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PREPARATION AND PHOTOLUMINESCENCE STUDY OF DIFFERENT CONCENTRATIONS OF EU^{3+} DOPED $NA_2SR(PO_4)F$ PHOSPHORS CO-DOPED WITH (SO_4) AND (VO_4) IONS FOR OPTICAL APPLICATIONS

Arati Duragkar
aratimduragkar@gmail.com
Department of Chemistry, Sevadal Mahila Mahavidyalaya, Nagpur 440024, India

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ABSTRACT:

Compound $Na_2Sr(PO_4)F$ doped with various concentrations as 0.1, 0.3, 0.5, 0.7 and 1 mol% of rare earth dopant Eu. These phosphors were synthesized by using combustion method. Photoluminescence (PL) emission spectra shows highest intensity emission peak at 592nm and 616 nm for 1mol% Eu doped phosphor. Preliminary studies showed that the phosphor might be a promising candidate for the optical system.To increase the luminescence intensity, $Na_2Sr(PO_4)F$: Eu 1mol% phosphor was further co-doped with various concentrations of (SO_4) and (VO_4) ions. Commission International deI'Eclairage (CIE) chromaticity colour coordinates of these phosphors were calculated. CIE- Chromaticity colour coordinates of this phosphor shows red emission.

Keywords:- Phosphors, Synthesis, Optical Properties, Combustion method.

INTRODUCTION:

Solid-state lighting sources are considered to be the next generation of white light- emitting diods (w- LED) due to their extraordinary optical properties, their extended lifetime, as well as mercury-free excitation. Europium ion is vigorously studied activator in the field of luminescence and extremely useful as a dopant in phosphors for LED and display devices, sensor [A. Rosendo et,al.(2003), S. Das et, al.(2017), Z.Gao et, al. (2017), G. Li et,al (2017)]. It can exist in Eu^{3+} as well as Eu^{2+}form. Eu^{3+}ion possess interconfigurational 4f-4f transitions which are forbidden transitions and Eu^{2+} ion possess 4f-5d transition which is allowed transition. 4f-4f transitions of Eu^{3+} ions are hypersensitive transition [G. Blasse et, al. (1966), G. Blasse et, al.(1966),G. Blasse et, al.(1966)]. Depending on the chemical environment present around the crystallographic site, which is available for doping of Eu^{3+}ions, some forbidden transitions become allowed and blue-green-red emissions can be seen. Site dependent emissions of Eu^{3+} ions is utilized by researchers to study local structural symmetry in host [T.Jansen et,al.(2017),M.Kaczkan et,al.(2016)]. W-LEDs show low colour rendering index and high correlated colour temperature due to the absence of a long- wavelengthred component in the visible spectrum. Red, Orange-red emitting phosphor can be used along with green and blue emitting phosphors to enhance the colourrendering indexof the W-LED[7S.K. Rautet et, al. (2014), R. Velchuri et, al. (2009)]. Eu $^{3+}$ dopant ion is one of the lanthanide element leads to produce red luminescence.

In this work, different concentrations of Eu^{3+} doped $Na_2Sr(PO_4)F$ phosphors with well chemical and physical stable performance were synthesized, which further codoped with different concentraions of (SO_4) and (VO_4) ions.

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The photoluminescence study of these samples revealed that the phosphors can be excited by blue and emit red light.

Synthesis of $Na_2Sr(PO_4)F$: Eu co-doped with (SO_4) and (VO_4)

The inorganic material $Na_2Sr(PO_4)F$ was synthesized by using solution combustion method. The A. R Grade chemicals, Sodium nitrate ($NaNO_3$), Strontium nitrate [Sr $(NO_3)_2$], Ammonium fluoride (NH_4F), Ammonium dihydrogen phosphate ($NH_4H_2PO_4$), Europium (III) oxide (Eu_2O_3), Ammonium sulfate [$(NH_4)_2SO_4$], Urea were used without further purification. All the chemicals weight as per stoichiometric ratio and dissolved in minimum quality of water and mix the content in china dish. Urea is used as a fuel. The content in the china dish then kept in preheated vertical muffle furnace at 600°C. 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 concentrate Eu^{3+} in the form of Eu_2O_3 dissolve in concentration nitric acid.

