Rare Earth (RE) doped color tunable phosphors for white light emitting diodes

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Abstract. Currently, white light-emitting diodes (WLEDs) are emerging lighting techniques and are highly desirable. These are one of the world's highest lighting technologies. LEDs have attracted special attention of research society, scholars and society through their benefits and applications. Phosphor-converting light-emitting diodes (PC-LEDs) show immense potential for the future and, in the coming years, it is expected that LEDs will be easily available at a cheaper price with lower energy consumption. Rare earth activated phosphors are an integral and important part of white light generation. In this review, we have studied numerous research papers about phosphor-converted light-emitting diodes (pc-LED). The main objective of this review paper is to analyze inorganic materials that have been synthesized by various methods and are suitable for eco-friendly lighting applications. In this review, we propose the plan and discovery of new LED phosphors from two alternate points of view, i.e., basic crystal structure in various host systems and co-doped activators through energy transfer. The color-tunable properties of rare-earth activated phosphors explain in detail. Numerous color-tunable phosphor materials have been studied and discussed in this review. In addition, utilizations, difficulties and development or improvement of pc-WLEDs will be discussed.

Keywords: pc-LEDs; Phosphor; Rare earth; Energy transfer; Color Tunable

1. Introduction
The present time is the time of modernization, in such a situation the demand for energy is increasing day by day. Modern technologies have fill up the gap between day and night. In this era of modernization, lighting technology is so powerful and efficient that can provide daylight even at night. Current light technology has changed human activity. The role of lighting has become important in human life. The electric lamp is an important invention used in everyday life. It was first invented in 1879 by Thomas Alva Edison [1]. In the modern era, energy-saving is important on this planet because if energy is not utilized judiciously and wisely then many problems will be created in front of us. The main goal of energy-saving is to decrease the amount of energy required to provide products and services. LED lighting technology reduces energy consumption, and provides an effective setup for the lighting sector. We have many choices in energy-efficient lighting. In the present time, pc-WLEDs are emerging as a high level solid state lighting in the future due to high quality and low cost technology [2]. According to the US Department of Energy (DOE) report in 2012, it was reported that if the use of WLED could be reduced by about 20% of total electricity consumption in the coming two decades (2010-2030) [3,4]. Figure 1 shows the progress in lighting technology [5].
Figure 1. Advances in lighting technology from Edison bulb (1879) to white LED (2010)

PC-WLEDs have various advantages such as low energy consumption, long lifespan, high efficiency, and environment friendly nature [2,3,6]. Light-emitting diodes (LEDs) have gained considerable popularity and the role of LEDs in the field of light will be important in the coming times. LEDs have more functionality and quality than other light sources, they have proved their ability through the maximum conversion of the electrical energy into light [7,8]. In the last few years, PC-WLEDs has grown popularity and it is already used commercially in various applications such as traffic lights, outdoor lighting, inside and outside lighting in airplane, vehicles, and transports, etc [9–11]. Generally three ways are known for white light generation based on LEDs as shown in Figure 2. (i) yellow-emitting phosphor excited by blue-emitting diode (ii) combination of Red, Green and Blue (RGB) LED lights and (iii) By combination of RGB phosphor with near UV LEDs [6,12–14].

Figure 2. Representation of white light generation through the combination of phosphors and LEDs.  
Adopted from Ref. [15]

The first commercially available White LED was invented in 1996 by Nichia Chemical Co., which emit white color light when blue InGaN LED chip coated with YAG:Ce3+ [16–18]. At present this method is used for White LED production. Nevertheless, this way has some drawbacks such as low CRI, high CCT value and absence of red component. White LEDs are more effective than other light sources because they have many controllable characteristics such as emission spectra, direction, color temperature, modulation, and polarization. LEDs have had very beneficial effects on the economy, the environment, and our lives. The impact is so evident that in the year of 2014, the Nobel Prize for Physics was awarded to the great physicists Isamu Akasaki, Hiro Amano, and Shuji Nakamura for the invention of efficient blue LEDs.

