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Structural and Characterization of Luminescence Material Sr_2SiO_4: Eu Phosphor: Literature/Experimental Review

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Abstract: In this paper, comparison made on different Concentrations of Eu as a dopant has been discussed. The influence of structural and luminescence properties on different preparation methods have been reviewed and compared. Amidst Sol-Gel, spray pyrolysis, combustion and solid-state ceramic route methods, the sol-gel method was found to be stable the single phase structure with improved luminescence properties. Eu doped Sr_2SiO_4 enable material to be a candidate for white light LEDs application. The structure were observed orthorhombic, monoclinic & single phase with different preparation technique. Photoluminescence result shows the excitation spectrum of x=0.1, 0.5, 1.0, 2.5 mol% Eu doped strontium silicate sample with 592 nm emission and Excitation 296 nm, 243 nm emission and Excitation 592 nm, 256 nm emission and Excitation 612 nm, 370 nm emission and Excitation 560 nm. The energy transfer from Eu(I) of a high-energy emitting centre to Eu(II) of a low-energy emitting centre increased as Eu concentration increased, resulting in emission with a long wavelength and also the yellow, yellow-orange, orange-red and blue-shift.

Keywords: Sr_2SiO_4, Phosphors, Luminescence, Photoluminescence.

1 Introduction

Light emitting diodes have changed the imagination of the display devices from the solid state cooled lighting technology with flexible possibility with 180^o viewing angle [1].Solid-state lighting through LEDs based on the phosphors has attracted the attention of researchers due to their applications to develop a new class of efficient display devices. These devices are eco-friendly and meet future sustainable development goals. LEDs with their capabilities of excellent brightness, durable, long life, radiation hazards and energy effectiveness are relied upon the suitable host and rare earth dopants. Recently, it was observed that the particle size also plays a critical role in the optical properties. With the reduced size, it has been found that the material becomes more significant in the area of application especially in display technologies [2].
The accomplishment of application area diversification and add-on features advancement in its application of display materials and relative growth could be seen in the LED marketplaces [3]. Likewise, they are more effective, compatible and useful as far as low manufacturing cost and greater performance are concerned. Figure 1 displays the use of LEDs and other conventional lighting and their role in the world market. It could be seen that most of the conventional lighting technologies are now converted in to LED applications with the time. As per the available report [3], is found that the share proportion is shown in percentage decade back is now doubled with the area of application.
In recent times M_2SiO_4: Eu (where M = Ba, Ca, Sr) phosphors have drawn attention in order to evolve the white light producing diodes [4][5]. The primary Eu^{2+} assisted glow was first accounted in 1968[6]. Sr_2SiO_4:Eu orthosilicate phosphors are found to be stable candidature for white LEDs as they display high light transformation effectiveness close to bright (NUV) and blue light [7] .
The fluorescence of Eu2+ activated alkaline earth orthosilicate phosphors was first described by Barry. M_2SiO_4:Eu (M = Sr, Ca, Ba) in 1968. Sr_2SiO_4:Eu^{2+} phosphors, among the silicate phosphors, have unique features that offer potential benefits in white light emitting diode and thermo chromic applications. This is due to the presence of two crystalline phases in the host lattice of Sr2SiO4: In white LEDs, in the high temperature α' phase

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(orthorhombic) with space group Pmnb is used, and (ii) the low temperature β' -phase (monoclinic) with space group P21/c, which is used in thermochromics. Eu doping is comparable system to introducing impurities into the host lattice in that it has the ability to change the crystallinity and phase of the samples, resulting in phase Sr_2SiO_4. As a result, pure phase Sr_2SiO_4:Eu^{2+} phosphors are difficult to obtain. There have been a some reports on β -phase Sr_2SiO_4:Eu^{2+} phosphors made by the traditional solid state reaction approach. Though, the impact of Eu doping, as well as its limits in terms of creating pure -Sr_2SiO_4:Eu^{2+} phosphors, has yet to be fully investigated [8]. Through a comprehensive understanding of the Eu^{2+} photoluminescence mechanism and structure–property connections, we aim to produce Eu^{2+}-doped silicate phosphors that can address application challenges [9].

Because of their energy-saving, dependability, maintenance, and safety benefits, white light emitting diodes (LEDs) for lighting have gotten a lot of attention in recent years [9]. Researchers are currently interested in the increasing demand for light emitting diodes. The phosphors used in white light emitting diodes are currently being researched. LED lights have risen to prominence as a potent source of illumination. They have quickly risen to popularity in the Indian lighting sector due to their advantages over traditional lighting technologies.

