



# Luminescent properties of $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$ red phosphor for solid state lighting



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**Abstract:** The  $\text{Eu}^{3+}$  activated  $\text{Lu}_2\text{MoO}_6$  phosphors were synthesized by high-temperature solid-state reaction method. The X-ray diffraction (XRD), excitation spectra, emission spectra and decay lifetime of the phosphors were measured to characterize the structure and luminescent properties. The XRD results show that all the prepared phosphors can be assigned to the monoclinic structure. The experimental results indicate efficient absorption of near ultraviolet light from the  $\text{Mo}^{6+}-\text{O}^{2-}$  group followed by intensive emission in the visible spectral range. The optimal content of  $\text{Eu}^{3+}$  is 10% (mole fraction). The critical distance  $R_c$  and energy transfer mechanism were also discussed in detail. This red emitting material may be applied as a promising red phosphor for the near ultraviolet excited white light emitting diodes.

**Key words:**  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$ ; luminescent properties; red phosphor

## 1 Introduction

White light-emitting diodes (WLED) have recently attracted great attention as promising candidates for next-generation lighting due to its low energy consumption, high efficiency, excellent reliability, long lifetime, etc [1,2]. In order to generate white light using LEDs, one approach is to combine a blue LED chip emitting at 465 nm with a broad band yellow  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  (YAG: $\text{Ce}^{3+}$ ) phosphor. However, it exhibits low color rendering index (CRI) due to lacking of red component in the emission spectra, which limits its application. Another approach is to combine a near-ultraviolet (NUV) InGaN-based LED chip (350–420 nm) with the red, green and blue phosphors. In this approach, the phosphors are excited by the NUV chips, and the tricolor emissions make an excellent white light, which usually offer better color rendering performance [3]. Unfortunately, the efficiency of currently commercially used red phosphor  $\text{Y}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  is much lower than that of green and blue phosphors [4]. Therefore, more efforts should be devoted to searching for efficient red phosphors that can efficiently absorb light in the NUV region (350–420 nm).

Trivalent europium ( $\text{Eu}^{3+}$ ) ions activated phosphors are considered as ideal red sources for WLED because of the sharp emission lines in the red spectral region [5]. For applications in NUV-based WLED, the phosphors should be able to efficiently absorb the emission from the LED chips. However, the  $f-f$  excitation of  $\text{Eu}^{3+}$  ions is narrow in bandwidth and weak in intensity due to the parity forbidden nature of the  $f-f$  transitions, which leads to the low efficiency. In order to enhance light absorption, sensitizers are introduced to absorb the excitation light and transfer the excitation energy to the  $\text{Eu}^{3+}$  ions, which is a feasible method to overcome this shortage and obtain promising red phosphors [6].

The  $\text{Eu}^{3+}$ -doped molybdates and tungstates were investigated because the  $\text{MoO}_4$  and  $\text{WO}_4$  groups can efficiently absorb ultraviolet light through the excitation of the Mo–O and W–O charge transfer states (CTS), respectively. This excitation is then followed by a transfer of the absorbed energy to  $\text{Eu}^{3+}$  ions for red emission [7,8]. Nevertheless, most of these materials exhibit low efficient absorption in the NUV region. Recently, much attention has been drawn to the  $\text{Eu}^{3+}$ -doped molybdate and tungstate  $\text{R}_2\text{MO}_6$  ( $\text{R}=\text{Y}, \text{Gd}, \text{La}$ ;  $\text{M}=\text{Mo}, \text{W}$ ) for their remarkable properties such as excellent thermal and chemical stabilities and a broad

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excitation band [9–12]. However, little attention has been attracted to investigate the  $\text{Eu}^{3+}$ -doped  $\text{Lu}_2\text{MoO}_6$  phosphors. LI et al [13] have synthesized the  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$  phosphors by sol-gel method and investigated the structure and photoluminescence properties of such phosphors. However, the structural, luminescent properties and energy transfer mechanism of  $\text{Eu}^{3+}$ -doped  $\text{Lu}_2\text{MoO}_6$  obtained by solid state reaction method were not studied. In this work,  $\text{Eu}^{3+}$ -doped  $\text{Lu}_2\text{MoO}_6$  phosphors were synthesized using the high-temperature solid-state reaction method. The effect of  $\text{Eu}^{3+}$  molar concentration on the structure, luminescence and decay lifetime of the  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$  phosphors was investigated. The energy transfer mechanism of  $\text{Eu}^{3+}$  in  $\text{Lu}_2\text{MoO}_6$  host was also discussed in detail.

