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Luminescence properties of YVO_4: Ln (Ln = Dy^{3+}, Eu^{3+}, Tm^{3+}) for white LED by hydrothermal method

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ABSTRACT

YVO_4: Dy^{3+}, Eu^{3+} / YVO_4: Dy^{3+}, Eu^{3+}, Tm^{3+} phosphors were synthesized by a simple hydrothermal method. The energy transfer from Dy^{3+} to Eu^{3+} was determined by photoluminescence (PL) spectra and fluorescence lifetime. The energy transfer mechanism from Dy^{3+} to Eu^{3+} was confirmed. The amount of Eu^{3+} in YVO_4: $0.01Dy^{3+}$, Eu^{3+} with strong emission and small size was determined by applied ratio, and finally a white light (0.300, 0.317) output was achieved in YVO_4: $0.01Dy^{3+}$, $0.01Eu^{3+}$, $0.03Tm^{3+}$ by adjusting the amount of activator ions. Thus the sample with the above characteristics has a promising application prospect in the light emitting diode (LED).

1. Introduction

In recent years, the global energy shortage and environmental pollution problems have become increasingly prominent, so the energy saving and emission reduction has become the theme of today's society [1]. Because lighting is one of the main forms of energy consumption, searching for a new environmental friendly lighting technology has become a hot spot of concern. The LED technology mainly concentrates on various aspects, including energy saving, environmental protection, long life expectancy, small size, fast response and so on [2]. Many efforts, therefore, have been committed to the synthesis of rare earth luminescent LED materials [3–5], such as rare earth vanadate [6], rare earth fluoride [7], rare earth phosphate [8], and rare earth silicate [9] *etc*. Among them, rare earth vanadate has a broader application prospect [10,11] because of its low price, high chemical stability, high thermal stability, large emission cross section and great absorption cross section. For instance, in 1964, YVO_4: Eu^{3+} luminescent material was developed as a red phosphor and used in color TV [12]. Wang et al. prepared the self-activated phosphor $Ba_2V_2O_7$ micro-rods with a high quantum efficiency of 47.5% by the hydrothermal reaction method [13]. Gan et al. prepared YVO_4: Ln^{3+} (Ln = Eu, Dy and Sm) by the microwave-assisted synthesis [14].

As is known, it is easy to obtain red emission by single-doping Eu^{3+}. Tm^{3+} usually emits blue light, and Dy^{3+} normally emits blue or yellow light. Adjustable light color was achieved in the three mixed systems by adjusting the amount of ratio [15,16]. For example, Song et al. studied 3D-hierachical spherical $LuVO_4$: Tm^{3+}, Dy^{3+}, Eu^{3+} [16]. Wang Y et al. synthesized YVO_4: Ln^{3+} and silica-coated YVO_4: Ln^{3+} (Ln^{3+} = Eu^{3+}, Dy^{3+}, Tm^{3+}) nanoparticles by microemulsion method [17]. All these phosphors have good luminescence properties. However, there are rare reports about small YVO_4: Ln^{3+} (Ln^{3+} = Eu^{3+}, Dy^{3+}, Tm^{3+}) nanoparticles with strong emission and color tunable emission. In this paper, the fluorescent nanoparticles of YVO_4: Dy^{3+}, Eu^{3+}, Tm^{3+} and YVO_4: Dy^{3+}, Eu^{3+} with strong emission and small particle size were synthesized by simple hydrothermal method. The mechanism of energy transfer from Dy^{3+} to Eu^{3+} in YVO_4: Dy^{3+}, Eu^{3+} phosphors is confirmed. Finally the pseudo white light output of YVO_4: Dy^{3+}, Eu^{3+}, Tm^{3+} was realized.

2. Experiment section

2.1. Materials and preparation

All samples were synthesized by hydrothermal method. The raw materials included NH_4VO_3 (A.R.), NaOH (A.R.), $NH_3{\cdot}H_2O$ (38%), Eu_2O_3 (99.99%), Dy_2O_3 (99.99%), Tm_2O_3 (99.99%) and Y_2O_3 (99.99%).

In the first step, 0.1461 g of ethylenediamine tetraacetic acid (EDTA) was added into 4 mL solution of $NH_3{\cdot}H_2O$ (38%) and water mixed by volume ratio of 1:1(solution A), and then the solution was the solution was stirred for 30 min. In the second step, a series of volume of 0.5 mol/L $Y(NO_3)_3$ solutions were added into the solution with different

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concentrations of Eu^{3+}, $Dy^{3+}$and Tm^{3+}. Then 0.12 g of NaOH and 0.117 g of ammonium hydroxide (NH_4VO_3) were dissolved in 8 mL of deionized water (solution B). Next, solution A and solution B was mixed to form solution C. In step four, the obtained solution C was adjusted to pH 9 with concentrated nitric acid, and stirred for 30 min to obtain solution D In the fifth stage, solution D was transferred into a 20 mL Teflon-lined stainless steel autoclave and then sealed. The autoclave was heated up to 180 °C for 48 h. Eventually, the autoclave aturally cooled to 25 °C was naturally. The obtained samples were washed with deionized water and ethanol in turn. Thereafter, they were centrifuged at 15,000 rpm for 5 min, and then dried for 6 h at 80 °C.

