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Research Paper

Efficient White Light Generation in $ZnMoO_4$ Phosphors Co-Doped with Eu^{3+} and Dy^{3+}

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Abstract

Eu^{3+}–Dy^{3+} co-doped $ZnMoO_4$ phosphors were synthesized using a high-temperature solid-state reaction method. The emission spectra were analyzed to determine the Commission Internationale de l'Éclairage (CIE) chromaticity coordinates (x, y), enabling precise evaluation of the emitted color. The results indicate that the luminescence color of the prepared phosphors is strongly dependent on the relative concentrations of Eu^{3+} and Dy^{3+} ions, demonstrating effective tunability of emission characteristics.

Keywords: Phosphors, Color-coordinates, Powder diffraction, Luminescence,

1. Introduction

White light–emitting diodes (W-LEDs) are widely regarded as next-generation light sources for both display technologies and general illumination. They offer several advantages over conventional lighting systems such as incandescent, fluorescent, and high-intensity discharge lamps, including longer operational lifetime, compact size, improved environmental compatibility, and resistance to moisture.A commercially successful approach has been developed by Nichia Corporation, which utilizes an indium gallium nitride (InGaN) blue LED ($\lambda_{em} \approx 470$ nm) combined with a yellow-emitting Y3Al5O12:Ce3+ phosphor. Another effective strategy for white light generation involves the integration of a UV LED chip with suitable white light–emitting phosphor materials, enabling broader spectral coverage and improved color rendering properties. [1]. The major problem of phosphors in the generation of white light is the lack of a red light component and the resultant low colour-rendering index (CRI). Commonly used Eu^{3+} and Ce^{3+} activator ions can efficiently absorb nUV radiation and emit the blue to

yellow wide band emissions. To achieve proper color dispersion in the visible region, phosphors doped with certain co-activators are very much essential [2]. In this approach many phosphors activated with Eu^{2+}-Mn^{2+} and Ce^{3+}-Mn^{2+} ions were investigated [3]. Single-component white light–emitting phosphors are typically realized by co-doping a sensitizer and an activator into a common host lattice. In such systems, energy transfer from the sensitizer to the activator plays a crucial role, and resonance-type multipolar interactions have been widely reported in various inorganic hosts, particularly for co-dopant pairs such as Eu^{2+}/Mn^{2+}, Ce^{3+}/Eu^{2+}, and Ce^{3+}/Mn^{2+}. In general, an individual rare-earth (RE) ion exhibits characteristic emission that spans only a limited region of the visible spectrum. Therefore, achieving white light emission requires the combination of emissions from multiple luminescent centers. One effective approach is the co-doping of suitable combinations of RE^{3+} ions within a single host matrix.Among the various combinations, Eu^{3+}–Dy^{3+} co-doping has attracted considerable attention in recent years due to its ability to generate complementary emissions that can be tuned toward white light, making such systems promising for solid-state lighting applications.

In this paper, a series of $ZnMoO_4$:: xEu_2O_3+yDy_2O_3 phosphors (x = 0.5, y = 0.5, 1.0 and 1.5 mol %) were synthesized by the solid-state reaction method and the luminescent characteristics under nUV excitation were examined. Also, the effect Dy^{3+} ions concentration on the luminescence intensities, energy transfer and chromatic properties has been discussed.

2. EXPERMENTAL

Different concentrations of Eu^{3+} - Dy^{3+} ions activated $ZnMoO_4$: xEu^{3+}, yDy^{3+} (x = 0.5, y = 0.5, 1.0 and 1.5 mol %) phosphors were prepared by the solid-state reaction method. The pre-weighted quantities of starting chemicals ZnO (quality A.R.), $NH_4H_2PO_4$ (99.99%), Eu_2O_3 (99.99%) and Dy_2O_3 (99.99%) were grinded in an agate mortar by adding a small amount of acetone to mix the materials homogeneously. The samples were fired at 200 and 400°C for 3h in each step with an intermediate grinding. Finally, the samples were sintered at 900°C for 3h in air using alumina crucible. After sintering, the samples were cooled to room temperature and grinded again for subsequent measurements.

