Investigation of Ce-doped $Gd_2Si_2O_7$ as a scintillator material

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Abstract

As a novel scintillation material, the use of Ce-doped $Gd_2Si_2O_7$ was investigated. In fact, $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ powder showed about five-times greater light output than Gd_2SiO_5 (GSO) powder samples for alpha-particles from ^{241}Am. Furthermore, a fast decay constant of 59 ns was obtained for alpha-particles from ^{241}Am. Relations between the Ce-concentration, crystal structure and luminescence characteristics were also elucidated. Results of these studies show that heavy Ce-doping alters the $Gd_2Si_2O_7$ crystal structure and that the luminescence intensity is dependent not on Ce-concentration but on the triclinic structure formed by heavy Ce-doping.

Keywords- gadolinium compounds, $Gd_2Si_2O_7$, neutron detectors, radiation detectors, scintillators

1. Introduction

Rare earth silicon oxide scintillators such as Gd_2SiO_5 (GSO) and Lu_2SiO_5 (LSO) are widely used for

radiological diagnosis because of their higher density, high light output, and fast decay constant [1–5].

Neutron scattering experiments for material sciences also require high-performance scintillators because higher intensity neutron sources such as J-PARC are under construction. For that reason, GSO is anticipated as a neutron scintillator because of the large neutron capture cross-section of Gd, greater light output [6], and good radiation-damage resistance [7]. However, Uozumi et al. reported higher sensitivity of GSO for detection of background gamma-rays in neutron scattering experiments [8]. Consequently, Reeder used thin GSO for neutron detection [9], and Konishi et al. used plastic as a moderator to reduce gamma-ray sensitivity [10]. Tokanai et al. also applied thin GSO single crystals to a position-sensitive detector for cold neutrons and described that position resolution would be improved using thinner GSO [11]. Another candidate of Gd-based scintillators is Gd_2O_2S (GOS), which is used in X-ray detection [12–14]. Although GOS has greater light output, its decay time of 4 μs is too long for application for neutron detection, which requires at least 1×10^6 cps count rate [15].

For those reasons, although we attempted fabrication of GSO film using evaporation or polishing aimed at decreasing gamma-ray sensitivity and improving the position resolution, the fragility of GSO [16] and its melting temperature of 1950°C prevented fabrication of GSO films.

Nevertheless, in the process described above, we found that Ce-doped $Gd_2Si_2O_7$ has a lower melting point < 1850°C than GSO and high luminescence intensity [17]. This paper presents results of a study of the relations between Ce-concentration, crystal structure, and luminescence characteristics;

we report a comparison of light-output measurements of Ce-doped $Gd_2Si_2O_7$, GSO single-crystals and GSO powder samples using a ^{241}Am alpha-particle source.

2. Experimental

2-1. Sample preparation

These experiments used a $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ powder sample and Gd_2SiO_5 (GSO) single-crystals purchased from Hitachi Chemical Co. Ltd. and $(Gd_{1-x}Ce_x)_2Si_2O_7$ (x=0–0.1) powders fabricated by solid state reaction. Then, Gd_2O_3, SiO_2, and CeO_2 powders whose respective purities were 5N, 5N, and 4N were mixed at a composition of $(Gd_{1-x}Ce_x)_2Si_2O_7$ (x=0–0.1). The mixed powders were sintered at 1500°C for 8 h in an alumina crucible using an electric furnace in an air atmosphere. The sintered powders were crushed using an agate mortar and passed through sieves. The size of each particle was estimated as 50-100um.

X-ray diffraction patterns were measured using Cu-Ka X-ray imaging. The X-ray patterns were compared with Joint Committee on Powder Diffraction Standards (JCPDS) data to determine the crystal structure; each crystal structure concentration was evaluated using Crystal Studio version 8.0 (Crystal Systems Co., Ltd.).

2-2. Luminescence spectra and alpha-particles spectra measurement

Photoluminescence spectra of each composition sample were measured under 240-nm wavelength light irradiation using a spectrometer (FP6500; Jasco Inc.). Samples of 100 mg were weighed out and coupled to a glass plate using optical grease.

