradiation is shown in Fig.3(b). An emission peak around 540 nm was observed, originating from the 5d-4f transition of Ce^{3+} ion [10].

Fig.4(a) presents the pulse-height spectra of 662 keV γ -rays from a ^{137}Cs source as measured with Ce:YAG and Ce:GSO single crystals. A clear photo-peak was observed in the pulse-height spectrum. The photo-peak positions in the pulse-height spectra were decided using the Gaussian function and the photoelectron yield (photoelectrons/MeV) of the shape-

controlled Ce:YAG and Ce:GSO single crystals. The photoelectron yield was recalculated to the light yield (photons/MeV) by using the average PMT quantum efficiency of 11 and 37%, respectively, for the emission spectrum of the Ce:YAG and Ce:GSO crystals.. The estimated light yield of the shape-controlled Ce:YAG single crystal was approximately 23,000 ph/MeV (2,530 phe/MeV) and the energy resolution was 16.7%. In a previous report about the scintillation properties of the polished Ce:YAG single crystal grown by the conventional method, the light yield under γ -ray irradiation (662 keV) was 20,300 ph/MeV [11]. Therefore, the shape-controlled Ce:YAG single crystal without cutting and polishing has comparable light yield to the polished Ce:YAG single crystal. On the other hand, the energy resolution of the shape-controlled Ce:YAG single crystal ($\sim 17\%$) was larger than that of the polished Ce:YAG single crystal grown by the conventional method (7 $\sim$ 11%) [11,12]. The decay curve of the shape-controlled Ce:YAG single crystal under γ -ray irradiation was investigated under the same condition (Fig.4(b)). The decay curve could be fitted by a double exponential decay function. The estimated decay times by the fitting was approximately 82.2 ns (83.4%) and 214 ns (16.6%) and the decay times are almost consistent with the previous reports of the polished Ce:YAG single crystal grown by the conventional method [11,12].

Conclusions

A shape-controlled Ce:YAG single crystal was grown by the μ -PD method using an Ir crucible. The grown square-shaped Ce:YAG single crystal exhibited high transparency with flat surfaces and near-net-shape crystal growth could be achieved by the μ -PD method for Ce:YAG scintillator single crystal. In addition, the square-shaped Ce:YAG single crystal without any cutting and polishing showed comparable optical and scintillation properties to the polished Ce:YAG single crystal grown by the conventional method as evidenced by the transmittance, light yield, energy resolution and decay time measurements. The near-net-shape single crystal growth for scintillator material can reduce the manufacturing cost of scintillator elements in the radiation devices by decreasing the production processes.

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Figure Captions

Figure 1 (a) Liquid-solid interface during the crystal growth. (b) Shape-controlled Ce:YAG single crystal grown by the μ -PD method.

Figure 2 X-ray rocking curve on the surface of the grown Ce:YAG single crystal. Inset is the Laue image on the surface.

Figure 3 (a) Transmittance spectrum and (b) radioluminescence spectrum under X-ray irradiation for the shape-controlled Ce:YAG single crystal.

Figure 4 (a) Pulse-height spectrum and (b) decay curve of shaped Ce:YAG single crystal under γ -ray irradiation.

Figure 1

Figure 2

Figure 3

Figure 4