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Luminescent Properties of Ce:Gd_3 (Al, Ga, Mg, M)$_5O_{12}$ Crystal (M=Zr, Hf)

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Abstract

In order to obtain a new scintillator with a higher effective-atomic number, we grew new scintillators; Ce:$Gd_3Al_2Mg_{0.5}Hf_{0.5}Ga_2O_{12}$ (Ce:GAGG-Hf) and Ce:$Gd_3Al_2Mg_{0.5}Zr_{0.5}Ga_2O_{12}$ (Ce:GAGG-Zr) grown by micro-pulling down method. The emission wavelengths of these crystals were almost the same as that of Ce:Gd_3Al_2 Ga_3O_{12}.

1. Introduction

Scintillation materials have been investigated and applied to many fields such as homeland security, astronomy and medical imaging system [1]. Ce-doped Gd_3Al_2 Ga_3O_{12} (Ce:GAGG) had found to be high light-yield (~ 46,000 photon/MeV) and good energy resolution (~ 4.9 % at 662 keV, FWHM) [2-4]. The GAGG crystal has garnet structure, and its space group is equivalent to *Ia3d* (No. 230). Ce^{3+}-dopant is crystallographically substituted for Gd3+ in the dodecahedral site coordinated by eight oxygen atoms. The scintillation emission is caused by 5d-4f transition of Ce^{3+} with an emission wavelength of approximately 500 to 550 nm [2].

Although Ce:GAGG has good properties, the effective atomic number (Z_{eff}) is smaller (51) than other conventional oxide crystals such as Ce:Gd_2SiO_5, Ce:$(Lu,Y)_2SiO_5$. Since the cross section of gamma-ray photo-absorption is proportional to Z_{eff}^{4-5}, gamma-ray stopping power of Ce:GAGG is generally smaller than other oxide scintillators.

In order to obtain a new crystal based on the Ce:GAGG with a higher effective atomic number, we replace Ga to Zr or Hf, which have higher atomic numbers of 40 and 72, respectively, compared with Ga. As a first step, we grown Ce:Gd_3 (Al, Ga, Mg, M)$_5O_{12}$ (M=Zr, Hf) crystals using micro-pulling down method (μ-PD method). This method is effective for materials research, because crystal growth occurs more rapidly (by 1–2 orders of magnitude) than the conventional methods such as Czochralski (Cz) and Bridgman (BG) methods. [5]. In this paper, we show the scintillator properties of these crystals.

2. Material and Methods

Ce:Gd_3 (Al, Ga, Mg, M)$_5O_{12}$ (M=Zr, Hf) crystals were grown by the μ-PD, and the powders of the crystal materials (Ce_2O, Gd_2O_3, alpha-Al_2O_3, HfO_2 ZrO_2, and Ga_2O_3) were of 4N purity. The μ-PD atmosphere was N_2 (flow). Crystals were cut and polished to the size of 6×6×1 mm^3. After the polishing process, optical properties, such as transmittance (V-530, JASCO), reflectance (UV-2550, SHIMADZU), emission and excitation spectra (FLS920, Edinburgh Instruments) were measured. Radio-luminescence spectra under alpha-ray (^{241}Am) excitation (FLS920, Edinburgh Instruments) were also obtained.

3. Results and Discussions

We succeeded in growth of Ce:$Gd_3Al_2Mg_{0.5}Hf_{0.5}Ga_2O_{12}$ (Ce:GAGG-Hf) and Ce:$Gd_3Al_2Mg_{0.5}Zr_{0.5}Ga_2O_{12}$ (Ce:GAGG-Zr) crystal as shown in Fig 1. Here, X-ray diffraction patterns were also measured, and these crystals had single phases.

Figure 2 shows transmittance and reflectively for the Ce:GAGG, Ce:GAGG-Hf and Ce:GAGG-Zr. Transmittance of Ce:GAGG-Hf and –Zr were different from Ce:GAGG around 300 nm. This result imply that band gap can be changed from Ce:GAGG.