White light emitting diodes, multicolor sensors, high-density optical storage systems, and high-energy radiation detection can be used rare earth ion-doped inorganic phosphors which is luminescent materials. Europium (Eu) is a rare earth element dopant with valence fluctuation between divalent (Eu^{2+}) and trivalent (Eu^{3+}) states, exceptionally effective phosphors, and narrow-band emission characteristics, allowing it to function as an emission centre in a host lattice.

Strontium silicate Sr_2SiO_4 is an excellent host material for phosphors due to the strength and thermal stability, stable crystal structure, high mechanical, which is provided by a tetrahedral silicate (SiO_4) 4-host matrix. It has potential application in the development of white light emitting diodes. Europium Eu is a common rare-earth metal that, when doped in hosts such as phosphates, silicates, and aluminates, exhibits a parity-allowed 4f-5d energy level transition from ultraviolet to red, depending on the host lattice and co-valence. . Eu^{2+} -activated Sr_2SiO_4 phosphor has two phases, i.e. α´ and β. Under NUV stimulation, the β -Sr_2SiO_4:Eu^{2+} produces a strong green-yellow band, making it an excellent choice for white LED phosphor.

Sr_2SiO_4:Eu^{2+},$Y_3Al_5O_{12}$: Ce (YAG:Ce) and $Tb_3Al_5O_{12}$:Ce(TAG:Ce) phosphors were mostly utilized as yellow-discharging phosphors for white light emitting diodes. Candidature of rare earth doped Sr_2SiO_4 were found and recommended by many research groups [2]. The expected variation in the luminescence properties with the percentage variation in the rare earth doping in the Sr_2SiO_4 sample required to be explored. In the present discussion the use of Sr_2SiO_4:Eu^{2+} with other rare earth doping is highlighted, compared and reviewed.

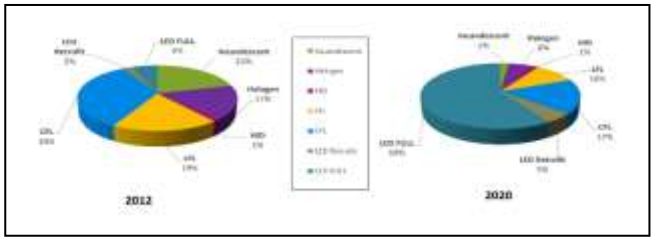

Fig.1: Comparison of Average percentage share of lighting technologies (Conventional to energy efficient lighting) between the two decades 2012 and 2020 [3].

2 Review of Literature

LED material to produce white light emission is a current trend in the field of lighting by the researchers. New materials are prepared and rigorously investigated to explore the new possibility to get the desired white light emission. Rare earth doped strontium orthosilicate (Sr_2SiO_4) phosphors have piqued interest due to their remarkable primary properties and potential uses in the creation of white light-emitting diodes, as discussed in the previous section.

Lee et al [10]. investigated Eu^{2+} doped Sr_2SiO_4 phosphors which were prepared through flux method. The study were conducted to compare the effect on photoluminescence of the samples with sintering temperatures (800^{o}C and 1300^{o}C) as well as an amount of flux used with variation of different dopant concentrations. Results suggested that the in the temperature of 800^{o}C the rare earth with the Sr ingredients were not reacted with each other. With the addition of more flux into the system makes the proper reaction in the material. NH_4Cl were used as flux which is considered as a prominent role in the reaction mechanism. XRD investigations suggested that upto 10% of flux concentration the peaks appeared but after 10% peaks are predominant. Eu^{2+} concentration with NH_4Cl as $Sr_{2-x}Eu_xSiO_4$ (x=0.01, 0.03, 0.05 and 0.07) showed a coexisted crystal phase upto 0.05 and after 0.07 peaks were identified as a single phase indications. Consequent emission band of 495 nm and 560 nm were observed in the PL spectra indicating presence of Eu. With increasing Eu^{2+} concentration, the energy transfer from Eu(I) of the high-energy emitting center to Eu(II) of the low-energy emitting center increased, leading to a longer wavelength emission, and also red-shift and phase change of β - to α'-Sr_2SiO_4 were observed.

The addition of NH_4Cl flux to the powder obtained by spray pyrolysis technique changed the hue from yellow green to yellow, according to a similar study (S. H. Lee et al., 2010).

