



## TAILORING PHOTOLUMINESCENT PROPERTIES THROUGH HOST MODIFICATION IN $\text{Sm}^{3+}$ DOPED $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$

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### ABSTRACT:

Compound  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  doped with various concentrations as 0.1, 0.3, 0.5, 0.7, 1 mol% of rare earth dopant  $\text{Sm}^{3+}$ . These phosphors were synthesized by using solution combustion method. Photoluminescence (PL) emission spectra shows highest intensity emission peak at 566nm and 604nm for 0.3 mol% Sm doped phosphor at 405 nm excitation wavelength. 0.3 mol% Sm doped phosphor further doped with calcium nitrate shows highest intensity emission peak at 566nm and 604nm for the phosphor  $\text{Sr}_{0.5}\text{Ca}_{2.5}\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3mol% where the emission intensity increased 1.5 times. The studies showed that these phosphors are promising candidate for the light application. Commission International de l'Eclairage (CIE) chromaticity colour coordinates of these phosphors were calculated. CIE- Chromaticity colour coordinates of this phosphor shows orange-red emission.

**Keywords:-** Phosphors, Photoluminescence, CIE, Solution Combustion Method

### INTRODUCTION:

Inorganic materials are converted to phosphors by doping with activators like rare earth metals. Samarium ion is vigorously studied activator in the field of luminescence and extremely useful as a dopant in phosphors for LED and display devices, sensors [D.K. Yim et,al.(2011),T. Murata et,al.(2005),S. Nakamura (1991)]. Samarium ion exists in  $\text{Sm}^{3+}$  form.  $\text{Sm}^{3+}$  ion usually shows orange-red emission arising from  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_J$  ( $J=5/2, 7/2, 9/2, 11/2$ ) transitions within the  $^4\text{F}_5$  electronic configuration [R. Yu, Y. Guo et, al.(2014),R. Shi et,al.(2016)].  $\text{Sm}^{3+}$  ions can be utilized to compensate deficiency of red component in white light-emitting diodes (LEDs) field emission displays (FEDs) and color rendering purposes as well [S. Selvi, K. Marimuthu, G. Muralidharan (2015)]. Furthermore, rare earth ions can be used as activators because they can be stabilized with the host material in their trivalent oxidation state. Trivalent- lanthanide ions doped phosphor materials have gained massive attention owing to their wide application in the field of fluorescent

lighting, white light emitting diodes (W-LED), display devices, bioimaging, photography, radiation dosimetry etc.[ R. Yu, H.M.et al. (2014), S.R.R. Anishia et al. (2011), J.Wang et al. (2016), Z. Zhang et al.(2016)]. Red, Orange-red emitting phosphor can be used along with green and blue emitting phosphors to enhance the colour rendering index of the W-LED [S.K. Rautet et,al.(2014), R. Velchuri et,al.(2009)].  $\text{Sm}^{3+}$  dopant ion is one of the lanthanide element producing red luminescence.

$\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  is the well known inorganic material used as a host to synthesize phosphors [X.M. Zhang et al.(2009), X.M. Zhang et al. (2010), D. Dutczak et al. (2013)]. In this work, different concentrations of  $\text{Sm}^{3+}$  doped  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  phosphors with well chemical and physical stable performance were synthesized. Normally, phosphors are prepared by the outmoded solid state reaction method. This method has some drawbacks such as it naturally needs high temperature, time-consuming heating process and consequent crushing for long time. The crushing

procedure recompenses the phosphor planes, subsequently results in the loss of emission intensity. Consequently the combustion synthesis has fascinated considerable attention since it is beneficial in attaining the unique chemical configurations with exclusive properties, outstanding purity and moderately low reaction temperature, ensuring more homogeneous products, and it is also probably prepares phosphors in small size [Y. Yonesaki, C. Matsuda (2011)]. To the best of our knowledge, the photoluminescence study of these samples revealed that the phosphors can be excited by blue light and emit orange-red light. In this work,  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  doped with samarium were synthesized by solution combustion synthesis and their luminescent behavior were investigated in detail. Therefore, in the present work it is tried to enhance intensity of emission wavelength by substituting the  $\text{Sm}^{3+}$  doped host material  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  by  $\text{Ca}^{2+}$  ions. Photoluminescence properties of these phosphors are also studied.

