

Luminescence properties of $Gd_2Si_2O_7:Ce$ and $Gd_2Si_2O_7:La,Ce$ under vacuum ultraviolet irradiation

M. Koshimizu^{1,*}, T. Yanagida², Y. Fujimoto¹, K. Asai¹

(1. Department of Applied Chemistry, Graduate School of Engineering, Tohoku University, 6-6-07 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan; 2. Graduate School of Materials Science, Nara Institute of Science and Technology, 8916-5 Takayama-Cho, Ikoma, Nara 630-0192, Japan)

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Abstract: The luminescence properties of Ce- or Ce and La-doped gadolinium pyrosilicate ($Gd_2Si_2O_7$, GPS) were characterized using vacuum ultraviolet (VUV) excitation light. A prominent emission band was observed in the luminescence spectra with excitation at 60 nm and ascribed to 5d-4f transition of Ce^{3+} . Because the excitation wavelength of 60 nm corresponded to the excitation in the host matrix, this result indicated that the excitation energy transfer occurred from the host matrix to Ce^{3+} ions. On the basis of the rise in the luminescence time profiles with excitation at 60 nm, the energy transfer occurred within 2 ns, which was much shorter than that of Ce-doped Gd_2SiO_5 . For Ce-doped GPS, the decay rate was slower for the host excitation than that for direct excitation of Ce^{3+} . In contrast, for Ce and La-doped GPS, no significant difference was observed for the host excitation and direct excitation of Ce^{3+} . This result indicated that the energy transfer from the host to Ce^{3+} ions led to a different radiative decay process, and that La doping had an effect on the energy transfer and decay process.

Keywords: energy transfer; scintillation; vacuum ultraviolet; rare earths

Gadolinium pyrosilicate ($Gd_2Si_2O_7$, abbreviated as GPS hereafter) doped with Ce has been attracting considerable attention owing to its excellent scintillation properties. At early stage, excellent scintillation properties were reported by Kawamura et al.^[1-3]. They reported structural, optical, and scintillation properties in detail. According to their papers, Ce-doped GPS crystals exhibit efficient scintillation owing to 5d-4f transition of Ce^{3+} , and the scintillation light yield is significantly larger than that of Ce-doped Gd_2SiO_5 (GSO). In addition, the scintillation decay is slightly faster than that of Ce-doped GSO. Subsequently, several groups reported the scintillation properties of Ce-doped GPS fabricated by various synthesis techniques and their applications for gamma-ray, α -ray, or thermal neutron detection^[4-7]. Recently, Kurosawa et al. reported the stabilization of the pyrosilicate phase by La doping and the excellent scintillation properties of Ce and La-doped GPS^[8,9].

The aim of this study is the analysis of the scintillation processes in Ce-doped and Ce and La-doped GPS. The scintillation processes can be divided into three stages^[10]: Stage 1: generation of excited states in the host matrix by energy deposition of ionizing radiation, Stage 2: excitation energy transfer from the host matrix to luminescence centers (dopants such as Ce^{3+}), and Stage 3: photon emission by radiative deexcitation of the luminescence centers. Among them, the energy transfer process is important; however, it has not been thoroughly analyzed.

As for the energy transfer process in Ce and La-doped GPS, a detailed analysis has been reported in a previous paper^[11]. They proposed an energy level diagram and discussed the photoluminescence properties based on the diagram.

In this study, we analyzed the energy transfer process using vacuum ultraviolet (VUV) spectroscopy. In order to analyze the energy transfer process, it is necessary to generate excited state in the host matrix. In a previous study^[11], the band-gap energy of La-admixed GPS is estimated to be 6.6–6.8 eV. VUV photons having an energy larger than the band-gap energy, as well as ionizing radiation, generate excited states in the host matrix. Thus far, Feng et al. have reported the excitation spectra of Ce-doped GPS in VUV region and discussed the contribution of higher 5d levels within the band-gap^[12]. In this study, we used VUV excitation photons with energies higher than the band-gap energy, and analyzed the energy transfer process. In addition to the measurements of luminescence spectra with VUV excitations, luminescence decay behavior was characterized using a single-photon counting method.

1 Experimental

The samples were GPS crystals doped with Ce at 1 mol.% or 10 mol.%, and GPS crystals doped with Ce at 1 mol.% and La at 10 mol.%. These crystals were grown

* Corresponding author: M. Koshimizu (E-mail: koshi@qpc.che.tohoku.ac.jp; Tel.: +81227157219)

with floating zone method. Raw material powders were Lu_2O_3 (99.999%), SiO_2 (99.999%), La_2O_3 (99.99%) and CeO_2 (99.99%) and they were mixed as the above molar ratios. After the mixing, powders were shaped to the cylinder by the hydrostatic pressure. The cylinders were sintered at 1600 °C for 6 h under an air atmosphere to obtain GPS ceramic. In the crystal growth, the rotation rate was 20 r/min and we pulled down the sintering rod to obtain a crystal with a rate of 3–8 mm/h. Hereafter the samples are referred to with their dopant concentrations in the melt. The X-ray-irradiated radioluminescence spectra were obtained using an X-ray generator. The applied voltage and the tube current was 80 kV and 2.5 mA, respectively. The spectra were recorded with a CCD detector (Andor, DU-420-OE) equipped with a grating monochromator (Oriel Instruments)^[13]. The characterization of the luminescence properties was performed at BL-7B of UVSOR, Institute for Molecular Science, Japan. The samples were placed in a vacuum chamber and excited with VUV or UV photons. The luminescence spectra were recorded with a CCD detector (Roper Scientific) equipped with a monochromator (Acton, 300i). The luminescence decay behavior was observed using time-correlated single photon counting technique. Single bunch operation of the UVSOR was used, and the time period between the successive excitation pulses was 176 ns. Emitted photons were monochromatized with the monochromator and detected with a multichannel-plate-mounted photomultiplier tube (Hamamatsu, R3809U-50). The time difference of the luminescence photon detection signal and the excitation pulse were accumulated into a histogram as luminescence time profile.