2. Rare earth ions

Rare earth ions are generally f block element it is also called lanthanide element. At present 17 rare earth element are founded specifically the 14 lanthanides (Ce–Lu), and 3 scandium, yttrium and Lanthanum. The electronic configuration of lanthanides are partially filled 4f shell. These three elements behave as like rare earth element. Generally rare earth ions are found in the form of trivalent elements such as Eu3+, Dy3+, Ce3+, Gd3+ etc. but some rare earth ions are found in the form of divalent element. Every element has their own physical, chemical and luminescence properties. The role
of rare earth ions in phosphors can be explained with the help of excitation and emission type spectra. The rare earth based nano phosphors are widely used in various fields such as solid state lighting, CR tubes, TV screen, display panels, biomedical field, TL dosimetric applications, solar cells, Phototherapy lamp etc [19–24]. At Now, a number of rare earth doped phosphor materials are prepared such as Gd$_2$Mo$_3$O$_9$:Eu$^{3+}$ [25], Ca$_8$Mg$_3$Al$_2$Si$_7$O$_{28}$:Eu$^{2+}$ [26], Ba$_2$YNbO$_6$:Mn$^{4+}$ [27], La$_2$(MoO$_4$)$_3$:Eu$^{3+}$[28], K$_2$Cu$_3$(SO$_4$)$_2$F:Eu$^{2+}$[29], Li$_6$Y(BO$_3$)$_3$:Eu$^{3+}$[30], KSrPO$_4$:Dy$^{3+}$[31], Lu$_2$MoO$_6$:Pr$^{3+}$ [32], Mg$_3$SiO$_4$:Tb$^{3+}$ [33], CaGd$_2$(MoO$_4$)$_4$:2xSm$^{3+}$[34], GdSr$_2$AlO$_5$:Ce$^{3+}$ [35], Ca$_3$Al$_2$O$_6$:RE (RE = Eu$^{2+}$,Ce$^{3+}$) [36], LaPO$_4$:Gd$^{3+}$[37] etc.

Table 1. Represents list of Rare earth ions and their expected excitation, Emission wavelength, emitting color and applications.

| Element                  | Excitation Wavelength (nm) | Emission wavelength (nm) | Emitted color      | Application                                                                 |
|--------------------------|----------------------------|--------------------------|--------------------|-----------------------------------------------------------------------------|
| Europium (Eu$^{3+}$)     | 395nm                      | 593nm & 615nm            | Orange & Red       | Red phosphor for LED, Mercury lamp, Fluorescent lamp.                        |
| Europium (Eu$^{2+}$)     | 354nm                      | 440nm                    | Blue               | Blue phosphor for LED, Laser, Mercury-vapor lamp.                           |
| Dysprosium (Dy$^{3+}$)   | 348nm                      | 480nm & 576nm            | Blue & Yellow      | Blue phosphor for LED, Laser, Mosquito’s repellent lamp.                    |
| Cerium (Ce$^{3+}$)       | 278nm & 295nm              | 355nm                    | UV emission        | Glass and ceramics, Polishing powder                                        |
| Samarium (Sm$^{3+}$)     | 405nm                      | 567nm, 604nm, 608nm      | Yellow & Red       | Yellow and Red phosphor,                                                    |
| Terbium (Tb$^{3+}$)      | 240nm, 379nm, 465nm        | 490nm & 545nm            | Blue & Green       | Blue and green phosphor for LED,                                             |
| Gadolinium (Gd$^{3+}$)   | 260nm, 280nm               | 313nm & 600nm            | UV & Red           | Red phosphor for LED, UV light for phototherapy lamp                        |
| Praseodymium (Pr$^{3+}$) | 448nm                      | 488nm, 522nm, 625nm & 633nm | Blue, green, Red   | White LEDs                                                                  |

3. Important features of pc-LEDs

3.1 Photoluminescence

Photoluminescence is type of luminescence. In Photoluminescence process, light is directed onto a materials, atoms are absorbs energy and goes to higher energy state (excited state) to ground state then radiate a photon as the electron returns to a lower energy state [38]. In other words, Photoluminescence is the process of light emission after from any form of matter after the absorption of photons. In the
photoluminescence process, energy release in the form of photons (light), this energy relates with the
distinction in energy levels between the excited state and the balance state. At present,
photoluminescence is very widely used for the characterization of white materials ranging from washing
powder to plasma screen. According to emission lifetime luminescence can be dived into parts one is
phosphorescence and another is fluorescence.
(i) Fluorescence: When a sample is absorbing photons and emits light with lifetime less than $10^{-8}$
seconds.
(ii) Phosphorescence: When a sample is absorbing photons and emits light with lifetime more than $10^{-8}$
seconds.
Generally, luminescence study gives information of defects which is presents in the radiative de-
excitation of the material. In phosphor, the defect states may move or the density may expand due to
doping of rare earth ions, which is observed by PL studies.

