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Research Article

SYNTHESIS AND OPTICAL PROPERTIES OF Ce^{3+} DOPED GADOLINIUM SILICATE PHOSPHORS

Atchyutha Rao Ch[1]*, Bujji Babu N[2] and Murthy K.V.R[3]

*[1]Department of Physics, SYKR&K GDC-Kovur-524137, SPSR NELLORE (DT), A.P, India
[2]Department of Chemistry, PR Govt Degree College (A), Kakinada- 533001, A.P, India
[3]Display Materials Laboratory, Applied Physics Department, Faculty of Technology and Engineering, M. S. University of Baroda, Vadodara-390001, India

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Gadolinium Silicates Phosphor, Rare earth ion, Conventional Solid state reaction method.

A B S T R A C T

Background: The phosphor is prepared by using the conventional Solid-state reaction method heated at 1200°C for 2 hrs. The received cakes are grounded for 30 minutes each. The present paper reports the optical properties of rare-earth-doped Gadolinium Silicate Phosphors. The Cerium Oxide (Ce^{3+}) was used as rare-earth-doped.

Method: Synthesis and optical properties of Ce^{3+} doped gadolinium silicate phosphors were prepared by conventional solid state reaction method. This synthesis route is very easy and does not require expensive as well as sophisticated equipment's. The major advantage of SSR method is, the final product in solid form is structurally pure with the desired properties depending on the final sintering temperatures. This method is environment friendly and no toxic or unwanted waste is produced after the SSR is complete. In this process the powders produced from SSR method is very fine as well as the cross contamination is very less. This method is also very convenient for large scale production on industrial scale.

Results: The phosphors are prepared and the received powders were characterized by X-ray diffraction (XRD), Scanning electron microscope (SEM), Photoluminescence study (PL) and Energy Dispersive through X-ray Elemental Analysis (EDAX) analysis. The following section discusses and the experimental results are mentioned in these phosphors. The present Phosphor can act as a host for blue light emission in many display devices and technological applications.

Conclusions: Synthesis and optical properties of Ce^{3+} doped gadolinium silicate phosphors were synthesized by simple conventional solid state reaction method. The crystallite size of synthesized phosphor powders was obtained in the approximate range of 27nm to 42nm. Also found that all the crystallite sizes are in nano form and we conclude majority phosphor crystallites are in nano form. From PL emission spectra is found very good broad peak at 438nm is due to transition $^5D_2 \rightarrow {}^7F_0$ with energy 2.6572ev. It is concluded that the formation of the phosphor is as per the empirical formula and weight percentage used to prepare the phosphors using a solid-state reaction (SSR) method. It is also concluded the SSR method is to synthesize the phosphors under study is a very good method.

INTRODUCTION

In the recent years, the phosphors development for three prime colours has attracted significant interest of researchers because of their possibility in technological applications like, high-resolution display devices and high-performance fluorescent lights. Recently, research related to the phosphors utilized for several forms such as LED TVs, LED lamps, simple lamps, TVs, signals, displays and mobile displays etc. Concerning many of these applications, the availability of systems consisting of uniform particles in size and shape is also an essential prerequisite for improved performance, and new synthetic routes are been developed in order to reach these systems. Rare earth ions are widely employed in the development of luminescent materials for exhibiting monochromatic emission colours due to their intrinsic optical properties. Rare earth applications in the field of display devices are still a warm topic much of the research around the world is to improve the phosphors efficiency and to improve the luminescence properties of the phosphors with discovery of blue light emitting devices. Recently various red phosphors materials have been actively investigated to improve their luminescent properties and to meet the development of different display and luminescence devices. Inorganic compounds doped with rare earth ions form an important class of phosphors as they possess a few interesting characteristics such as excellent chemical stability, high luminescence efficiency, and flexible emission colors with different activators [1-6]. Rare earth ion-doped hosts have demonstrated

