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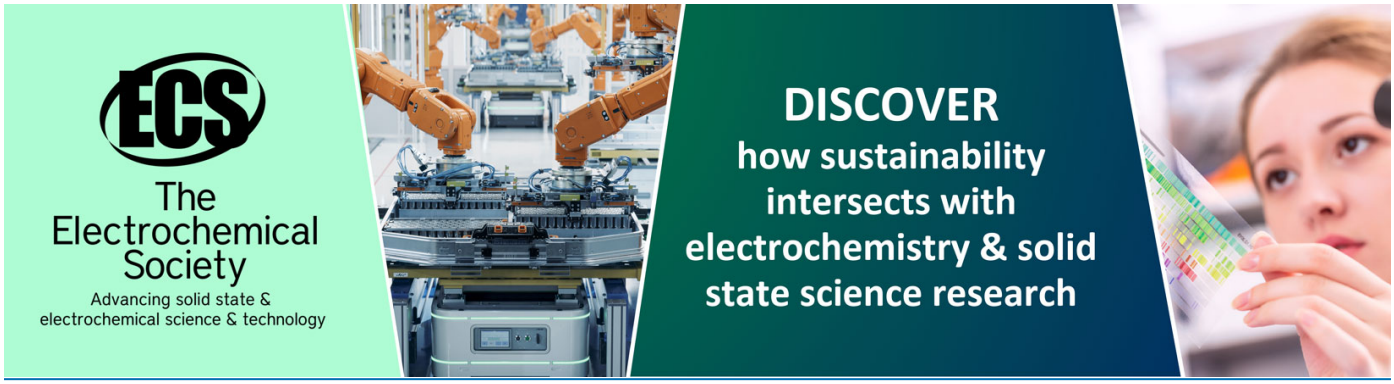

Growth and scintillation properties of Ce doped $Gd_2Si_2O_7/SiO_2$ eutectics

[1]**Kei Kamada**[1,2], **Shunsuke Kurosawa**[1,3] , **Rikito Murakami**[3], **Yuui Yokota**[1], **Jan Pejchal**[1,] **Yuji Ohashi**[3] **and Akira Yoshikawa**[1,2,3]
[1]New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aoba, Sendai 980-8579
[2]C&A corporation, 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
[3]Institute for Material Reseach, Tohoku University, 2-1-1, Sendai,980-8577 Japan
E-mail: kamada@imr.tohoku.ac.jp

Abstract. Ce:$Gd_2Si_2O_7/SiO_2$ eutectic was grown by the μ-PD method. The square-shape sample with a side of 5 mm and a length of 15 mm was obtained. Two phases of orthorhombic $Gd_2Si_2O_7$ and SiO_2 was observed. Rod-phase was SiO_2 and matrix phase was $Gd_2Si_2O_7$. Ce^{3+} 4f5d emission have been observed at 400nm. The sample showed light yield of around 16,000 photons/MeV. Scintillation decay time was 46.3ns(21%) 249ns(79%).

1. Introduction

A number of halide and oxide scintillator materials have been developed and they are applied to various fields such as astronomy, medical imaging, and homeland security. Although halide scintillators, such as Tl:NaI, Tl:CsI and Ce:$LaBr_3$ [1], have high light outputs of more than 30,000 photons/MeV, they have hygroscopic nature. In the X-ray imaging, indirect flat panel detector (FPD) composed of a scintillator layer such as CsI:Tl and photo-sensor array requires improvements in their spatial resolution. But, CsI:Tl columnar films reduce the spatial resolution, because the thicker it becomes the more light scattering occurs [2]. Recently submicron-diameter phase-separated scintillator fibers (PSSFs) were reported and they possessed both the properties of an optical fiber and a radiation-to-light conversion. The PSSFs were fabricated using a directionally solidified eutectic (DSE) system. The DSE systems have been discovered in various materials for many applications [3-6]. In PSSFs, the light emitted from the scintillator fibers is confined and transported along the fiber direction by a total reflection mode as shown in Fig.1, so that high-resolution radiation imaging can be achieved. CsI/NaCl [6] and GAP/Al_2O_3[5] have been already reported as PSSFs.
Ce:$Gd_2Si_2O_7$ (Ce:GPS) had a good light output of 30,000 photons/MeV and FWHM energy resolution of 6.0% at 662 keV at room temperature. However, according to the previous report [7-10], Ce:GPS crystals need to be grown with heavy Ce-doping (approximately, 10 mol%) in order to stabilize the crystal growth process. Such high Ce-concentration would lead to lower light output because of self-absorption or concentration quenching. The main reason is the fact that $Gd_2Si_2O_7$ is not congruently melt. When the chemicals with stoichiometric composition is melted, initial melt has apatite phase.

