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Luminescence and structural properties of $Gd_2SiO_5:Eu^{3+}$ phosphors synthesized from the modified solid state method

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ABSTRACT

This paper reports the preparation of Eu^{3+} doped Gadolinium oxyorthosilicate ($Gd_2SiO_5:Eu^{3+}$) phosphor with different concentration of Eu^{3+}(0.1–2.5 mol%) using the modified solid state reaction method. The synthesis procedure of the $Gd_2SiO_5:Eu^{3+}$phosphor using inorganic materials such as Gd_2O_3, silicon dioxide (SiO_2), europium oxide (Eu_2O_3) and boric acid (H_3BO_3) as flux is discussed in detail. The prepared phosphor samples were characterized by using X-Ray Diffraction (XRD), Field Emission Gun Scanning Electron Microscopy (FEGSEM), Transmission Electron Microscopy (TEM), Fourier Transform Infrared Spectroscopy (FTIR), Photoluminescence (PL) and Thermoluminescence (TL). The Commission Internationale de l'Eclairage(CIE) coordinates were also calculated. The PL emission was observed in the 350–630 nm range for the $Gd_2SiO_5:Eu^{3+}$ phosphor. PL excitation peaks were observed at 266, 275, 312 and 395 nm while the emission peaks were observed at 380, 416, 437, 545, 579, 589, 607, 615 and 628 nm. The emission peak at 615 nm was the most intense peak for all the different Eu^{3+} concentration samples. From the XRD data, using the Scherrer's formula, the average crystallite size of the $Gd_2SiO_5:Eu^{3+}$ phosphor was calculated to be 33 nm. TL was carried out for the phosphor after both UV and gamma irradiation. The TL response of the $Gd_2SiO_5:Eu^{3+}$ phosphor for the two different radiations was compared and studied in detail. It was found that the present phosphor can acts as a single host for red emission (1.5 mol%) for display devices and light emitting diode (LED) and white light emission for Eu^{3+}(0.1 mol%) and it might be used as a TL dosimetric material for gamma dose detection.

1. Introduction

Rare-earth silicates are an important group of inorganic compounds that can be applied in microwave devices, semiconductors, ferroelectrics, ferromagnetic, lasers and phosphors [1,2]. Several methods have been used for obtaining rare-earth silicates in various systems that are used for preparing devices for different applications. Gadolinium oxyorthosilicate, Gd_2SiO_5 (GSO), is a good host material of a prominent scintillator, GSO:Ce, used for gamma-ray detection [1–4]. GSO with its wide band gap (~6.1 eV) is a good candidate for VUV excited phosphor use, may be even leading to a usable quantum cutter [4]. The development of plasma display panels (PDP) and mercury-free fluorescent tubes demands new phosphors to efficiently convert the vacuum ultraviolet (VUV) radiation that is generated by noble gas discharges into visible light [5]. In the quantum cutting process, every one photon absorbed by the VUV excited phosphors, two or more visible photons should be emitted. In the Gd^{3+}-Eu^{3+} [6] system the use of energy transfer between two kinds of rare earth ions can open a new possible route of visible quantum cutting [5].

The present paper reports on the synthesis, characterization and effects of a variable concentration of Eu^{3+} on the luminescence of $Gd_2SiO_5:Eu^{3+}$ phosphor. The photoluminescence (PL) of a variable concentration of Eu^{3+}(0.1–2.5 mol%) are reported. The presence of the Gd^{3+} absorption in the Eu^{3+} excitation spectra was monitored to determine if any energy transfer from Gd^{3+} to Eu^{3+} occurred.

2. Experimental

Phosphor of Gd_2SiO_5 doped with Eu^{3+} ions with a variable molar concentration of Eu^{3+}(0.1–2.5 mol%) was prepared by the solid state

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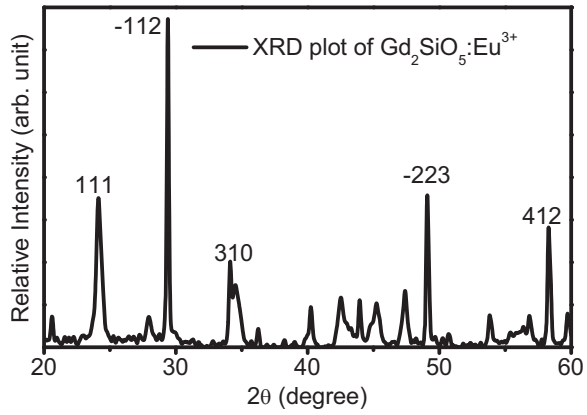

Fig. 1. XRD spectra of $Gd_2SiO_5:Eu^{3+}$(1.5 mol%) phosphor each peak is indexed in Table 1.

reaction method. The precursors Gd_2O_3, SiO_2, Eu_2O_3 and H_3BO_3 (as flux) were used for the synthesis of the $Gd_2SiO_5:Eu^{3+}$. The composition of each chemical was weighed in a proper stoichiometric ratio and then mixed thoroughly for 45 min using a mortar and pestle. The ground sample was placed in an alumina crucible and subsequently fired at 1000 °C for 1 h for calcinations and then at 1250 °C for 3 h for sintering in a muffle furnace. Every heating was followed by intermediate grinding. Finally the samples were cooled slowly to room temperature in the furnace and ground again into powder for subsequent characterization.

