Recent development of Eu$^{3+}$-doped phosphor for white LED application: A review

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Abstract. The phosphor converted w-LEDs gain crucial attention in solid state lighting (SSL) for generation of illumination owing to their numerous meritorious advantages such as superior lifetime, excellent efficiency, compactness, reliability and power saving consumption as well as environmental friendly. The quality of w-LEDs in lighting and display is influence by host phosphor and the choice of activator. So greatly attempt were dedicated to developed inventive un-nucleiluminescent phosphor materials compose of chromatic stability, optimum CRI and low correlated color temperature. This review elaborate the introduction of Eu$^{3+}$ activated red emitting phosphor assigned to 5D0→7FJ (J= 1,2,3,4) energy levels and its fundamental merit for w-LEDs. This article represent the analysis of combination of different types of Eu$^{3+}$ activated luminescent materials by traditional and novel methods and its impact on photoluminescence for SSL.

Keywords: Phosphor; Europium rare earth ions; Energy transfer mechanism; photoluminescence; w-LED.

I. Introduction

In the modern society and digitalization, energy saving and environmental protection gain world-wide attention. In the past decades, luminescence materials shows crucial utilization for development of display technology [1]. On the basis of absorption and emission of radiant energy luminescence are classified into categories namely up-conversion and down conversion emission mechanism[2]. The active centre and active medium plays very important role in order to visualize and realize efficient luminescence. The phosphors materials applicable for commercial purpose must possess certain meritorious characteristics like long operational life, remarkable energy efficiency, high CRI, considerably environmental friendly. Moreover the conventional source of light includes incandescent lamp, halogen and xenon lamp owing to their large energy consumption and environmental issue are replace by light emitting diode[3]. It should be expected that widespread utilization of LED source of light in comparison with traditional light source reduce worldwide consumption of electricity and exclusion of mercury support the environment which gives additional boost to the development of LEDs for lighting[4]. The development of this eco-friendly technology help in reduction of global power requirements as well reduces utilization of fossil fuels. There are various chronology reported for the development optimum luminescence performance of phosphor for w-LED application required for solid state lighting applicable for lighting and display backlight sources [5]. Commercially, phosphor converted w-LEDs can be obtained when blue emitting InGaN based semiconductor chip coated with yellow light emitting YAG:Ce phosphor results generation of white light [6]. However, it shows poor color rendering indices (CRI~ 70-80) along with high correlated color temperature (CCT ~4000-7500 K) but due to deficiency of red component in the spectral region hinder its utilization for indoor lighting[7]. To overcome the flows alternative method is adopted involving the combination of near ultraviolet (n-UV) LED chip with tri-chromatic red, green and blue (RGB) light emitting phosphor has been proposed[8]. Unfortunately phosphor mixture revealed strong re-absorption of the blue light by the green and red phosphor as well as non-uniformity in luminescent properties, resulting deficient in
luminescent efficiency, but at the same time achieve high color rendering index (\(> 90\)), broad band emission spectrum with quite stable color output [9]. It can be further resolved by development of single phase phosphor assemble with UV or n-UV LEDs [10,11].

Rare earth ions play a very important role in the world of display technology, due to their remarkable luminescence characteristics gives extremely sharp peak emission band due to presence of numerous allowed energy levels. The various novel phosphors activated with rare earth ions exhibits significance attention in the field of solid state lighting, radiation detection [12]. The pronounce luminescence efficiency, excellent stability, long durability and environmental-friendly are the main parameter of phosphor required for its commercialization for the w-LEDs application, which can be realized by suitable doping of rare earth and hence gain extensive attention in the field of research [13]. Normally the rare earth ions exhibits two type of emission (i) broad band emission (ii) narrow band emission [14].

Among the various rare earth the europium (Eu) most commonly found in +3 and +2 oxidation state, having second lowest melting point and lower density of all lanthanides. The present review elaborate the basic aspect of transition mechanism involved in trivalent Eu\(^{3+}\) ions along with luminescence properties of Eu\(^{3+}\) doped activated inorganic materials also focused on reliable invention and utilization of rare earth Eu\(^{3+}\) phosphor for w-LEDs applications.

2. Basic of transition mechanism of Eu\(^{3+}\) in the phosphor
Europium is one of the rarest of rare earth elements on earth and is the most reactive rare-earth element. It exists in trivalent oxidation state under most conditions. Eu\(^{3+}\) is a one of potential activator responsible for the development of red phosphors owing to \(^5\)D\(_o\)\(\rightarrow\)^7F\(_{J}\) energy levels and hence consider as promising candidate for red emitting w-LEDs phosphor. The schematic energy level diagram of Eu\(^{3+}\) ions is shown in figure 1.