[1] Kei Kamada, New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aoba, Sendai 980-8579

Scintillator materials combined with photodetectors are used to detect high energy photons and accelerated particles in medical imaging techniques, high energy and nuclear physics detectors, high-tech industrial applications and most recently also in the advanced homeland security related techniques. Recently, Ce:$Gd_2Si_2O_7$ (Ce:GPS) belonging to the pyrosilicate group has been investigated, and this crystal was found to have much higher light output and shorter decay time than Ce:GSO. [6–8] Toropov et al. found that the composition Gd_2O_3-$2SiO_2$ is not congruent in the Gd_2O_3–SiO_2 system,[9] and Kawamura et al. reported that the GPS phase can be grown by heavy Ce-doping (approximately10 mol %).[9] Furthermore, substituting Ce^{3+} site with La^{3+} whose ionic radius is similar to Ce^{3+} is also able to stabilize the congruent phase, and Ce:(La, Gd)$_2Si_2O_7$ (Ce:La-GPS) scintillator with a good energy resolution was developed [10-13].
In this study, Ce 1mol% doped $Gd_2Si_2O_7/SiO_2$ eutectics were grown by the micro pulling down (m-PD) method and their DSE system has been investigated. Investigation of obtained eutectic structure , luminescence and scintillation performances were performed.

2. Experimental

2.1. Crystal growth procedure

A stoichiometric mixture of 4N CeO_2, Gd_2O_3 and glass-SiO_2 powders (High Purity Chemicals Co.) was used as starting material. Ce:$Gd_2Si_2O_7/SiO_2$ eutectics were grown by the micro-pulling-down (μ-PD) method with an RF heating system at the eutectic composition (25.6mol%$Gd_{1.98}Ce_{0.02}O_3$ - 74.4mol%SiO_2). Here, the volume ratio is about $Gd_{1.98}Ce_{0.02}Si_2O_7$:$SiO_2$=0.809:0.191, theoretically. Typical pulling rates were 0.3-0.8mm/min. Crystals were grown from an Ir crucible under N_2 atmosphere. A Ir rod was used as a seed. Plates of 5mm x 5mm x 0.5mm were cut and polished for the measurements. The eutectic phase structure was investigated by back scattered electron image (BEI). The phases in the eutectic crystal determined by powder XRD patterns.

2.2. Measurements of scintillation properties

Radio-luminescence spectrum at room temperature was measured with the above spectrometer (EI FLS920) excited by 5.5MeV alpha rays from an 241Am source. To determine the light output, we obtained the pulse height spectrum of this crystal under excitation with 662 keV gamma rays from a ^{137}Cs source. The scintillation photons were detected using by a photomultiplier (PMT; Hamamatsu R7600U-200), and then the signals from the PMT were amplified (ORTEC 113), shaped (ORTEC 572A), and read out with a multi-channel analyzer (AMPTEK 8000A). The pulse height of $Bi_4Ge_3O_{12}$ (BGO) crystal, which has a light output of 8,500 photons/MeV was also measured as a reference. Additionally, its scintillation decay time excited by the gamma rays (^{137}Cs) was measured with the PMT and an oscilloscope (Tektronix TDS 3052B). The scintillation decay time was calculated from exponential approximation.