The highest emission intensity is shown by $Na_2Sr(PO_4)F$: Eu in 1 mol% therefore by keeping this dopant concentration constant we further added co-dopant like (SO_4) and (VO_4) by using A. R Grade chemical Ammonium sulfate [$(NH_4)_2SO_4$], Strontium nitrate $Sr(NO_3)_2$, Ammonium fluoride (NH_4F), Sodium nitrate ($NaNO_3$) in different concentration. These co-doped phosphors were also synthesized by using the same solution combustion method.

The chemical reaction is-

$$\mathbf{2NaNO_3 + Sr(NO_3)_2.9H_2O + NH_4H_2(PO_4) + NH_4F + (NH_4)_2SO_4}$$

$$\downarrow$$

$$\mathbf{Na_2Sr(PO_4)F + 8N_2 + CO_2 + 13H_2O}$$

Molecular weight of chemicals:

Na_2NO_3 = 84.9947 g

$Sr(NO_3)_2$ = 211.63 g

NH_4F = 37.037 g

Urea = 7.5 g

1.1 Table -1: Determination of optical density of Cu (II) solution before and after adsorption on
Table: Stoichiometric weight of chemicals in gram to synthesize $Na_2Sr(PO_4)F$: Eu (0.1,

	$NH_4H_2(PO_4)$	$NaNO_3$	$Sr(NO_3)_2$	Eu_2O_3	Urea	NH_4F
Pure	1.5	2.2168	-	-	7.5	0.4830
0.1	1.5	2.2168	2.7571	0.0046	7.5	0.4830
0.3	1.5	2.2168	2.7516	0.0138	7.5	0.4830
0.5	1.5	2.2168	2.7461	0.0229	7.5	0.4830
0.7	1.5	2.2168	2.7406	0.0321	7.5	0.4830
1	1.5	2.2168	2.7323	0.0458	7.5	0.4830

0.3, 0.5, 0.7, 1 mol%) phosphors.

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1.2 Table: Stoichiometric weight of chemicals in gram to synthesize $Na_2Sr(PO_4)(SO_4)F$: Eu (0.1, 0.3, 0.5, 0.7, 1 mol%) phosphors.

SO_4	$NaNO_3$	$SrNO_3$	NH_4F	Eu_2O_3	Urea	$NH_4H_2PO_4$	$(NH_4)_2SO_4$
Pure	1.4777	1.8397	0.3219	0.0305	5	-	-
0.1	1.4777	1.8397	0.3219	0.0305	5	0.9	0.1148
0.3	1.4777	1.8397	0.3219	0.0305	5	0.7	0.3446
0.5	1.4777	1.8397	0.3219	0.0305	5	0.5	0.5743
0.7	1.4777	1.8397	0.3219	0.0305	5	0.3	0.8041
1	1.4777	1.8397	0.3219	0.0305	5	0	1.1487

1.3 Table: Stoichiometric weight of chemicals in gram to synthesize $Na_2Sr(PO_4)(VO_3)F$: Eu (0.1, 0.3, 0.5, 0.7, 1 mol%) phosphors.