![Figure 1](image-url) Schematic representation of transition level of Eu\(^{3+}\) ion

The PLE spectrums Eu\(^{3+}\) doped phosphor normally exhibits broad excitation in range of 250 nm-350nm attributes to charge transfer band corresponds to O\(^2-\)\(\rightarrow\)Eu\(^{3+}\) transitions in the host [15]. Moreover existence of several excitation peak corresponds to \(^7\)F\(_o\)\(\rightarrow\)^5D\(_{J}\),\(^5\)L\(_J\) transition. Figure 1 depict some excitation centred at 395 nm, 466 nm and 535nm attributes to \(^7\)F\(_o\)\(\rightarrow\)^5L\(_6\), \(^7\)F\(_o\)\(\rightarrow\)^5D\(_2\), \(^7\)F\(_o\)\(\rightarrow\)^5D\(_1\) transition [16]. In Eu\(^{3+}\) ions Under suitable excitation wavelength PL spectrum consist of several emission peaks centred at 595 nm, 618 nm, 656 nm, 700 nm attributes to \(^5\)D\(_o\)\(\rightarrow\)^7F\(_J\) \((J=1, 2, 3, 4)\) transition[17]. Moreover the intense red of Eu\(^{3+}\) activated phosphor (613 nm, 702 nm) is owing to \(f-f\) orbital transition forbidden the sharp emission is purely host independent. As the trivalent
3. Synthesis Technique for the development of Eu$^{3+}$ activated Phosphor

The various synthesis methods are adopted for the development novel inorganic phosphor materials in order to obtain excellent luminescent performance of rare earth activated phosphor for w-LEDs application few of them are elaborate in details

3.1 Solid State diffusion Method

This technique also term as high temperature synthesis method. This conventional technique is the highly popular and favourable route for the manufacture of rare earth activated phosphors such as oxide, aluminosilicates, vanadates, fluorides, chlorides and nitrides based phosphors for the mass production. [18] The traditional synthesis of several phosphates, sulphates phosphors requires temperature greater than 1000°C, because it cannot be prepared smooth by the SSD at the temperatures below 1000 °C. This method involves the precursors in solid form which are mixed to form solid solution. The heat treatment generally ranging from 500°C to 2000°C required to carried out homogeneous diffusion between cation and the anion by overcoming the lattice energy.

3.2 Combustion Method

The combustion method is the use method for development of oxide-based phosphors with nano range particle size. This method composed of precursors normally taken in nitrate form and urea, carbohydrazide or hydrazine-based compounds and glycine etc are used as reducer (fuel). All starting materials are keep into stoichiometrically amount with the calculation of oxidizer and reducer valences which are further crushed by mortar and pestle for 1hr. The resultant past is kept in the furnace at the temperature of 550°C-600°C. The vigorous flame is produced with a temperature of about 1600°C. The resultant material is in the form of homogeneous and foamy phase.

3.3 Sol-gel method

It is techniques widely used for the fabrication of nano structured phosphor materials and also thin films or ceramics prepared by alkoxide materials which are generally organometallic compound. This process involved formation of sol by hydrolysis and polymerization reaction result in the formation of porous gel and hence term as sol-gel method. The resultant material is obtained by drying and annealing. This method is a very attractive method because of less time duration, low process temperature, low cost, good quality products and obtained maximum purity to making a chemical material.

4. Development of novel phosphor for w-LED

4.1 $Sr_3Eu_2B_4O_{12}$: Eu$^{3+}$

Li et al. reported novel Eu$^{3+}$ activated Sr$_3$Eu$_2$B$_4$O$_{12}$ phosphor synthesized by conventional SSR technique [19]. It crystallizes as an orthorhombic cell in space group $Pnma$ (no.62) confirm from Rietveld method. The emission of $^5D_0 \rightarrow ^7F_2$ transition is more pronounce than that of $^5D_0 \rightarrow ^7F_1$ reveals that illumination in red region only because of Eu$^{3+}$ among the three crystallographic positions.In addition to this it excellent thermal stability of 85.6% with quantum efficiency of 94.7% along with CCT= 4102K, CRI=83.4, and CIE= (0.3736, 0.3639) which are prime factor enhance credential of prepared phosphor for the fabrication of red emitting w-LED [20].