Complete reaction is given as:

$$Gd_2O_3 + SiO_2 + Eu_2O_3 \xrightarrow[H_3BO_3(Flux)]{\Delta 1000^{\circ}} Gd_2SiO_5 : Eu^{3+} \xrightarrow{\Delta 1250^{\circ}C} Gd_2SiO_5 : Eu^{3+}$$

The X-ray diffraction (XRD) measurements were carried out using a Bruker D8 Advance X-ray diffractometer. The X-rays were produced using a sealed tube and the wavelength of the X-ray was 0.154 nm (Cu K-alpha). The X-rays were detected using a fast counting detector based on a Silicon strip technology. Rietveld fitting method [7] was used to analyse the XRD patterns. The Fourier Transform Infrared Spectroscopy (FTIR) spectrum of a sample was recorded at room temperature in the wave number range of 4000–400 cm^{-1} on a Bruker spectrophotometer. FEGSEM (Field emission gun scanning electron microscopy) TEM (transmission electron microscopy) used for determination of surface morphology and exact particle size. The PL emission and excitation spectra were recorded at room temperature by using a Shimadzu RF-5301 PC spectrofluorophotometer. The excitation source was a Xenon lamp. The obtained phosphor was expose to UV light using a 254 nm UV source and gamma rays (Co^{60} source) with a dose of 0.1–2.0 kGy radiation, respectively. TL glow curves were recorded at room temperature by using a TLD reader I1009 supplied by Nucleonix Sys. Pvt. Ltd. Hyderabad [8–13].

Table 1b
Summary of diffraction angle, FWHM, d spacing and crystallite size of $Gd_2SiO_5:Eu^{3+}$.

2θ°	β (FWHM)°	d spacing Å	D (Crystallite size) nm
20.450	0.236	4.339	34.293
23.918	0.334	3.717	24.316
29.102	0.216	3.065	37.986
33.764	0.216	2.652	36.775
39.791	0.216	2.263	39.076
43.434	0.216	2.081	39.548
46.829	0.295	1.938	29.369
48.495	0.256	1.875	34.106
53.152	0.295	1.721	30.129
56.120	0.256	1.637	35.231
57.554	0.295	1.600	30.738
58.968	0.315	1.565	29.014
60.369	0.236	1.532	34.293

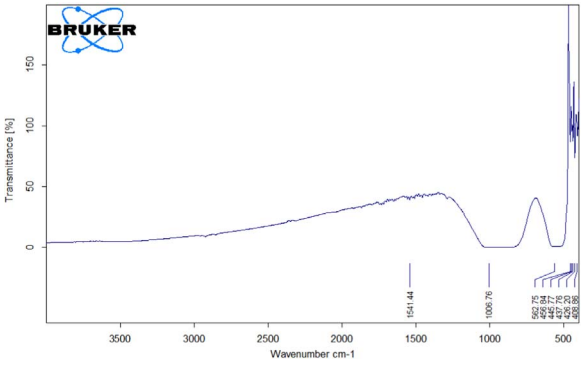

Fig. 2. FTIR Spectrum of $Gd_2SiO_5:Eu^{3+}$(1.5 mol%).

Table 1a
Indexing and lattice parameters of $Gd_2SiO_5:Eu^{3+}$.

Lattice parameters of reference pattern of $Gd_2SiO_5:Eu^{3+}$ reported by Dramicanin et al. [15]							
Lambda	**a**	**b**	**c**	**Alpha**	**Beta**	**Gamma**	**Vol.**
1.5418 Å	9.1105 Å	6.9783 Å	6.8544 Å	90 Å	107.14 Å	90 Å	416.4 $Å^3$
0	1	1	1	0	1	0	
Lattice parameter of $Gd_2SiO_5:Eu^{3+}$ (1.5 mol%) after refinement: (Standard errors on 2nd line)							
1.5418 Å	9.0682 Å	6.957 Å	6.85 Å	90 Å	106.88 Å	90 Å	413.5 $Å^3$
0	0.1103	0.0772	0.0882	0	1.404	0	
h	**k**	**l**	**2θ(Obs)**	**2θ(Cal)**	**Dif**		
2	0	0	20.45	20.4689	−0.0189		
1	1	1	23.918	23.0939	0.8241		
−1	1	2	29.102	29.1583	−0.0563		
3	1	0	33.764	33.5521	0.2119		
3	1	1	39.791	39.7071	0.0839		
0	1	3	43.434	43.4076	0.0264		
2	2	2	46.829	47.1992	−0.3702		
−2	2	3	48.495	48.511	−0.016		
1	2	3	53.152	53.0851	0.0669		
2	3	2	56.12	56.1744	−0.0544		
−3	1	4	57.554	57.5617	−0.0077		
4	1	2	58.968	58.9908	−0.0228		
5	1	1	60.369	60.4948	−0.1258		