Corresponding author:* **Atchyutha Rao Ch
Department of Physics, SYKR&K GDC-Kovur-524137, SPSR NELLORE (DT), A.P, India

good photoluminescence (PL) properties and chemical-physical stabilities. Ce^{3+} in such kinds of a host may emit various colors demanded by blue lighting. Rare earth ion-doped phosphors have been used in varied fields based on their electronic and optical characters arising from their $^4f_2$ electrons. Among the rare earth elements, europium is a special element as dopant, because it exhibits the property of valence fluctuation, i.e., the valence state is divalent or trivalent. And it exhibits different characteristics luminescence due to the different valence. The blue light emission of Ce^{3+}at 438nm is due to transition $^5D_2 \rightarrow {}^7F_0$ with energy 2.6572ev [7-10]. The rare-earth is usually incorporated in these materials as divalent or trivalent cation for the realization of optically active materials in photonics and optoelectronic applications. The cerium is efficiently used as a luminescent center in phosphors for various purposes. Phosphors doped with cerium ions are of greater importance for observing blue colors on the monitors of various display devices [11-14]. In this research paper, we have studied the optical properties of Ce^{3+} doped Gadolinium Silicates Phosphors prepared by the conventional solid-state reaction method fired at 1200°C for 2h. The prepared phosphors were characterized by subjected to PL, XRD, SEM, and EDAX analysis.

Experimental Method

The conventional Solid state reaction method was utilized for preparing these phosphors, which is the simpler and standard method. The inorganic compounds like Gadolinium oxide (Gd_2O_3), Silicon dioxide (SiO_2), and Cerium Oxide (Ce_2O_3) of high purity (99.9%) chemicals were used as starting materials. First, we prepared Gd_2SiO_4 phosphor, without adding any dopants, as a host material, by weighing Gadolinium oxide (Gd_2O_3), Silicon dioxide (SiO_2) in stoichiometric proportions of 2:1. The compounds were mixed with a spatula and then ground into a fine powder using an agate mortar and pestle manually about an hour at room temperature. The grounded sample was placed in an alumina crucible and heated at 1200°C in the air for 3 hours in a muffle furnace with a heating/cooling rate of 5°C/min.

RESULTS AND DISCUSSION

Crystal structure analysis

To determine the crystal structure and phase purity of the phosphors, XRD analysis was carried out. The crystal structure of the prepared silicate phosphor was determined by using X-ray diffraction analysis. The XRD pattern of Gd_2SiO_5: Base phosphor and Gd_2SiO_5: Ce^{3+} (0.5mol %) phosphors are as shown in Fig.1a & 1b. From the XRD pattern analysis, it was found that the prominent phase formed is Gd_2SiO_5, after the diffraction peaks are well indexed based on the JCPDS card No.40-0287 [15]. The XRD pattern confirms the formation of the phosphor it may be majority is in single phase, since the sintering temperature required for silicate phosphor is around 1300°C. This may be the reason many peaks are observed in the XRD pattern of prepared Gd_2SiO_5: Base phosphor and Gd_2SiO_5: Ce^{3+} (0.5mol %) phosphors. Table 1 shows the calculated crystallite sizes of the phosphors from the XRD pattern using Scherer's formula. $D = K\lambda / \beta \cos\theta$, Where D = crystallite size, K = constant, λ = X-ray wavelength, β = Full width at half maxima (FWHM), θ = Angle of the big peak [16, 17]. From table 1 it is found that all the crystallite sizes are in nano form and we conclude majority phosphor crystallites are in nano form. It is also observed that as Ce^{3+} concentration increases the average crystallite size gradually increases. Fig. 2 shows the relation between Ce^{3+} (mol %) percentages concentration in the present phosphor vs crystallite size. It is concluded that as Ce^{3+} (mol %) percentages concentration increases in Gd_2SiO_5 phosphor crystallite size is also increasing.

Table 1 Calculated crystallite sizes of the phosphors

S. No	Name of the phosphor	Crystallite size (nm)
1	Gd_2SiO_5: Base	26.73
2	Gd_2SiO_5: Ce^{3+} (0.5mol %)	29.34
3	Gd_2SiO_5: Ce^{3+} (0.2mol %)	30.58
4	Gd_2SiO_5: Ce^{3+} (0.5mol %)	33.14
5	Gd_2SiO_5: Ce^{3+} (1.0mol %)	34.41
6	Gd_2SiO_5: Ce^{3+} (1.5mol %)	34.49
7	Gd_2SiO_5: Ce^{3+} (2.0mol %)	34.62
8	Gd_2SiO_5:Ce^{3+} (2.5mol %)	42.31