The pulse-height spectrum and decay curves of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ were also measured to compare the light output of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ with other scintillators using a ^{241}Am alpha-particle source. The light output was compared by evaluating the MCA channel of the total energy peak. In this experiment, GSO single crystals and GSO powder sample which was obtained by crushing GSO single crystals were used as references. Figure 1 shows that $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ and GSO powder sample were coupled to a glass plate and set. Ce-concentration of the GSO single-crystals was …... GSO single crystals were coupled to photomultiplier by optical grease. The H7195 photomultiplier tube (Hamamatsu Photonics KK), Ortec 113 pre-amplifier, and Ortec 672 spectroscopy amplifier were used for spectroscopy. A digital oscilloscope (LT584; Iwatsu Co.) was used at 1 GHz frequency range and 2 GS/s sampling for decay-curve measurements.

3. Results and discussion

3-1. Crystal structure of the samples

Figure 2 shows XRD patterns of $(Gd_{1-x}Ce_x)_2Si_2O_7$ powder samples with various compositions. Results showed that XRD patterns varied with increased Ce concentration. The crystal structure determined

by JSPDS and the relative abundance of each crystal structure are shown in Table 1, which shows that triclinic structure abundance increased with Ce concentration. According to Felsche's study of rare earth disilicate, $Gd_2Si_2O_7$ forms an orthorhombic structure and space group $Pna2_1$ or Pnam [18]. It is considered that heavy Ce-doping changed the GPS lattice parameter; subsequently, the triclinic structure was formed.

3-2. Luminescence spectrum of $(Gd_{1-x}Ce_x)_2Si_2O_7$ (x=0–0.1)

Fig3 shows the comparison of photo luminescence spectra of $(Gd_{1-x}Ce_x)_2Si_2O_7$ (x=0–0.1) samples under 240nm wavelength light irradiation. Increase of luminescence intensity with Ce-concentration was observed. The relation between luminescence intensity and abundance of triclinic structure are plotted in Fig4. Of course the luminescence intensity varies from crystal orientation or surface condition etc, however, remarkable increment of luminescence intensity was obtained in large triclinic structure abundance part. It is considered that $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ with triclinic structure contribute to luminescence. The peak wavelength 374nm and 397nm of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ are due to 5d - 4f transition in Ce^{3+} ion of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$.

3-2. α-Particle measurements

The $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ powder with the most abundant triclinic structure was used in pulse height

spectrum measurements using a ^{241}Am alpha-particle source. To compare the light output of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$, a GSO powder sample was also used in measurements. The total energy peak channel in Fig. 5 indicated that $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ had five-times greater light output than GSO. However, two peaks exist in the spectrum of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$.

A typical decay curve of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ was plotted in Fig. 6. The decay constant obtained from exponential function fitting was 52 ns and ….. This decay constant is 1/67 of the decay time of GOS, so higher counting rate is expected than GOS.

4. Conclusion

As the first step toward novel scintillator development, the relations between Ce-concentration of $Gd_2Si_2O_7$, crystal structure, and luminescence characteristics were examined in this study. Results showed variation of the crystal structure by Ce-doping and a relation between the abundance of triclinic structure and luminescence intensity. According to Felsche, a triclinic structure is not formed in $Gd_2Si_2O_7$ [18], but we considered that heavy Ce-doping changed the lattice parameter of $Gd_2Si_2O_7$ and that the triclinic structure was formed. Pulse-height spectrum measurements using an ^{241}Am alpha-particle source showed that $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ has five-times greater light output than GSO and a fast decay time of 59 ns. These results support the possibility of the use of Ce-doped$Gd_2Si_2O_7$ as a scintillation material.

References

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Figure caption

Table 1 Abundance of triclinic structure in $(Gd_{1-x}Ce_x)_2Si_2O_7$

Fig.1 Experimental set-up of alpha-particle spectra measurement.

Fig.2 XRD patterns obtained for $(Gd_{1-x}Ce_x)_2Si_2O_7$.

Fig.3 Photoluminescence spectra of $(Gd_{1-x}Ce_x)_2Si_2O_7$ under 250 nm irradiation

Fig.4 Relation between triclinic abundance and luminescence intensity.