#### **EXPERIMENTAL:**

##### **SOLUTION COMBUSTION SYNTHESIS (Self-propagating high- temperature synthesis)**

With the development of technologies on materials, several kinds of chemical synthesis techniques such as co- precipitation, sol- gel, reverse micro emulsion and solution combustion methods have been applied to prepare phosphors. Among these, combustion synthesis was an interesting and ideal technique to prepare the fine, chemically

homogenous, pure and single phase phosphor powders in these synthesise condition. Smaller particle size was produce from reaction which liberates the greatest amount of gas. The highest luminescence emission intensity was correlated with the highest measured flame temperature, indication that thermal effect as important for producing optimally performing phosphor powders. This synthesis technique used the heat energy released by the redox exothermic reaction between the metal nitrates and urea or other fuels at a relatively low-igniting temperature. Practical way of lowering the operating temperature is to use the heat

generated in exothermic chemical reaction itself for the synthesis. The understandable advantage of this synthesis is that the heat is supplied within the reaction, which reduces the use of insulators, crucible and refractory muffles, consequently synthesis becomes cost effective. In generals this method may be known as Self- propagating high-temperature synthesis (SHS). The process was not only safe but also time and energy saving.

The exothermic reaction started spreads across the length of the bar and elevated temperature about 600 degrees Celsius can be achieved in such reaction to produce nitride and carbide materials, for that the oxidizing and reducing valencies of the oxidizers and fuel must be balanced. Nitrate to fuel ratio can used for be adjusted to vary the temperature generated in the reaction [D. Curie (1963)].



**Fig.1.1 Schematic representation of solution combustion method**

### Synthesis of $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ : $\text{Sm}^{3+}$ co-doped with calcium:

The inorganic material  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  was synthesized by using solution combustion method. The A. R. Grade chemicals, Aluminum nitrate [ $\text{Al}(\text{NO}_3)_3$ ], Strontium nitrate [ $\text{Sr}(\text{NO}_3)_2$ ], Ammonium chloride ( $\text{NH}_4\text{Cl}$ ), Samarium (III) oxide ( $\text{Sm}_2\text{O}_3$ ), Urea were used without further purification. All the chemicals weight as per stoichiometric ratio and solution of these contents mix in China dish. Urea is used as a fuel. The content in the China dish then kept in preheated vertical muffle furnace at 600 Degree Celsius. where auto combustion took place accompanied with evolution of brown fumes. After completion of combustion process the white solid material, we crushed it in fine powder using mortar and pestle. In the same manner by adding different concentration  $\text{Sm}^{3+}$  in the form of  $\text{Sm}_2\text{O}_3$  dissolve in concentration nitrate acid. The

highest emission intensity is shown by  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm in 0.3 mol% therefore by keeping this dopant concentration constant we further added co- dopant like calcium by using A. R Grade Chemical Aluminium nitrate [ $\text{Al}(\text{NO}_3)_3$ ], Strontium nitrate [ $\text{Sr}(\text{NO}_3)_2$ ], Calcium nitrate [ $\text{Ca}(\text{NO}_3)_2$ ], Ammonium chloride ( $\text{NH}_4\text{Cl}$ ), in different concentration. These co- doped phosphors were also synthesized by using the same solution combustion method.

#### Molecular weight of chemicals:

$\text{Al}(\text{NO}_3)_3 = 212.996 \text{ g/mol}$

$\text{Sr}(\text{NO}_3)_2 = 211.63 \text{ g/mol}$

$\text{NH}_4\text{Cl} = 53.491 \text{ g/mol}$

Urea = 60.06 g/mol

$\text{Ca}(\text{NO}_3)_2 = 164.1 \text{ g/mol}$

**1.1 Table: Stichiometric weight of chemicals in gram to synthesize  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm (0.1, 0.3, 0.5, 0.7, 1 mol%) Phosphors.**

Sm mol%	Wt. Of Al ( $\text{NO}_3$ ) <sub>2</sub>	Sr ( $\text{NO}_3$ ) <sub>2</sub>	$\text{NH}_4\text{Cl}$	Urea	$\text{Sm}_2\text{O}_3$
Pure	3	2.5375	0.4277	6	-
0.1	3	2.5367	0.4277	6	0.0013
0.3	3	2.5350	0.4277	6	0.0041
0.5	3	2.5333	0.4277	6	0.0069
0.7	3	2.5316	0.4277	6	0.0097
1	3	2.5290	0.4277	6	0.0139

**1.2 Table: Stichiometric weight of chemicals in gram to synthesize  $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3 mol% (where, X= 0.5, 1, 1.5, 2, 2.5, 3)**

