



STUDY OF TUNABLE MULTICOLOR EMISSION BY RARE EARTH DOPED Sr_2CeO_4 FOR PHOSPHOR-CONVERTED WHITE LIGHT

¹R. S. Ukare, ²A. S. Patle

^{1,2}Department of Physics C. J. Patel College Tirora-441911, Dist. Gondia, India.

Corresponding author email: rajshekharukare26@gmail.com.

ABSTRACT

Sr_2CeO_4 is well-known host lattice for efficient multicolor emitting luminescence material with different activators, keeping this view we doped rare earth ions Dy^{3+} , Gd^{3+} , Eu^{3+} in Sr_2CeO_4 material and prepared all these samples by the easy combustion method. Prepared sample shows flexible intensity of multicolor optics region for phosphor-converted white light application. Sr_2CeO_4 shows the interesting optical properties in powder form due to the presence of two phases and doubly ionized oxygen vacancies. The crystal structure was ascertained through X-ray diffraction analysis. Prepared $\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+}$, $\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+}$ and $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$ samples are examined by emission and excitation spectra for solid state lighting applications (SSL) under commercial available NUV exciting source. SEM images and XRD pattern of prepared sample are taken to study surface morphology which shows particles of irregular size and average size in nanometer range. Prepared luminescence materials can also be used to improvement of efficiency of solar cells, field emission display panels and light emitting diodes under commercially available NUV (350-420 nm) and blue (430-470 nm) light.

Keywords: Dy^{3+} , Gd^{3+} , Eu^{3+} , Sr_2CeO_4 , Phosphor-converted white light.

1. INTRODUCTION

Sr_2CeO_4 gives excellent high luminescence efficiency, flexible emission colors with different activators thus used to improvement of efficiency of solar cells, field emission display panels and light emitting diodes, when excited by the UV light [1,2].

Near-UV LEDs when coupled with white light emitting phosphor materials which emit red, green and blue light in a specific ratio are very economical and interesting for intense WLEDs [3,4]. Today light emitting materials which efficiently absorbed NUV (350-420 nm) and blue (430-470 nm) light are easily available with efficiency of 2 Watt/m² to 8 Watt/m² [5,6]. Inorganic Sr_2CeO_4 is one of the smart blue light emitting luminescence materials and also a very good ceramic materials [7], used as a scintillations, which convert high energy ionizing photons, and used in medicine, oil logging environmental monitoring and particle physics [8]. Sr_2CeO_4 nanophosphors with non rare earth elements (Ca, Mg, Ba, Zn) doping also produce tunable photoluminescence [9, 10]. Due to the presence of SrCeO_3 and SrCO_3 phases, which have an impact on the deconvoluted bands in the emission spectra, Sr_2CeO_4 exhibits fascinating optical features in powder form. The band gap of Sr_2CeO_4 material is approximately 3.4 eV, thus it's a good candidate for use as a scintillation or in the improvement of efficiency of solar cells, when excited in the UV and visible region. The (PL) emission peaks of both Eu^{3+} and Eu^{2+} activated Sr_2CeO_4 phosphors at 467 (blue), 537 (green) and 616 nm (red) generate white light under 260, 280 and 350 nm [9,10] but intensity of red light is not sufficient with deficiency yellow region light. Santosh K. Gupta et al., prepared Bluish white emitting Sr_2CeO_4 and red emitting $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$ nanoparticles by polymeric precursor route [11,12], which consist of infinite edge-shearing CeO_6 octahedral chains separated by Sr atoms, it is also found that emission radiation of $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$ nanoparticles

depends on temperatures thus it can be used for nanothermometry technology [13]. The broad blue emission band of Sr_2CeO_4 makes it suitable for the doping of rare earth ions so that it emits tunable different radiation in the visible region, that can be used to obtain day sun light emission for solid state lighting. Rare earth ions doped Sr_2CeO_4 nano phosphor shows good luminescence intensity and good crystalline structure at high calcination temperature [14-17].