3.2 Energy transfer
The Energy Transfer (ET) process provides basic information about rare earth doped materials for
various applications. In this type of process, there are two particles in nearness, one with an absorption
band at a wavelength moved to the more extended wavelength than another, light energy consumed by
the one retaining at the more limited wavelength is generally moved to the one that absorbs at the more
drawn-out frequencies. That is, one atom goes about as a donor of excitation energy, and the other as an acceptor of this energy. In this type of process, energy transfers from an excited (donor) to the neighbouring unexcited activator (acceptor). The process of energy transfer is important for some applications such as solar energy conversion cells, solid state lasers, displays, amplifier, fluorescent lamps, up-converters, studies of DNA dynamics and conformational analyses of proteins [39–42]. A number of theories and formulas has been established for the rate of energy transfer by electric-dipole-
dipole interaction, electric-dipole-quadrupole interaction, and the exchange interactions.

3.3 CIE Chromaticity coordinates
The Commission Internationale de l'Eclairage (CIE) was founded in 1913 by international autonomous
body, which creates international standard related to the science of light, color, vision and lighting [43].
At present commission has found three CIE models namely as CIE1931, CIE 1960, CIE 1976. All three
models are presented in Figure 3. But CIE 1931 was widely used compare to other CIE models.

![Figure 3. CIE Chromaticity Models of CIE 1931, CIE 1960 and CIE 1976, respectively](http://company235.com/tools/colour/cie.html)
3.4 Correlated color temperature (CCT)

Generally, CCT value is an important parameter used to measure the coldness and heat present in light, it is also described the characteristics of visible light from various sources. As the literature shows that the measurement of coldness and heat of any lighting device can be difficult and the value of CCT can be different for different lighting source. With the help of CCT the color temperature can be determined for the light of use, which does not approximate any blackbody radiator. In this measurement temperature measured in the Kelvin (K) unit. Generally, CCT values are found to be between 2200K and 7200K. According to the literature, if the CCT value is less than 3200, the light source emits warm light, which means that the maximum part of the light is present in the red, orange and yellow range. But if the value of CCT is from 3200K to 7800K, the light source emits cold light i.e., the maximum part of the light is present in the blue color range. However, CCT does not tell you anything about the color rendering capability of the LED.

3.5 Colour Rendering Index (CRI)

In addition to CCT, CRI is an important parameter to determine the lighting quality of a light source, which is indicating the color quality and ability of color emission of the lighting source. The value of CRI value ranges from 0 to 100. The color rendering index value should be higher for white light source because the color shift is very less in this situation. If the value of the CRI is 0 then the quality of the light source is poor, similarly, if the value of the CRI is between 55 – 65, 65 – 75 and 75 – 100, then the quality of the light source is fair, good and excellent, respectively.

3.6 Quantum Efficiency

When planning about LEDs, it is important that we pay equal attention to all the parameters, which is necessary for the development of high-quality LEDs. Quantum efficiency is an important parameter for LEDs. The development of LED requires equal attention to quantum efficiency among other parameters. Using high-efficiency phosphors can be very effective for the conversion of electric power into light. After studying a lot of research papers, it has been found that the quantum efficiency is not mentioned in the new LED conversion phosphor. Quantum Efficiency can be determined by the following equation

\[ Q = \frac{\eta_{\text{con}}}{\eta_{\text{abs}}} = \frac{\eta_{\text{con}}}{\eta_{\text{exc}} - \eta_{\text{reff}}} \] (1)

Where, \( \eta_{\text{exc}} \) is the number of photons, which is directly towards the phosphor, \( \eta_{\text{con}} \) is the number of emitted photons and \( \eta_{\text{reff}} \) is the number of reflected photons.

4. Synthesis methods

Synthesis methods are a key factor for the development of materials for various applications. The numerous synthesis route is adopted for the preparation of luminescent materials. Different synthesis methods are also used to manufacture materials, as well as the ability to provide controllable pigments according to different applications. In contrast, the development of luminescent materials with preferred characteristics becomes somewhat difficult. We face many problems during material synthesis and think of many aspects such as the role of rare-earth ions, pure formation, removal of impurities, particle size, morphology behavior, low cost, behavior with the environment, reproducibility, possible applications, etc. In the literature, various synthesis routes are available for synthesized rare earth activated phosphors for WLEDs applications some of them are elaborate in details:

4.1 Solid-state reaction Method

The solid-state reaction (SSR) method is also known as solid state diffusion method. This method is very popular in the research society for the preparation of rare earth phosphors especially for solid state
lighting applications. In this method such as oxide, carbonate, silicate, vanadates, fluorides, chlorides-based host materials are used for mass production. This method involves the precursors in solid form which are mixed to form solid materials. The heat treatment generally ranging from 700°C to 2000°C required to carried out the homogeneous diffusion between cation and the anion by overcoming the lattice energy.