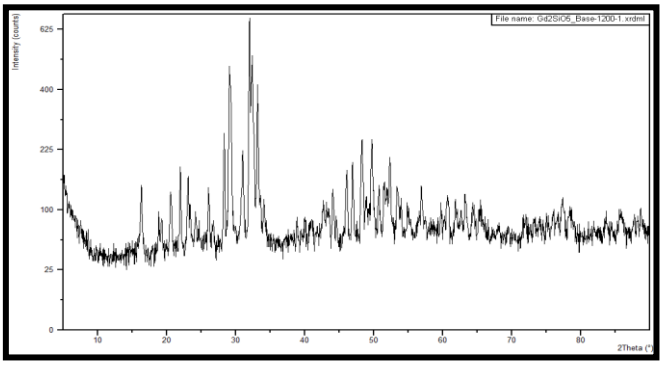


Fig 1(a) XRD pattern of Gd_2SiO_5: Base phosphor

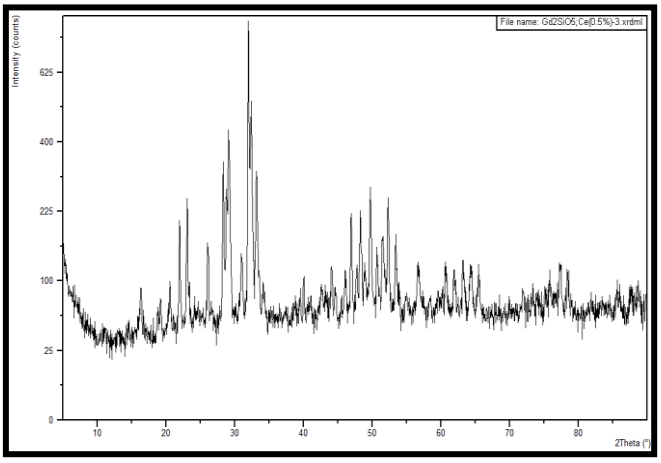


Fig. 1(b) XRD pattern of Gd_2SiO_5: Ce^{3+} (0.5mol %) phosphor

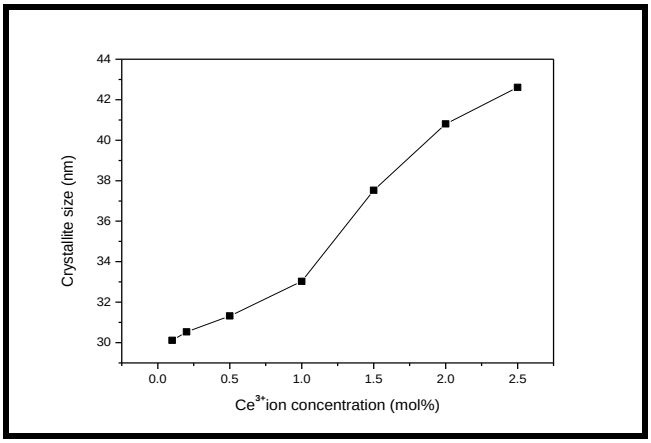


Fig 2 Ce^{3+} (mol %) percentages concentration Vs crystallite size

2 PL studies

The phosphors of various (mol%) concentration of Ce^{3+} doped Gd_2SiO_5 were prepared by using conventional solid state reaction method and heated at 1200°C for 2 hrs in open air atmosphere. Fig. 3.1 –3.4 are the excitation and emission spectrum of Ce^{3+} doped Gd_2SiO_5 phosphor with different concentrations (0.1, 0.2, 0.5, 1.0, 1.5, 2.0 and 2.5 mol %). From the figures it is observed that two excitations were considered i.e., 347 and 700nm and emissions are recorded accordingly. The excitation spectra were recorded by monitoring 440nm wavelength. From PL emission spectra is found very good broad peak at 438nm is due to transition $^5D_2 \rightarrow {}^7F_0$ with energy 2.6572ev. It is also observed that the Ce^{3+} ion concentration increases the emission at 438nm is gradually increases and the 700nm excitation gives the emission around 440nm nearly. Same intensity was observed at 340nm excitation [18]. Therefore, it is concluded this phosphor with less concentration Ce^{3+} ion may be a good blue emitting phosphor when excited with 350nm and also the high concentration of Ce^{3+} ion gives rise very good blue emission when excited with 350nm is a good LED material when excited with nUV LED chip (350nm).

Table 2 shows that the emission wavelength vs emission intensity for various concentrations of Ce^{3+} doped Gd_2SiO_5 phosphor with different concentrations and different excitation wavelengths at 347 and 700nm.