VO_4	$NaNO_3$	$SrNO_3$	NH_4F	Eu_2O_3	Urea	$NH_4H_2PO_4$	$(NH_4)_2VO_4$
Pure	1.4777	1.8397	0.3219	0.0305	5	-	-
0.1	1.4777	1.8397	0.3219	0.0305	5	0.9	0.1016
0.3	1.4777	1.8397	0.3219	0.0305	5	0.7	0.3050
0.5	1.4777	1.8397	0.3219	0.0305	5	0.5	0.5084
0.7	1.4777	1.8397	0.3219	0.0305	5	0.3	0.7118
1	1.4777	1.8397	0.3219	0.0305	5	0	1.0169

RESULT AND DISCUSSION –

Photoluminescence Study of $Na_2Sr(PO_4)F$ doped with different concentrations of Eu^{3+}-

Excitation and emission spectra of $Na_2Sr(PO_4)F$: Eu (0.1, 0.3, 0.5, 0.7 & 1 mol%) –

Fig. 1.1 revealed PL excitation spectra of $Na_2Sr(PO_4)F$: Eu^{3+}phosphors under 591 nm emission wavelength. PL excitation spectrum shows five excitation band around 363 nm,383 nm and 395 nm, 417nm and 466nm which arised from $^7F_0 \rightarrow {}^5D_4$,$^7F_0 \rightarrow {}^5G_2$, $^7F_0 \rightarrow {}^5L_6$,$^7F_1 \rightarrow {}^5D_3$,$^7F_0 \rightarrow {}^5D_2$ transitions that are quite notable and can be well-matched with commercial InGaN-based LEDchips. Characteristically, emission spectra of Eu^{3+}doped $Na_2Sr(PO_4)F$ [Fig. 2.2] shows numerous sharp emission peaks between 500 and 700 nm, which are ascribed to the 4f→4f transitions of Eu^{3+} ions. The characteristic peaks are obtained at 593 nm ($^5D_0 \rightarrow {}^7F_1$), 616 nm ($^5D_0 \rightarrow {}^7F_2$). From Fig. 1.2 ,it is clear that the $^5D_0 \rightarrow {}^7F_2$ transition peak at 616 nm is considerably weaker than the $^5D_0 \rightarrow {}^7F_1$

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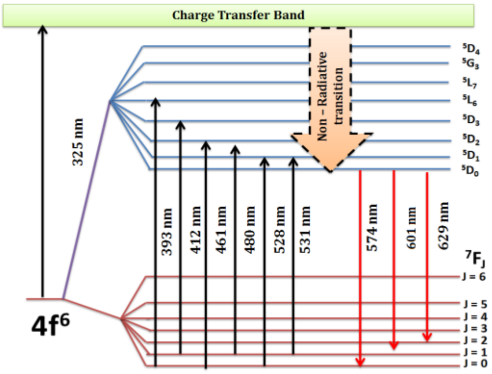


transition peak at 591 nm. It is well known that the transition $^5D_0 \rightarrow {}^7F_1$ is due to magnetic dipole and it is unresponsive to the crystal field environment. The highest intensity emission peak is observed for $Na_2Sr(PO_4)F$: Eu 1mol% phosphor. Therefore, further co-doping is done with this phosphor.

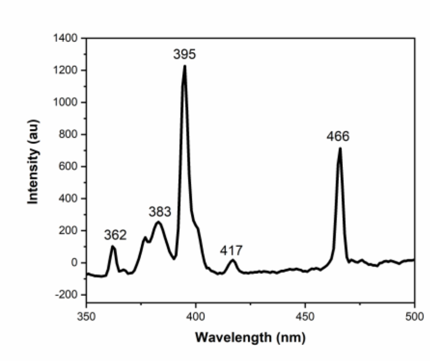


Fig. 1.1

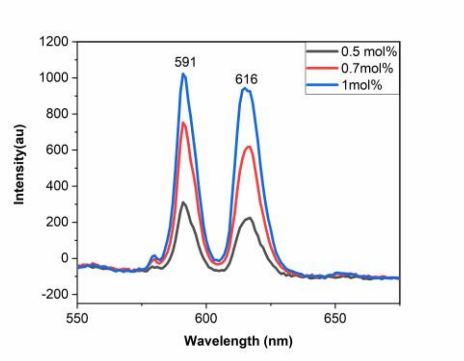


Fig. 1.2

Fig. 1.1 Excitation Spectra of $Na_2Sr(PO_4)F:Eu^{3+}$ phosphor monitored at 591 nm emission wavelength
Fig. 1.2 Emission Spectra of $Na_2Sr(PO_4)F:Eu^{3+}$ phosphor monitored at 395nm excitation wavelength
Fig. 1.3: Energy Level Diagram of Eu^{3+} ion doped in $Na_2Sr(PO_4)F$ host