4.2 **Sol Gel Method**

In the sol gel method, nitride, chloride, sulfate forms or water removable materials are used as starting precursors. This method permits a superior control of the entire reactions participated during the preparation of solids. Homogenous multi-component systems can be effectively acquired. This method carried out via series of chemical process includes hydrolysis, gelation, drying and thermal treatment. 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. The major benefits of this method are homogeneity of synthesized material, high purity of the samples, morphological control, variety of precursors, etc.

4.3 **Combustion method**

Combustion synthesis is one of the easiest ways for preparation of luminescent materials. This method can be divided into two categories i.e., Simple combustion synthesis method and solution combustion synthesis method. In this method, various type of fuels is used such as urea, citric acid, glycine, ethanol, DFH, ethylene glycol, Hydrazine, corbohydrazide, etc. It is also known as self-propagation method. In this method, a high amount of gas is emitted, resulting in the formation of a flame. This method required 500 to 550°C temperature for fired of the sample. The resultant is obtained in the form of homogeneous and foamy phase. This method gained more popularity and attention due to its amazing advantages such as low cost, effective, power saving, short time consumption, and easily done in labs.

4.4 **Hydrothermal Method**

The hydrothermal approach is a procedure that utilizes high temperature and pressure to precipitate materials straightforwardly from solution. This approach utilizes the expanded solubility of practically all inorganic substances in water at raised temperatures and pressures and resulting crystallization of the dissolved material from the solution. Hydrothermal processing is used to synthesize powders, single crystals, fibers and ceramic materials. Today, Modified microwave assisted hydrothermal synthesis is in trend for preparing these phosphor materials which reduces the processing time and energy required for carrying out the reaction. This method also has a potential to improve the material characteristics like morphology and crystallite size of particles.

5. **Previously reported phosphor materials**

In recent few years, White Light Emitting Diodes (WLEDs) as we better know them as, are fast revolutionizing the lighting scenario and White LEDs are emerging as efficient, reliable, & eco-friendly light source with demonstrated capabilities to take the place of FTL’s/CFL’s let alone GLS bulbs. Power LEDs are geared up to revolutionize the lighting systems and are drawing a lot of attention from architects, interiors/exteriors lighting designers, municipalities, housing colonies and automobile lighting designers. At present, combination of yellow-emitting Y3Al5O12:Ce3+ phosphor with blue-emitting InGaN LED chip produce white light, this is most popular way for the generation of white light [31]. But this combination has some serious drawbacks such as low CRI value, high CCT, and deficiency of red component. Due to these drawbacks this combination is not suitable for indoor lighting. This trouble can be totally overwhelmed by adding an appropriate blue-light-activated red phosphor to the LEDs [45,46]. At present a number of rare earths doped phosphors are synthesized. As known that numerous phosphors materials have been extensively reported and their stable physical and chemical properties, crystal structure, PL study studied. But only few materials are available which is used for commercial LEDs. Therefore, researchers and scholars concentrate to develop new phosphors for
WLEDs. Here we are discussed some color tunable phosphors, which are used for White Light Emitting Diodes (WLEDs).

5.1. KAlGeO4:Bi3+, Eu3+ Colour Tunable Phosphor

Wenzhi Sun et al. [47] reported photoluminescence properties of color tunable KAlGeO4:Bi3+, Eu3+ phosphor for WLEDs application. Wenzhi Sun et al. synthesized KAlGeO4:Bi3+, Eu3+ phosphor via high temperature solid state method. The band gap energy of synthesized KAlGeO4:Bi3+, Eu3+ phosphor was found to be 5.90 eV. The Rietveld refinement and lattice parameter shows hexagonal structure, their photoluminescence properties were examined by Hitachi F-7000 spectrophotometer. Then Lifetime and efficiency of synthesized phosphor were also investigated. The PL emission spectra of KAlGeO4:Bi3+ phosphor shows two emission band at 434 nm and 513 nm under 320 nm excitation wavelength, mentioned emission bands are ascribed due to spin-allowed 5S0→5P1 transition of Bi3+. PL emission spectra of KAlGeO4:Eu3+ phosphor shows two emission peaks at 593 nm and 613 nm, which are ascribed due to 5D0→7F1 (MD transition) and 5D0→7F2 (ED transition) of Eu3+ ions. In the energy transfer study, Bi3+ is fixed at 0.01 and concentration of Eu3+ ions vary from 0-0.09, the PL emission intensity increasing after increasing concentration of Eu3+ ions. Wenzhi Sun et al. energy transfer efficiency of KAlGeO4:0.01Bi3+, yEu3+ were also calculated. The CIE color coordinate of KAlGeO4:0.01Bi3+, yEu3+ (y=0, 0.01, 0.03, 0.05 and 0.09) phosphor lies in the blue to pink region after energy transfer it is found in the orange-red region. The PL emission spectra for KAlGeO4:0.01Bi3+, 0.05Eu3+ on temperature (T=290-473K). The PL emission intensity decreased when temperature increased, it is possible thermal quenching effect. Therefore all these results show KAlGeO4:0.01Bi3+,0.05Eu3+ phosphor have potential features, which may potential color tunable phosphor for WLEDs [47].

