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# Optical and radioluminescence properties of Ce-doped $\text{Sr}_2\text{RE}_8(\text{SiO}_4)_6\text{O}_2$ ( $\text{RE} = \text{Y, Gd, Lu}$ )



Takuya Igashira\*, Naoki Kawano, Go Okada, Noriaki Kawaguchi, Takayuki Yanagida

Graduate School of Materials Science, Nara Institute of Science and Technology (NAIST), 8916-5 Takayama, Ikoma, Nara, Japan

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## ABSTRACT

Ce-doped  $\text{Sr}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ ,  $\text{Sr}_2\text{Gd}_8(\text{SiO}_4)_6\text{O}_2$  and  $\text{Sr}_2\text{Lu}_8(\text{SiO}_4)_6\text{O}_2$  (SrYS, SrGS and SrLuS, respectively) apatite crystals were grown by the Floating Zone method, and their optical and radioluminescence (RL) properties were characterized. In the photoluminescence (PL) and RL spectra, we observed an emission band between 400 and 500 nm for all the samples, which were due to the 5d-4f transition of  $\text{Ce}^{3+}$ . The RL decay time profiles were approximated by a sum of two exponential decay functions, and the origin of faster decay component was due to the 5d-4f transition of  $\text{Ce}^{3+}$ . Among all the samples, SrGS sample showed the lowest afterglow level. In the pulse height spectrum, all the samples showed a full energy absorption peak under  $^{241}\text{Am}$  5.5 MeV  $\alpha$ -ray irradiation. The absolute scintillation light yields of SrYS, SrGS and SrLuS were around 31, 76 and 940 ph/5.5 MeV- $\alpha$ , respectively.

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## 1. Introduction

Scintillator is a fluorescent substance which converts a high energy (keV-GeV) radiation photon or particle to thousands of visible-ultraviolet photons immediately [1]. Demanded characteristics of ideal scintillator are high light yield, fast response and adequate effective atomic number ( $Z_{\text{eff}}$ ) for the target radiation type [2–4]. In the past decades, a number of successful scintillators have been developed for ionizing radiation detectors used in a wide range of applications such as high energy physics [5], medical imaging [6], security system [7] and well-logging [8]. In recent years, scintillators which consist of Gd as a host material and  $\text{Ce}^{3+}$  as a luminescent center such as Ce-doped  $\text{Gd}_2\text{SiO}_5$  [9],  $\text{Gd}_2\text{Si}_2\text{O}_7$  [10,11] and  $\text{Gd}_2\text{O}_2\text{S}$  [12] have attracted much attention. One reason is because they have a large degree of cross-section with high energy photons. Another reason is that the ionic radii of  $\text{Gd}^{3+}$  and  $\text{Ce}^{3+}$  are well-matched, and the dopant ion can easily substitute the host atom. In addition, a past study has reported that one of this class of scintillators show a high light yield of 40,000–50,000 ph/MeV under  $\gamma$ -ray irradiation via efficient energy transportation from Gd to Ce [10].

Motivated by the earlier studies, we have been investigating on Ce-doped apatite crystals consisting of Gd as one of the host material components. Apatite materials have been developed for laser applications in 1970's [13], and they have been, in recent years, applied for artificial bone [14] and other applications [15]. However, apatite single crystals as a scintillator have not yet actively investigated despite the fact that they have a great potential to become effective scintillators. According to a recent study, chemical compositions of such apatite crystals for scintillator usages can be expressed as  $\text{RE}_{9.33}(\text{SiO}_4)_6\text{O}_2$

\* Corresponding author.

E-mail address: [igashira.takuya.il4@ms.naist.jp](mailto:igashira.takuya.il4@ms.naist.jp) (T. Igashira).

and  $AE_2RE_8(SiO_4)_6O_2$ , where  $RE$  and  $AE$  denote rare earth and alkaline earth elements, respectively [16]. Yanagida et al. have initiated the development of apatite crystals scintillator in 2010 [16]. They doped  $Yb^{3+}$  and  $Ce^{3+}$  as luminescent centers and observed a clear full energy-deposited peak of  $^{241}Am$   $\alpha$ -rays in the pulse height spectrum. Following this study, we synthesized and evaluated optical and radioluminescence (RL) properties of  $AE_2Gd_8(SiO_4)_6O_2$  ( $AE = Ca$  and  $Sr$ ) doped with different concentrations of  $Ce^{3+}$  [17,18]. Among them, 0.5% Ce-doped  $Sr_2Gd_8(SiO_4)_6O_2$  showed the highest light yield. We also confirmed that the RL decay time was longer than those of generally reported Ce-doped Gd-based scintillators. In the reason of this phenomenon, we conclude that the emission of  $Ce^{3+}$  can be influenced via energy migration processes from  $Gd^{3+}$  which has a much longer lifetime than that of  $Ce^{3+}$ ; therefore, the emission of  $Ce^{3+}$  appears slower than that without  $Gd^{3+}$ . However, there is still a large room for studying apatite scintillators, and one of the investigations to be conducted is to replace the  $RE$  sites from Gd to other element.

In the present study, we synthesized 0.5% Ce-doped  $Sr_2Y_8(SiO_4)_6O_2$ ,  $Sr_2Gd_8(SiO_4)_6O_2$  and  $Sr_2Lu_8(SiO_4)_6O_2$  (SrYS, SrGS and SrLuS, respectively) single crystals by the Floating Zone (FZ) method. We chose Y and Lu to substitute the  $RE$  site because the ionic radius seems appropriate, and some outstanding scintillators consist of Y or Lu such as Ce-doped  $Y_3Al_5O_{12}$  [19],  $Lu_{0.4}Gd_{1.6}SiO_5$  [20] and Pr-doped  $Lu_3Al_5O_{12}$  [21]. After the crystal growths were conducted, we evaluated their optical and radioluminescence properties including photoluminescence (PL) emission and excitation spectra, PL decay time profiles, X-ray induced RL spectra and decay times, afterglow as well as pulse height spectra.

