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Synthesis and luminescent properties of Eu^{3+} doped Y_2WO_6 nanophosphors

HAO Ruonan (郝若男), MENG Qingyu (孟庆裕)*, LIU Wei (刘 威), LIU Hongling (刘宏亮)

(Key Laboratory for Photonic and Electronic Bandgap Materials, Ministry of Education, School of Physics & Electronic Engineering, Harbin Normal University, Harbin 150025, China)

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Abstract: Novel nanosized Y_2WO_6:Eu^{3+} phosphors were synthesized via a co-precipitation reaction. The crystal structure of Y_2WO_6:Eu^{3+} sample was monoclinic phase characterized by using X-ray diffraction (XRD). The particle size was about 80 nm observed by field emission scanning electron microscopy (FE-SEM). The photoluminescence properties of Y_2WO_6:Eu^{3+} nanophosphors were studied. The results indicated that Eu^{3+} $^5D_0 \rightarrow {}^7F_2$ red luminescence at 611 nm could be effectively excited by 394 nm near-UV light and 465 nm blue light in Y_2WO_6 host. The luminescence intensity was the strongest while the Eu^{3+} doping concentration was 20%. And the chromaticity coordinates of this concentration is (0.651, 0.348). The energy transfer type between the Eu^{3+} was determined to be the exchange interaction and the critical energy transfer distance (D_c) was calculated to be about 0.81 angstrom. The J-O parameters, quantum efficiencies of Eu^{3+} $^5D_0$ energy level and Huang-Rhys factor of Y_2WO_6:Eu^{3+} nanophosphors were calculated. The calculated values indicated that Y_2WO_6:Eu^{3+} had a high capacity for activators and the electron-phonon coupling was weak. Therefore, the Y_2WO_6: Eu^{3+} nanophosphor is a nice red luminescent material and it may have a potential application in white LED.

Keywords: red nanophosphors; tungstate; luminescence; white LED; rare earths

Nowadays, phosphors have been widely used for applications such as in radiation detectors and visual displays. The introduction of trivalent rare-earth ions (RE^{3+}) in matrices as luminescent centers has been an improvement in the field of luminescent materials[1–4]. Eu^{3+} doped tungstates possess strong Eu^{3+} 4f-4f transition absorption in near UV and blue region, and can transform the absorption energy to red luminescence of high color purity[5–10], so it may have a potential application for white LED red phosphors. Because nano-materials have special properties that are different from the bulk materials, there has been much interest in exploring the elemental physical mechanism and in finding potential applications as new engineering materials in many aspects relative to luminescence[11–15]. Therefore, preparation and luminescent properties of Eu^{3+} doped tungstate nano-materials have attracted more and more interest. In addition, Eu^{3+} doped $M_x(WO_4)_y$ systems have been widely studied, but less attention has been paid to the other types of tungstates (for example, Y_2WO_6), while other types of tungstate systems may have more excellent luminescent properties, so it should also be paid enough attention to.

In this paper, we synthesized Eu^{3+} doped Y_2WO_6 nanophosphors using a co-precipitation method and studied their photoluminescence properties in detail.

1 Experimental

1.1 Synthesis

Eu^{3+} doped tungstates were prepared by a co-precipitation method. The co-precipitation method is usually adopted for preparing nanosized rare-earth oxides[16–19]. All reagents used in this work were of analytical grade without any further purification. A series of Y_2WO_6:Eu^{3+} phosphors were synthesized by co-precipitation method. First, the $Eu(NO_3)_3{\cdot}6H_2O$ and $Y(NO_3)_3{\cdot}6H_2O$ of required molar ratio (total mole of RE^{3+} is 0.004 mol) were dissolved in 20 mL distilled water, and the acidity of the solution was adjusted to pH 2.0 with HNO_3. Suitable molar amount of $Na_2WO_4{\cdot}2H_2O$ was dissolved in 50 mL distilled water, and the pH was adjusted to 11–12 with ammonium hydroxide. Second, the rare-earth solution was dropped to the aqueous Na_2WO_4 solution slowly under stirring, and then a white precipitate formed at once. After all rare-earth solution was dropped into Na_2WO_4 solution, the white suspension solution was stirred for 40 min to ensure a complete reaction. The precursor solution was centrifuged at 4500 r/min for 30 min. The resulting white precipitate was washed several times with distilled water and dried at 80 °C for 3 h. Finally, the red phosphors were obtained when resultant

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* **Corresponding author:** MENG Qingyu (**E-mail:** qingyumeng163@163.com; **Tel.:** +86-451-88060526)

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precursors were calcined at 900 °C for 2 h. Eu^{3+} doped Y_2WO_6 were synthesized via the above method. The Eu^{3+} doping concentrations were 1 mol.%, 5 mol.%, 10 mol.%, 15 mol.%, 20 mol.%, 25 mol.%, 30 mol.%, 35 mol.% and 40 mol.%. The calcination temperature was decided via temperature-optimized experiments.