Photoluminescence Study of $Na_2Sr(PO_4)F$: Eu 1mol % co-doped with different concentrations of SO_4 ions-Excitation and emission spectra of $Na_2Sr(PO_4)_{1-x}(SO_4)_xF$: Eu 1mol% (where x = 0.1, 0.3, 0.5, 0.7 & 1) –

In previous section 1.1 it has been reported that the maximum PL emission intensity for $Na_2Sr(PO_4)F$: Eu^{3+} phosphor was observed at 1 mol% of Eu^{3+} ions(Fig 1.2). By considering this static in the scheme we developed the $Na_2Sr(PO_4)_{1-x}(SO_4)_xF$: Eu^{3+} phosphor by keeping Eu^{3+} concentration 1 mol% constant and varying the (SO4) concentration from 0.1 to 1.The PL excitation and emission spectra of $Na_2Sr(PO_4)_{1-x}(SO_4)_xF$: Eu^{3+} are shown in fig 4.4 and 4.5 respectively.

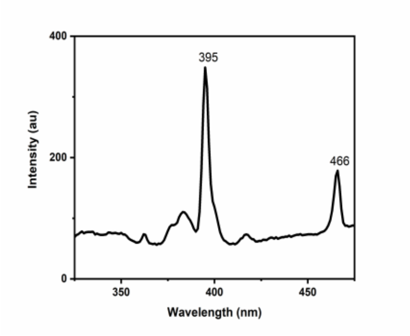


Fig. 1.4

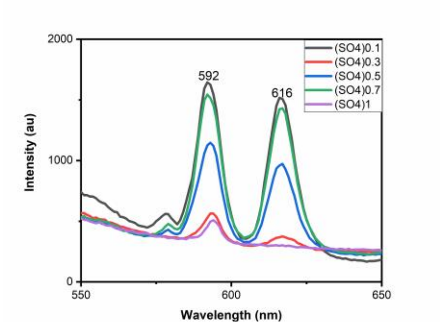


Fig. 1.5

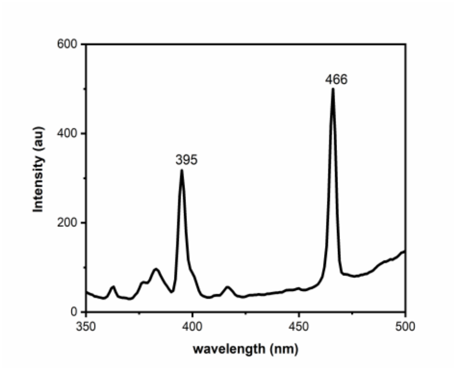
Fig. 1.4 Excitation Spectra of $Na_2Sr(PO_4)_{1-x}(SO_4)_xF$: Eu 1mol% phosphor monitored at 592 nm emission wavelength

Fig. 1.5 Emission Spectra of $Na_2Sr(PO_4)_{1-x}(SO_4)_xF$: Eu 1mol% phosphor monitored at 395 nm

The excitation band monitored at 616nm emission is observed at 395 nm as highest intensity excitation peak. At this excitation wavelength when emission spectra are observed it showed two emission peaks with nearly similar intensity at 592nm and 616 nm due to $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ electronic transition. The highest emission intensity is observed for $Na_2Sr(PO_4)_{0.9}(SO_4)_{0.1}F$: Eu 1mol% phosphor. As compared to emission intensity of $Na_2Sr(PO_4)F$: Eu 1mol%, the emission intensity of $Na_2Sr(PO_4)_{0.9}(SO_4)_{0.1}F$: Eu 1mol% phosphor is nearly increased by nearly 1.5 times. The emission peaks corresponds to red region emission.