I. Sr3LaNa((PO4)3):F: Tb3+, Eu3+ phosphors

Shuo Li et al. [48] studied energy transfer mechanism and color tunable emission in Tb3+, Eu3+ co-doped Sr3LaNa((PO4)3):F phosphors. In the present work, Sr3LaNa((PO4)3):F: Tb3+, Eu3+ phosphors were successfully prepared via SSR route. The PL emission spectra of Tb3+ activated Sr3LaNa((PO4)3):F phosphor shows several emission bands at 492 nm, 545 nm, 586 nm, and 623 nm, which are ascribed due to 5D4→7F7 (J=6, 5, 4, 3) transitions. And Eu3+ activated Sr3LaNa((PO4)3):F phosphor shows three emission peaks at 592 nm, 618 nm, and 702 nm under 394 nm excitation wavelength. Energy transfer efficiency also calculated by authors, Sr3LaNa((PO4)3):F: 0.20Tb3+, 0.10Eu3+ phosphor shows 43.71% energy transfer from Tb3+ to Eu3+. The CIE chromaticity coordinate revealed colour tunable emission of Sr3LaNa((PO4)3):F: 0.20Tb3+, 0.10Eu3+ phosphor, it shows colour tuning from green-yellow to orange reddish emission. All these results indicate that under UV excitation blue emitting phosphor combined with Sr3LaNa((PO4)3):F: 0.20Tb3+, 0.10Eu3+ phosphor synthesized phosphor can fabricate white LED lamp with high chromaticity [48].

5.2. Sr3La(BO3)3: Ce, Tb phosphors

Xiulan Wu wt al. [49] reported Sr3La(BO3)3:Ce, Tb phosphors, which is successfully synthesized by solid state reaction method. The synthesized phosphor sample were characterized by various techniques such as XRD, PL, DRS, and PL Decay etc. Their energy transfer and color tunable properties also studied in details. In the present work Xiulan Wu wt al. reported, Ce3+ activated Sr3La(BO3)3 phosphor shows blue emission under near UV excitation. Xiulan Wu wt al. found that Ce3+ activated Sr3La(BO3)3 phosphor shows maximum emission intensity at x=0.03 after co-doping of Tb3+ ions in the Sr3La(BO3)3:0.03Ce3+, yTb3+ (y = 0.05) phosphor shows PL emission intensity decreased when concentration of Tb3+ ions increased. The CIE diagram shows that color tuning from blue to blue greenish then green. All these properties shows Sr3La(BO3)3: Ce3+, Tb3+ phosphor may be a suitable green emitting phosphor for generation of WLEDs [49].

5.3. Ba3La((PO4)3):Ce3+, Mn2+ phosphors

Weigang Liu et al [50] reported, multi-color emitted Ba3La((PO4)3):Ce3+,Mn2+ phosphors, which are synthesized by gel-combustion and subsequent calcination in an Ar/H2 atmosphere at 1250°c. The PL
emission spectra of Ba$_3$La$_{1-x}$ (PO$_4$)$_3$: $xCe^{3+}$ ($x = 0.01-0.10$) phosphors shows broad and asymmetric emission band in the ~300-550 nm under 278nm excitation, which are ascribed $4f_0^5d_1 \rightarrow 4f_1^5d_0$ transition of Ce$^{3+}$ ions. Highest emission intensity observed at $x = 0.05$, then PL emission intensity decreased due to concentration quenching effect. After co-doping of Mn$^{2+}$ ions, PL emission spectra shows two broad emission band one is observed at near ultraviolet band (~370 nm) and red band (~591-605 nm) under 278nm excitation, which are ascribed f → d transition of Ce$^{3+}$ ions and $4T_1 \rightarrow 6A_1$ transition of Mn$^{2+}$ ions. The maximum energy transfer efficiency observed 43.2% at y = 0.30. The synthesized Ba$_3$La (PO$_4$)$_3$:0.03Ce$^{3+}$, yMn$^{2+}$ phosphor shows good thermal stability. This emission spectra depicts when temperature increased emission wavelength of Ce$^{3+}$ does not shifted but emission wavelength of Mn$^{2+}$ shifted [50].