3. Result and discussion

3.1. Photoluminescence studies

Figure 1 presents the excitation spectra of singly doped Eu^{3+} and Dy^{3+} phosphors, revealing a significant overlap of excitation bands in the near-UV region. Notably, the 365 nm excitation wavelength is capable of efficiently exciting both Eu^{3+} ions and the co-doped Eu^{3+}–Dy^{3+} system.

Figure 2 illustrates the emission spectra of $ZnMoO_4$ phosphors containing 0.5 mol% Eu^{3+}, 0.5 mol% Dy^{3+} (singly doped), and the co-doped Eu^{3+}–Dy^{3+} composition under 365 nm excitation. It is evident that the emission intensity of Eu^{3+} ions in the co-doped ($0.5Eu^{3+}$–$0.5Dy^{3+}$) phosphor is significantly enhanced compared to that of the singly doped 0.5 mol% Eu^{3+} sample. In contrast, the emission intensity of Dy^{3+} ions in the co-doped phosphor is markedly reduced relative to the singly doped Dy^{3+} sample. This behavior suggests the occurrence of energy transfer from Dy^{3+} to Eu^{3+} ions in the co-doped system.

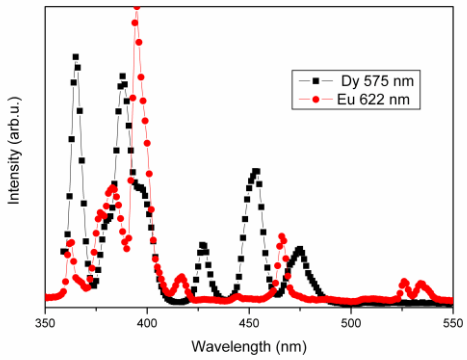

Fig.1.Excitation spectra of Dy^{3+} monitored at 575 nm and Eu^{3+} monitored at 622 nm.

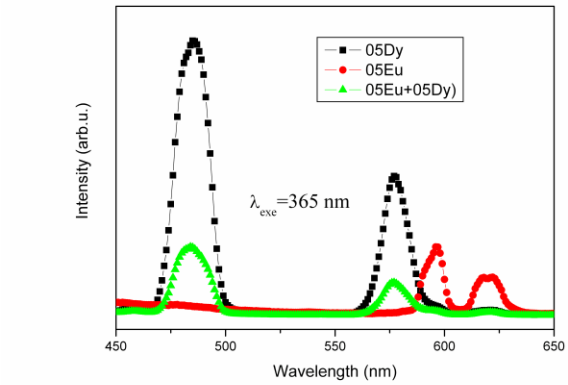

Fig.2.Emission spectra of singly doped Dy^{3+} , Eu^{3+} ions and co-doped Eu^{3+}- Dy^{3+} ions in $ZnMoO_4$ phosphor

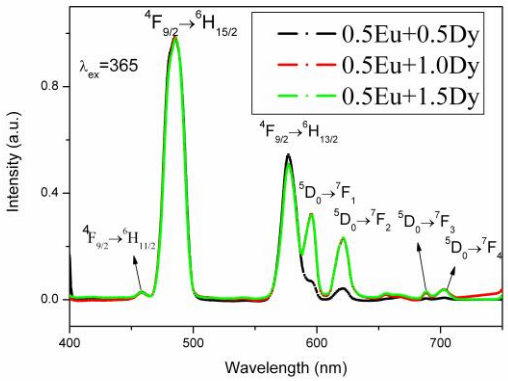

Fig.3.Emission spectra of the Eu^{3+}-Dy^{3+} co-doped $ZnMoO_4$ phosphor

Fig.3. shows the emission spectra of Eu^{3+}-Dy^{3+} co-doped $ZnMoO_4$ phosphors under 365 nm excitation. The spectra revealed a total of seven emission bands, out of which three bands correspond to $^4F_{9/2} \rightarrow {}^6H_{11/2}$ (458 nm, blue), $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (484 nm, greenish blue), $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (577 nm, yellow) transitions of Dy^{3+} ion and remaining four bands correspond to $^5D_0 \rightarrow {}^7F_1$ (596 nm, greenish yellow) , $^5D_0 \rightarrow {}^7F_2$ (620 nm, yellowish red), $^5D_0 \rightarrow {}^7F_3$ (688 nm, red), $^5D_0 \rightarrow {}^7F_4$ (703 nm, red) transitions of Eu^{3+} ions. These blue, green,