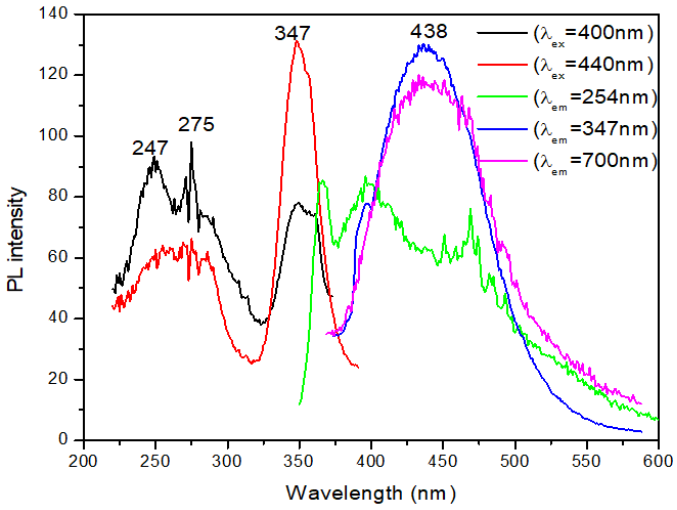


Fig. 3.1 PLE & PL spectrum of Gd_2SiO_5: Ce^{3+} (0.1mol %) phosphor

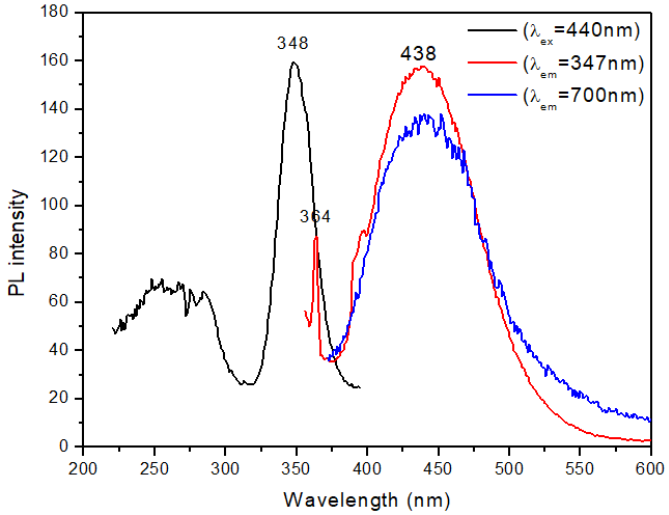


Fig. 3.2 PLE & PL spectrum of Gd_2SiO_5: Ce^{3+} (0.5 mol %) phosphor

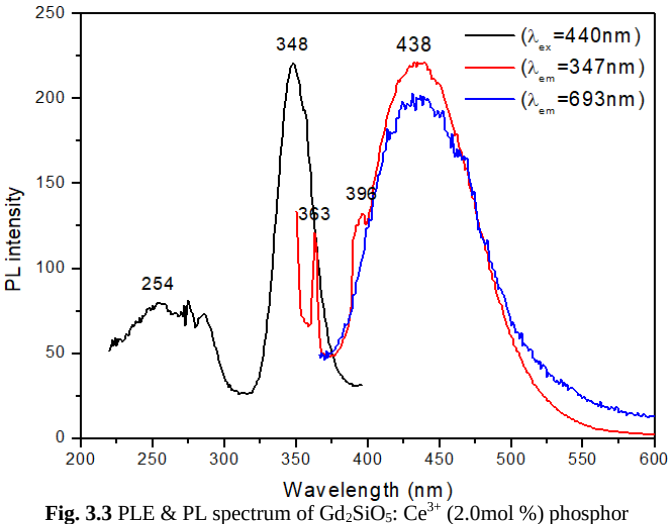


Fig. 3.3 PLE & PL spectrum of Gd_2SiO_5: Ce^{3+} (2.0mol %) phosphor

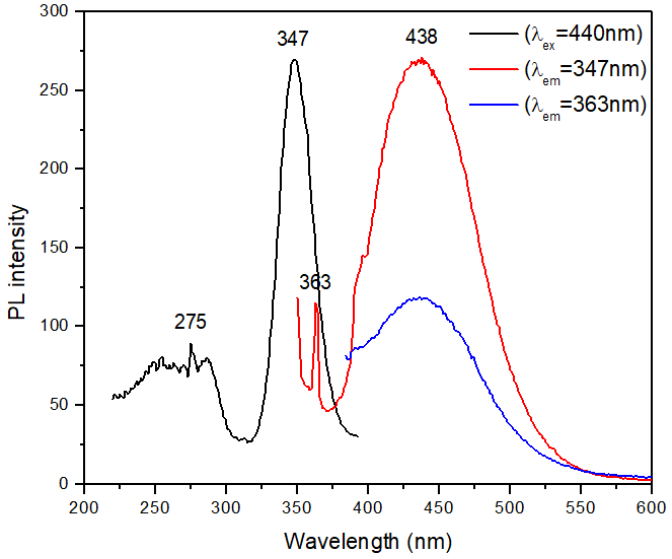


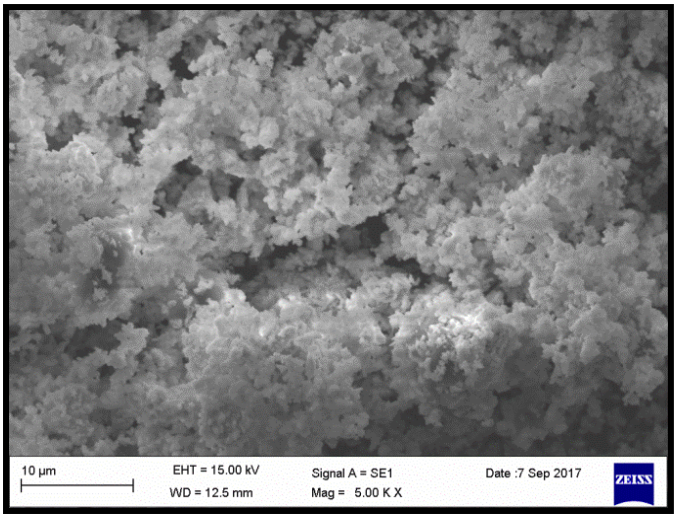
Fig. 3.4 PLE & PL spectrum of Gd_2SiO_5: Ce^{3+} (2.5mol %) phosphor

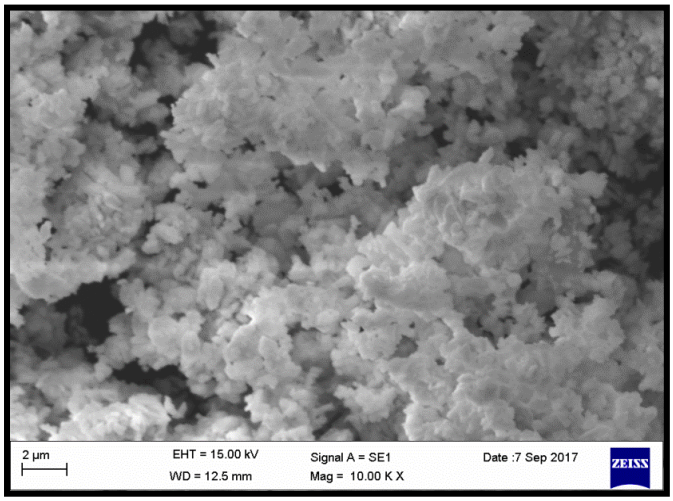
Table 2 Emission wavelength vs Emission intensity

S. No	Name of the Phosphor	Emission intensity of 438nm peak under	
		λ_{ex}=347nm	λ_{ex}=700nm
1	Gd_2SiO_4: Ce^{3+} (0.1mol %)	130	120
