Gurukul International Multidisciplinary Research Journal (GIMRJ)*with*
International Impact Factor 8.357
Peer Reviewed Journal
https://doi.org/10.69758/GIMRJ/2505I5VXIIIP0015

e-ISSN No. 2394-8426
Monthly Issue
MAY-2025
Issue–V, Volume–XIII

Luminescence Properties of Europium(III) Ion Doped Host Matrices

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Abstract:- Europium ion-doped phosphors have garnered significant attention due to their potential applications in solid-state lighting, dosimetry,optoelectronics and display technologies. These phosphors are known for their ability to emit light in the red to orange spectrum, which is crucial for creating white light in LEDs. Eu^{+3} ion shows photoluminescence from $^5D_0$ excited state to ^7F_J term which is represented as $^5D_0 \rightarrow {}^7F_J$. The luminescence of Eu3+ doped phosphors is influenced by factors such as europium concentration and site occupancy, which can affect the emission colour and efficiency. The structural and luminescent properties of Eu-doped phosphors can be optimized by co-doping with other elements.Moreover, the charge compensation effect of alkali metal ions in some phosphors leads to enhanced luminescence and thermal stability, indicating their potential for white-light-emitting diode applications. Our objective is to explore the development of Eu (III) doped phosphors for their colour tunability and thermal stability. Building on recent advancements, we highlight the research prospects of this phosphors.

Keywords :Luminescence, Rare earth, Europium, Phosphors

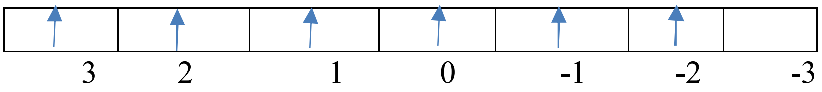
Introduction:

In daily life, there is enormous use of solidstate light because of their high energy efficiency, low energy consumption, and environmental friendliness[1–10]. Generally, the different colours of LEDs are due to the doping of rare earth elements in phosphors. When rare earth ions specially Ln^{3+} introduced into the host material it gives the characteristic colours which are arise due to spectroscopically allowed electronic transitions of 4f electrons from ground state to excited state, giving maximum absorption in the visible wavelength range (700–400 nm)[11].The rare earth activated phosphors have attracted much attention due to their high luminescence efficiency, colour purity, long emission lifetimes values [12]. In present study, we focus on Eu^{3+} doped phosphors which gives red emission on irradiation with UV light having various application in solid state lighting.Eu ion doped phosphors such as Phosphates, Aluminates, Tungstate, Oxides and Borates are synthesized using various methods, including high-temperature solid-state reactions, combustion method, Hydrothermal method and sol-gel techniques. For instance, Eu^{3+} doped $Ca_9Gd(PO_4)_7$, $LaBWO_6$, $BaLaO_4$ phosphors are synthesized by a high-temperature solid-state reaction, resulting in a Rhombohedral and Orthorhombic crystal structure. Similarly, Eu^{3+} doped $ZnWO_4$ phosphors are prepared using a hydrothermal method, achieving a pure monoclinic phase [13]. The effectEu ion doping on structural integrity and phase purity of these phosphors are confirmed using X-ray diffraction (XRD) and other spectroscopic techniques[14][15].

Eu ion doped phosphors are characterized by orange red photoluminescence (PL) spectra.These spectra reveal transitions such as $^5D_0 \rightarrow {}^7F_J$ (J = 0, 1, 2, 3, 4).For example, in$KBaScSi_3O_9:Eu^{3+}$ phosphors the magnetic dipole transition ($^5D_0 \rightarrow {}^7F_1$) predominates the red emission due to $^5D_0 \rightarrow {}^7F_2$[16]. The luminescence of these phosphors can be further enhanced by co-doping with other metal ions by energy transfer mechanism. Tunable luminescence is achieved by manipulating the valence states of Eu ions; in Ba_2InTaO_6 phosphors, both Eu^{2+} and Eu^{3+} ions contribute to a broad emission spectrum, enabling color tuning from greenish-yellow to red[17]. This tunability is advantageous for applications requiring multi-color emissions, such as LEDs and decorative lighting.Therefore Eu ion doped phosphors are in focus for current researchers for their various application in solid-state lighting, anti-counterfeiting, Biomedical and optoelectronics [17]–[26]. Their ability to emit a wide range of colours and thermal stability makes them valuable in various technological fields.

1. Luminescence mechanism in Eu^{3+}

Europium is f block element having atomic number 63. the number of electrons present in Eu^{3+} ion is 60 thus electronic configuration of Eu^{3+} can be represented as [Xe] $4f^6$ or $4f^6$ in short.