Fig.5 Comparison of light outputs $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ with GSO. Pulse height spectra were measured using a ^{241}Am alpha-particle source.

Fig.6 Typical decay curve of $(Gd_{0.9}Ce_{0.1})_2Si_2O_7$ for alpha particles.

Table 1. Abundance of triclinic structure in $(Gd_{1-x}Ce_x)_2Si_2O_7$

Composition	Abundance of triclinic structure (%)	Other crystal structure (%)
$Gd_2Si_2O_7$	0	100
$(Gd_{0.99}Ce_{0.01})_2Si_2O_7$	0	100
$(Gd_{0.95}Ce_{0.05})_2Si_2O_7$	17	83
$(Gd_{0.90}Ce_{0.10})_2Si_2O_7$	35 86	65 14
$(Gd_{0.90}Ce_{0.10})_2Si_2O_7$ (Hitachi Chemical Co. Ltd.)	96	4

Figure caption

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Fig. 2. XRD patterns obtained for $(Gd_{1-x}Ce_x)_2Si_2O_7$.

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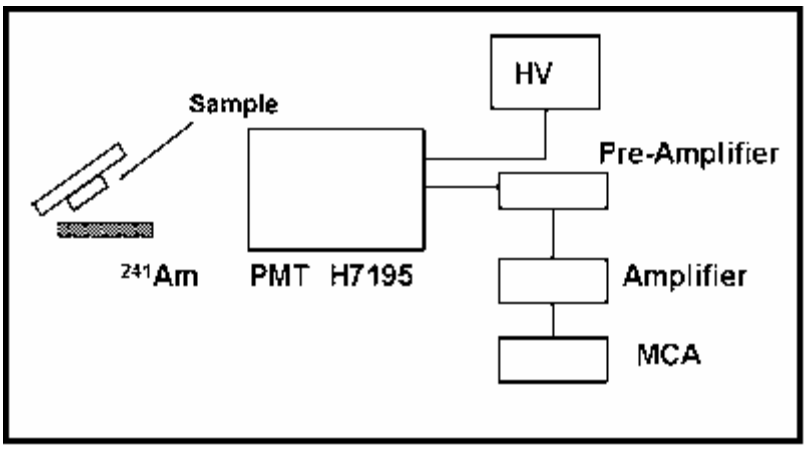

Fig1

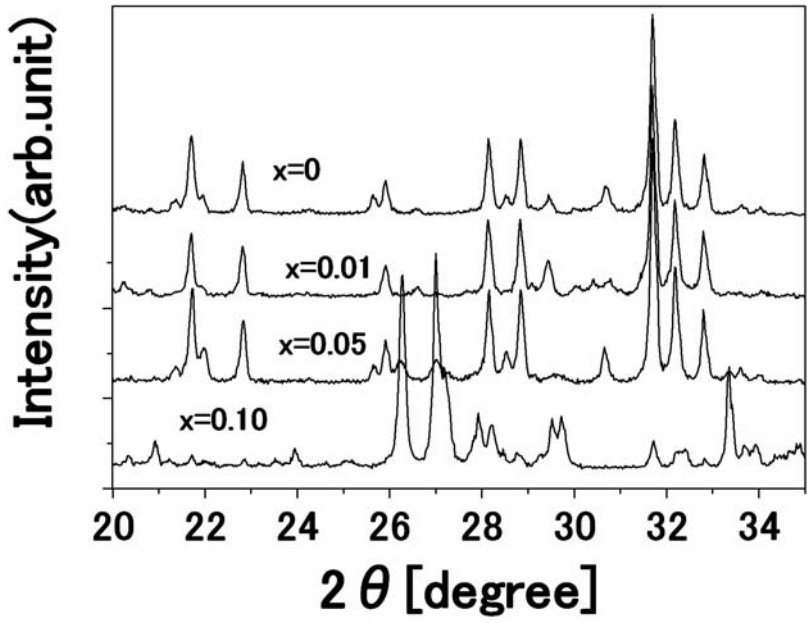
Intensity(arb.unit)
x=0
x=0.01
x=0.05
x=0.10
20
22
24
26
28
30
32
34
2θ [degree]

Fig. 2..

