Radiation Measurements 109 (2018) 8–12

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Structure, photoluminescence, and scintillation characteristics of a $Gd_{1.9}Y_{0.1}SiO_5$:0.5%Ce (GYSO:Ce) single crystal scintillator

He Feng[a,*], Jianyu Chen[b], Zhijun Zhang[a], Yong Wang[c], Zhan Xu[a], Jingtai Zhao[a], Rihua Mao[c]

[a] School of Material Science and Engineering, Shanghai University, Shanghai 200444, PR China
[b] Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Science, Shanghai 201800, PR China
[c] Shanghai Institute of Ceramics, Chinese Academy of Science, Shanghai 201800, PR China

ARTICLE INFO

Keywords:
Crystal structure
Energy transfer
Luminescence mechanism
Scintillator

ABSTRACT

A single crystal of $Gd_{1.9}Y_{0.1}SiO_5$:0.5%Ce (GYSO:Ce) was grown using the Czochralski method and its structure, photoluminescence, and scintillation properties were analyzed. Based on Rietveld structure refinement, we found that mixing with Y5at% changed the host structure very little. Two typical types of cerium emission, i.e., Ce1 and Ce2, were confirmed based on the photoluminescence excitation and emission spectra at 77 K. The energy transfer of Gd $^6P_J \rightarrow$Ce1 probably explains the slow component in the photoluminescence and scintillation processes. The light yield of the GYSO:Ce sample was determined as 11200 ph/MeV and the energy resolution was 8.6%. The afterglow intensity of GYSO:Ce was relatively low.

1. Introduction

Inorganic single crystals scintillators can convert high energy particles or neutrons into ultraviolet–visible light, and thus they are used widely in the detection of high energy particles and neutrons (Nikl and Yoshikawa, 2015). The Gd_2SiO_5:Ce (GSO:Ce) single crystal obtained by Takagi and Fukazawa (1983) is well known as an effective inorganic scintillator with a high light output, relatively low refractive index, and excellent high temperature scintillation performance. Therefore, GSO:Ce scintillators can be used in various radiation detection fields, such as positron emission tomography and high well logging.

Due to its structural anisotropy, a GSO single crystal is inclined to crack along the (100) cleavage plane (Sidletskiy et al., 2012). One method that can overcome this cleavage problem is Lu or Y mixing. In previous studies of GSO:Ce-based mixed crystal scintillators, Lu mixing with GSO:Ce (GLSO:Ce) was studied intensively (Jarý et al., 2014; Shimizu et al., 2006; Usui et al., 2007). However, few studies have investigated the crystal growth and performance of GSO:Ce single crystals with Y mixing (Jie et al., 2005).

To obtain insights into Y mixed GSO:Ce single crystals, we produced a single crystal of $Gd_{1.9}Y_{0.1}SiO_5$:0.5%Ce (GYSO:Ce) using the Czochralski method, where we determined its structural characteristics and fundamental photoluminescence (PL) and scintillation properties.

2. Experimental

The nominal composition of the raw material was $Gd_{1.8905}Y_{0.0995}Ce_{0.01}SiO_5$ and a crystal boule was obtained via the Czochralski method. The detailed preparation process and an image of the as-grown crystal boule were provided in previous study (Jie et al., 2005). The single crystal sample was ground to a powder to obtain X-ray diffraction (XRD) measurements. The dimensions of the sample used to obtain PL and scintillation measurements were 7 × 6 × 2 mm and two sides were polished. The XRD pattern was recorded with a Huber imaging plate Guinier camera (G670; Cu-Kα1 radiation, λ = 1.54056 Å, 2θ interval = 3–100°, and step = 0.005°). The X-ray excited luminescence (XEL) spectrum was recorded using an in-house manufactured XEL spectrometer with a W target. The density of the GYSO:Ce sample was measured with the Archimedes drainage method. The transmittance spectrum was measured using a Hitachi U-3900H spectrophotometer. The PL properties comprising the excitation (PLE), emission (PL), and PL decay curves were recorded with an Edinburgh FLS920 time resolved and steady state fluorescence spectrometer. The γ-ray pulse spectrum was measured under a ^{137}Cs source and a photomultiplier tube (PMT Hamamatsu R1306) was used to detect the luminescence of the GYSO:Ce. The sample was wrapped in Tyvek tape and silicone oil was placed between the sample and PMT to increase the luminescence collection efficiency. The voltage of the PMT was −650 V and the shaping time was 1 μs. To estimate the light yield in units of ph/MeV, a cerium-doped lutetium yttrium orthosilicate (LYSO:Ce)