4.2 $Ca_3Y(AlO)_{12}(BO_3)_3$: Eu$^{3+}$

Guo et al. developed red color emitting Eu$^{3+}$ doped Ca$_3$Y(AlO)$_3$(BO$_3$)$_3$ phosphors for w-LEDs application by SSR method [21].Preparedmaterial reveals intense emission at 621nm in the excitation wavelength 397nm ascribe to CBT of $Eu^{3+} \rightarrow O^{2-}$ attributed to $4f \rightarrow 4f$ transition of Eu$^{3+}$ ions. The emission intensity is influence by the doping concentration of Eu$^{3+}$ ions revealed the desired emission at 5.0 mol% of Eu$^{3+}$ ions. This phosphor, shows the color purity of 90% with excellent thermal stability of 76.5% at 150°C from room temperature.

4.3 $K_3Ln(PO_4)(WO_4)$: Tb$^{3+}$, Eu$^{3+}$

A novel $K_3Ln(PO_4)(WO_4)$ ($Ln=$ y, Gd, and Lu), Tb$^{3+}$, Eu$^{3+}$codoped n-UV excitable phosphor was reported by conventional SSR technique[22]. The emission colour of these phosphors could be tuned
from green to yellow, orange and subsequently red by controlling the ratio of Tb$^{3+}$ and Eu$^{3+}$ confirm from shifting of CIE co-ordinates from $(0.35, 0.58)$ to $(0.65, 0.34)$ attributes to energy transfer from Tb$^{3+}$ to Eu$^{3+}$ ions in the host. The IQE and EQE of prepared phosphor reported to be 76.45% and 35.24%, respectively at the excitation 394nm with excellent thermal efficiency of 92.2% at temperature of 150°C.

IV. $Pb_3Bi(PO_4):Eu^{3+}$

Yu et al. described a synthesized eulytite structure of Eu$^{3+}$ doped red emitting Pb$_3$Bi(PO$_4$) Phosphor developed by solid state diffusion method[23]. This phosphor shows body-centered cubic (BCC) structure with space group I43d (002) confirm from XRD data and Rietveld study. PL characteristics exhibits that at excitation wavelength of $394\text{nm}$ shows strong emission peaks however at $593\text{nm}$ ascribe to $^5\text{D}_0\rightarrow^7\text{F}_1$ transition of Eu$^{3+}$ ions. Hence, the reported Pb$_3$Bi(PO$_4$)$_2$: Eu$^{3+}$ red phosphor has potential applications in decoration lighting and LED devices.

V. $KBaScSi_3O_9:Eu^{3+}$

Nagaraj et al. reported Eu$^{3+}$ doped novel red emitting silicate based KBaScSi$_3$O$_9$ (KBSS) phosphor by SSR technique[24]. The XRD study exhibits monoclinic structure of prepared material in nature with $P2_{1}/n$ space group. The PLE spectra recorded in region $350$-$500\text{nm}$ at $\lambda_{em} = 61\text{inm}$, high intensity peak observed at $466\text{nm}$ corresponds to $^7\text{F}_0\rightarrow^5\text{D}_2$ transition of Eu$^{2+}$ ions, shows emission spectra consisting several emission peaks centred at $611\text{nm}$ corresponds to $^5\text{D}_0\rightarrow^7\text{F}_2$ transition of Eu$^{3+}$ ions in the host suggested best suitability for laser application.

VI. $CaBiVO_3:Eu^{3+}$

Kaur et al. reported Eu$^{3+}$ singly doped Calcium bismuth vanadate CaBiVO$_3$ (CBV) phosphor via solid state reaction method [25]. The prepared phosphor reveals orthorhombic crystal with space group $D_{2h}^{3} - pbca(61)$. Under UV absorption ($\lambda_{ex} = 342\text{nm}$), undoped CBV shows weak broad band emission in the range of $400$-$680\text{nm}$ ascribe to $^3\text{T}_{1g}\rightarrow^1\text{A}_{1g}$ transition of $[\text{VO}]^{2+}$ group. Whereas Eu$^{3+}$ activated CBV material revealed intense red emission in blue excitation may be due to energy transfer from $[\text{VO}]^{2+}$ groups to the $^5\text{D}_0$ energy level of Eu$^{3+}$ ions in the host. At the excitation wavelength $464\text{nm}$ CBV: xEu$^{3+}$ (0.1 $\leq$ x $\leq$ 6.0) phosphor shows most intense emission at $593\text{nm}$ (orange region) attributes to magnetic dipole $^5\text{D}_0\rightarrow^7\text{F}_1$ and other located at $613\text{nm}$ (red region) ascribe to electric dipole $^5\text{D}_0\rightarrow^7\text{F}_2$ transition. The concentration quenching beyond 4 mole% in Eu$^{3+}$ ions in host. The CIE co-ordinates at excitation wavelength $464\text{nm}$ are obtained at $(0.639, 0.358)$ which are resemble to that of commercially available red phosphor.

