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Effect of annealing in hydrogen atmosphere on the photoluminescence properties of phosphor-in- glass in tellurate glass

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ABSTRACT

A series of phosphor-in-glasses (PIGs) was prepared by one-step melt-quenching method sintering the mixture of tellurate raw material powder and YAG:Ce^{3+} phosphors at 450–700 °C, of which some was annealed at 250 °C for 3 h under the hydrogen atmosphere. The test results show that the relative photoluminescence (PL) intensity of phosphor-in-glass sintered at 450–700 °C was significantly strengthened from 42.29% to 174.72% after the hydrogen annealing, and the relative photoluminescence excitation (PLE) intensity from 21.00% to 207.90%. However, the relative photoluminescence intensity of the PIG sintered at 700 °C before and after hydrogen annealing was nearly zero because of the destruction of crystal structural surrounding Ce^{3+} ion. The concentration of Ce^{3+} and Ce^{4+} measured by X-ray photoelectron spectrum analyser (XPS) in PIGs before and after hydrogen annealing obviously happened to change, which was associated with the variation of PL and PL excitation intensity. Besides, the X-ray diffraction (XRD) pattern of YAG:Ce^{3+} phosphor and PIGs sintered at different temperatures further presented the destruction of YAG:Ce^{3+} lattice as the sintering temperature increased. Therefore, the appropriate sintering temperature and hydrogen annealing can increase the PL and PLE intensity and the Ce^{3+} concentration.

1. Introduction

White light-emitting diodes (WLEDs), the new generation of solid-state lighting source, have been extensively used in display backlighting, automotive headlamps, street lamps, and interior lighting because of the features of long lifetime, high luminous efficiency, energy saving, environment-friendly and so on [1,2]. The current commercial WLED is obtained by combining a gallium nitride (GaN) based blue chip with yellow phosphor (yellow YAG: Ce^{3+} phosphor), which was packaged by organic encapsulants, such as resin or silicone [3,4,5]. Nevertheless, resin and silicone encapsulation are colored by the heat from the blue GaN chip and easily aging after long-term service, ascribing to poor thermal conductivity and thermal stability, which may reduce the secular reliability of the WLED [6,7,8]. Therefore, inorganic materials with excellent thermal conductivity and thermal stability, such as phosphor-ceramics and phosphor-in-glasses (PIGs), are the pretty good choice to replace polymer sealant as potential phosphor carrier applied in light-emitting diodes encapsulation [9,10,11]. Compared with phosphor-ceramics, the luminescence of PIG can be easily controlled through incorporating of different phosphors into glass composition and the low-temperature synthesis of PIG (< 800 °C) can keep the high performance of doped phosphor in the course of the fabrication process, Consequently, PIG has attracted more attention than phosphor-ceramics [9].

YAG:Ce^{3+} PIG, manufactured by co-sintering of the uniform mixture of commercial YAG:Ce^{3+} phosphors and glass raw material powders at a suitable temperature [12], shows yellow emission because of the Ce^{3+}: 5d → 4f transition by 460 nm excitation [13]. The efficient luminescence of YAG: Ce^{3+} PIG can be achieved by decreasing the corrosion of melting glass to phosphor and matching the refraction index between glass matrix and phosphor to keep PIG transparent and reduce light scattering [11]. Meanwhile, the luminescent properties of the YAG: Ce^{3+} PIG can be also significantly enhanced by controlling the reducing atmosphere diminishing the oxidation of Ce^{3+} of the PIG [6]. Recently, tellurate glasses are considered as the attractive glass matrix to prepared the phosphor-in-glass due to the low melting temperature and high refraction index [14,15].

In this paper, TeO_2-based PIGs, prepared by varying the sintering temperature from 450 °C to 700 °C, were annealed at 250 °C for 3 h in the condition of hydrogen atmosphere. The photoluminescence (PL) and photoluminescence excitation (PLE) of the PIGs before annealing were compared with those after annealing. In addition, the XRD and