2. EXPERIMENT

We synthesized rare earth doped Sr_2CeO_4 phosphor by the combustion method for that we first dissolved $\text{Sr}(\text{NO}_3)_2$ (99.99%) and $\text{Ce}(\text{NO}_3)_3$ (99.99%) in dilute HNO_3 (AR) in separate beakers. Then Gd_2O_3 (99.99%), Dy_2O_3 (99.99%), Eu_2O_3 (99.99%) are separately dissolved in dilute HNO_3 solutions to form nitrate solutions in proper stoichiometric ratio. Then to form $\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+}$ the nitrate solutions of $\text{Sr}(\text{NO}_3)_2$, $\text{Ce}(\text{NO}_3)_3$ and Dy_2O_3 are poured in a beaker and stirred well to form a uniform mixture. Now urea is used as a reducing agent keeping urea to nitrate molar ratio as 2:1. Then the mixture was set aside in a crucible and heated at 500 to 550°C in a furnace for 8-15 minutes then it translated to white powder, which after grinding a consistent powder was achieved. Lastly, powder was kept in furnace at 800°C for

2 hours to eradicate other impurities. Likewise, other samples are created and used for characterization.

3. RESULTS & DISCUSSION:

3.1 POWDER X-RAY DIFFRACTION

To obtain phase purity and crystallite structure we take X-ray diffraction of Sr_2CeO_4 phosphor which is shown in figure 1. The XRD peak positions are well matched with JCPDS card No.22-1422 [18], which belongs to the triclinic crystal structure. Diffraction patterns shown in figure 1 were obtained using $\text{Cu K}\alpha$ wavelength ($\lambda = 1.5404 \text{ \AA}$), at 40 kV and 30 mA applied to x-ray tube. and glancing angle 2θ varies from 10° to 75° keeping scan step time = 4.8450 second. The average size of the powder particles was calculated using the calculated from X-ray peak broadening of the diffraction using Scherrer's formula.

$$d = \frac{0.9\lambda}{\beta \cos \theta} \dots \dots \dots (1)$$

Where, β denotes the full width at half maximum (FWHM) of XRD peaks, λ is wavelength of X-ray and θ denotes angle of diffraction. The average particle size of rare earth doped $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$ is found to be 174 nm.

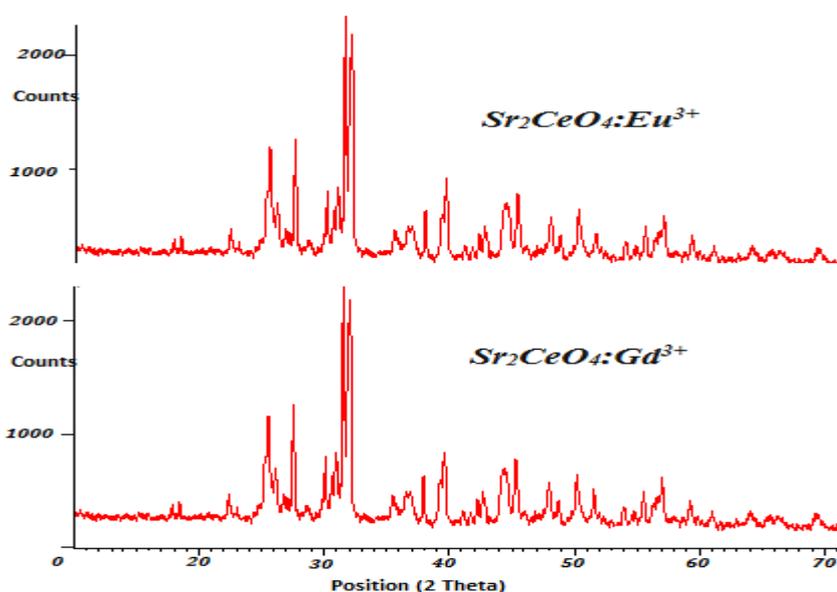


Fig. 1: powder X-ray diffraction pattern of $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$ and $\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+}$.