Pratibha et al. synthesized Eu^{3+} (1 mol%) doped Sr_2SiO_4 nanophosphor by a low temperature combustion method using citric acid as a fuel. The nanophosphor achieves an orthorhombic structure without any secondary phase, according to PXRD. SEM micrographs of the nanophosphor shows highly porous with large void and agglomeration. According to the UV–Vis investigation, the maximum absorption at 200–270 nm can be generated due to the transition between the valence and conduction bands. The $Sr_2SiO_4:Eu^{3+}$ has the most red emission, as well as blue and green emission. The TL characteristics of - irradiated nanophosphor were studied, and a single peak at 1730C was discovered.it is seen The intensity of the light peak grows linearly with dose, indicating that the $Sr_2SiO_4:Eu^{3+}$ is suited for radiation dosimetry applications [11].

Chaudhari et al. For the formation of Sr_2SiO_4, reaction mechanics and kinetic analysis were examined. It is reported that there is a direct process between $SrSO_3$ and SiO_4 through TG/DTA and XRD analysis for the formation of Sr_2SiO_4. They have considered a three-dimensional solid-state reaction method by involving of $SrCO_3$ and SiO_4 it is also studied XRD showed that the structure of Sr_2SiO_4 phosphor is Orthorhombic in its crystal size is 22 nm. Thus, doping Eu^{2+} on -Sr_2SiO_4 phosphor emits white light at x=0.64 and x=0.33. Which can be used in white light emitting diode [12].

Verma et al. reported Eu^{2+} activated Sr_2SiO_4 nanophosphors. Prepared in low temperature solution combustion method using urea $[CO(NH_2)_2]$ as a fuel it is also studied its Photoluminescence (PL) spectroscopy, X-ray diffraction (XRD), UV-Spectroscopy properties. The photoluminescence properties of nanoscale Sr_2SiO_4: Eu^{2+} phosphors activated at 256 nm indicated a strong red emission.The Sr_2SiO_4: Eu^{2+} phosphors exhibited white emission ranging from 500 to 750 nm when it was excited by near-ultraviolet light, it is seen that rare earth doped Sr_2SiO_4 the majority of phosphors are employed in light conversion phosphors for near-UV chips [13].

Yanmin et al. has been reported the structural and optical properties of Sr2SiO4:Eu3+ and $Sr_2SiO_4:Eu^{2+}$ phosphors produced using a traditional solid-state reaction technique were examined .The phosphors were characterized by X-ray diffraction, Photoluminescence (PL) spectroscopy. The produced phosphors were composed of orthorhombic'-Sr_2SiO_4 and monoclinic'- Sr_2SiO_4 phases, according to the X-ray diffraction data. The phosphor excited under 256 nm, red light and the phosphor Sr_2SiO_4 exhibited white emissions (x=0.30, y=0.40, Tc=6500k) ranging from 420 to 625 nm under excitation by near-ultraviolet light. Sr2SiO4: Eu^{3+} was chosen as the phosphor for white LEDs pumped by near-UV chips because of its good luminous characteristics [14].

3 Results and Discussion

Various researches were conducted to observe the aspect of the rare earth doped Sr_2SiO_4 and found very interesting remarks on the system. Europium (Eu) as a rare earth dopant for Sr_2SiO_4 are studied and compared for the structural variations. The comparison of the preparation

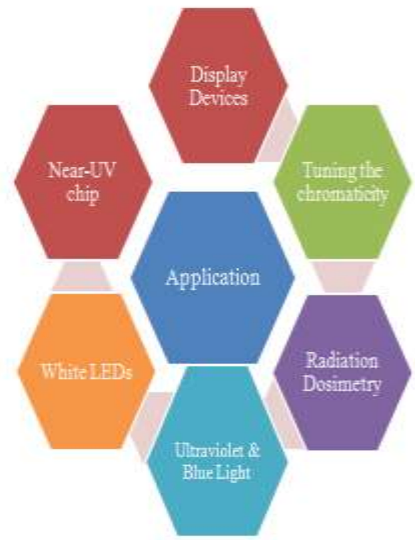

Fig. 2: Proposed Application areas proposed by various researchers [2][10][11] [12][15][16].

methods and corresponding crystal structure is illustrated in the Fig.3. Various methods like spray pyrolysis, combustion, standard solid state and sol gel methods are reviewed with various references [1] [13][14][15]. From Figure 3 it is clear that the sol gel method is the most suitable method to obtain a single phase structure of the material. It also to be noted that except sol gel method, all available methods are producing mixed phase structures with regular and irregular surface morphology. Variation in the structures is also observed as a function of preparation methods as shown in the Figure 4. Aspect of the flux addition by NH_4Cl is also studied and is observed that with moderate addition (between 2-5 %) in the Sr_2SiO_4 material, the structure were found polyhedron with regular morphology and with the excess of NH_4Cl (above 5%), the crystal structure deviated to the irregular morphology [1]. Effect of sintering temperature is also having a crucial role in the structural variation. Sintering temperature at higher temperatures showed a red shift in the Photoluminescence spectroscopy [10].