2	Gd_2SiO_4: Ce^{3+} (0.5 mol %)	160	140
3	Gd_2SiO_4: Ce^{3+} (2.0mol %)	225	200
4	Gd_2SiO_4: Ce^{3+} (2.5 mol %)	275	--

SEM study

Fig. 4 shows the SEM images of Ce^{3+} doped Gd_2SiO_5 phosphor with different concentrations and different resolutions. It is observed that from the SEM images of Ce^{3+} doped Gd_2SiO_5 phosphors particles are highly agglomerated with irregular size shape distribution look bunch of the flowers [19], and also just like as cutting of the cauliflower flakes. The particle sizes are in the size range is 2-10µm is seen.





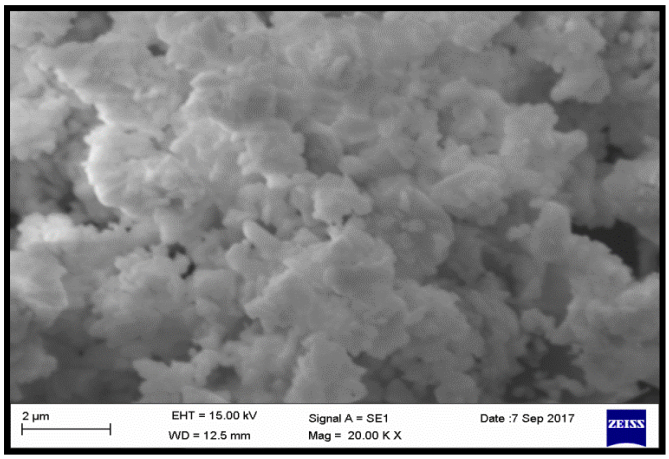


Fig 4 SEM images of Gd_2SiO_5: Ce^{3+} phosphor with different concentrations and different resolutions