3.2 SEM RESULT

SEM images of rare earth doped Sr_2CeO_4 phosphor prepared by the combustion method are

shown in figure 2. SEM images of figure 2(a) $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+} = 1 \text{ mol\%}$ before sintering shows more agglomeration than figure 2(b), these

agglomerated particles have irregular surfaces and non uniform morphology. Figure 2(c) $\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+} = 1 \text{ mol}\%$ shows the irregular

particles and average size of 705 nm. Figure 2(d) shows the surface morphology of $\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+} = 1 \text{ mol}\%$.

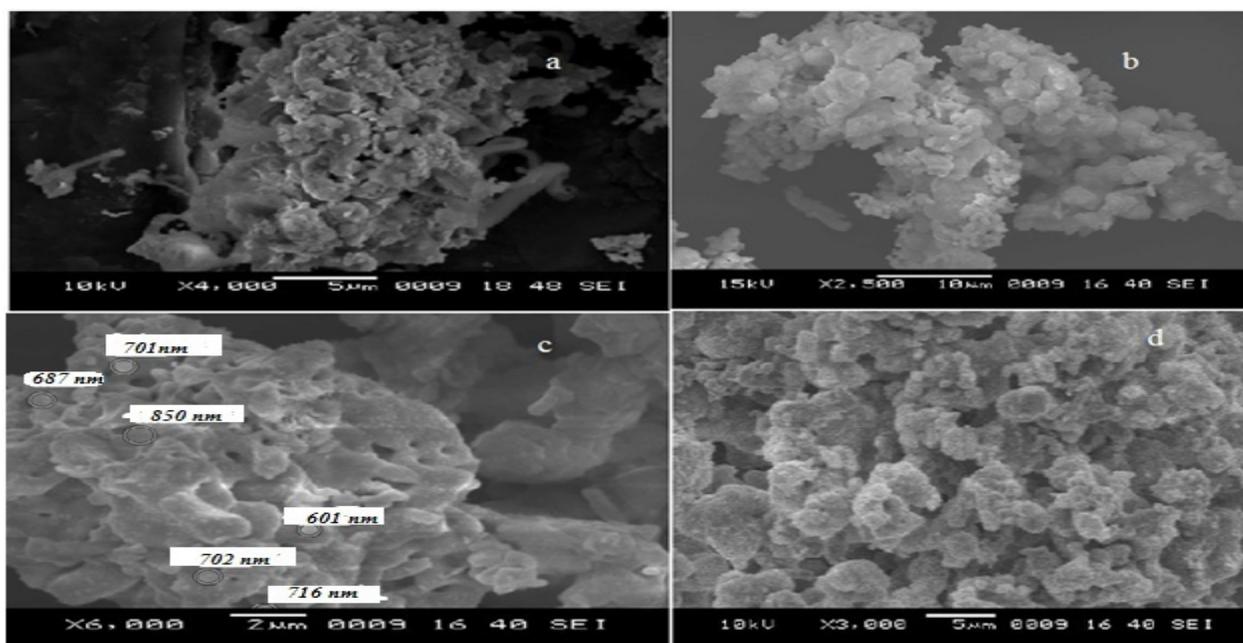


Fig. 2 SEM images of (a) $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+} = 1 \text{ mol}\%$ before sintering (b) $\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+} = 1 \text{ mol}\%$ after sintering at 800°C (c) $\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+} = 1 \text{ mol}\%$ and (d) $\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+} = 1 \text{ mol}\%$.

3.3 PHOTOLUMINESCENCE RESULTS

The excitation spectra of prepared rare earths (Eu^{3+} , Dy^{3+} , Gd^{3+}) ions doped Sr_2CeO_4 shows extending from 220 nm to 380 nm in the blue-green region with the maximum (FWHM~86) with peak at about 280 exciting at 570 nm visible radiations (Fig.3). The emission spectra recorded on exciting at 254, 260, 280 as well as 350 nm show a broad band with the maximum (FWHM~146) with extended emission into longer wavelengths.