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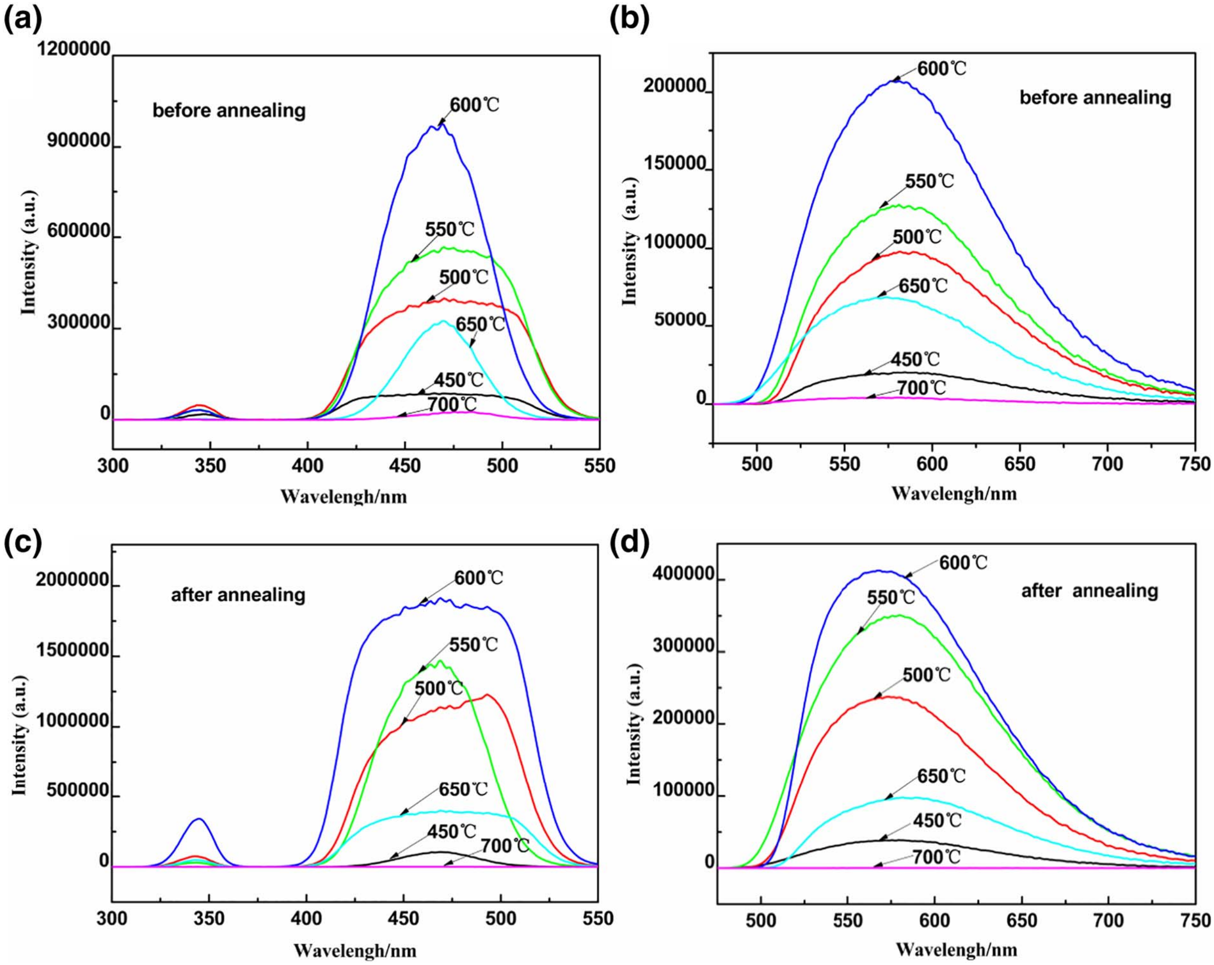

Fig. 1. PLE and PL of the PIGs sintered at different temperature: (a) PLE spectra before annealing; (b) PL spectra before annealing; (c) PLE spectra after annealing; (d) PL spectra after annealing.

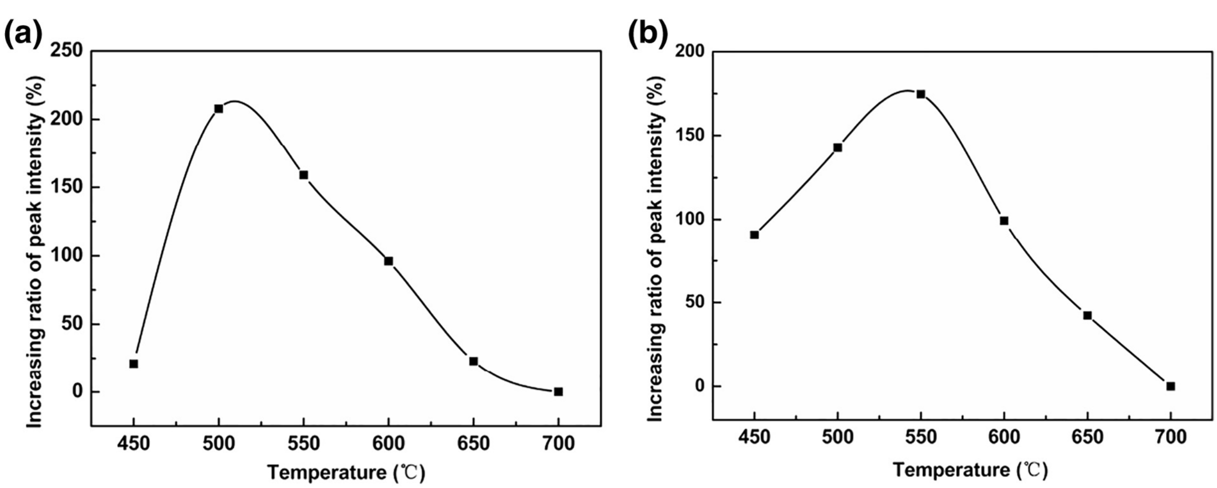

Fig. 2. Increasing ratio of spectral peak intensity of the PIGs sintering at different temperature after annealing: (a) increasing ratio of PLE spectra peak intensity; (b) increasing ratio of PL spectral peak intensity.

XPS were carried out to analyses the crystal structure and the Ce^{3+} concentration in the samples.

2. Experiments

The uniform mixture of 70%TeO_2-20%ZnO-10%Na_2O (mol%) -based glass raw materials and 7% YAG phosphor was sintered at the temperature between 450 °C and 700 °C for 0.5 h by one-step melt-quenching method [16], and the glass melt was fleetly poured into a cold copper mould, which gradually cooled to room temperature. Part of the produced were cut and polished into plates with 1 mm thickness was annealed at 250 °C for 3 h under the mixture atmosphere of H_2 and N_2 (5%H_2, 95%N_2).

The photoluminescence (PL) and PL excitation (PLE) spectra were