1.2 Characterization

The crystal structure of the samples was examined by using powder X-ray diffraction (XRD) performed on an XRD-6000 (Shimadzu) diffractometer by using Cu $K\alpha_1$ radiation (λ=0.15406 nm). The XRD data were collected by using a scanning mode in the 2θ range from 15° to 70° with a scanning step of 0.02° and a scanning rate of 2.0 (°)/min. Silicon was used as an internal standard. The particle size, shape, and morphology were examined by using Hitachi S-4800 Field emission scanning electron microscopy (FE-SEM). Emission and excitation spectra were recorded with an EDINBURG GH-LFS920 spectrophotometer equipped with a 450 W xenon lamp as excitation source. In the measurements of fluorescent decay curve, the threefold frequency output (355 nm) of a Quantel Brillant B pulsed YAG:Nd laser was used to excite the samples, with repetition frequency of 10 Hz and pulse duration of 5 ns. The fluorescent signals were obtained from an ANDOR-SR750 three grating monochromator. And an STANFORD-SR430 Multichannel Scaler/Averager was used for recording data. All measurements were carried out at room temperature.

2 Results and discussion

2.1 Structural and morphology characterization of the products

The XRD patterns for all the samples were measured, and a similar diffraction pattern was observed for each sample. The unchanged diffraction pattern indicates that incorporating different amounts of Eu^{3+} does not affect the crystal structure of the products. Fig. 1 shows the XRD patterns of Y_2WO_6 nanocrystallines doped with 5 mol.%, 10 mol.%, 20 mol.% and 40 mol.% Eu^{3+}, respectively. The Y_2WO_6:Eu^{3+} sample is primitive lattice which appears in JCPDS card #730118. All the samples obtained are monoclinic phase. Fig. 2 shows the FE-SEM images of samples mentioned in Fig. 1. It can be seen from Fig. 2 that the average particle size is about 80 nm.

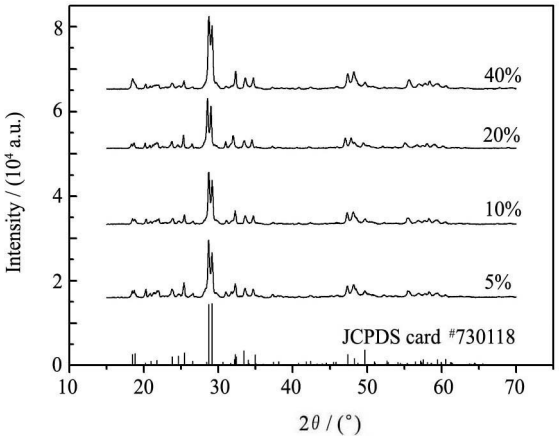

Fig. 1 XRD of the Y_2WO_6:Eu^{3+} phosphors with different Eu^{3+} doping concentrations

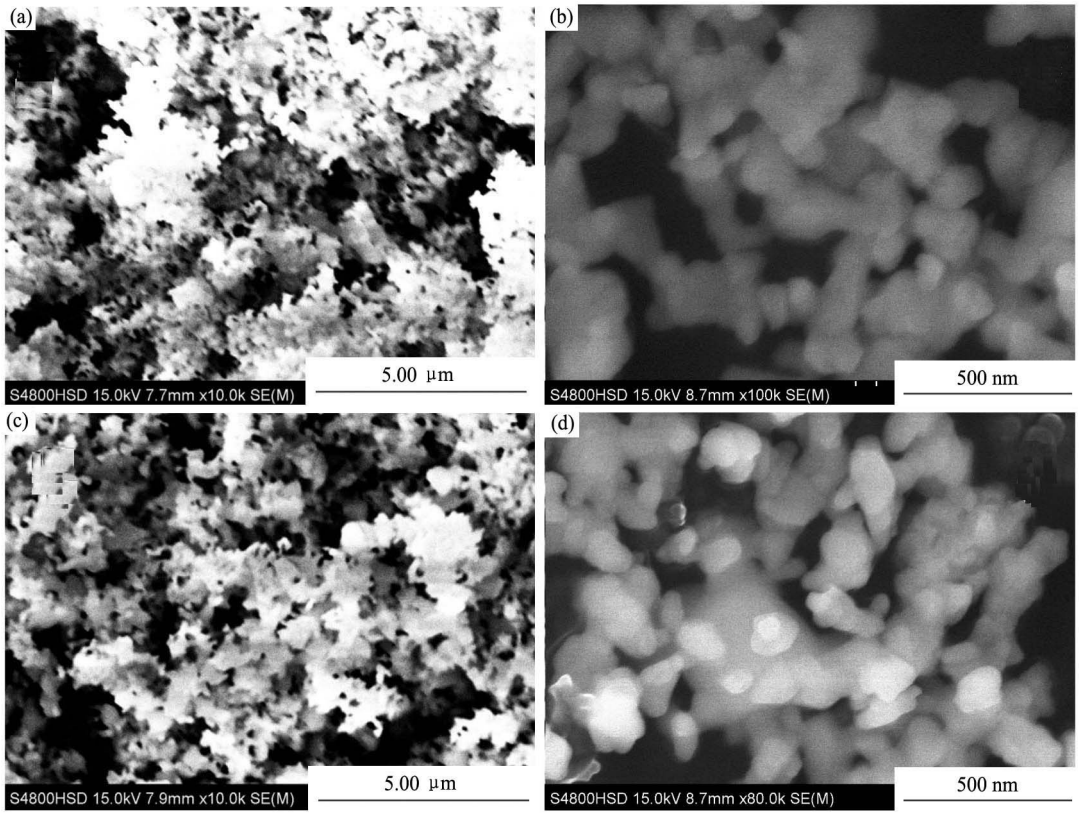

Fig. 2 SEM images of Y_2WO_6 nanophosphors doped with 5 mol.% Eu^{3+} (a, b) and 20 mol.% Eu^{3+} (c, d)