The energy level of these ions is well explained by the term symbol derived by R-S coupling denoted as $^{2S+1}$ L. Where L is the total angular momentum quantum number, S is the total spin quantum number and 2S+1 is the spin multiplicity. For Eu^{3+} ion, highest spin multiplicity term is septate (7) which corresponds to six unpaired electrons. S = (1/2+1/2+1/2+1/2+1/2+1/2) =3 or spin multiplicity 2S+1 = 7 for which value of L=3 or F therefore, term symbol represented as ^7F. The term symbol does not account for the true complexity of the energy levels of the lanthanoid ions which arise from interaction between spin and orbital momentum called spin orbit coupling for this J value is needed called total angular momentum quantum number. The possible values of J are |L+S| up to |L-S| . For ^7F term of Eu^{3+} possible values of J are 6,5,4,3,2,1,0. So, each free ion labelled as $^{2S+1}L_J$. For Eu^{+3} ion J=7 the term described by Russell Saunders's coupling are represented as below.

Ion	Term	Energy level
$Eu^{3+}(f^6)$	^7F	$^7F_0{}^7F_1{}^7F_2{}^7F_3{}^7F_4{}^7F_5{}^7F_6$

For Europium ion only ^7F and ^5D term are allowed for transition. According to Hund's rule, $^7F_0$ is the ground state of $4f^6$ electronic configuration and excited state is ^5D. Therefore Eu^{+3} ion shows photoluminescence from $^5D_0$ excited state to ^7F_J term which is represented as $^5D_0 \rightarrow {}^7F_J$ as shown in Figure 1.The various wavelength range of transition from $^5D_0$ to ^7F_J term are shown in

Table-1[28]. The intensity peak are observed by photoluminescence spectra of sample with various transition.

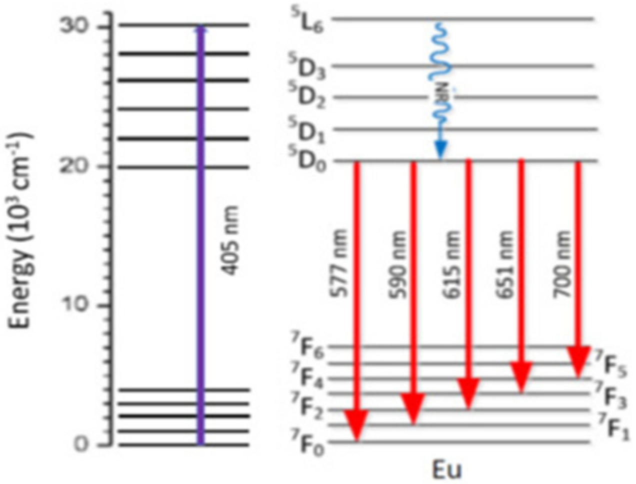


Fig 1 :Energy level diagram of Eu +3 ion [27].

Transition	Wavelength range
$^5D_0 \rightarrow {}^7F_0$	570–585
$^5D_0 \rightarrow {}^7F_1$	585–600
$^5D_0 \rightarrow {}^7F_2$	610–630
$^5D_0 \rightarrow {}^7F_3$	640–660
$^5D_0 \rightarrow {}^7F_4$	680–710
$^5D_0 \rightarrow {}^7F_5$	740–770
$^5D_0 \rightarrow {}^7F_6$	810–840

Table 1: Wavelength range of transition from $^5D_0$ to ^7F_J term

2. Luminescence Mechanism of Europium Ion in Different Host Phosphors

The luminescence of europium ions in various host phosphors is a subject of significant interest due to its applications in lighting and display technologies. Europium can exist in two oxidation states, Eu^{2+} and Eu^{3+}, each exhibiting distinct luminescent properties. The host matrix and synthesis conditions play crucial roles in determining the luminescence characteristics of europium-doped phosphors.

2.1 Coexistence and Conversion of Eu^{2+} and Eu^{3+}

In several host matrices, both Eu^{2+} and Eu^{3+} ions coexist, contributing to the luminescence properties. For instance, in $SrBPO_5$ phosphors, the presence of gadolinium enhances the luminescence of Eu^{3+} while quenching Eu^{2+} luminescence through valence conversion and energy transfer mechanisms[29]. Similarly, in $Ba(Eu)MgSiO_4$ phosphors, Ba^{2+} substituted by Eu^{3+}creates vacancies to maintain charge neutrality. These vacancies help facilitate the reduction of Eu^{3+} to Eu^{2+}, enhancing the luminescence of $Eu^{2+}$with broad-band green emission[30].

2.2 Influence of Host Matrix and Synthesis Conditions

The host matrix significantly affects the luminescence properties of Eu^{3+}as shown in Table 2. The synthesis atmosphere and co-doping with other elements can also influence the valence state and luminescence.For example in $BaAl_2O_4$:Eu phosphors, the sintering temperature affects longer persistent luminescence due to efficient charge trapping and release [31].