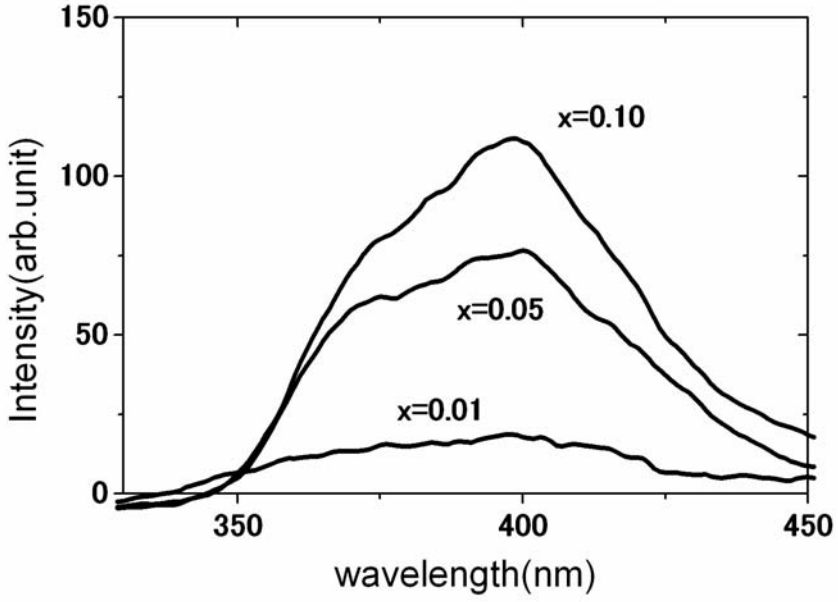
150
100
50
0
Intensity(arb.unit)
x=0.10
x=0.05
x=0.01
350
400
450
wavelength(nm)

Fig. 3

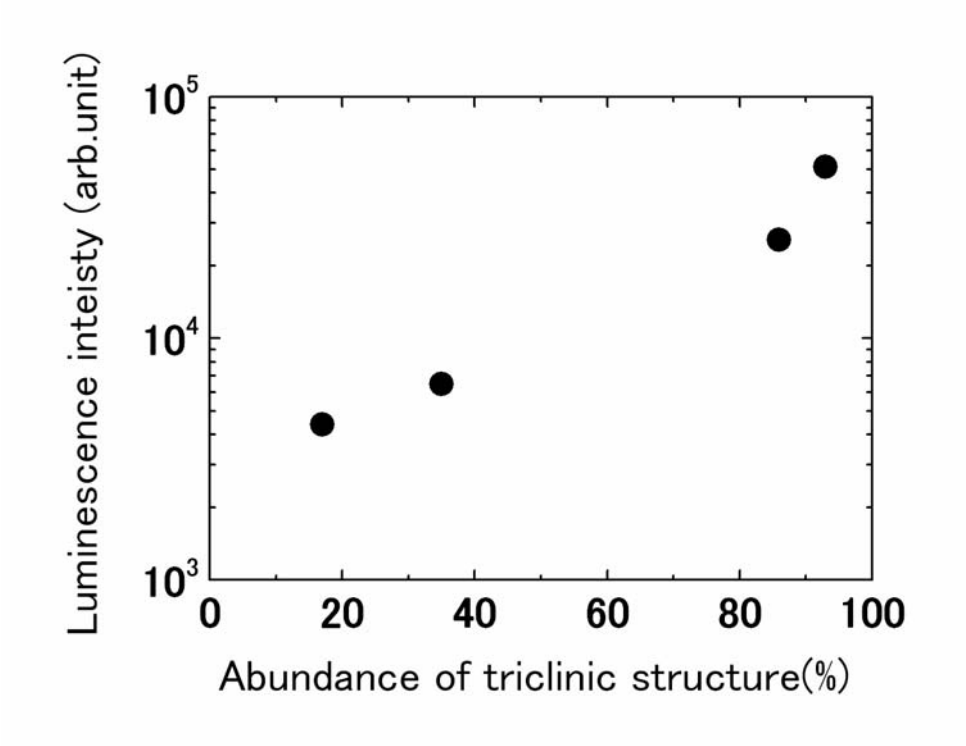
Luminescence inteisty (arb.unit)
10^5
10^4
10^3
0
20
40
60
80
100
Abundance of triclinic structure(%)

