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Design of laser-driven SiO_2-YAG:Ce composite thick film: Facile synthesis, robust thermal performance, and application in solid-state laser lighting

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In recent times, there have been rapid advances in the solid-state laser lighting technology. Due to the large amounts of heat accumulated from the high flux laser radiation, color conversion materials used in solid-state laser lighting devices should possess high durability, high thermal conductivity, and low thermal quenching. The aim of this study is to develop a thermally robust SiO_2-YAG:Ce composite thick film (CTF) for high-power solid-state laser lighting applications. Commercial colloidal silica which was used as the source of SiO_2, played the roles of an adhesive, a filler, and a protecting agent. Compared to the YAG:Ce powder, the CTF exhibits remarkable thermal stability (11.3% intensity drop at 200 °C) and durability (4.5% intensity drop after 1000 h, at 85 °C and 85% humidity). Furthermore, the effects of the substrate material and the thickness of the CTF on the laser lighting performance were investigated in terms of their thermal quenching and luminescence saturation behaviors, respectively. The CTF with a thickness of 50 μm on a sapphire substrate does not show luminescence saturation, despite a high-power density of incident radiation i.e. 20 W/mm^2. These results demonstrate the potential applicability of the CTF in solid-state laser lighting devices.

1. Introduction

During the past decades, solid-state lighting (SSL) devices have been attracting increasing attention, and have become tough competitors to the conventional light sources (e.g., incandescent, fluorescent, and high intensity discharge lamps) [1–3]. Among all SSL devices, the phosphor-converted white-light emitting diode (PC-wLED) has emerged to be the most successful due to its cost-effectiveness, compact size, energy efficiency, superior lifetime, and environmental friendliness [2–6]. However, there exists a significant challenge in the development of high-power LEDs, known as the "efficiency droop" of the blue InGaN chip. When the input power density of the LED system reaches 10 kW/cm^2, the power-conversion efficiency (PCE) of a current state-of-the-art 450 nm InGaN chip drops to a mere ~10%, which is much lower than the value corresponding to an input powder density of 0.5 kW/cm^2, i.e. ~40%. So far, no suitable solution has been proposed to overcome the efficiency drop of LEDs [7–9].

An alternative technique to resolve the issue of LEDs' efficiency drop is by using a laser diode (LD). LDs are essentially electrically pumped semiconductors with a *p-n* junction based active laser medium. Owing to the direct proportionality between their input power and resulting PCE, they can achieve high efficiencies. For example, the current state-of-the-art 450 nm LDs have a PCE of ~20% at an input power density of 20 kW/cm^2, which is approximately two fold larger than that of LEDs under the same input power density [7]. Additionally, the color purity of LDs is superior compared to that of LEDs, indicating their wider color gamut, suitable for flat display screens (e.g. liquid crystal displays). Thus, PC-wLDs have a tremendous potential to be widely used for applications in vehicle headlights, street lamps, and display backlights etc. [10–13].

Organic embedding materials (e.g. silicone) which are used in conventional color converters, are not suitable for PC-wLDs due to

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their tendency to degrade under the exposure to heat generated by laser irradiation [7,10,13]. Xie et al. reported a Phosphor-in-Glass (β-Sialon:Eu in ZnO-B_2O_3-BaO-Al_2O_3 glass) material for application in PC-wLDs, as a result polymeric encapsulation materials can be avoided for this purpose. However, when the power density of the laser reached ~1 W/mm^2, an obvious luminescence saturation occurred which led to a significant decrease in the power efficiency of the device [10]. In order to improve the luminescence saturation behavior of the color converter, an Al_2O_3/YAG:Ce composite phosphor ceramic was fabricated, in which a polycrystalline α-Al_2O_3 with a high thermal conductivity (~32 W/m K) was used as a heat transfer medium, and YAG:Ce particles were embedded into it. The Al_2O_3/YAG:Ce composite showed superior thermal stability and zero luminescence saturation, even under the influence of a high laser power density of 50 W/mm [13]. However, the high fabrication costs and difficult processing methods (*i.e.*, spark plasma sintering) can dramatically limit its application and reduce its potential for commercialization.

In this work, a SiO_2-YAG:Ce composite thick film (CTF) was synthesized via a facile and cost-effective spin-coating process. Commercial colloidal silica was used as the source of SiO_2 and played the roles of an adhesive, a filler, and a protecting agent. The microstructure, luminescence, and thermal quenching properties of the CTF were investigated and their luminescence saturation behavior was evaluated using a packaged prototype laser lighting lamp.