VII. $Ba_2GdVO_6:Eu^{3+}$

A series of white emitting Eu$^{3+}$ doped Ba$_2$GdVO$_6$ luminescent material was reported by SSD technique[26]. The band gap Eg of prepared material is $3.78\text{eV}$ obtained from diffuse reflection spectra. The excitation spectrum of prepared phosphor monitor at $612\text{nm}$ shows broad band in region $200$-$300\text{nm}$ ascribe to the Eu$^{3+}$-$\text{O}^{2-}$ charge transfer transition and host absorption with sharp excitation peak at $395\text{nm}$ and $465\text{nm}$ corresponds to $^7\text{F}_0\rightarrow^5\text{L}_6$ and $^7\text{F}_0\rightarrow^5\text{D}_2$ respectively transition of Eu$^{3+}$ ions in the host. Moreover, excitation spectra consist of two sharp peak at $553\text{nm}$ and $612\text{nm}$ ascribe to MD transition $^5\text{D}_0\rightarrow^7\text{F}_1$ along with ED transition $^5\text{D}_0\rightarrow^7\text{F}_2$ respectively of Europium element in the host at excitation wavelength $465\text{nm}$. The as-prepared Ba$_2$GdVO$_6$:20Eu$^{3+}$ exhibit 72% of thermal stability at 150°C.

VIII. $(Y_1_xEu_x)_2WO_6:Eu^{3+}$

Shi et al. reported the synthesis of $(Y_{1-x}Eu_x)_2WO_6$ red emitting phosphor via hydrothermal method. Under UV excitation, the prepared phosphor exhibits intense emission centered at $611\text{nm}$ corresponds to $^5\text{D}_0\rightarrow^7\text{F}_2$ transition of Eu$^{3+}$ ions in the host. Moreover, bluish luminescence of $[\text{WO}_6]^{6-}$ complex ions was observed in broad range of $350$-$650\text{nm}$. The depletion in intensity with enhancement of Eu$^{3+}$ ions reveals energy transfer from $[\text{WO}_6]^{6-}\rightarrow\text{Eu}^{3+}$ in the host[27].

IX. Sr$_4$Al$_2$O$_{15}:Eu^{3+}$

The photoluminescence and thermoluminescent properties of Sr$_4$Al$_2$O$_{15}$: Eu$^{3+}$ phosphor was studied by Emen et al [28]. As-synthesize materials display orthorhombic structure with $Pnmm$ (space group) confirm from XRD analysis. This phosphor monitor at $616\text{nm}$ gives four excitation band. The intense broad excitation band located at $308\text{nm}$ ascribe to charge transfer transition of Eu$^{2+}$-$\text{O}_2$ and other three
are position at 360nm, 383nm, 411nm corresponds to $^7F_o \rightarrow ^5L_J$ (J=6, 3, 2) transition of Eu$^{2+}$ ions respectively in the host. The emission spectrum corresponds to excitation wavelength 308nm exhibits four emission peak cantered at 592nm, 616nm, 651nm, 707nm assigned to $^5D_o \rightarrow ^7F_J$ (J=1, 2, 3, 4) transition of Eu$^{3+}$ ions. The emission peak at 616 nm and 595nm referred as ED and MD transition etc.

The larger emission of electric dipole (ED) as compare to magnetic dipole (MD) reveals that lower symmetry around the Eu$^{3+}$ ions does not contain in inversion symmetry centre [29]. Activation energy range from 0.562-0.886 eV indicate the creditability of phosphor for display and LED application.

X. $\text{Ba}_2\text{YAlO}_5$: Eu$^{3+}$

Duan et al. studied Eu$^{3+}$ singly doped $\text{Ba}_2\text{YAlO}_5$ red emitting phosphor[30]. PLE excitation spectrum shows prepared phosphor is not only UV excitable but also gives excellent absorption in blue region. The prepared phosphor is highly sensitive to Eu$^{3+}$ ions leads to dominance of electric dipole transition exhibits intense emission in red region at 614nm attributes to $^5D_0 \rightarrow ^7F_2$ transition with CIE co-ordinate shift to (0.590, 0.398) and addition to shows 74.4% thermal stability at 473k.