* Corresponding author.
E-mail address: fh117@shu.edu.cn (H. Feng).

https://doi.org/10.1016/j.radmeas.2017.12.001
Received 14 July 2017; Received in revised form 17 November 2017; Accepted 11 December 2017
Available online 14 December 2017
1350-4487/

standard sample with similar dimensions was also recorded under the same measurement conditions. The energy resolution was calculated according to the Gaussian fit result for the full energy peak. The scintillation decay curve for the sample was obtained with the PMT R1306 and recorded using a DSO Agilent 9254 digital scope. The afterglow curve was recorded with a PMT R2059 under −1200 V after 2 s of continuous X-ray (15 kV and 15 μA) irradiation.

3. Results and discussion

3.1. Structural determination

The structure of GYSO:Ce was determined by Rietveld refinement. The raw XRD data were automatically indexed with the Dicvol program (Werner, 1990), which indicated the presence of monoclinic cells with parameters of $a = 9.12$ Å, $b = 7.03$ Å, $c = 6.74$ Å, and $\beta = 107.49°$. The previously reported XRD pattern based on the atomic coordinates of GSO (Smolin and Tkachev, 1969) was very similar to the experimental pattern, so this structural model was used in the subsequent calculations. Rietveld refinement was performed using FULLPROF (Rodriguez-Carvajal, 1998). According to the fitted results, the structure of the as-grown GYSO:Ce crystal was monoclinic with the space group of $P2_1/C$. The cell parameters were $a = 9.12$ Å, $b = 7.03$ Å, $c = 6.74$ Å, and $\beta = 107.49°$. The refined atomic coordinates are shown in Table 1. The observed, calculated, and difference profiles obtained for GYSO are shown in Fig. 1.

Fig. 2 shows the transmittance spectrum obtained for the GYSO:Ce sample and an image of the sample is also shown in the inset (a). Absorption was observed simultaneously from Gd^{3+} and Ce^{3+}. The absorption bands in the 200–375 nm range were ascribed to the 4f→5d electron transition of Ce^{3+}, as also found in GSO:Ce (Tanaka et al., 1998) and LYSO:Ce (Qin et al., 2005). The small absorption peak around 315 nm as shown in the inset (b) was due to the $^8S_{7/2}\rightarrow{}^6P_J$ electron transition of Gd^{3+}. The transmittance at about 82% above 433 nm indicated the good optical quality of the as-grown GYSO:Ce crystal sample.

3.2. PL properties of GYSO:Ce

The fundamental PL properties were investigated based on the PLE, PL, and PL decay curves. The PLE and PL curves recorded for GYSO:Ce at 77 K are shown in Fig. 3. It has been reported that the cerium ions occupy two types of sites (Ce1 and Ce2) in several rare earth orthosilicate hosts, including LSO, GSO, YSO, and their mixed crystals (Feng et al., 2010; Jarý et al., 2014; Suzuki et al., 1994a; Zorenko et al., 2013). The results were similar for GYSO:Ce, where the peak positions of the excitation and emission spectra were located at 350 and 430 nm for Ce1, and 380 and 501 nm for Ce2, respectively. The two small peaks located at 274 and 315 nm in the PLE curve for Ce1 emission as shown in the inset corresponded to the $^8S_{7/2}\rightarrow{}^6I_J$ and $^8S_{7/2}\rightarrow{}^6P_J$ electron transition of Gd, thereby indicating that the energy transfer of Gd (^6I_J and ^6P_J)→Ce1 occurred during the PL process.

The appearance of the Gd→Ce related excitation peaks indicated that two types of Ce emission occurred: prompt Ce and Gd→Ce energy

Table 1
Refined atomic coordinates obtained for GYSO.

