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Eu substitution and particle size control of Y₂O₂S for the excitation by UV light emitting diodes

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Abstract

Yttrium oxysulfide doped with europium $(Y_2O_2S:Eu^{3+})$ red phosphor is used in UV light emitting diodes (LEDs) by mixing with blue and green phosphors to generate white light which are important for the application in general lighting. Here, we demonstrate the effect of shape and size and the concentration of activator (Eu) of red Y_2O_2S phosphor. © 2005 Elsevier Ltd. All rights reserved.

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1. Introduction

Fluorescent materials have variety of applications in lighting and display industries. Improvement in the field of luminescence has been made by doping the rare earth ions as luminescent centers. For example, Eu activated materials as red component have potential applications in color television displays. The flat panel display types include electroluminescent (EL), field emission (FED) and plasma displays (PDP). In FEDs and PDPs, the excitation mechanisms are cathodo-luminescence and photo-luminescence, respectively. EL devices are the solid state analogs to cathodoluminescent vacuum tubes (CRT) [1].

Now-a-days light emitting diodes (LEDs) have been emerged as an important class of source for white light. Green and red light emitting diodes were used in traffic light. And many kinds of products have applications in mobile phones as display devices and outdoor lighting. Phosphor is

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defined as a material that emits photons with high luminescence efficiency. The cathodoluminescent phosphors convert electron energy into visible light and are used in the display devices as the component in screen material [2,3].

Most industries manufacturing white light sources by combining blue chip with yellow phosphor. Its lighting efficiency is pretty good in the low current condition, but the color rendering is poor, as it only contains blue and yellow and other colors are excluded. So UV light emitting diodes (UV LEDs) would be an important choice in the future because of various kinds of lights can be mixed in the UV excitation. With these LEDs the problem of low color rendering for the blue LED with yellow phosphor can be solved. Now-a-days most researchers have concentrated on the excitation of phosphors by using the UV source.

Blue, green, and red light are three important elements in producing white light. In these three elements, red phosphor is the most important material to increase the overall white light efficiency [1]. Because emission of red phosphor often very sharp and low intensity relative to blue or green

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phosphors. So our objective is to study the red phosphor to increase their emission characters.

Y₂O₃ and Y₂O₂S doped with Eu are the well-known red PL (photo-luminscent) and CL (cathodo-lumiscent) phosphors. Y₂O₂S:Eu is the most important red primary extensively applied in cathode-ray tubes (CRTs) due to its high luminous efficiency and better color saturation compared with Y₂O₃:Eu red phosphor [4]. Y₂O₂S:Eu³⁺ has the sharper emission lines leading to better colorimetric definition and higher luminescence efficiency than other red phosphors [5]. The red phosphor Y₂O₂S:Eu with its sharp line for good calorimetric definition and high luminescence efficiency is useful in the manufacture of UV excited devices [6]. Literature survey shows Eu doped in oxysulfide compound was more efficient than doped in oxide compound. The effect of Eu ion on the luminescence properties of yttrium oxysulfide has been studied and well established. As known, shape and size of phosphor particles would affect the efficiency [9,10]. In the recent study, sphere particle or like was fabricated by flux fusion method [7,11]. The effect of various flux compositions on the particle shape and size distribution has also been studied [12].

In the present investigation, we demonstrate the synthesis of europiumyttriumoxysulfide phosphor by sulfurization of rare earth oxide in the flux [7,8]. This process is harmless and eco-friendly compared to other synthetic techniques. We analyzed Eu occupancy in place of Y by using XRD data refinement by Rietveld's method. SEM and CL spectral were performed to investigate the particle size and spectral properties of the samples.

2. Experimental

Europium doped yttrium oxysulfide was synthesized by sulfide fusion method. Y₂O₃ and Eu₂O₃ were taken as raw materials and mixed with a mixture of flux materials Na₂CO₃, K₂CO₃, Li₂CO₃, Li₃PO₄ in the ratio of 4:1:4:1. Mixture was ground well and sintered in the range of 1100–1200 °C under nitrogen atmosphere to avoid oxygen during reaction. Sintered powders were washed with nitric acid to remove the residuals and dried at 100 °C [13]. Several samples were synthesized in the same way by changing the europium mole percentage to ascertain the strong red emitting material.

The overall reaction process for the formation of red phosphor $Y_2O_2S:Eu^{3+}$ can be represented as

$$Y_2O_3 + Eu_2O_3 + flux \rightarrow Y_2O_2S : Eu^{3+} + flux residue$$
 (1)

XRD patterns were recorded for all the samples by X-Ray diffractometer (Philips PANalytical, Germany) with Cu K α radiation (λ =1.5406 Å) to determine the crystal structure and phase. Rietveld refinement method was applied to refine XRD date to investigate atomic parameters [14].

Luminescence measurements were conducted by photoluminescent instrument (Yvon–Spex, Spex Fluorolog-3, USA) which used Xe lamp as the light source and PMT as the detector. The scanning rate was kept at 2 nm/min and the emission range was 380–780 nm. Emission spectra were transferred to CIE (Commission Internationale de l'Eclairage) color coordinates. The sintered phosphor powder was examined with a scanning electron microscope (recorded by FESEM-JSM6700F) to investigate the particle size, particle shape and surface morphology.

3. Results and discussion

Fig. 1 shows the XRD patterns for $(Y_{2-x}Eu_x)O_2S$ (x=0.04-0.13) samples sintered at 1150 °C for 2 h. Y_2O_2S pure phase could be obtained for all x values by using optimized flux ratio Na₂CO₃:K₂CO₃:Li₂CO₃:Li₃PO₄=1.2:0.3:1.2:0.3.

In order to derive the position of Eu^{3+} in the crystal structure and to calculate lattice constants for the sample $(Y_{2-x}\mathrm{Eu}_x)\mathrm{O}_2\mathrm{S}$ (x=0.04) we applied GSAS structure refinement method. In the refinement process P-3m1 space group has been taken as the model and thermal vibration for each atom set as isotactic. Occupation parameter for Y and Eu $(Y+\mathrm{Eu})$ has been taken as 1.

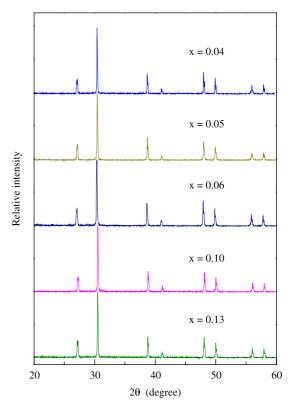


Fig. 1. X-ray diffraction patterns of different Eu doped in Y_2O_2S samples sintered at 1150 °C for 2 h.

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