Preparation of a Novel Red $Y_2\text{MoSiO}_8$: Eu$^{3+}$, Dy$^{3+}$ Phosphor

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Abstract. A novel Eu$^{3+}$ ion-doped and Dy$^{3+}$ ion-doped yttrium silicomolybdic ($Y_2\text{MoSiO}_8$) phosphor was prepared by traditional high temperature solid state reaction at 1200°C. The $Y_2\text{MoSiO}_8$ monodoped Eu$^{3+}$ phosphors were excited at 395nm, the peak emission of red light is at 618nm and 595nm. The concentration of Eu$^{3+}$ ions affects the emission intensity of the fluorescent substance. $Y_2\text{MoSiO}_8$: Eu$^{3+}$, Dy$^{3+}$ phosphors excited at 370 and 395nm, it can emit red light at peaks of 617nm and 611nm, respectively, widen the absorption band of Eu$^{3+}$ in single doping, and improve the luminous intensity. This phosphor has the advantages of simple preparation, high luminous efficiency and easy industrialization.

1. Introduction
LED, which convert electricity into light, are the most valuable light source of the 21st century. Currently, commercial white LEDs are made by using a combination of blue LED chip and yellow phosphor (YAG: Ce$^{3+}$). The process is simple, low cost, and the materials produced are highly efficient. However, the emitting light of this LED is short of red and cold white light, with poor color rendering and low color rendering index [1-3]. If you want to achieve indoor luminous conditions, to obtain high color rendering index, color temperature stability of the product, the appropriate amount of red phosphor should be added into YAG: Ce$^{3+}$. Therefore, the exploration of new red phosphors has become the focus of current research, and it is of great significance to improve the performance of white LED [4-6]. In order to develop the efficient and stable red phosphor required by the white LED for lighting and further improve the performance of red light, researchers have done a lot of research work. At present, the widely studied and mature red phosphor systems mainly include: garnet system, sulfide system, nitride system, wolframomolybdic acid system system, etc. The phosphor based on sulfide is easy to decompose the toxic gas SO$_2$, the synthesis condition of silicon-based nitrous oxide phosphor is difficult to reach, and the vague understanding of crystal structure restrict the widespread application of this phosphor [7].

Molybdate phosphor is a kind of LED phosphor developed in recent years. Molybdate is used as the substrate of luminescent material preparation met. Molybdate is a kind of self-activated luminescent material, which has strong absorption in the near ultraviolet region and can effectively transfer energy [8].

Phosphors based on molybdate system also have good luminescence performance, stability and color purity, so they have attracted wide attention in recent years. $Y_2\text{MoSiO}_8$: Eu$^{3+}$ red fluorescence has been reported, which can emit red light of 618nm and 595nm under excitation of 395nm. When the same substrate is used, the luminescence intensity and luminescence efficiency are the key factors.
However, the red phosphors activated by Eu\(^{3+}\) will be limited by the narrow band absorption of Eu\(^{3+}\), so the red phosphors activated by Eu\(^{3+}\) still have the problem of low luminous intensity. Therefore, it is an important problem for us to broaden the absorption zone of Eu\(^{3+}\), improve the luminous intensity, and develop efficient and stable phosphors. Molybdenate has unique self-activation characteristics, and its spectral band is very wide. Pure phase structure Y\(_2\)MoSiO\(_8\): Eu\(^{3+}\) phosphors can effectively excite phosphors at 395nm, and emit red light at 595nm and 618nm, with the main peaks at 595nm and 618nm, respectively [9]. Doped Eu\(^{3+}\) and Dy\(^{3+}\) ions in molybdate matrix lattice can make it have good luminous intensity and luminous efficiency. When excited below 395nm, the phosphor Y\(_2\)MoSiO\(_8\): Eu\(^{3+}\), Dy\(^{3+}\) can emit red light at a higher peak, so the concentration of Eu\(^{3+}\) and Dy\(^{3+}\) ions has an important effect on the luminescence intensity of the fluorescent body [10].

2. Experiment

2.1. Preparation of Y\(_2\)MoSiO\(_8\): Eu\(^{3+}\), Dy\(^{3+}\)

In this experiment, Y\(_2\)MoSiO\(_8\): Dy\(^{3+}\) and Eu\(^{3+}\) phosphors were prepared with different raw material formulas, sintering temperature and sintering time. Y\(_2\)O\(_3\), MoO\(_3\), SiO\(_2\), Eu\(_2\)O\(_3\) and Dy\(_2\)O\(_3\) as the raw material were weighed to a specific ratio (Y: Mo: Si: Eu: Dy = 2: 1: 1: 0.03-0.09: 0.03-0.09). The raw materials were mixed and then 1.5-2.0wt % NH\(_4\)F flux was added. Different proportions of fluxes and charge compensators were added to the raw materials for sample synthesis. The medicine was placed into the agate mortar. A small amount of anhydrous ethanol was added to it, and it was fully ground for 40 min to evenly mix the raw materials. The evenly ground medicine is then transferred to the corundum crucible and placed in the muffle furnace. The mixture was heated at 1200°C for four hours. After the sample is cooled to room temperature in the muffle furnace, it is ground again evenly to obtain the phosphor sample.