The formation process of YVO_4: Ln^{3+} is as follows:

$$3OH^{-} + (1 - X)Y^{3+} + XLn^{3+} \rightarrow (Y_{1-X}Ln_X)(OH)_3 \quad (1)$$

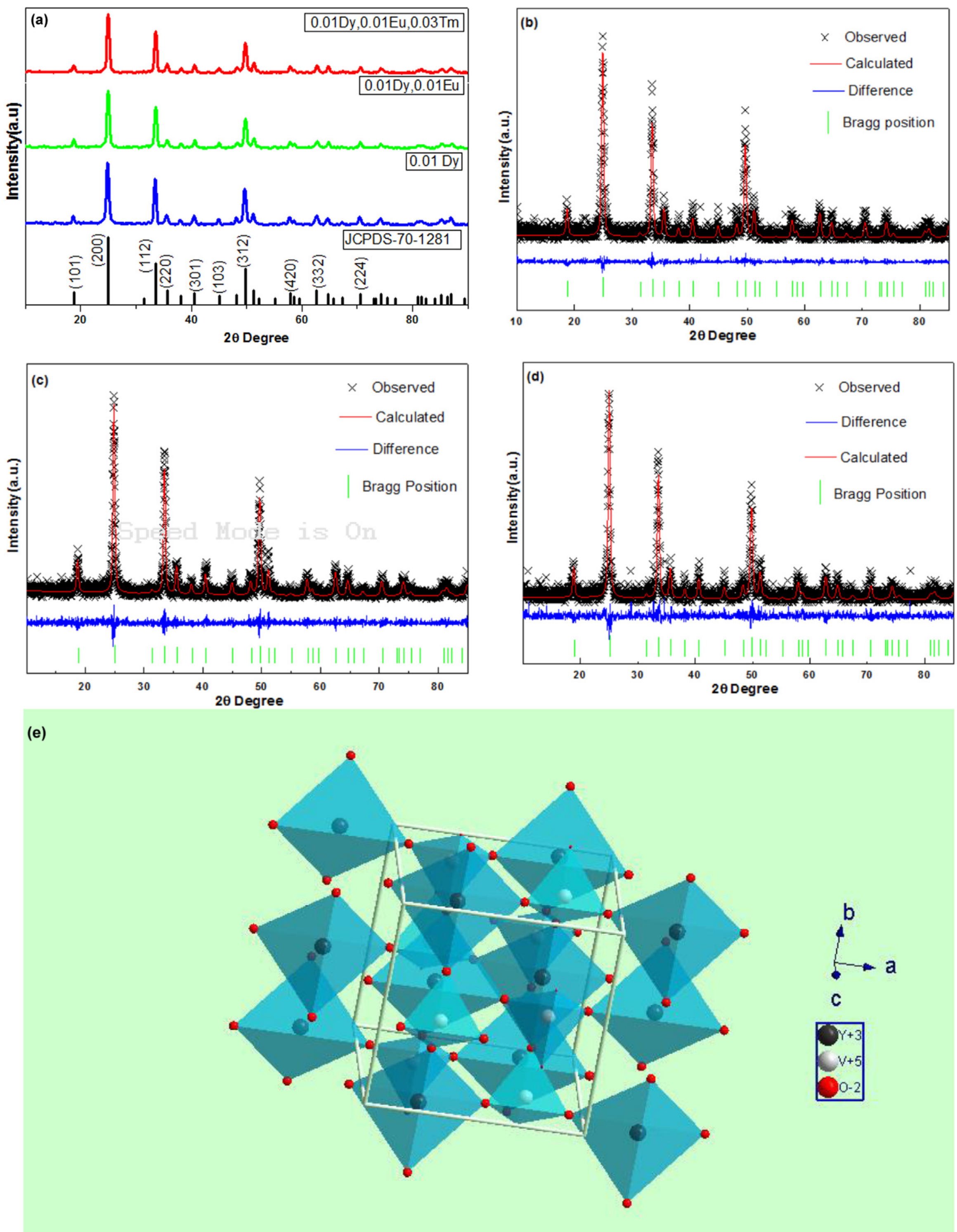

Fig. 1. (a) XRD patterns of YVO_4:Ln^{3+} ($Ln^{3+} = Dy^{3+}$, Eu^{3+}, Tm^{3+}/Dy^{3+}, Eu^{3+}/Dy^{3+}) samples. (b) Rietveld refinement of the powder XRD pattern of YVO_4: Dy^{3+} (c) Rietveld refinement of the powder XRD pattern of YVO_4: Dy^{3+}, Eu^{3+} (d) Rietveld refinement of the powder XRD pattern of YVO_4: Dy^{3+}, Eu^{3+}, Tm^{3+}. (e) Crystal structure of YVO_4.