Sr	Wt. of Al ( $\text{NO}_3$ ) <sub>2</sub>	Wt. of Sr ( $\text{NO}_3$ ) <sub>2</sub>	Wt. of Ca ( $\text{NO}_3$ ) <sub>2</sub>	Urea	$\text{NH}_4\text{Cl}$	$\text{Sm}_2\text{O}_3$
2.5	3	2.1120	0.3261	6	0.4277	0.0348
2	3	1.6891	0.6542	6	0.4277	0.0278
1.5	3	1.2662	0.9823	6	0.4277	0.0209
1	3	0.8433	1.3104	6	0.4277	0.0139
0.5	3	0.4203	1.6385	6	0.4277	0.0069
0	3	0	1.9666	6	0.4277	0

**3. RESULT AND DISCUSSION-**

**3.1 Photoluminescence Study of Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub> doped with different concentrations of Sm<sup>3+</sup>-**

**Excitation and emission spectra of Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub>: Sm (0.1, 0.3, 0.5, 0.7 & 1 mol%) -**

Sm<sup>3+</sup> ion usually shows orange-red emission and Sm<sup>3+</sup> ions can be utilized to compensate deficiency of red component in white light-emitting diodes (LEDs). Fig. 4.1 shows the excitation spectra of Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub>: Sm monitored at 598 nm emission

wavelength which is composed of several sharp peaks in the range of 340-420 nm wavelength. The peaks at 345nm, 362nm, 376nm, 405 and 418nm are allocated to the electronic transitions resulting from <sup>6</sup>H<sub>5/2</sub>→<sup>4</sup>K<sub>17/2</sub>, <sup>6</sup>H<sub>5/2</sub>→<sup>4</sup>H<sub>7/2</sub>, <sup>6</sup>H<sub>5/2</sub>→<sup>4</sup>P<sub>7/2</sub>, <sup>6</sup>H<sub>5/2</sub>→<sup>4</sup>K<sub>11/2</sub> and <sup>6</sup>H→<sup>6</sup>P<sub>5/2</sub>. The strongest peak is centered at 405 nm. Therefore, the emission spectra were monitored at 405 nm excitation wavelength.

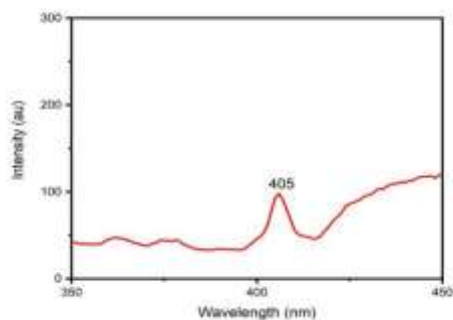


Fig. 2.1

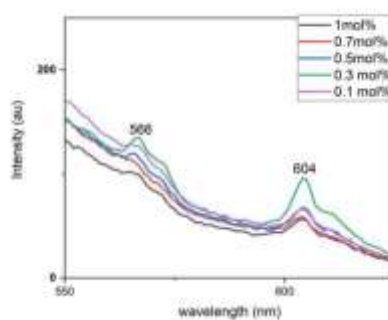


Fig. 2.2

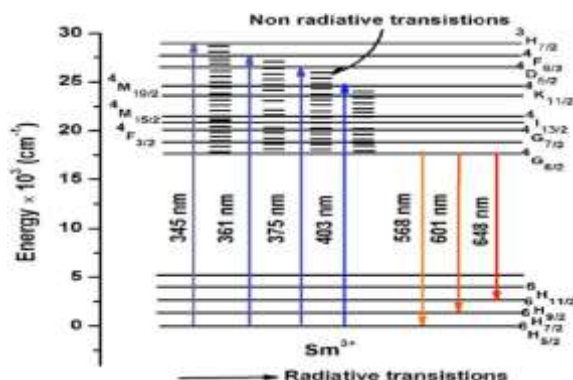
**Fig. 2.1 Excitation Spectra of Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub>:Sm<sup>3+</sup> phosphor monitored at 566 nm emission wavelength**

**Fig. 2.2 Emission Spectra of Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub>: Sm<sup>3+</sup> phosphor monitored at 405 nm excitation wavelength**

The emission spectra show two emission peaks at 566nm and 604nm wavelength corresponding to the <sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>5/2</sub> and <sup>4</sup>G<sub>5/2</sub>→<sup>6</sup>H<sub>7/2</sub> transitions of Sm<sup>3+</sup> ions respectively as shown in Fig. 2.2 at 405nm excitation wavelength.