5.4. Ca$_3$Mg$_8$(PO$_4$)$_4$ (CMP): Ce$^{3+}$, Dy$^{3+}$, Li$^+$ phosphors

For generation of white light Govind B. Nair et al. [51] reported Ce$^{3+}$ to Dy$^{3+}$ ions doped CMP phosphor. In the present study, authors are synthesized Ce$^{3+}$ to Dy$^{3+}$ ions doped CMP phosphor by urea assisted combustion method. The synthesized phosphor was successfully characterized by XRD, Rietveld refinement, SEM, FT-IR, DRS, and PL techniques. FT-IR spectra revealed present vibrations on the synthesized material. Band gap of prepared phosphor were also calculated by DRS technique, un-doped Ca$_3$Mg$_8$(PO$_4$)$_4$ phosphor shows value of band gap to be 3.23eV. The PL properties of synthesized phosphors are also analysed. The Ce$^{3+}$ doped CMP:Li$^+$ phosphor revealed broad emission spectra in the range from 325nm to 425nm under 313nm excitation, the emission band ascribed due to f → d transition of Ce$^{3+}$ ions. PL emission spectra for Dy$^{3+}$ doped CMP phosphor exhibits blue (482nm) and yellow (572nm) emission under 351nm excitation. Further, energy transfer mechanism from Ce$^{3+}$ to Dy$^{3+}$ ions were also studied. The CIE coordinate were also found in the white region with excellent color purity. All these results revealed synthesized materials have potential application for UV excited WLEDs [51].

Some of the reported phosphors are shortly described in table 2, which are also used for White Light Emitting Diodes (WLEDs).

Table 2. List of recently reported phosphors

| Ref. No | Name of synthesized phosphor | Synthesis Method | Remarks |
|---------|-------------------------------|------------------|---------|
| [52]    | Ca$_{14}$Al$_{10}$Zn$_6$O$_{35}$:Mn$^{4+}$ phosphor | Sol gel method | Cubic crystal structure with F23 space group, Band gap energy $E_g=$2.092eV, Strong red emission under 450nm excitation |
| [53]    | CsGd(MoO$_4$)$_2$: Dy$^{3+}$/Eu$^{3+}$ phosphor | Solid State Method | Blue, yellow, Red emission under 395nm excitation, particle size 10nm – 200nm by HRTEM and 1µm to 10µm by SEM, ET process from Dy$^{3+}$$\rightarrow$Eu$^{3+}$, CIE coordinate yellow and red region, expected to be a promising material for warm white emitting LEDs |
| [54]    | BaZrGe$_2$O$_{29}$:Eu$^{3+}$phosphor | Solid State Method | Calculated band gap 3.982eV, red emission at 612nm under 394nm excitation, CIE color coordinate (0.395, 0.343), CCT = 3234 K, and $R_a = 84.2$ |
| [55]    | La$_5$Ga$_6$Ge$_{14}$: Eu$^{3+}$, Bi$^{3+}$ phosphors | Solid State Method | Particle size around 1–2 µm, band gap $E_g=$ 5.23eV, Bright red emission, |
| Reference | Material | Co-doping | Reaction Method | Crystal Structure | Emission Band | Applications |
|-----------|----------|-----------|----------------|------------------|--------------|--------------|
| [56]      | Ce\(^{3+}\) → Tb\(^{3+}\) → Eu\(^{3+}\) triactivated Ca\(_3\)Gd(GaO)\(_3\)(BO\(_3\))\(_4\) | - | High-temp. solid-state reaction technique | Hexagonal |  | CIE chromaticity coordinates close to red-emitting phosphor Y\(_2\)O\(_2\)S: Eu\(^{3+}\). |
| [57]      | K\(_2\)MgGeO\(_4\):Bi\(^{3+}\) | - | solid-state reaction technique | Hexagonal | 290-450 nm | Intense and broad emission (440 to 800 nm) under Near UV. Quantum efficiency = 66.6\%, Ra = 93.8 |
| [58]      | NaBaY(BO\(_3\))\(_2\):Ce\(^{3+}\), Tb\(^{3+}\) | - | solid state reaction under the reductive atmosphere | Hexagonal |  | Energy transfer from Ce\(^{3+}\) to Tb\(^{3+}\), Lifetime is 65-40ns. Under 350 mA current function, CRI=80.16, CCT=5511, excellent degradation property and favourable color stability |
| [59]      | NaAlSiO\(_4\): Ce\(^{3+}\), Mn\(^{2+}\) phosphor | - | Solid-state reaction technique | Hexagonal | 290-350 nm | Hexagonal crystal structure with space group P63, under 430nm emission, NaAlSiO\(_4\): Ce\(^{3+}\) shows two emission peaks around at 290 and 350 nm. Energy transfer process from Ce\(^{3+}\) → Mn\(^{2+}\), \(R_c\) is 13.53Å, CIE coordinates (0.371, 0.333) very close to standard White Light Coordinate, |
| [60]      | KBaScSi\(_2\)O\(_7\):Eu\(^{2+}\) phosphor | - | Solid-state reaction technique | Hexagonal | 290-450 nm | Strong and Broad excitation band (290-450nm) and emission is also broad and strong in cyan blue region, QE = up to 91.3\%, thermal stability = 90\%, CRI = 88.6, CCT = 3770K, and LE = 21 lm/W, under 365nm excitation, potential candidate for generation of WLEDs |
| [61]      | Ce\(^{3+}\)/Dy\(^{3+}\) co-doped Sr\(_3\)SiO\(_5\) | - | Citrate assisted sol-gel method | Tetragonal | 470 nm | Tetragonal phase with P4/ncc space group, Particle size around 1µm energy transfer between the Ce\(^{3+}\) and Dy\(^{3+}\) ions, Lifetime is 40ns-30ns, CIE Chromaticity coordinate found to be Bluish White region, can be used UV excitable WLEDs |
| [62]      | Sr\(_3\)GdNa(PO\(_4\))\(_3\):F:Eu\(^{2+}\),Mn\(^{2+}\) | - | Solid state reaction method | Hexagonal | 470 nm | Sr\(_3\)GdNa(PO\(_4\))\(_3\):F:Eu\(^{2+}\),Mn\(^{2+}\) phosphor have hexagonal crystal structure with \(I\bar{P}\) (no. 147) space group, PL emission spectra shows broad blue emission centered at 470 nm, by co-doping Mn\(^{2+}\) in the host material PL emission shifted blue to yellow region under 390nm excitation wavelength, Sr\(_3\)GdNa(PO\(_4\))\(_3\):F:Eu\(^{2+}\),Mn\(^{2+}\) phosphor might have potential application in WLEDs.
[63] $\text{Ba}_5\text{Zn}_4\text{Y}_8\text{O}_{21}:\text{Eu}^{3+}$ nanophosphor Combustion Method Phase formation and structural study by Rietveld refinement, Judd-Ofelt intensity parameters, suitable candidate for white LEDs under near UV excitation.