## 2 Experimental

### 2.1 Sample preparation

The  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$  phosphors were synthesized by the high-temperature solid-state method. The starting materials are analytical reagent (AR) grade molybdenum trioxide ( $\text{MoO}_3$ ), lutetium oxide ( $\text{Lu}_2\text{O}_3$ ) (99.99%) and europium oxide ( $\text{Eu}_2\text{O}_3$ ) (99.99%). The stoichiometric amounts of reactants were ground thoroughly in an agate mortar and preheated at 600 °C for 1 h. Subsequently, the products were removed from the muffle furnace, cooled, finely ground and sintered at 1200 °C for 4 h in air. Finally, the products were cooled to room temperature and then ground into white powder to form the final products.

### 2.2 Characterization

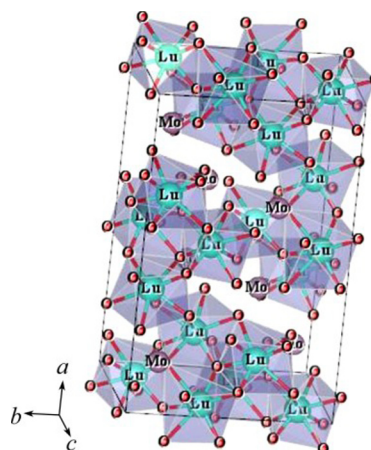
The crystal structures were analyzed by X-ray diffractometer (Persee, XD-2, Beijing Purkinje General Instrument Co., Ltd, Beijing, China) with  $\text{Cu K}\alpha$  radiation ( $\lambda=0.15406$  nm). The excitation, emission spectra and the decay curves were measured by FLSP920 (Edinburgh Instrument Ltd, Livingston, UK) fluorescence spectrophotometer equipped with 450 W xenon lamp or a pulse xenon lamp as light sources and Shimidazu R9287 (Hamamatsu Photonics K.K., Hamamatsu, Japan) photomultiplier (200–900 nm) along with a liquid nitrogen-cooled InGaAs (Hamamatsu Photonics K.K.) (800–1700 nm) as the detectors. All spectra were collected at room temperature under identical experimental conditions so that the emission intensities of the samples with different  $\text{Eu}^{3+}$  doping concentrations can be compared.

## 3 Results and discussion

### 3.1 Structure analysis of $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$ phosphors

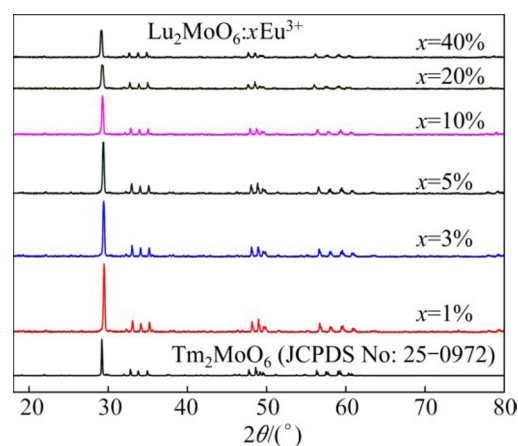
Figure 1 shows unit cell of  $\text{Lu}_2\text{MoO}_6$  drawn with

VESTA [14]. This material is crystallized in the  $I2/a$  space group (No.15). The  $\text{Lu}^{3+}$  ions occupy three nonequivalent crystallographic sites, namely, 4e, 4e (with  $\text{C}_2$  site symmetry) and 8f (with  $\text{C}_1$  site symmetry), and all are coordinated to eight O atoms, whereas the Mo atoms are coordinated to five O atoms with four of which at a short distance and the other at a long distance [15].



**Fig. 1** One unit cell of  $\text{Lu}_2\text{MoO}_6$  with 8-fold coordination of  $\text{Lu}^{3+}$  ions

Figure 2 shows the XRD patterns of  $\text{Lu}_2\text{MoO}_6$  phosphors doped with  $x\text{Eu}^{3+}$  ( $x=1\%$ , 3%, 5%, 10%, 20%, 40%). It indicates that all the diffraction peaks coincide well with the data from the JCPDS card No. 25–0972 ( $\text{Lu}_2\text{MoO}_6$ ), and  $\text{Lu}_2\text{MoO}_6$  can be assigned to the monoclinic structure [13]. No additional peaks of other phases have been found, indicating that the  $\text{Eu}^{3+}$  ions are effectively doped into the host lattice. Additionally, the diffraction peaks of  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$  samples are found to shift a little to lower angles with the increase of  $\text{Eu}^{3+}$  concentration, this is because the radius of  $\text{Eu}^{3+}$  (0.95 Å) is larger than that of  $\text{Lu}^{3+}$  (0.85 Å) in  $\text{Lu}_2\text{MoO}_6$  host. When  $\text{Lu}^{3+}$  is substituted by  $\text{Eu}^{3+}$ , the interplanar distance  $d$  increases, the diffraction angles decrease according to



**Fig. 2** X-ray diffraction patterns of  $\text{Lu}_2\text{MoO}_6:\text{Eu}^{3+}$  phosphor contrasted with standard pattern of JCPDS 25–0972

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