Fig. 3: Various method of synthesis adopted by researchers [1][13][14][15].

Table – 1. Comparative study of rare earth activated Sr_2SiO_4 phosphor synthesis method, characterization and luminescence

SNo	Study	Author	Synthesis Method	Application	Structure	Particle Size	PL Study
1	Photoluminescence properties of Sr_2SiO_4:Eu^{2+} Phosphors	Lee et al.	Solid State Reaction Method	Display Devices		Narrow size crystal particles	
2	Colour tunable Sr_2SiO_4:Eu^{2+}Phosphors through themodification of crystal structure	Cheng et al.	Solid-State Reaction Method	Blue-yellow, good candidates for tuning the chromaticity in application	α-Sr_2SiO_4(orthorhombic) to β-Sr_2SiO_4(Monoclinic).		The photoluminescence emission band is λ=320nm of Sr_2SiO_4:Eu^{2+}
3	Photo and thermo luminescence study of Sr_2SiO_4:Eu^{3+}	Prabha et al.	Combustion method	Sr_2SiO_4:Eu^{3+} is suitable for radiation dosimetry application	the standard pattern of orthorhombic α- Sr_2SiO_4 available in JCPDS No. 39-1256		The PL emission spectra are composed at peaks in all the three regions blue, green and red.
4	Characteristics of α',β-Sr_2SiO_4:Eu^{2+} phosphor powders prepared by spray pyrolysis.	Lee et al.	Spray Pyrolysis	Phosphor was useful for ultraviolet and blue light.	β -Sr_2SiO_4 phosphor powders with regular polyhedron structure	The mean size of the β-Sr_2SiO_4 phosphor powders with regular polyhedron structure was5.2 micrometer.	The wavelength of the emission spectra showing the maximum peak intensity is 543.2 to 561.8 nm
5	GaN-Based white light emitting diodes fabricated with a mixture of $Ba_3mgSi_2O_8$:Eu^{2+} and Sr_2SiO_4:Eu^{2+} Phosphors	Kim et al.	Solid-State Reaction Method.	The GaN-Based white-light emitting diode fabricated using a mixture of $Ba3mgSi_2O_8$:Eu^{2+} phosphors has a broad-band spectrum, higher color rendering index and higher color stability against forward bias currents than $Y_3Al_5O_{12}$:Ce^{3+} based white-LEDs.	The Ba_3MgSiO_8:Eu^{2+} and Sr_2SiO_4:Eu^{2+} phosphors are polycrystalline and reproducible.		The PL spectra of Ba3mgSi2O8:Eu2+ show one peak at 442 nm and two unresolved peaks at 505 nm.
6	Synthesis and Optical properties of Eu^{3+} doped Sr_2SiO_4 phosphor for Solid-State lighting applications.	B.et al.	Solid-State diffusion method.	Phosphors applied in white LEDs.	The crystal structure of Sr_2SiO_4 is orthorhombic.	The average crystallite size of the Sr_2SiO_4 phosphor is 22nm.	The PL emission spectra are 589-593 and 613-619 nm.
7	Preparation of Eu-Activated Sr_2SiO_4 phosphor by a Combustion method and its Orptical Properties.	Verma et al.	Combustion method	Good light-conversation phosphor for near-UV chip.	The crystal structureof α-Sr_2SiO_4 is orthorhombic and β -Sr_2SiO_4 is(monoclinic)		The photoluminescence properties of the nano-size Sr_2SiO_4:Eu^{3+} phosphors excited under 256nm, showed intense emission in red region. Sr_2SiO_4:Eu^{3+} phosphor exhibited white emissions ranging from 500 to 750 nm when it was excited by near-ultraviolet light.
8	Role of synthesis method and α, β-$Sr_{(2-x)}SiO_4$:xEu^{2+} phases on the photoluminescent properties of $Sr_{(2-x)}SiO_2N_2$:xEu^{2+}phosphors	Dhia A.et al.	Solid-State Reaction Method.	Phosphors applied in white LEDs	Strontium orthosilicate has two crystal structure is orthorhombic and monoclinic. with high and low temperature.	The crystal size of α, β- $Sr_{(2-x)}SiO_4$:xEu^{2+} of SSO and SSONprepared by four methods co- 47.34nm and 47.55,PSG-49.83 &50.11,SG-53.99 & 52.05, SS-56.89 &56.19 nm	The photoluminescence results of SSON show an emission spectrum band between 460 and 640 nm with green emission peaks at 530 nm.
9	Effect of Mg Substitution in Sr_2SiO_4:Eu^{2+} nanophosphors for blue and white emission at near UV excitation.	Dubey et al.	Solution Combustion Method.	Phosphors applied in white LEDs.	The crystal structure of Sr_2SiO_4:$Eu^{2+}$is belong to Orthorhombic phase of α-Sr_2SiO_4		The emission peak for Eu^{2+} doped Sr_2SiO_4 nanophosphor is observed at ~490 nm and ~553 nm corresponding to two Sr^{2+} sites Sr(I) and Sr(II) respectively for 395 nm excitation but the addition of Mg^{2+} dopant in Sr_2SiO_4 leads to suppressionof ~553 nm emission peak due to absence of energy levels of Sr(II) sites which results in a single broad emission at ~460 nm.
10	photoluminescent properties of Sr_2SiO_4:Eu^{3+} and Sr_2SiO_4:Eu^{2+}phosphors Prepared by solid-state reaction method.	Yanmin et al.	Solid-State Reaction method.	Good light-conversion phosphor candidate for near-UV chip.	The obtained phosphors were composed of orthorhombic α'-Sr_2SiO_4 and monoclinic β-Sr_2SiO_4 phase.		The PL emission spectra is for red region excited under 256 nm & for white region is 425 nm to 650 nm
11	Structure & site selective luminescence of sol-gel derived Eu: Sr_2SiO_4	Gupta et al.	Sol-gel method	Red phosphor	Single Phase Structure		The emission spectrum of the species getting selectively excited at 296 nm