Radio-luminescence spectra of the all samples excited by

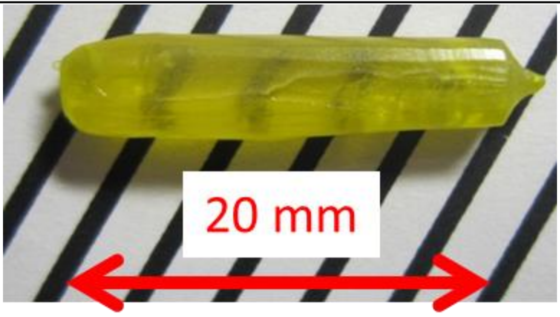

Fig. 1 Photograph of as-grown Ce:Gd_3 Al_2, $Mg_{0.5}$, $Zr_{0.5}$ Ga_2O_{12} crystal

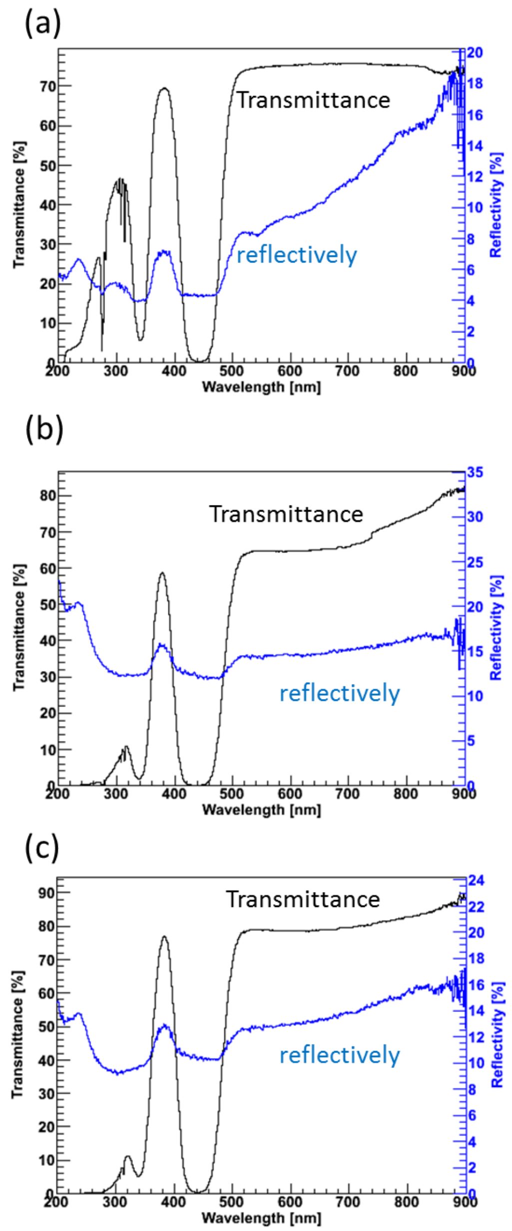

Fig. 2 Transmittance and reflectively for the Ce:GAGG (a), $Ce:Gd_3Al_2Mg_{0.5}Hf_{0.5}Ga_2O_{12}$ (b) and $Ce:Gd_3Al_2Mg_{0.5}Zr_{0.5}Ga_2O_{12}$ (c).

^{241}Am alpha rays (5.5 MeV) at room temperature are shown in Fig 3. Emission peaks of all samples were almost the same positions.

Here, we also measured the light output. However, light output of Ce:GAGG-Hf and Ce:GAGG-Zr excited by gamma rays decreased compared with Ce:GAGG due to the change of the band gap.

3. Conclusions

$Ce:Gd_3Al_2Mg_{0.5}Hf_{0.5}Ga_2O_{12}$ (Ce:GAGG-Hf) and $Ce:Gd_3Al_2Mg_{0.5}Zr_{0.5}Ga_2O_{12}$ (Ce:GAGG-Zr) crystal were grown by the μ-PD method. Although the emission wavelengths of Ce:GAGG-Hf or Zr were almost the same as that of Ce:GAGG, light outputs of Ce:GAGG-Hf or Zr was

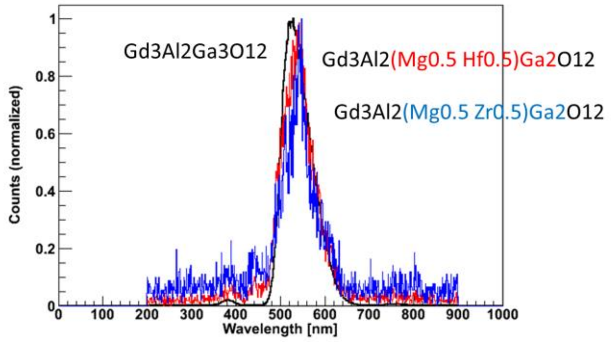

Fig. 3 Radio-luminescence spectra of the all samples excited by ^{241}Am alpha

small due to the change of the band structure.

In this presentation, we show the optical and scintillation properties including the above results and other results such as excitation and emission wave length, pulse height spectra of these samples. In addition, we also show the some optical properties for non-doped Ce:GAGG-Hf and –Zr.

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