2.3 Site Occupancy and Substitution Effects

The site occupancy of europium ions within the host lattice play crucial role in determining luminescence properties. In $Sr_{0.8}Li_{0.2}Ti_{0.8}Nb_{0.2}O_3$:$Eu^{3+}$ phosphors, the substitution of Eu^{3+} ions at different cationic sites affects the intensity and colour purity of the emitted lightdue to changes in the local crystal field environment and energy transfer dynamics[32]. Similarly, in $Sr_{8-x}La_{2+x}(PO_4)_{6-x}(SiO4)_xO_2$ phosphors, the substitution of cations allows for full-colour tuning by altering the luminescence behaviour of Eu^{2+} ions[33].

3. Energy transfer process

Even though Europium ion show photoluminescence property towards red emission but the intensity of emitted light is weak which is due to the weak light absorption. To increase the intensity of Eu^{3+} the energy transfer process is observed which is explained by “Antenna effect” i.e. energy transfer effectproposed by Crosby and Whan. Table 1 shows the Synthesis and photometric parameters of Eu^{3+} doped phosphors.

Phosphors	Crystal Structure	Synthesis method	Sintering temperature	λ_{ex} (nm)	λ_{em} (nm)	Emitted colour	Ref.
$LiLaMgWO_6$:Eu^{3+}	Monoclinic	Solid state method	900^0C for 6h	393	615	Strong red	[34]
$BaGd_2O_4$:Bi^3,Eu^{3+}	Orthorhombic	Solid state method	1300 °C for 10 h	337	611	tuneable emission including blue, amaranth and orange red emission	[35]
$Ca_3La(GaO)_3(BO_3)_4$:Eu_{3+}		Solid state method	1000 °C for 4 h	397	621	red	[36]
$Ca_9Gd(PO_4)_7$:Eu^{3+}	Rhombohedral	Solid state method	1250 °C for 6 h	395	615	Intense red	[37]
$ZnWO_4$:$Eu^{3+,}Dy^{3+}$	Monoclinic	hydrothermal method	700 °C for 1 h	295	615	white light emission	[38]
YVO_4:Eu^{3+}	Wakefieldite-(Y)- tetragonal	soft template assisted microwave synthesis		325	619	Strong red	[39]

$LaBWO_6$:Tb^{3+}, Eu^{3+}	Orthorhombic	Solid state method	1000 °C for 4 h.	380	616	tunability from green to yellow, then to red.	[40]
$LiLaMo_2O_8$:Eu^{3+} R (R = Bi^{3+} and Sm^{3+})	orthorhombic	Solid state method	700 °C for 6 h	395	616	red	[41]
Ca_2BO_3Cl: Eu^{2+}	monoclinic	Solid state method	1173 K for 3 h	395	570	Bright yellow	[42]
Ca_3-xAl_4ZnO_{10}: xEu^{3+}	orthorhombic	Solid state method	1100 to 1300 ^{0}C for 5 h	391	615	red	[43]
BYSO:Bi^{3+},Eu^{3+}	trigonal	Solid state method	1150 °C for 4 h	337	611	white	[44]
SrY2O4:Bi^{3+},Eu^{3+}	orthogonal	Solid state method	1400 C for 5 h	330	611	red	[45]
$Ba_9Lu_2Si_6O_{24}$:Bi^{3+}, Eu^{3+}	rhombohedral	Solid state method	1150 °C for 4 h	335	611	blue-green	[46]
$ZnLa_{0.75}Tb_{0.20}Eu_{0.05}B_5O_{10}$		Solid state method	930 °C for 15 h.	364	611	Color-tunable emissions from green to yellow and to orange were realized in these phosphors.	[47]
$BaLaAlO_4$:Eu^{3+}	orthorhombic	Solid state method	1000 °C for 1 h	396	626	Intense red	[48]

Table 2 : Synthesis and photometric parameters of Eu^{3+} doped phosphors.

3.1 Energy transfer process from organic ligand complex.

When UV radiation incident on organic ligand of the complex, ligand absorbs UV radiations and excited to first singlet state ($S_0 \rightarrow S_1$). To reach ground state from excited state, two types of conversion exhibits which is explained by Jablonsky diagram.

1. As ligand come back to the lowest excited state from intermediate excited state the process is called internal conversion. After internal conversion,ligand come back to the ground state and the process is called fluorescence[49].
2. Ligand can undergo nonradiative transition (intersystem crossing) from first singlet excited state to triplet state. Ligand come back to the ground state from triplet state by radiative transition called phosphorescence.

The triplet state is an energy transferlevel, ligand can transfer the energy from triplet state to 4fenergy level of lanthanide and by transferring energy they come back to the ground state.

The condition for energy transfer from ligand to lanthanide are as follows.