## 2. Experimental procedures

Ce-doped SrYS, SrGS and SrLuS samples with the compositions of  $Sr_2(Ce_{0.005}RE_{0.995})_8(SiO_4)_6O_2$  ( $RE = Y, Gd, Lu$ ) were grown by the FZ method using an FZ furnace (FZD0192, Canon Machinery Inc.). We used  $CeO_2$ ,  $SrCO_3$ ,  $SiO_2$ ,  $Gd_2O_3$ ,  $Y_2O_3$ ,  $Lu_2O_3$  powders with the purity of 99.99% as the starting materials. The  $SrCO_3$  powder was heated at  $1100^\circ C$  for 10 h in order to release  $CO_2$  and to obtain SrO. The raw compounds were mixed at stoichiometric ratio by mortar and pestle. Next, the powder mixture was formed to a cylinder rod by applying a hydrostatic pressure. Further, the cylinder rod was sintered at  $1400^\circ C$  for 12 h to make into a solid ceramic rod. Finally, the ceramic rod was loaded into the FZ furnace, and we grew a single crystal. The crystal growth was conducted in air. The heat source of the FZ furnace was a halogen lamp, and the rotation rate and pull-down rate was 20 rpm and 3–8 mm/hr, respectively. The actual chemical compositions of the obtained crystal samples were measured by X-ray fluorescence (XRF; MESA-500W, Horiba). In addition, X-ray diffraction (XRD; MiniFlex600, Rigaku) was conducted in order to identify the crystal structure.

PL emission/excitation spectra and PL quantum yields (QY) were evaluated by using Quantaaurus-QY (C11347, Hamamatsu). The excitation and emission wavelength ranges were 250–500 nm and 200 nm–900 nm, respectively. PL QY was defined by  $QY (\%) = N_{emit}/N_{absorb}$  where  $N_{emit}$  and  $N_{absorb}$  are the numbers of emitted and absorbed photons, respectively. PL decay time profiles were characterized by Quantaaurus- $\tau$  (C11367, Hamamatsu).

Measurements of RL properties were conducted at room temperature as well as optical properties. RL spectra were measured by our original setup under X-ray irradiation [22]. The excitation source used here was a conventional X-ray tube (XRBP&N200X4550, Spellman), which was supplied with 2 mA of tube current and 80 kV of bias voltage. RL from the sample was guided to a monochromator (SR163, ANDOR) and then a charge coupled device (CCD) detector (DU920-BU2NC, ANDOR). The RL decay time profiles were evaluated by using our original setup [23], equipped with a pulse X-ray tube. A voltage of 30 kV was applied to the tube and repetitive X-ray pulse of a few ns was delivered at a frequency of 200 kHz. The X-ray induced afterglow profiles were also evaluated by using the same setup with 2 ms irradiation. We evaluated absolute scintillation light yields by using pulse height spectroscopy techniques with  $^{241}Am$   $\alpha$ -rays as a radiation source. A sample covered by layers of Teflon tape was placed on a window of photomultiplier tube (PMT; R7600, Hamamatsu) with optical grease (OKEN6262A, OKEN). We formed a small hole in the Teflon tape at the center of sample in order to allow  $\alpha$ -rays to make it to the sample surface. A bias voltage of 700 V was supplied to the PMT by using a DC power supply (556, ORTEC), and the electrical signal output was amplified by a pre-amplifier (113, ORTEC). Besides, a shaping amplifier (ORTEC572, ORTEC) with 0.5  $\mu s$  shaping time processed the amplified signal to obtain the light intensity per event, which was then statistically accumulated by a multichannel analyzer (MCA-8000A, Amptek) in order to obtain a pulse height spectrum.

## 3. Results and discussion

The synthesized Ce-doped SrYS, SrGS and SrLuS samples are shown in Fig. 1. As seen in the figure, the typical size of the obtained samples was approximately 4 mm in diameter and 20 mm in length. The color of SrYS and SrGS samples were yellow due to a strong absorption in the blue region by the 4f-5d transitions of  $Ce^{3+}$  [24] while SrLuS sample looks white. Under UV light (302 nm for SrYS and SrGS; 364 nm for SrLuS), we observed bright blue luminescence by naked eye. The emission seems to be the typical emission of  $Ce^{3+}$  due to the 5d-4f transitions. According to the XRF measurements, the actual Ce concentrations of  $Sr_2(Ce_{0.005}RE_{0.995})_8(SiO_4)_6O_2$  ( $RE = Y, Gd, Lu$ ) were 0.5%, 0.5% and 0.4%, respectively. Therefore, the nominal and actual Ce concentrations were very close. Fig. 2 represents XRD patterns of all the samples and the standard card pattern (JCPDS No: 28-0212) of  $Ca_2Gd_8(SiO_4)_6O_2$  as a reference. We confirmed that the YSrS and GSrS samples have mainly the apatite phase in the XRD patterns. However, it is likely that a greater part of the LuSrS sample was formed into ceramic, judging from the appearance and XRD patterns which exhibited some impurity phases. This result should be caused by the difference of ionic radius between the host ion ( $Lu^{3+}$ ) and substitution ions ( $Ce^{3+}$  and  $Sr^{2+}$ ). In the general

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