2.2. Representation of Y\(_2\)MoSiO\(_8\): Eu\(^{3+}\), Dy\(^{3+}\)

XRD, TEM, laser particle size meter and other instruments were used to study the influence of combustion synthesis temperature on the composition, particle size and particle size distribution of powder products. Their absorption spectra, emission spectra, color temperature and color coordinate values of the pure phase Y\(_2\)MoSiO\(_8\): Dy\(^{3+}\) and Eu\(^{3+}\) phosphors were characterized. The surface and cross-section structure of the green slab and the surface morphology of the ceramic block were observed by the field emission scanning electron microscope of Hitachi Japan.

3. Results and discussion

The XRD patterns of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\) (x = 0.03, 0.05, 0.07, and 0.09) phosphors were measured and the results are displayed in Fig. 1. The diffraction peaks of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\) are well matched with those of JCPDs card 23-1485.

The morphology of Y\(_{1.94}\)MoSiO\(_8\): 0.03Eu\(^{3+}\), 0.03Dy\(^{3+}\) was investigated by SEM patterns as shown at Fig. 2. The sample was agglomerated and the crystal size was about 5μm. The particle size diameter of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\) was shown at Table 1. Fig. 3 shows the fluorescence intensity of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\). The concentration of Eu\(^{3+}\) ions has an important effect on the emission intensity of the fluorescent substance. When the doping amount of Eu\(^{3+}\) and Dy\(^{3+}\) is 3%, the strongest excitation wavelength is 395nm and the strongest emission wavelength is 611nm. When the doping amount of Eu\(^{3+}\) and Dy\(^{3+}\) is 5%, the strongest excitation wavelength is 395nm and the strongest emission wavelength is 611nm. When the doping amount of Eu\(^{3+}\) and Dy\(^{3+}\) is 7%, the strongest excitation wavelength is 370nm and the strongest emission wavelength is 617nm. When the doping amount of Eu\(^{3+}\) and Dy\(^{3+}\) is 9%, the strongest excitation wavelength is 370nm and the strongest emission wavelength is 617nm. When Y\(_2\)MoSiO\(_8\): Eu\(^{3+}\) and Dy\(^{3+}\) are excited at 370nm and 390nm, red light can be emitted at 617nm and 611nm, respectively. The absorption band of Eu\(^{3+}\) in single doping can be widened and its luminescence intensity can be improved. It can be seen from the excitation spectra of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\) and xDy\(^{3+}\) that there is a wideband absorption peak in the range of 350-
400nm, which is caused by the transfer of electrons from the 2p full electron sublayer of O\(^{2-}\) to the 4f full electron sublayer of Eu\(^{3+}\) ions. Dy\(^{3+}\) has obvious absorption peaks at 360nm, which is the classical characteristic peak of Dy\(^{3+}\) ion's 4f level transition. In addition, a number of narrow-band peaks can be seen from the excitation spectra, which are caused by the transition of electrons in the 4f shell of Eu\(^{2+}\) from the ground state to the excited state.

**Figure 1.** XRD patterns of series of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\).

![XRD patterns](image1)

**Figure 2.** SEM image of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\).

![SEM image](image2)

**Table 1.** Particle diameter of Y\(_{2-2x}\)MoSiO\(_8\): xEu\(^{3+}\), xDy\(^{3+}\).

|oping amount| x=0.03 | x=0.05 | x=0.07 | x=0.09 |
|-------------|--------|--------|--------|--------|
| Particle diameter (μm) | 2.3 | 2.5 | 2.6 | 2.8 |
4. Conclusion
In conclusion, a series of pure phase structures $\text{Y}_2\text{MoSiO}_8$: $\text{Dy}^{3+}$ and $\text{Eu}^{3+}$ were synthesized successfully. The phosphors can be excited effectively below 395 nm, and emit the red light of the main peak at 617 nm and 611 nm. When $\text{Eu}^{2+}$ and $\text{Dy}^{3+}$ ions were in the matrix, the luminescence intensity of $\text{Y}_2\text{MoSiO}_8$: $\text{Dy}^{3+}$ and $\text{Eu}^{3+}$ phosphors was significantly enhanced through the synergistic luminescence effect. When $\text{Eu}^{2+}$ and $\text{Dy}^{3+}$ ions were in the matrix, the luminescence intensity of $\text{Y}_2\text{MoSiO}_8$: $\text{Dy}^{3+}$ and $\text{Eu}^{3+}$ was significantly enhanced through the synergistic luminescence effect. This phosphor is low in temperature and cost, it has high luminous intensity and good luminous performance. The phosphor has good stability and excellent physical and chemical properties, which can be applied to white LED. The preparation method of the phosphor powder is a high temperature solid phase method, process simple, without special equipment, good production environment friendly, small damage to the operator and the environment, the total organic matter is low, the production cost is low, easy to control, the core of this method is to grind mill, need to make the raw material mix for reaction. This method made brighter phosphor and craft simple, suitable for mass production.

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