The leading peak 280 nm and another peak at around 340 nm may be allocate to two availability of two sides of Ce^{4+} ions in Sr_2CeO_4 . There are two different $\text{Ce}^{4+}-\text{O}^{2-}$ bond lengths in the lattice and hence two unlike charge transfer transitions take place. The emission spectra of rare earth doped Sr_2CeO_4 beneath viable available NUV light source is shown in figure 4, where we see that Dy^{3+}

doped Sr_2CeO_4 gives high strength peak at 573 nm and a wide peak at about 481 nm and Gd^{3+} doped Sr_2CeO_4 gives high intensity peak at 527 nm and a wide peak at about 480 nm whereas D. L. Monika, et al; prepared Gd^{3+} doped Sr_2CeO_4 by Solid State Diffusion method which gives a broad band with peak at 470 nm[17]. Eu^{3+} doped Sr_2CeO_4 gives as many peaks at 468, 492, 512, 538, 557, 586, 596, 615 nm. From earlier research paper study we see that $\text{Sr}_2\text{CeO}_4:\text{Sm}^{3+}$ shows emission peaks at 387, 474, 556, 611, 653 nm [13, 15]. The broad band emission originates from Sr_2CeO_4 host itself and Eu^{3+} , Sm^{3+} co doped Sr_2CeO_4 shows peaks at 387, 467, 474, 491, 511, 538, 556, 611, 586, 616, 653 nm. Thus emission lines in the blue, green, and red region coexist. Thus Sr_2CeO_4 can be a good phosphor material for lamps and display devices [19-25].

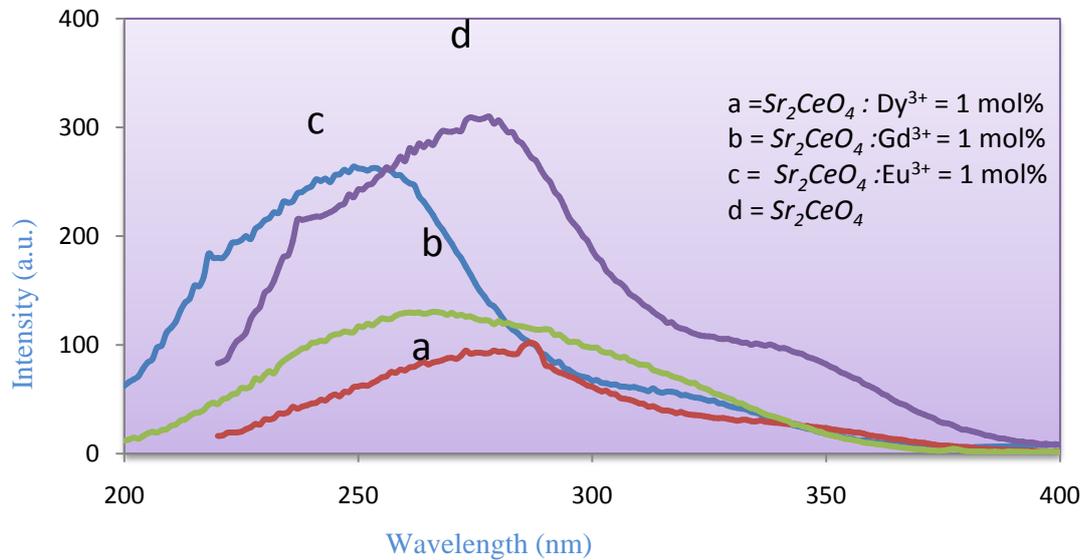


Fig.3: Excitation spectra of Sr_2CeO_4 at different rare earth ions.