[64] Trivalent lanthanide (Eu$^{3+}$, Tb$^{3+}$ and Dy$^{3+}$) ions activated $\text{BaGd}_2\text{O}_4$ (BG) phosphors Facile Pechini-type sol–gel process The synthesized $\text{BaGd}_2\text{O}_4$ (BG) phosphors have orthorhombic phase, TEM image shows needle type nanorods structure, synthesized phosphor shows excellent with PL properties after doping Eu$^{3+}$, Tb$^{3+}$ and Dy$^{3+}$ ions. Cathodoluminescence properties also investigated, which is shows similar results as PL. CIE Chromaticity coordinate shows red, green, and white emissions as for individual ion doped $\text{BaGd}_2\text{O}_4$ (BG) phosphors, after co-doping Eu$^{3+}$/Tb$^{3+}$ or Eu$^{3+}$/Dy$^{3+}$ ions show colour tunable emission.

[65] $\text{ZnGa}_2\text{O}_4:\text{Tb}^{3+}$ Wet Chemical Method Blue-green tunable zinc gallate phosphor was successfully prepared via Wet chemical route. TEM image shows quantum dots, PL properties shows blue to green colour emission, which is confirmed by CIE Chromaticity diagram, Critical distance found to be 18Å. All these results show colour tunable properties for display applications.

[66] $\text{CaCO}_3$: Eu$^{3+}$, Tb$^{3+}$ microwave co-precipitation method The PL emission spectra of $\text{CaCO}_3$: Eu$^{3+}$, Tb$^{3+}$ phosphor shows red and green color emission, respectively. After co-doping of Tb$^{3+}$ ions emission color could be gradually tuned, which is confirmed by CIE chromaticity coordinate.