Photoluminescence Study of $Na_2Sr(PO_4)F$: Eu 1mol % co-doped with different concentrations of VO_4 ions

Excitation and emission spectra of $Na_2Sr(PO_4)_{1-x}(VO_4)_xF$: Eu 1mol% (where x = 0.1, 0.3, 0.5, 0.7 & 1) –



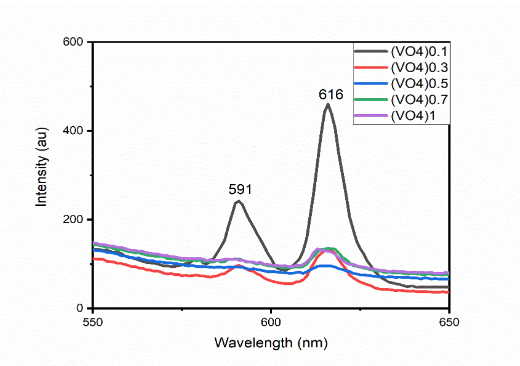


Fig. 1.6

Fig. 1.7

Fig. 1.6 Excitation Spectra of $Na_2Sr(PO_4)_{1-x}(VO_4)_xF$: Eu 1mol% phosphor monitored at 592 nm emission wavelength

Fig. 1.7 Emission Spectra of $Na_2Sr(PO_4)_{1-x}(VO_4)_xF$: Eu 1mol% phosphor monitored at 466 nm

In previous section 1.1 it has been reported that the maximum PL emission intensity for $Na_2Sr(PO_4)F$: Eu^{3+} phosphor was observed at 1 mol% of Eu^{3+} ions(Fig 1.2). By keeping this Eu concentration constant we developed the $Na_2Sr(PO_4)_{1-x}(VO_4)_xF$: Eu^{3+} phosphor by varying the (VO_4) concentration from 0.1 to 1. The

PL excitation and emission spectra of $Na_2Sr(PO_4)_{1-x}(VO_4)_xF: Eu^{3+}$ are shown in fig 4.6 and 4.7 respectively.

The excitation spectra when monitored at 616nm emission wavelength shows the band of peaks in the range of 350nm to 470nm. The prominent peaks are observed at 395nm and 466nm. Out of which the excitation peak at 466nm due to $^7F_0 \rightarrow {}^5D_2$ transition is dominant one. When we monitored the emission spectra at 466nm excitation wavelength two peaks are observed at 592nm and 616 nm. Out of which the peak at 616 is dominant which is contradictory as compared to the previous studied $Na_2Sr(PO_4)F: Eu^{3+}$ and $Na_2Sr(PO_4)_{1-x}(SO_4)_xF: Eu^{3+}$ phosphors. Therefore, $Na_2Sr(PO_4)_{0.9}(VO_4)_{0.1}F: Eu^{3+}$ phosphor can be used as red phosphor.

CONCLUSION –

Adsorption is one of the cheaper methods for removal of heavy metal ions from contaminated water. Various adsorbents are used for removal of heavy metal ions. Many times agricultural waste material is used for the preparation of adsorbent. If such material is used then problem of solid agricultural waste will solved up to some extent. Coconut shells are easily available. We have tried to prepare adsorbent from coconut shell.The adsorbent prepared from coconut shell was used for removal of Cu (II), Ni (II) and Co (II). It was observed that the adsorbent prepared from coconut shell was very good adsorbent for removal of Cu (II), Ni (II) and Co(II) metal ions. Out of three metal ions it removes Cu (II) up to greater extent as compared to Ni (II) and Co(II). Thus instead of wasting coconut shell and throwing anywhere it can be used for the preparation of very good adsorbent.

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