3. Results

3.1. Crystal growth procedure

Ce:$Gd_2Si_2O_7/SiO_2$ eutectic was grown by the μ-PD method. The square-shape sample with a side of 5 mm and a length of 15 mm was obtained (Fig.1). The BEI image of transverse cross-section of the eutectic is shown in Fig. 2. The phases were determined as orthorhombic $Gd_2Si_2O_7$ and SiO_2 glass by the EDX measurement and powder XRD pattern of the grown eutectic. Rod-phase was SiO_2 and matrix phase was $Gd_2Si_2O_7$ according to the EDX measurement result. The diameter of SiO_2 scintillation fiber was approximately 700 nm (Fig.2) and dispersed well on the transverse cross-section.

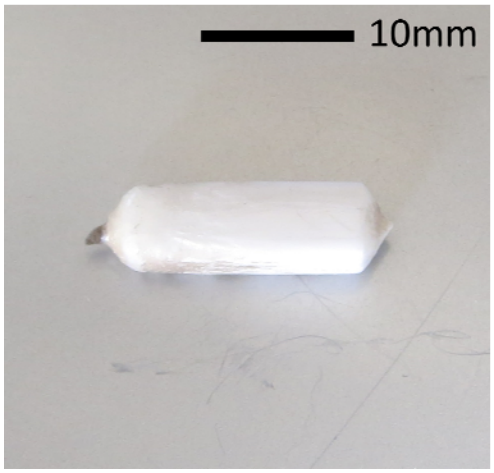

Figure 1. Photograph of the grown Ce:$Gd_2Si_2O_7/SiO_2$ eutectic

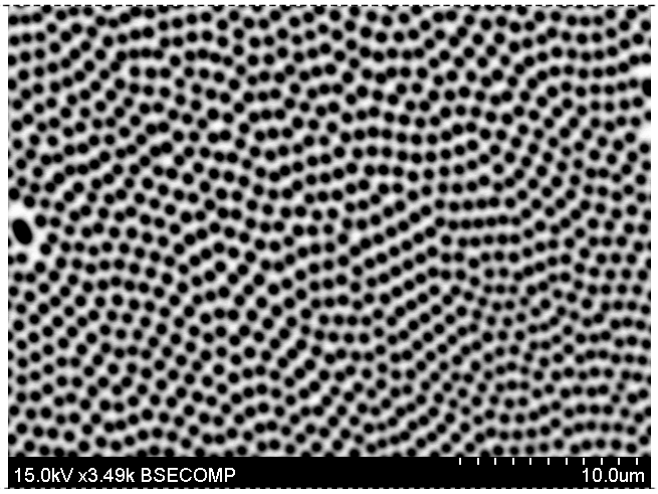

Figure 2. BEI of transverse cross-section of the grown Ce:$Gd_2Si_2O_7/SiO_2$ eutectic. White phase was Ce:$Gd_2Si_2O_7$ and brack phase was SiO_2.

3.2. Luminescence and gamma-ray response measurement procedurembering

Figure 3 shows radioluminescence spectra of the Ce:$Gd_2Si_2O_7/SiO_2$ eutectic samples. Expected Ce^{3+} 4f5d emission have been observed in 350-440nm peaking at 400nm. Figure 4 exemplifies energy spectra of the Ce:$Gd_2Si_2O_7/SiO_2$ eutectic samples. The samples irradiated by ^{137}Cs, where 662 keV. The sample showed light yield of around 14,000-16,000 photons/MeV considering with the emission wavelength of the samples and quantum efficiency of the PMT (45%@390nm, 32%@480nm). Scintillation decay curves were obtained by using the PMT and digital oscilloscope and the sample was irradiated by ^{137}Cs (Figure 5). Scintillation decay time was 46.3ns(21%) 249ns(79%).