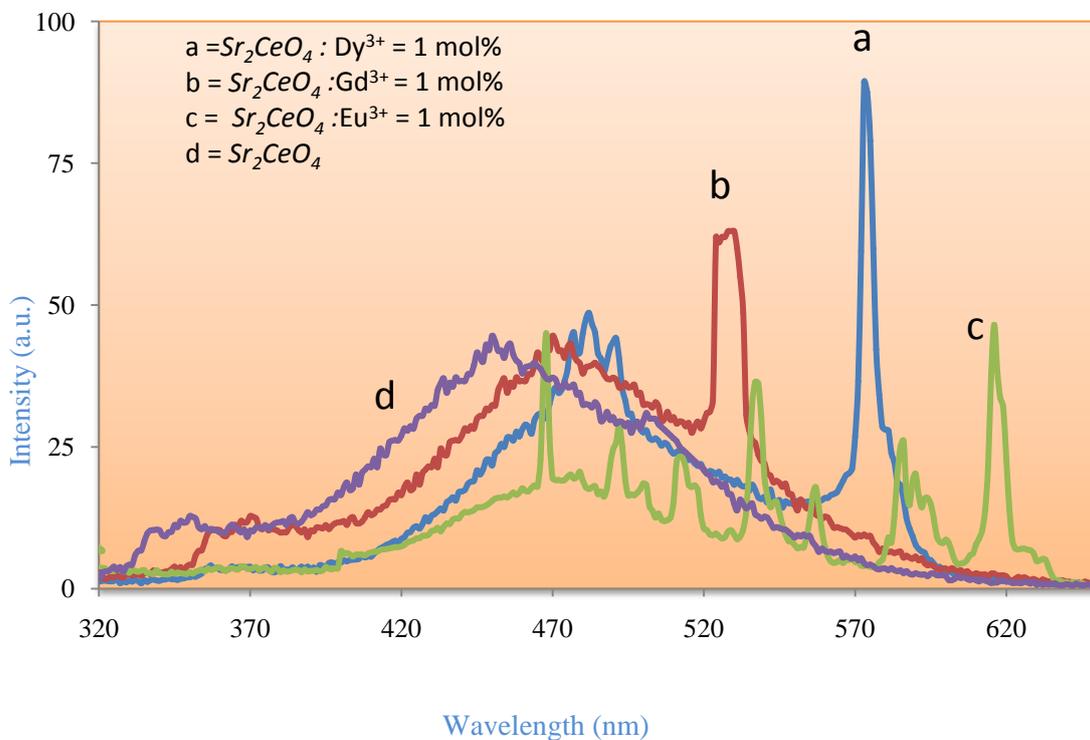


Fig.4: Emission spectra of Sr_2CeO_4 at different rare earth ions doping, under 340 nm.

Prepared all three sample shows same position peaks at blue region at around 470 nm, where as Dy^{3+} doped Sr_2CeO_4 gives elevated intensity extra peak at 573 nm at orange-yellow region, Gd^{3+} doped Sr_2CeO_4 gives additional high intensity peak at 527 nm at green region and Eu^{3+} doped Sr_2CeO_4 gives one high intensity peak at 615 nm. So using these three samples we can form a day light emitting LEDs under commercial available NUV stimulating source.

By combining primary colors, red, blue and green, we can form secondary colors yellow, cyan, and magenta [26]. Thus by blending above three samples which emit various flexible colors, we can produce better white light using phosphor-converted conventional RGB method. We also can control the exact requirement of white light which one can use according to the need also.

The luminescence peaks of different rare earth doped Sr_2CeO_4 are summarized in table(1), which shows that Sr_2CeO_4 is one of the smart material which shows different crystalline size from nanometer to micrometer range and tunes to different emission depends on types of impurities and method of

preparation. Since, rare earth doped Sr_2CeO_4 doped sample emit light in blue to yellow-orange red spectral, which is sweet spot for photosynthesis, thus such white light emitting material may use for horticulture applications[10,25].