yellow and red emissions are observed in the Eu^{3+}-Dy^{3+} co-doped phosphors by the excitation of UV light. Thus the white light emission can be achieved by the optimization of Dy^{3+} and Eu^{3+} ions concentrations. Most strikingly, the intensities of blue and yellow emissions of Dy^{3+} ions decrease with the increase of Dy^{3+} ion contraction, indicating the energy transfer from Dy^{3+} ions to Eu^{3+} ions. The yellow to blue emission intensity ratios (I_{yellow}/I_{blue}) of 0.62 and 0.28 corresponding to $ZnMoO_4$: $0.5Eu^{3+}$- $0.5Dy^{3+}$ and $ZnMoO_4$: $0.5Eu^{3+}$-$1.5Dy^{3+}$ phosphors respectively conforms the energy transfer from Dy^{3+} to Eu^{3+} ions.

3.3. Decay analysis

The decay profiles of $^4F_{9/2}$ level of single doped $ZnMoO_4$: Dy^{3+} and co-doped $Na_3Gd(PO_4)_2$: Eu-Dy phosphors under 365 nm excitation for various Dy^{3+} concentration by monitoring the emission at 575 nm are shown in Fig.4. The experimental lifetimes (τ_{exp}) are determined from decay profiles according to the equation

$$I = I_0 \exp(-t/\tau) \qquad (2)$$

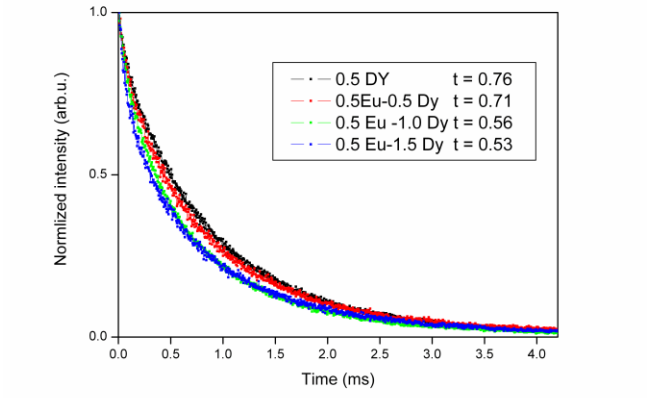

Fig.4. The decay curves of $^4F_{9/2}$ level in singly doped Dy^{3+} and Eu^{3+} - Dy^{3+} co-doped ions in $ZnMoO_4$ phosphors.

also are included in Fig.4. It is clearly seen that the lifetimes of $^4F_{9/2}$ emission level at 575 nm in the Eu^{3+}- Dy^{3+} co-doped $ZnMoO_4$phosphors are shorter than in the singly doped Dy^{3+} phosphors and decreases gradually with the increase of Dy^{3+} ions concentration. These results give additional support for the evidence of energy transfer from Dy^{3+} to Eu^{3+} ions.

3.4. Chromatic properties

Most lighting specifications refer to the colours in terms of 1931 CIE chromatic colour coordinates which are used recognize the three primary colours: red, green and blue by the human visual system [4-5]. In general, the color coordinates of any light source can be represented in the (x, y) coordinate system of the CIE chromaticity diagram. The chromaticity coordinates (x, y) of $ZnMoO_4:xEu^{3+}–yDy^{3+}$ phosphors, calculated using a color analysis program and listed in Table 1, are found to lie within the white light region, as illustrated in Fig. 5. Furthermore, the Commission Internationale de l'Éclairage (CIE) chromaticity coordinates of $ZnMoO_4:0.5Eu^{3+}–1.0Dy^{3+}$ phosphor under different excitation wavelengths are shown in Fig. 6. Upon excitation at 365 nm and 385 nm, the corresponding coordinates were determined to be (0.21, 0.34) and (0.26, 0.36), respectively. In contrast, excitation at longer wavelengths of 395 nm and 425 nm resulted in coordinates of (0.51, 0.32) and (0.59, 0.37), respectively.