Fig.4

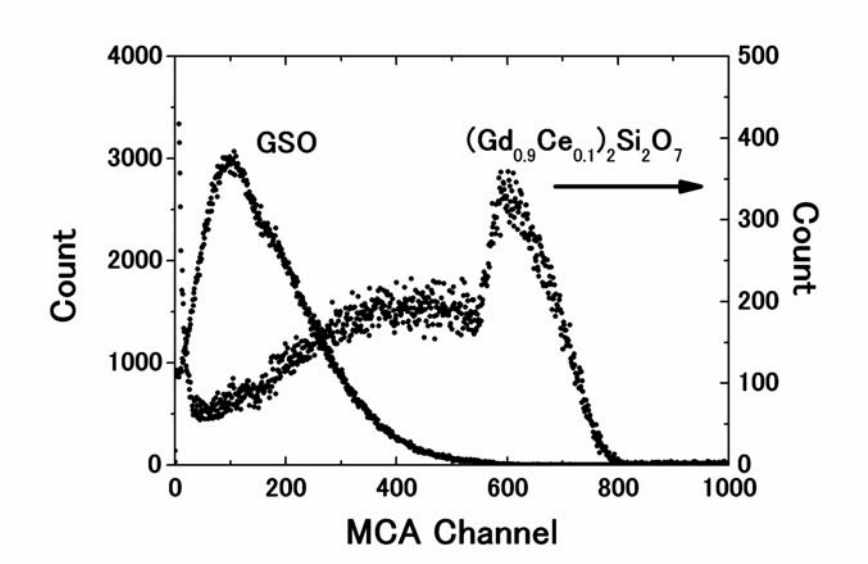
GSO
(Gd0.9Ce0.1)2Si2O7
Count
Count
4000
3000
2000
1000
0
500
400
300
200
100
0
0
200
400
600
800
1000
MCA Channel

Fig. 5

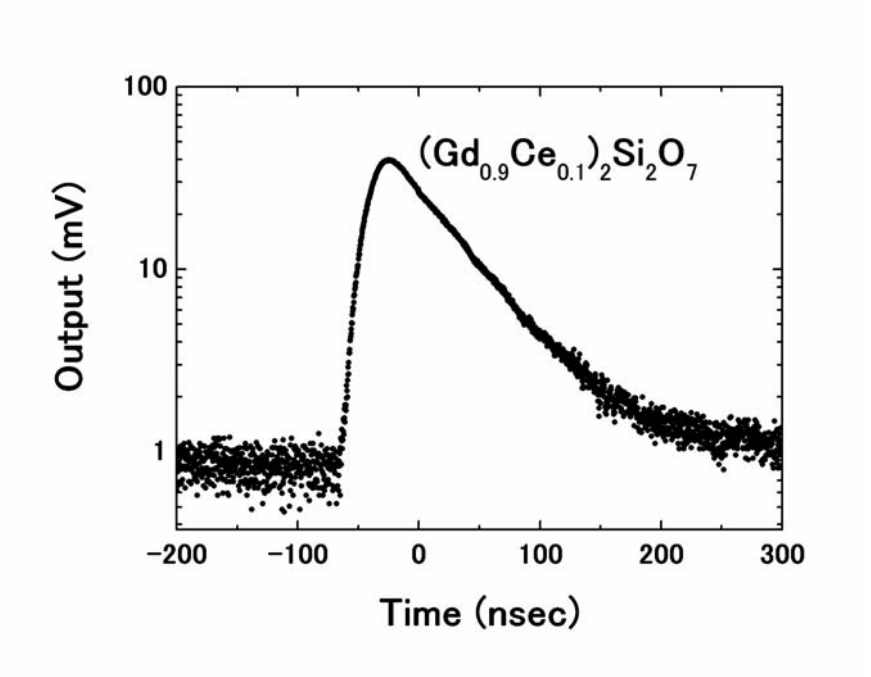
100
10
1
Output (mV)
(Gd0.9Ce0.1)2Si2O7
−200
−100
0
100
200
300
Time (nsec)

Fig.6.