1. Triplet state of ligand should located at 2000 to 3000 cm^{-1} above the emitting level of lanthanides.
2. If the triplet state of ligand is lower the emitting level of lanthanides, no energy transfer process take place.

In $Eu(DBM)_3L\text{-}pCH_3$ complex there is energy transfer process is observed fromdibenzoyl methane (DBM) ligand to Eu^{3+} ion.Devi at al confirmed the energy gap between triplet state of ligand to the excited state of Eu3+ ion ($^5D_0$)] should be in the range 2500−4000 cm^{-1} for an efficient energy transfer [50]. This energy transfer process also observed in inorganic complex.

3.2 Energy transfer process in d- block element

This type of sensitization also observed in Lanthanide-d block elements. As d-d absorption band of d-block elements having lower energy than $^5D_0$ excited state of Eu^{3+}, it quenches the luminescence of Eu^{3+}. As d-d transition depends upon ligand environment it acts as sensitizer as well as quencher[51].

3.3 Energy transfer process in p block elements

Some p-block elements also sensitization towards Eu^{3+} photoluminescence. Bi^{3+} itself is an efficient activator in luminescent material. The co-doping of Bi^{3+} containing phosphor with Eu^{3+} there is energy transfer from Bi^{3+} to $Eu^{3+}$is observed which intense the red photoluminescence of Eu^{3+}.The examples of Eu^{3+}doped phosphors co-doped with $Bi^{3+}$are shown in table 2.Pb^{2+} and Sb^{3+} also act as a sensitizer of Eu^{+3} luminescence.

3.4Energy transfer process in f block elements

Sensitization process is also observed in trivalent lanthanide ion co-doped with Eu doped phosphors. As lanthanide ion has energy level which is resonant with $^5D_0$ state of lanthanide ion, Eu^{3+} ion easily transfer its energy to other lanthanide ion and luminescence of Eu^{3+} ion will be quenched. Dy^{3+}, Nd^{3+}, Yb^{3+}, Er^{3+}, Tm^{3+} partially quench the Eu^{3+} luminescence. The energy transfer process is observed in Sm^{3+} and Tb^{3+} to Europium which give enhancement of luminescent intensity of Eu^{3+}. $^5D_4$ energy level of Tb^{3+} is well above the $^5D_0$ level of Eu^{3+} so that energy transfer process is observed from Tb^{3+} to Eu^{3+} [15]. The energy transfer not only increases the intensity of Eu^{3+}but also it gives the extra peaks in the excitation spectrum.

4. Applications of Europium Ion-Doped Phosphors

Europium ion-doped phosphors have a wide range of applications, particularly in the field of lighting and display technologies. These phosphors are known for their ability to emit light in various colors, primarily in the red and orange spectrum, which makes them suitable for use in white light-emitting diodes (WLEDs) and other lighting applications.

4.1 Solid-State Lighting (SSL):

Europium-doped phosphors are extensively used in solid-state lighting due to their efficient luminescent properties. For instance, europium-activated borophosphate phosphors exhibit reddish-orange emission, making them suitable for warm white LEDs when combined with near-ultraviolet LED chips[52].Similarly, europium-doped bismuth silicate phosphors can be excited by near-ultraviolet light, providing a high quantum efficiency, which is advantageous for solid-state lighting applications[53].

4.2 White Light-Emitting Diodes (WLEDs):

$BaLaGaO_4:Eu^{3+}$ phosphors are used in WLEDs, where they contribute to a high colour rendering index and colour purity, essential for high-quality lighting solutions. These phosphors also have applications in plant growth lighting due to their deep-red emission[54]. Bin Yu at al synthesized Ba_2InTaO_6 phosphors in which combination of Eu^{3+} and Eu^{2+} ions in allows for tunable luminescence, which is beneficial for creating multi-colour lighting solutions in WLEDs.

4.3 Thermal Stability and Efficiency:

The thermal stability of europium-doped phosphors is crucial for their performance in high-temperature environments. For example, the co-doping of Mn^{2+} with Eu^{3+} in $ZnMoO_4$ phosphors enhances thermal stability and reduces electron lifetime, making them suitable for high-power SSL applications[55].

4.4 Specialized Applications:

There are lots of specialized application of Europium ion doped phosphors. For example Europium-doped $NaGd(WO_4)_2$ nanophosphors have been developed for pH sensing applications, showcasing the versatility of europium-doped materials beyond traditional lighting uses[56].In the field of UV sensing, europium-doped $LaPO_4$ phosphors demonstrate high quantum efficiency, making them suitable for UV detection applications[26]

Conclusion

Europium ion-doped phosphors exhibit significant potential for various applications due to their tunable luminescent properties and stability. The choice of host material and doping concentration are crucial for optimizing their performance in lighting and display technologies. These materials continue to be a focus of research for enhancing the efficiency and color quality of light-emitting devices.

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