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The synthesis and photoluminescence property of $YPO_4:Eu^{3+}$ hollow microspheres

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ABSTRACT

The $YPO_4:Eu^{3+}$ hollow microspheres have been successfully prepared via a simple two-step route. First, the core–shell structured MF@$Y(OH)CO_3$:Eu precursor was fabricated by a urea-based homogeneous precipitation method using colloidal melamine formaldehyde (MF) microspheres as template. Then the $Y(OH)CO_3$:Eu precursor was transformed into hollow $YPO_4:Eu^{3+}$ by a subsequent solvothermal method, and MF microspheres were dissolved in the solvent simultaneously. The as-prepared hollow microspheres were well characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM) and luminescence spectroscopy. It is found that the template can be removed without additional calcination or etching process. The YPO_4 hollow microspheres may have potential applications in drug delivery, cell biology and diagnosis.

1. Introduction

Hollow structures have gained special attention in the field of drug storage and release, because they simultaneously possess large voids inside the shells and mesopores at the shells [1]. The hard template strategy is an effective approach to prepare hollow spheres with well-defined shape and good dispersity. In the hard template strategy, many compounds, such as polymeric, inorganic

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nonmetallic, and metallic particles, have been extensively used as templates [2–7]. In addition, because the melamine formaldehyde resin is very cheap, hollow spherical phosphors would achieve a reduction in the amount of expensive rare earth materials, and thus lower the cost of the rare earth luminescent materials [8].

As an important class of lanthanide inorganic compounds, rare earth orthophosphates ($REPO_4$) have attracted much attention due to their unique photo-luminescent properties and various potential applications in various areas, including color displays, light sources, field-effect transistors, solar cells, and biomedical labels [9–14]. Recently, in order to realize the controlled morphology and structure of the YPO_4 luminescent materials, dramatic efforts have been dedicated to develop new approaches [15–20], including polyol, precipitation, hydro-/solvothermal, and combustion method, surfactant-mediated solution route and so on. However, to the best of our knowledge, only a few researchers have reported the synthesis of YPO_4 hollow spheres. Zhang and co-workers recently reported that large-scale good-quality submicrometer-sized YPO_4:Eu^{3+} hollow spheres were synthesized by utilizing the colloidal spheres of $Y(OH)CO_3$:Eu^{3+} as a sacrificial template [21]. Jiu et al. reported the synthesis of YPO_4 hollow microspheres from PS microspheres. In addition, the desired materials can be coated on the templates to form core–shell structured spheres. Subsequently, the templates are removed by calcination at elevated temperature in air or by selective etching with a solvent to generate hollow structured spheres [22]. The above method generally includes acid erosion or calcination to remove the temple. Herein, we describe a method to synthesize the MF@$Y(OH)CO_3$:Eu^{3+} core–shell structure and the hollow structure of the YPO_4:Eu^{3+} phosphors. In the hydrothermal process, the $Y(OH)CO_3$:Eu^{3+} of the shell part of reacted with $NH_4H_2PO_4$ to create YPO_4:Eu^{3+} composites, and the temple was simultaneously removed to obtain the hollow structure. The phase compositions, morphologies, and optical properties of the as-synthesized hollow YPO_4:Eu^{3+} microspheres are systematically investigated.

2. Experimental

All other chemicals were analytical-grade reagents and were purchased and used without further purification. The monodisperse MF colloidal microspheres were prepared according to the process report [23]. Then, the MF/$Y(OH)CO_3$:Eu^{3+} microspheres were also synthesized using the same procedure [8]. The as-obtained 0.2 g of MF/$Y(OH)CO_3$ precipitation was dispersed into 20 mL deionized water by ultrasonic for 5 min. A total of 0.2 g of $NH_4H_2PO_4$ dissolved in 20 mL of deionized water was dripped into the dispersion followed by further stirring. The reaction mixture was transferred into a 100 mL Teflon-lined autoclave and kept at 180 °C for 12 h. The obtained products were carefully collected after washing with deionized water and alcohol and drying at 60 °C for 2 h in the air. Finally

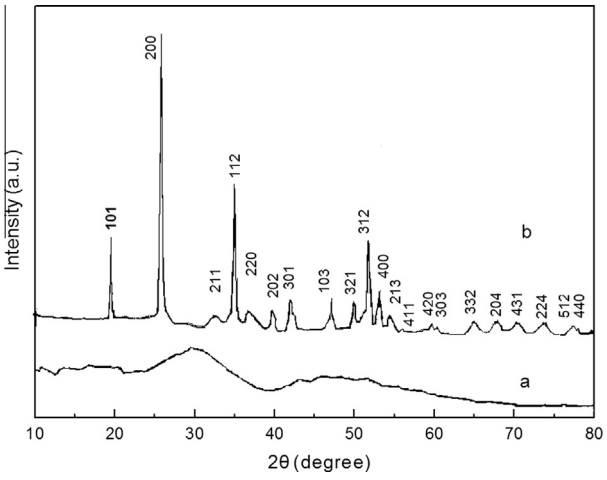

Fig. 1. The X-ray diffraction patterns of the precursor MF/$Y(OH)CO_3$:Eu (a) and the final product (b).