Sample	Method of preparation	Excitation wavelength (nm)	Emission peaks (nm)	Average crystalline size	Reference
$\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$	Combustion	340	462, 492, 513, 538, 557, 586, 613	720 nm	Fig.(3)
$\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+}$		340	482, 574		
$\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+}$		340	471, 530		
$\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}, \text{Tb}^{3+}$	Solid state method	254	467, 491, 511, 538, 557, 586, 616	32 nm	[19]
$\text{Sr}_2\text{CeO}_4:\text{Sm}^{3+}$	Solid state method	320	387,474,556, 611, 653	68 nm	[20,21]
$\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}, \text{Sm}^{3+}$	Solid state method	300	467, 492, 511, 538, 556, 567, 585, 616, 650	--	[22]
$\text{Sr}_2\text{CeO}_4:\text{Er}^{3+}$	Solid state method	226	527, 536, 552, 558	35 nm	[21]
$\text{Sr}_2\text{CeO}_4:\text{Gd}^{3+}$	Solid state Diffusion	254	A broad band with peak at 470 nm	38 nm	[23]
$\text{Sr}_2\text{CeO}_4:\text{Ho}^{3+}$	Solution combustion	450	Broad band from 524 to 574 and low intensity broad band from 630 to 670	nanocrystalline particles	[25]
$\text{Sr}_2\text{CeO}_4:\text{Eu}^{3+}$	Precipitation	360	468, 492, 512, 538, 557, 586, 596, 616	6 to 9 μm	[11]

Table(1): Effect of rare earth on emission peaks in Sr_2CeO_4 phosphors.

4. CONCLUSION

PL studies of Sr_2CeO_4 phosphors with rare earths Gd^{3+} , Dy^{3+} , Eu^{3+} doped separately prepared by modified easy combustion method are presented in this article. XRD pattern of Sr_2CeO_4 phosphor reveals the triclinic single phase crystal structure of Sr_2CeO_4 . SEM images of Sr_2CeO_4 shows agglomerated particles with irregular surfaces and non uniform morphology and average size of 705 nm. Sr_2CeO_4 with different rare earths doping shows same position peaks at blue region at around 470 nm, whereas, $\text{Sr}_2\text{CeO}_4:\text{Dy}^{3+}$ gives

high intensity extra peak at 573 nm at orange-yellow region, Gd^{3+} doped Sr_2CeO_4 gives extra high intensity peak at 527 nm at green region and Eu^{3+} doped Sr_2CeO_4 gives one high intensity peak at 615 nm. So by blending these three samples which emit blue-green-orange-red color, one can be use as a good efficient phosphor-converted white light emitting material under commercial available NUV exciting source.

ACKNOWLEDGMENTS

The authors are very thankful to Prof. S. J. Dhoble to provide the lab facility. They are also

thankful to Dr. A. C. Sharma, the Principal C. J. Patel college Tirora for continuous encouragement.