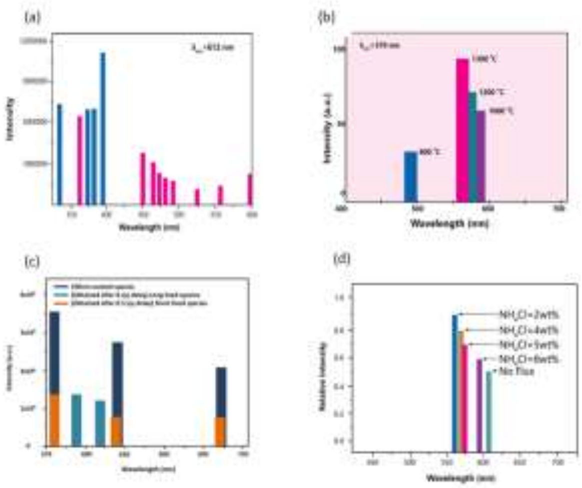

Fig. 4: Comparison of Various PL Emission Spectrum of Sr_2SiO_4:Eu^{2+} &Eu^{3+} nanophosphor [1][12][14][15].

Figure 4 shows the PL study of the samples prepared by different methods. In Figure 4.(a), the phosphor Sr_2SiO_4 is prepared by Combustion method, whose PL study shows that its PL emission spectra is λ_{emf} = 612 nm; which indicates a peak around 361 nm. Peaks in three blue, green, and red regions make up the PL emission spectra [14]. Figure 4(b) shows that the Sr_2SiO_4: Eu^{2+} (2.5 mol%) The solid state reaction method was used to make phosphor.in which Sr_2SiO_4:Eu^{2+} powders sintered at various temperatures excited PL spectrum at 560 nm with an increase in sintering temperature upto 370 nm that increased PL intensity with increase in firing temperature and also observed blue shift [12].
The sample Sr_2SiO_4:Eu Phosphor in Figure 4(c) is synthesized by the sol-gel method. Excitation and emission spectra of a strontium silicate doped Eu sample at λ_{em} = 575nm and λ_{ex} = 296nm. The emission spectrum of the two rare earth ions species has been detected at 296 nm shortly after the time resolved data measurement [15].
Figure 4(d), demonstrates that when α'- and β- Sr_2SiO_4:Eu^{2+} phosphor powder has been prepared by Spray Pyrolysis method with the addition of NH_4Cl containing 2 wt%, 4wt%, 5wt% and 6wt%. When the phosphor was prepared with 2wt% and 5wt% of NH_4Cl Flux, the phosphor powder had a regular polyhedron structure and the main crystal structure of β - Sr_2SiO_4. When phosphor powder was prepared from spray solutions with an additional 6% of NH_4Cl Flux, the phosphor had an irregular morphology with main crystal structure of α'- Sr_2SiO_4. The PL emission spectrum of the powder prepared by spray-pyrolysis with NH_4Cl is shown in Figure 4(d). When the NH_4Cl flux in phosphor powder is increased from 2 wt% to 6 wt%, the maximum wavelength intensity of its PL emission spectrum is found to be from 543.2 to 561.8 nm [1].

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