2. Experimental procedure

2.1. Preparation

Commercial colloidal silica (SiO_2, 35 wt%, Beijingsaide Co, Ltd), commercial YAG:Ce powder (YH-Y552M), and PVP (Polyvinyl Pyrrolidone, $M_w = 1.3$ M, AR) were used as the starting materials and were weighed in a weight ratio of SiO_2/YAG:Ce/PVP~1/2/0.1 and placed in an Erlenmeyer flask followed by the addition of deionized water. The flask was heated to 65 °C under continuous stirring to completely dissolve the PVP powder. After 6 h, the solution became a viscous sol (~7200 mPa s), with the YAG:Ce particles suspended inside it. The process of spin-coating was employed at 1200 rpm for 30 s to deposit the suspension on the substrates. After coating, the samples were placed in a vacuum oven to impregnate the open pores, and dried at 65 °C for 12 h. Subsequently, they were sintered at 500 °C for 2 h in a muffle furnace at a heating rate of 10 °C/min.

2.2. Characterization

The morphology of the sample was captured using a scanning electron microscope (SEM, Su-70, Hitachi, Japan) and the luminescence was examined using a fluorescence spectrometer (Fls-980, Edinburgh, UK) equipped with a 450 W Xenon lamp and an integrating sphere. The luminescence saturation behavior was evaluated using an intensified multichannel spectrophotometer (Spectro-320, Instrument Systems, Germany). The durability study was conducted in a testing chamber maintained at a constant temperature (85 °C) and humidity (85%) (ASLi, TH-225, China) for a certain time (100–1000 h). The intensity in durability measurement was based on the relative photoluminescence intensity that was examined using the fluorescence spectrometer.

3. Results

Fig. 1 (a) and (b) demonstrate the fabrication procedure of the CTF and its cross-sectional SEM image, respectively. It can be seen that there exists a strong interfacial adhesion between the substrate and the SiO_2-YAG:Ce film which could be ascribed to the adhesion function of the colloidal silica. Generally, the film formed is compact, with a few open pores which is probably formed during the organic removal process. The regions corresponding to both the

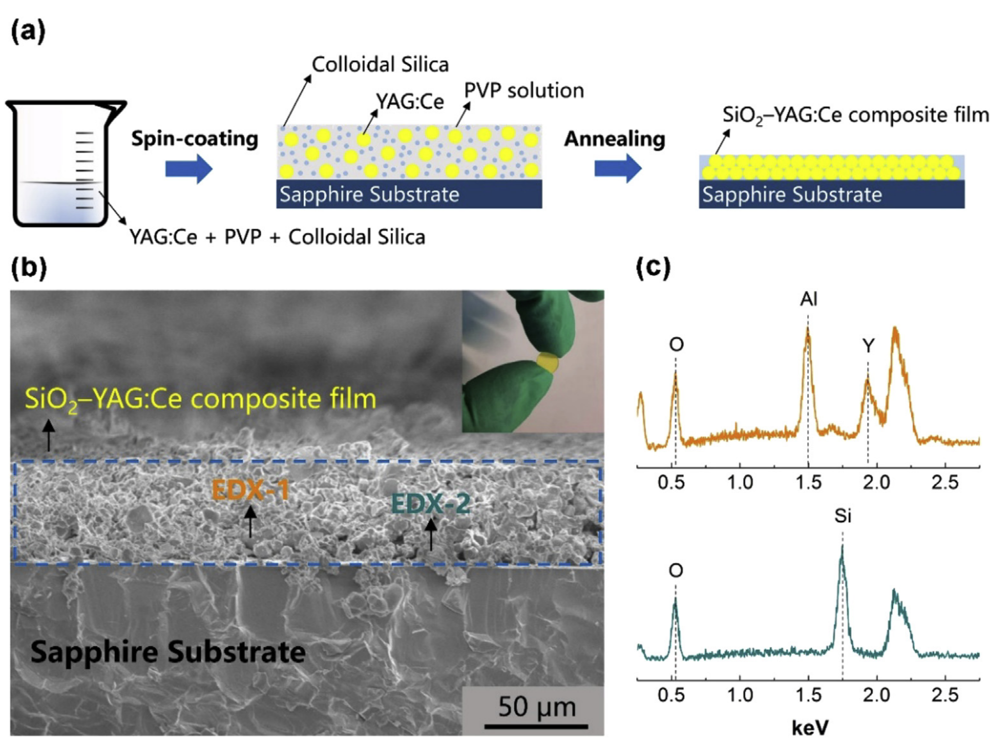

Fig. 1. (a) Schematic diagram depicting the fabrication of the SiO_2-YAG:Ce film; (b) Cross-sectional SEM image of the SiO_2-YAG:Ce, inset is the photograph of the SiO_2-YAG:Ce film; (c) EDX spectra of the positions marked corresponding to (b).