5. REFERENCES

- [1]. E. Talik, L. Lipińska, D. Skrzypek, A. Skuta, P. Zajdel, A. Guzik, H. Duda, *Material Research Bulletin*, 47 (2012) 3107-3113.
- [2]. C H Atchyutha Rao, N Bujji Babu and KVR Murthy, *International Journal of Physics and Applications* 2023; 5(1): 13-18 DOI: <https://doi.org/10.33545/26647575.2023.v5.i1a.57>.
- [3]. K.R. Ashwini , H. B. Premkumar, G.P. Darshan, R.B. Basavaraj , H. Nagabhushana, B. Daruka Prasad,(2020), *Journal of Science: Advanced Materials and Devices*, 5, 111-118.
- [4]. Siti Sarina Binti Nasir, Kouta Yakura, Noriyuki Horiuchi, Masaya Tsuta, Ariyuki Kato, (2019), *J. Phys. Chem. Solid*. 133, 135-141.
- [5]. S. Ye, F. Xiao, Y. X. Pan, Y. Y. Ma, Q. Y. Zhang. (2010), *Mat. Sci. and Eng. R* 71, 1-34.
- [6]. San Jose, (2013), Osram to acquire California-based LED supplier LED Engin Inc. Purchase to boost Osram's specialty lighting business, CA 95134 USA, LED Engine, 651 River Oaks Parkway, sales@ledengine.com(2013).
- [7]. <https://www.sciencedirect.com/science/article/pii/S0272884222018880>.
- [8]. Takumi Kato , Naoki Kawano , Go Okada , Noriaki Kawaguchi , Masanori Koshimizu and Takayuki Yanagida, Version of Record: <https://www.sciencedirect.com/science/article/pii/S0925346718302167>
- [9]. Tomasz Grzyb, Agata Szczeszak, Justyna Rozowska, Janina Legendziewicz, and Stefan Lis,(2012), *Nanophosphors. Phys. Chem.* 116, 5, 3219–3226.
- [10]. Leonardo A. Rocha , Marco A. Schiavon , Clebio S. Nascimento Jr. , Luciana Guimarães , Márcio S. Góes, Ana M. Pires, et al; (2014), *J. of Alloy and Comp.* 608 ,73-78.
- [11]. K SURESH, N V POORNACHANDRA RAO and K V R MURTHY, (2014), *Bull. Mater. Sci.*, 37, pp. 1191–1195.
- [12]. Christian Maak, Philipp Strobel, Volker Weiler, Peter J. Schmidt and Wolfgang Schnick, (2018), *Chem. Mater.* 2018, 30, 15, 5500–5506.
- [13]. Chunxiang ZHANG, Chunxiang ZHANG, Jianshe SHI, Xujie YANG, Lude LU, (2010), 28(4):513-518.
- [14]. Dharmendra Yadav, Upendra Kumar, Shail Upadhyay. (2019), *Journal of Advanced Ceramics*, 8(3): 377-388.
- [15]. Jefferson L. Ferrari, Ana M. Pires, Osvaldo A. Serra, Marian R. Davolos, (2011), *J. of Lumin.* 131, 25–29.
- [16]. Paleena Thulimilli, (2021), *ES Mater. Manuf.*, 2021, 14, 73-78.
- [17]. R. S. Ukare, Vikas Dubey , G. D. Zade & S. J. Dhoble ,(2016), *J. Fluoresc.* 26, 791-806.
- [18]. D Xing, ML Gong, X Qiu, D Yang, KW Cheah , (2006), *Journal of Rare Earths*,24, 289–293.
- [19]. Pradip Z. Zambare,(2019) *IJRAR*, 6, i570-i574.
- [20]. Pradip Z. Zambare and O. H. Mahajan (2013), *Mat. Sci.*, 1, 8-13.
- [21]. Savali Ravi Kumar, Ch. Atchyutha Rao, K.V.R. Murthy, B. Subba Rao, (2018), *IJTRD* , 5(2), 755-757.
- [22]. Pradip Z. Zambare , A. P. Zambare and, O. H. Mahajan, (2013), *Journal of Luminescence and its applications*, 6(4), 249-252.
- [23]. Dr. Ch. Atchyutha Rao, K. V. R. Murthy,(2020), *IJSDR* , 5, 112-116.
- [24]. Pradip Z. Zambare, K. S. Chaudhari, O.H. Mahajan, *IJPAP* 2,(2014), 1-5.
- [25]. D. L. Monika, H. Nagabhushana, B. M. Nagabhushana, S.C.Sharma, K. S. Anantharaju, B. Daruka Prasad, C. Shivakumara, (2015), *J. of Alloys and Compounds*, 648, 1051-1059.
- [26]. https://www.google.com/search?q=production+of+white+light&rlz=1C1CHBD_enIN806I_N806&oq=production+of+white+light&aqs=chrome..69i57j0i22i30j0i39014.26336j0j7&source=id=chrome&ie=UTF-8.
- [27]. Aroh Shrivastva, Deepak yadav, Paritosh Chaudhari and Amit Sircar, *J Mater Sci: Mater Electron* (2023) 34:455 <https://doi.org/10.1007/s10854-023-09900-y>