Recently reported some more color tunable phosphors:

- LuVO$_3$:Bi$^{3+}$,Eu$^{3+}$ [67], Ca$_{10}$Li(PO$_4$)$_7$:Ce$^{3+}$,Mn$^{2+}$ [68], ScPO$_4$:Tb$^{3+}$,Eu$^{3+}$ [69], NaLa(MoO$_4$)$_2$:Sm$^{3+}$/Dy$^{3+}$ [70], SrLaMgTaO$_6$:Dy$^{3+}$/Eu$^{3+}$ [71], MgYSi$_2$O$_5$:Ce$^{3+}$,Tb$^{3+}$ [72], Na$_6$Ca$_3$Si$_6$O$_{18}$:Ce$^{3+}$,Tb$^{3+}$ [73], Sr$_2$LiAlO$_4$:Eu$^{2+}$/Ce$^{3+}$ [74], BaY$_2$Si$_3$O$_{10}$:Tm$^{3+}$,Dy$^{3+}$ [75], C$_{30}$ZnK(PO$_4$)$_7$:Ce$^{3+}$/Tb$^{3+}$/Mn$^{2+}$ [76], $\text{Ba}_5\text{Lu}(\text{PO}_4)_3$:Tb$^{3+}$, Mn$^{2+}$ [77], Ba$_3$La$_2$(BO$_3$)$_4$: Ce$^{3+}$/Dy$^{3+}$ [78], NaCa$_2$GeO$_4$:F:Mn$^{2+}$, Yb$^{3+}$ [79], Li$_3$Ba$_2$Y$_3$(WO$_4$)$_6$:Tb$^{3+}$,Eu$^{3+}$ [80], Ca$_{20}$Al$_{36}$Mg$_3$Si$_3$O$_{68}$: Eu$^{2+}$,Eu$^{3+}$ [81], LaBWO$_6$:Tb$^{3+}$,Eu$^{3+}$ [82].

6. Challenges and Future Advances

In the last few years, phosphor converted White LEDs have tremendous advantages such as low cost, ecofriendly, higher efficiency, low energy consumption, and long operating lifetime etc. At present efficiency of WLEDs is 25-50%, but is expected that it efficiency of WLEDs increased up to 90%
near future [83]. It is well known that YAG:Ce³⁺ is used as a commercial phosphor but it has some drawbacks, we are trying to develop new phosphor materials for WLEDs and improvement in LED luminous and further cost reductions. The range of uses will surely continue to increase, not only for white LEDs but also for LEDs of other colours, within the context of saving energy and the need for thinner designs. Our research and industrialist are trying to develop new technique for generation of WLEDs. New technique of wafer composed of gallium-nitride-on-silicon is being used to produce White LEDs. This has condensed the price of product. We are trying to change all GaN LEDs will be made with GaN-on-Si. As a future possibility of white LEDs, visible light is a district of the electromagnetic range which a new age of lighting gadgets can use for establishing connection. The quick selection of indoor and outdoor lighting device will set the strong stage for conveying information, visible light connection (VLC) or Li-Fi [84, 85].

7. Summary

Solid-state lighting (SSL) is emerging as an alternative to the pre-existing lighting technologies and is highly desirable. This type of lighting involves (LEDs) semiconductors light emitting diodes, OLEDs (organic light emitting diodes) and PLEDs (polymer organic light emitting diodes) as the source of illumination. As a new solid state light source, LEDs are considered as the next generation lighting device because of their long-life time, energy saving, reliability, safety and environment friendliness characteristics. To fulfil the need of light emitting diodes applications, silicate phosphors have been developed and designed from the past years. Recently, a number of phosphor materials are prepared for WLEDs applications. In this review, we systematically discussed color tunable phosphor materials, which shows excellent results for production of WLEDs. By co-doping of rare earth ions as a sensitizers, we can not only improve efficiency of phosphor materials, we can tune the color of emission in the wide range by using energy transfer. For Color tuning and energy transfer mechanism some sensitizers and activators couples are used such as Ce³⁺-Eu²⁺, Ce³⁺-Tb³⁺, Ce³⁺-Eu³⁺, Ce³⁺/Eu²⁺–Tb³⁺, Ce³⁺–Dy³⁺, Ce⁴⁺/Eu²⁺–Tb³⁺–Sm³⁺, Ce³⁺/Eu²⁺–Tb³⁺–Eu³⁺, Ce³⁺–Eu³⁺, and Ce⁴⁺/Eu²⁺/Tb³⁺–Mn²⁺ [6]. Numerous improvements in the device construction pooled with -innovative progress ion in technology have enhanced the optical and electrical characteristics of white LEDs. The quest for novel solutions for strong state lighting and WLEDs will invigorate future examinations in the field of colour tunable phosphor materials.

8. References

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