These observations clearly indicate that the emission color of $ZnMoO_4:xEu^{3+}–yDy^{3+}$ phosphors can be effectively tuned by varying the excitation wavelength, demonstrating their strong potential for use in color-adjustable solid-state lighting applications.

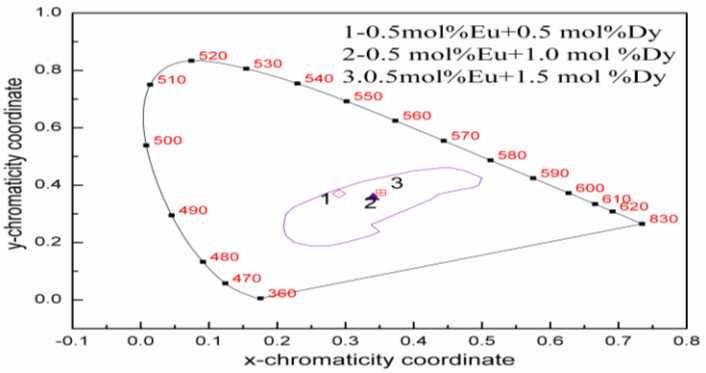

Fig.5. CIE chromaticity diagram for $ZnMoO_4$: xEu^{3+}- yDy^{3+} phosphors at different concentrations.

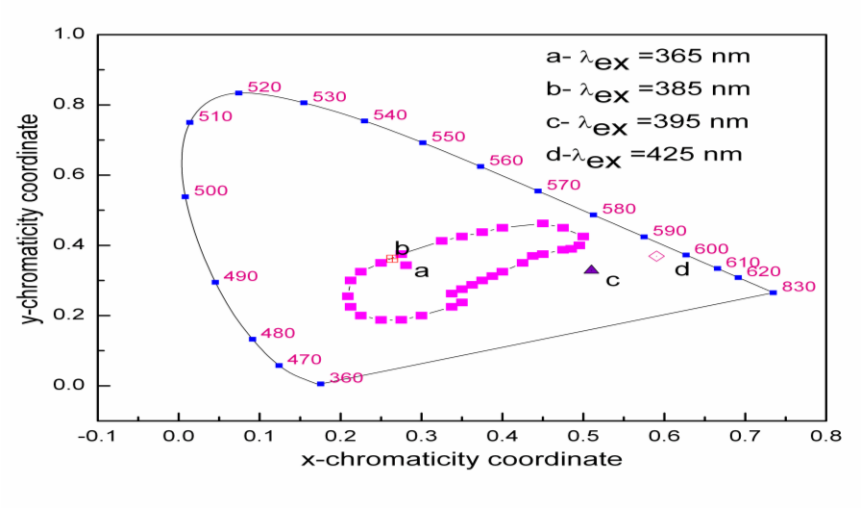

Fig.6. CIE chromaticity diagram for $ZnMoO_4$: xEu^{3+}- yDy^{3+} phosphors at different excitation wavelengths

Table 1 Chromaticity colour coordinates of $ZnMoO_4$:xEu^{3+}-yDy^{3+} phosphors at different concentrations (λ_{ex}=365 nm)

Concentration of Eu^{3+} (*x* %)	Concentration of Dy^{3+} (*y* %)	x-coordinate	y-coordinate
0.5	0.5	0.26	0.33
0.5	1.0	0.29	0.34
0.5	1.5	0.30	0.35

4.Conclusions

Eu^{3+}–Dy^{3+} co-doped $ZnMoO_4$ phosphors were successfully synthesized via a conventional solid-state reaction method. The excitation and emission studies confirmed that the phosphors can be efficiently excited under UV irradiation. The Commission Internationale de

l'Éclairage (CIE) chromaticity coordinates for the optimized composition were determined to be (0.32, 0.37), which are in close proximity to the ideal white light coordinates (0.33, 0.33) under 365 nm excitation.These results indicate that Eu^{3+}/Dy^{3+} co-doped $ZnMoO_4$ phosphors exhibit promising luminescent properties, making them potential candidates for application in solid-state lighting and multicolor three-dimensional display devices.

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