2 Results and discussion

Fig. 1 shows the X-ray-irradiated radioluminescence spectra of the Ce-doped and Ce and La-doped GPS crystals. Fig. 2 shows the luminescence spectra of the Ce-doped and Ce and La-doped GPS crystals. The excitation wavelength was 60 nm. Judged from the host excitation band peaking at 126 nm in a previous paper^[12], the excitation wavelength of 60 nm corresponds to a

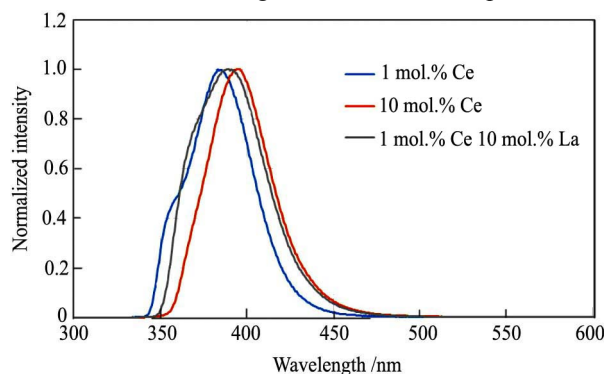


Fig. 1 X-ray-irradiated radioluminescence spectra of the Ce-doped and Ce and La-doped GPS

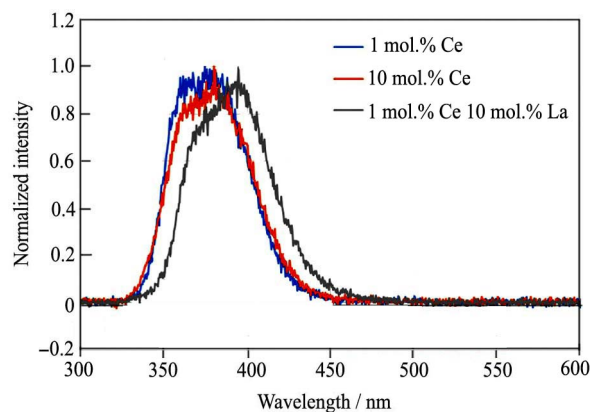


Fig. 2 Luminescence spectra of the Ce-doped and Ce and La-doped GPS (The excitation wavelength was 60 nm)

photon energy much higher than the band-gap energy, and excited states are generated in the host matrix. In both spectra, a prominent band is observed at approximately 390 nm and ascribed to 5d-4f transition of Ce^{3+} . Thus, Ce^{3+} emission is observed in the case of photoexcitation of the host matrix, as well as in the case of scintillation. The result in Fig. 2 indicates that the emission occurs after the energy transfer from the host matrix to Ce^{3+} ions.

Figs. 3–5 show the luminescence time profiles of Ce-doped GPS at 1 mol.%, Ce-doped GPS at 10 mol.%, and Ce and La-doped GPS crystals, respectively. The excitation wavelengths were 60 and 300 nm, which correspond to excitation in the host matrix and of Ce^{3+} ions, respectively. The excitation wavelength of 60 nm corresponds to the photon energy of about three times the

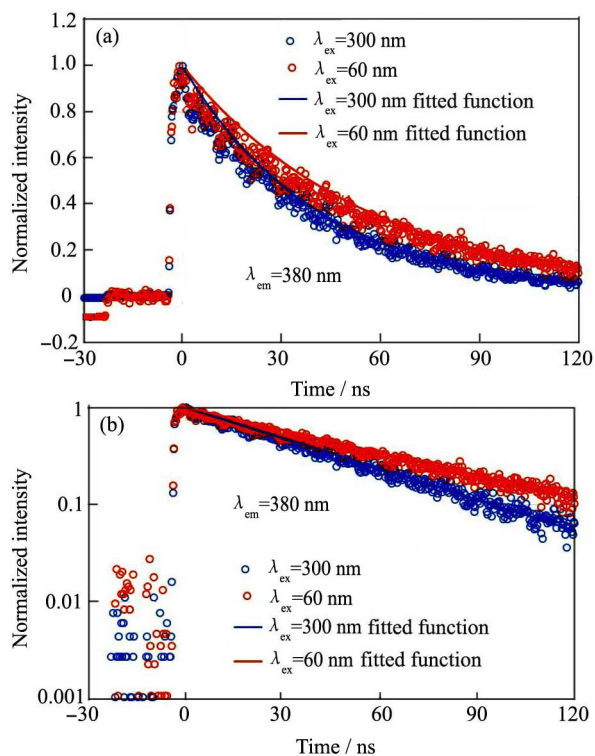


Fig. 3 Luminescence time profiles of GPS doped with Ce at 1 mol.% (a) In linear scale; (b) In semi-log scale

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