EDAX analysis of Gd_2SiO_5: Ce^{3+} phosphor

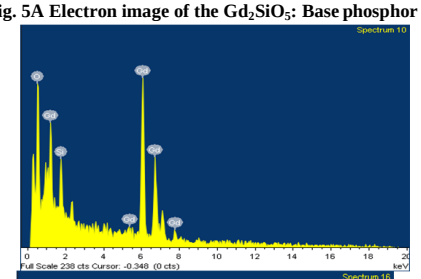
Gd_2SiO_5: Ce^{3+} phosphor subjected to another optical property which is "Energy Dispersive through X-ray Elemental Analysis" (EDAX). Fig. 5A and 5B are the electron images of EDAX analysis of Gd_2SiO_5 base phosphor and Ce^{3+} doped Gd_2SiO_5 phosphor, and the table containing the element, weight %, and atomic % of the phosphors under study [20]. It is also concluded the SSR method is to synthesize the phosphors under study is a very good method.

Fig. 5A Electron image of the Gd_2SiO_5: Base phosphor

Spectrum processing:

Peaks possibly omitted: 0.274, 2.316 keV
Processing option: All elements analyzed (Normalized)
Number of iterations = 2
Standard:
O SiO2 1-Jun-1999 12:00 AM
Si SiO2 1-Jun-1999 12:00 AM
Gd GdF3 1-Jun-1999 12:00 AM

Element	Weight %	Atomic %
OK	22.77	68.69
Si K	5.38	9.25
Gd L	71.85	22.05
Totals	**100.00**	



Spectrum processing:
Peaks possibly omitted: 0.266, 2.312 keV
Processing option: All elements analyzed (Normalized)
Number of iterations = 2
Standard:
O SiO2 1-Jun-1999 12:00 AM
Si SiO2 1-Jun-1999 12:00 AM
Ce CeO2 1-Jun-1999 12:00 AM
Gd GdF3 1-Jun-1999 12:00 AM

Element	Weight%	Atomic%
O K	27.86	74.02
Si K	5.20	7.87
Ce L	0.65	0.20
Gd L	66.29	17.92
Totals	**100.00**	

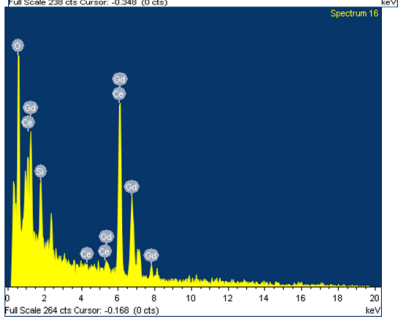


Fig. 5B Electron image of the Gd_2SiO_5:Ce^{3+} phosphor

CONCLUSIONS

- From the XRD studies the phosphor it may be majority is in single phase, since the sintering temperature required for silicate phosphor is around 1300°C. This may be the reason many peaks are observed in the XRD pattern of prepared Gd_2SiO_5: Base phosphor and Gd_2SiO_5: Ce^{3+} (0.5mol %) phosphors.
- From the PL studies the excitation spectra were recorded by monitoring 440nm wavelength.
- From PL emission spectra is found very good broad peak at 438nm is due to transition $^5D_2 \rightarrow {}^7F_0$ with energy 2.6572ev.
- It is also observed that the Ce^{3+} ion concentration increases the emission at 438nm is gradually increases and the 700nm excitation gives the emission around 440nm nearly.
- Therefore, it is concluded this phosphor with less concentration Ce^{3+} ion may be a good blue emitting phosphor when excited with 350nm.
- It is also concluded the high concentration of Ce^{3+} ion gives rise very good blue emission when excited with 350nm is a good LED material when excited with nUV LED chip (350nm).
- It is concluded from all the EDAX figures and tables the dopant Ce^{3+} ion, as well as Si and Oxygen of various percentages, are seen which are compared with the calculations made while preparing the phosphors.
- Therefore, it is mainly concluded the formation of the phosphor is as per the empirical formula and weight percentage used to prepare the phosphors using a solid-state reaction (SSR) method. It is also concluded the SSR method is to synthesize the phosphors under study is a very good method.

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