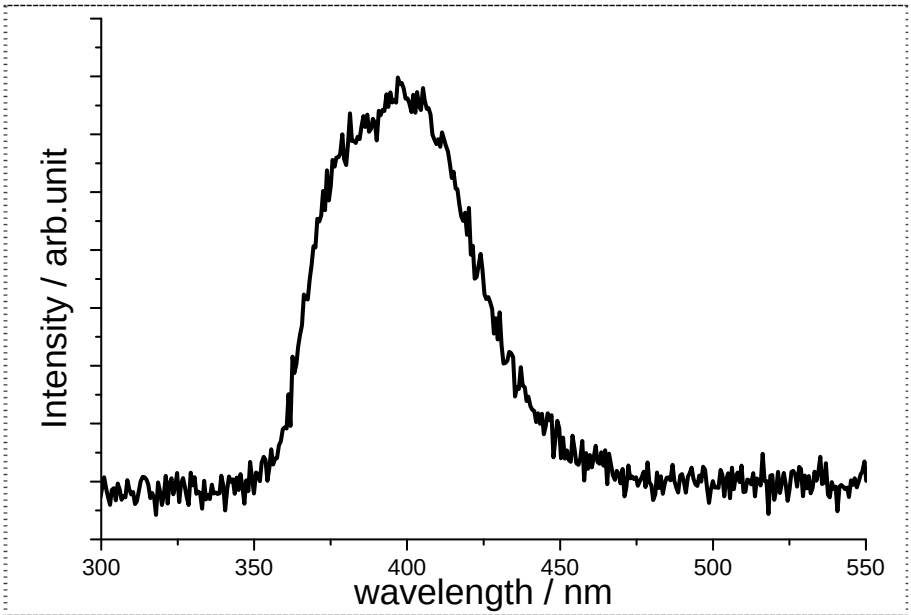

Figure 3. Radioluminescence spectra of the Ce:$Gd_2Si_2O_7/SiO_2$ eutectic sample.

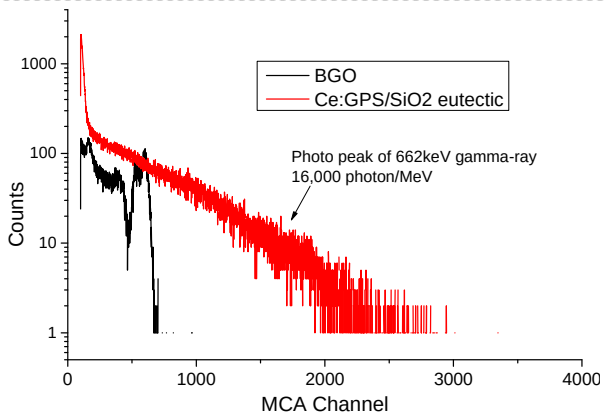

Figure 4. Energy spectra of the Ce:$Gd_2Si_2O_7/SiO_2$ eutectic sample and BGO standard excited by a 662 keV gamma ray.

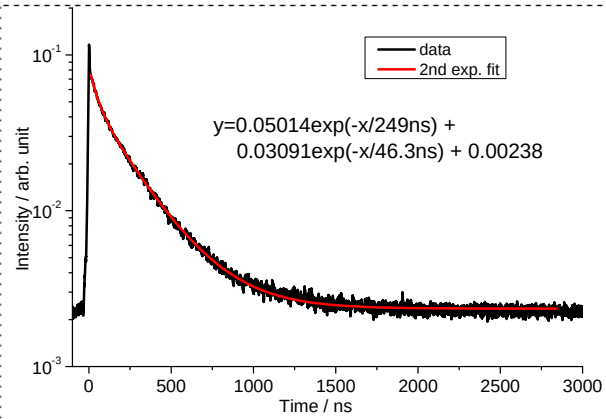

Figure 2. Scintillation decay curve of the Ce:$Gd_2Si_2O_7/SiO_2$ eutectic sample excited by a 662 keV gamma ray.

4. Conclusion

The sub-micron diameter SiO_2 glass fibers surrounded with Ce:GPS matrix from directionally solidified eutectics were fabricated. In this crystal, the length of longest fiber of SiO_2 along the growth direction was about 200 μ m or so. Expected Ce^{3+} 4f5d emission have been observed in 350-440nm peaking at 400nm. The sample showed light yield of around 16,000 photons/MeV. Scintillation decay time was 46.3ns(21%) 249ns(79%). By using DSE system of Ce:$Gd_2Si_2O_7/SiO_2$ eutectic, high resolution X-ray imaging can be expected.

5. References

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