Atom	x/a (Å)	y/b (Å)	z/c (Å)
Gd1	0.11537 (13)	0.14819 (25)	0.41482 (19)
Gd2	0.52811 (14)	0.62436 (28)	0.23552 (19)
Si	0.19292 (74)	0.57863 (81)	0.47045 (85)
O1	0.22873 (136)	0.45308 (120)	0.65177 (233)
O2	0.16162 (124)	0.45648 (137)	0.23468 (159)
O3	0.37001 (119)	0.65021 (235)	0.49526 (142)
O4	0.07912 (119)	0.77739 (139)	0.38477 (184)
O5	0.34274 (111)	0.38822 (194)	0.03625 (166)
Y1	0.11537 (13)	0.14819 (25)	0.41482 (19)
Y2	0.52811 (14)	0.62436 (28)	0.23552 (19)
Ce1	0.11537 (13)	0.14819 (25)	0.41482 (19)
Ce2	0.52811 (14)	0.62436 (28)	0.23552 (19)

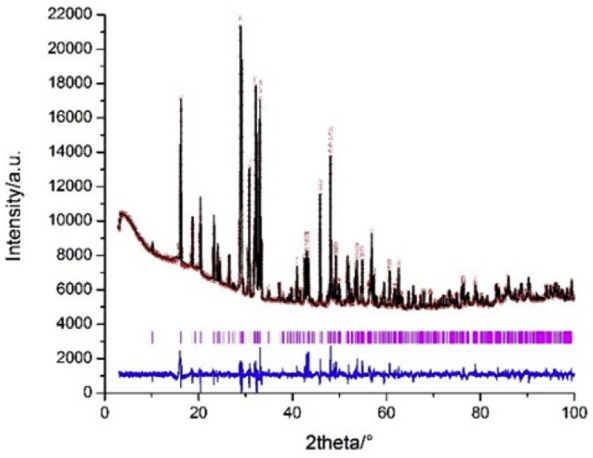

Fig. 1. Observed (o), calculated (full line), and difference profiles (lower trace) obtained for the GYSO:Ce sample.

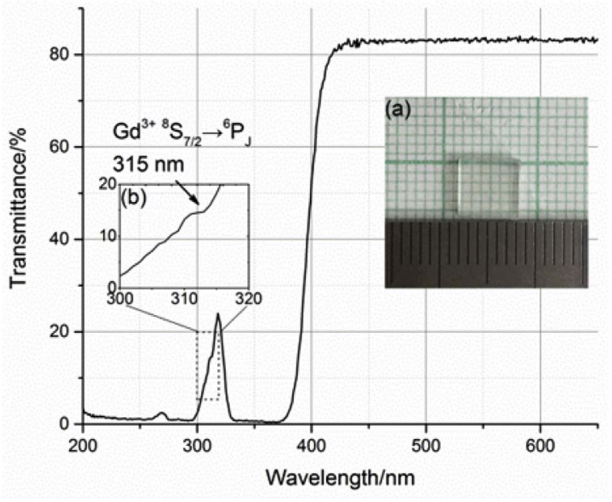

Fig. 2. Transmittance spectrum obtained for the GYSO:Ce sample. An image of the sample is shown in the inset (a) and an enlargement picture in the 300–320 nm range of this spectrum is shown in the inset (b).

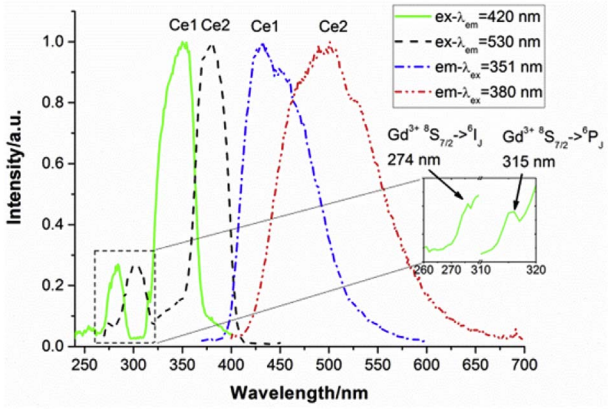

Fig. 3. PLE and PL curves recorded for the GYSO:Ce sample at 77 K. An enlargement picture in the 260–320 nm range of PLE spectrum monitored at 420 nm is shown in the inset.