Fig 2.2 revealed the maximum photoluminescence intensity is for 0.3 mol% of Sm<sup>3+</sup> doped phosphor. Thus, the best doping concentration for Sm<sup>3+</sup> ions in Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub> host lattice is 0.3mol%. The fig 2.3 indicated the representative energy level transitions of Sm<sup>3+</sup> ions.

In this study, the concentration of Sm<sup>3+</sup> ions is varied from 0.1 to 1 mol% in Sr<sub>3</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub> host



**Fig. 2.3 Representative energy level transitions of Sm<sup>3+</sup>ions**

**3.2 Photoluminescence Study of Sr<sub>3-x</sub>Ca<sub>x</sub>Al<sub>2</sub>O<sub>5</sub>Cl<sub>2</sub>: Sm 0.3mol% sPhosphors where x= 0.5,1,1.5,2,2.5,3**

### Excitation and emission spectra of $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3mol% phosphors

As the maximum emission intensity in different concentrations of Sm doped  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  phosphors were observed for  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ :  $\text{Sm}^{3+}$  0.3mol%, by

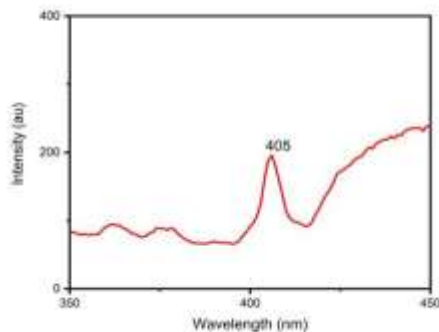


Fig. 2.4

**Fig. 2.4 Excitation Spectra of  $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3mol% phosphor at 566nm emission wavelength**

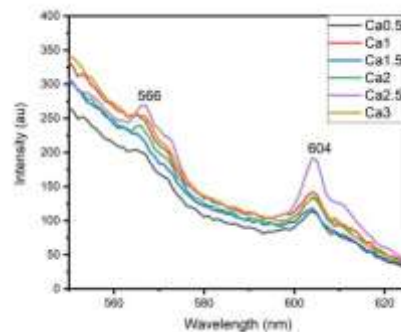


Fig. 2.5

**Fig. 2.5 Emission Spectra of  $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3mol% phosphors at 405 nm excitation wavelength**

The emission spectra of  $\text{Sr}_{3-x}\text{Ca}_x\text{Al}_2\text{O}_5\text{Cl}_2$ : $\text{Sm}^{3+}$  0.3 mol%(where,  $x = 0.5, 1, 1.5, 2, 2.5$  and 3) phosphors when observed at 405 nm excitation wavelength, two emission peaks are observed at 566nm and 604nm. These two peaks corresponds to  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ ,  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$  transition of  $\text{Sm}^{3+}$  ions as shown in fig. 2.5. The maximum emission intensity is observed for  $\text{Ca}_{2.5}$  concentration after that as we increased the concentration of  $\text{Ca}^{2+}$  ions, the emission intensity decreases means here we found concentration quenching after  $\text{Ca}_{2.5}$  concentration.

#### 4. CONCLUSION:

$\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  phosphors doped with Samarium ion were prepared by solution combustion method using urea as a fuel. The prepared phosphors have been characterized for photoluminescence study. They are excited by near UV or blue light. By regulating the doping concentration of dopants and sensitizer, phosphors with high luminescent intensities were acquired.  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$  host material doped with different concentrations of

keeping this concentration of Sm ions constant we further co-doped this phosphor by introducing the different concentration of  $\text{Ca}^{2+}$  ions. Fig 2.4 shows the single excitation peak at 405nm when monitored at 604 nm emission wavelength.

$\text{Sm}^{3+}$  ions, out of which  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3 mol% showed highest emission intensity hence this phosphor was

co-doped with different concentrations of  $\text{Ca}^{2+}$  ions. In present study,  $\text{Sr}_3\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3 mol% host when co-doped with  $\text{Ca}^{2+}$  ions, showed emission peak in orange-red region with adequate emission intensity. Highest emission peak was observed for  $\text{Ca}_{2.5}$  concentration phosphor. So all prepared phosphors can be served as orange-red phosphor for LED. Out of all the prepared phosphors,  $\text{Sr}_{0.5}\text{Ca}_{2.5}\text{Al}_2\text{O}_5\text{Cl}_2$ : Sm 0.3mol% phosphor was found to be the best phosphor to be used as orange-red component for white LED phosphor.

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