XI. $\text{Ca}_9\text{La}(\text{PO}_4)_{5-x}\text{(SiO}_4)_x\text{FCl}:\text{Eu}^{3+}$

A Eu$^{3+}$ activated $\text{Ca}_9\text{La}(\text{PO}_4)_{5-x}\text{(SiO}_4)_x\text{FCl}$ crystal phosphor was developed via solid state diffusion technique [31]. XRD pattern of as synthesized material reveals highly crystalline in nature. The fluoride-based phosphors doped with different rare earth activators have gain significant attraction due to excellent properties like various application fingerprint, light-emitting diodes, electroluminescence cell and many others [32]. The PLE spectra of modified $\text{Ca}_9\text{La}(\text{PO}_4)_{5-x}\text{(MoO}_4)_x\text{(SiO}_4)_x\text{FCl}:\text{Eu}^{3+}$ monitored at excitation wavelength 395 nm exhibits the emission centred at 593nm($^5D_0 \rightarrow ^7F_1$) and 613nm($^5D_0 \rightarrow ^7F_2$). The $^5D_0 \rightarrow ^7F_2$ transition is much stronger than the $^5D_0 \rightarrow ^7F_1$ energy level. As compared to the other ions (MoO$_4^{3-}$), (VO$_4^{3-}$), (WO$_4^{3-}$), and (SO$_4^{3-}$), $\text{Ca}_9\text{La}(\text{PO}_4)_{5-x}\text{(MoO}_4)_x\text{(SiO}_4)_x\text{FCl}$ 1 mol\%Eu$^{3+}$ phosphor is a promising red candidate for w-LEDs.

Table 1: Summary of some investigations carried out on various types of Red emitting phosphors.

| Host Materials         | Preparation method | Crystal structure | Excitation wavelength (nm) | Emission wavelength (nm) | Colour coordinates | Applications                      | Ref. |
|------------------------|--------------------|-------------------|----------------------------|--------------------------|--------------------|------------------------------------|------|
| $\text{KBaScSi}_3\text{O}_9$: Eu$^{3+}$ | solid state reaction method | monoclinic system (P21/n) | 395 nm, 466 nm             | 593 nm, 611 nm          | (0.655, 0.3447) | solar cells, solid state lighting, lasers & field emission displays, | [24] |
| $\text{SrAl}_2\text{O}_4$: Eu$^{3+}$    | Combustion technique | Monoclinic system | 393 nm                     | 618 nm                  | (0.6313, 0.3683) | display device                     | [33] |
| $\text{NaSrB}_3\text{O}_5$: Eu$^{3+}$  | solid state reaction method | Monoclinic system P21/c | 395 nm, 466 nm             | 611 nm                  | (0.631, 0.368)   | white LED applications              | [34] |
Applications and future prospective

White light emitting diode received prime position and attention which are in position to replace the convention source of light include incandescent and fluorescent lamps in the field of lighting industry and made tremendous revolution in next generation light source. The world-wide utilization of w-LEDs from house hold to street lighting support environmental issue and heavy power consumption shows great the impact on economic and ecological aspect of society. The various Eu\(^{3+}\) activated phosphor is commercialize for industrial application for the development of w-LEDs. Raju G. et al. reported Ba\(_4\)La\(_6\)(SiO\(_4\))O:Eu\(^{3+}\) UV excitable phosphor exhibits red emission cantered at 395 nm ascribe to \(^{7}F_0\rightarrow^{5}L_6\) transition of Eu\(^{3+}\) reveals significance in development of FED systems for lighting applications as red light emission[38]. The TRZNB:Eu\(^{3+}\) phosphor was successfully synthesized by conventional melt quenching technique, reveals deep red emission at 616 nm under n-UV excitation have potential applicable in the optoelectronic luminescent display devices and laser materials[42]. Recently Zhang et
al. synthesized Eu$^{3+}$ doped Y$_2$Mg$_2$Al$_2$Si$_2$O$_12$:phosphor via high temperature SSD technique[43]. It exhibits the PL emission in the range of 600nm to 720nm shows its probable materials for red emission.

5. Conclusion

The exclusion of conventional light sources such as incandescence lamp, fluorescent triphosphor, tungsten halogen, xenon arc lamp etc with the development of low power consumption pc-wLED green light source for the next generation illumination is in crucial progress owing to their numerous characteristics like exceptional luminous efficiency, low power consumption, desired CCT and CRI adjustable by change of physical properties of selected phosphor are elaborate which is fundamental aspect for economical and ecological growth. This paper discusses with the europium- (Eu$^{3+}$) doped luminescent phosphors materials for SSL application in various field of technology. The phosphor must avoid blue color reabsorption by green and red component for fabrication of LED. The emission from the rare earth activated phosphor is describe the various parameter such as desirable excitation and emission spectra, excellent luminescent efficiency.

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