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Preparation and properties of the flexible remote phosphor film for blue chip-based white LED

Jing Jia [a,b], Aiqin Zhang [a,c,*], Dongxin Li [a,b], Xuguang Liu [a,d], Bingshe Xu [a,e], Husheng Jia [a,b,**]

[a] Key laboratory of Interface Science and Engineering in Advanced Materials, Ministry of Education, Taiyuan University of Technology, Taiyuan 030024, PR China
[b] College of Materials Science and Engineering, Taiyuan University of Technology, Taiyuan 030024, PR China
[c] College of Textile Engineering, Taiyuan University of Technology, Taiyuan 030600, PR China
[d] College of Chemistry and Chemical Engineering, Taiyuan University of Technology, Taiyuan 030024, PR China
[e] Shanxi Research Center of Advanced Materials Science and Technology, Taiyuan 030024, PR China

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ABSTRACT

A simple and effective method for larger area flexible and relatively uniform phosphor film layer for blue chip-based white LED is presented in this paper. Besides having a certain ultraviolet resistance and thermal stability, the proposed yttrium aluminum garnet (YAG) phosphor film also achieved applicable color temperature (T_c of 5480 K and 4900 K), color rendering index (CRI of 68.4 and 70) and luminous efficiency (121.7 and 77 lm/W) when simply assembled with single blue chip and high-power COB separately. The preparation process and its test results suggested flexible phosphor film consists of YAG phosphor particles and common silicone without additional converter devices, which has positive practical significance for the packaging of remote phosphor-converted white LEDs.

1. Introduction

White light-emitting diodes (WLEDs), which are regarded as potential replacements in future display and solid-state lighting, have been widely applied in solid-state lighting, outdoor information display, liquid crystal display (LCD) backlighting, landscape lighting and even the automotive field, owing to their superior advantages such as energy saving, long lifetime, free pollution and low weight, etc. [1,2]. Currently, WLEDs can be mainly achieved through light converting techniques which are low-cost, simple and easy to realize industrialization, while two other means, known as multi-chip and quantum well technology, have not reached industrial scale because of high cost, instability and immature technologies [3–5].

One of conventional light converting systems for WLEDs is comprised of blue LED chip and yttrium aluminum garnet (YAG) yellow phosphor excited by blue light, in which process phosphor powders mixed with transparent epoxy resin or silicone are dropped directly onto the surface of blue nitride-based LED chip [see Fig. 1a]. To improve the differences among LEDs such as color deviation caused by uneven distribution of YAG phosphor, surface mounted devices (SMD) LEDs have been developed, in which a caky mixture of phosphors and epoxy resin or phosphor sprayed onto the surface of some kind of substrate is stick on blue chip to generate white light. For instance, Joong-yeon Cho et al. reported the preparation by direct spin-on glass (SOG) printing of nanopatterned yttrium aluminum garnet phosphor-incorporated film to improve the light output power of white LEDs [6]. However, almost 60% of the blue light would be backscattered by the phosphor and lost in the chip because the phosphor is tightly close to the LED die in both of those WLED packages [7,8]. To increase luminance efficiency, some efforts have been made, showing that placing phosphor away from the chip die [Fig. 1b] and using a diffuse reflector cup significantly reduce the absorption of backscattered blue light for LED chip and thus improve the luminance efficiency [9–12]. Moreover, remote phosphor structure can reduce the decline of conversion efficiency and high operating temperature for phosphors resulted from chip heat. Currently, the studies on remote phosphor configuration for phosphor-converted white LEDs used in lighting applications have become popular. Huang et al. proposed a planar lighting system using array of blue LEDs to excite a YAG:Ce^{3+} yellow phosphor remote film fabricated through depositing the slurry of phosphor onto a polyethylene terephthalate (PET) film [13]. Tsai et al. reported a glass phosphor layer with ultra-high thermal stability appropriate for phosphor-converted white light-emitting diodes (PC-WLEDs) [14]. Wang et al. presented a new method for fabricating fluorescent film by the rare-earth phosphor powder filled polycarbonate (PC) resin was prepared

* Corresponding author.
** Corresponding author at: Key Laboratory of Interface Science and Engineering in Advanced Materials, Taiyuan University of Technology, Ministry of Education, No. 79 Yingze Street, Taiyuan 030024, PR China.
E-mail addresses: ZAQ6014567@126.com (A. Zhang), jiahusheng_tyut@163.com (H. Jia).

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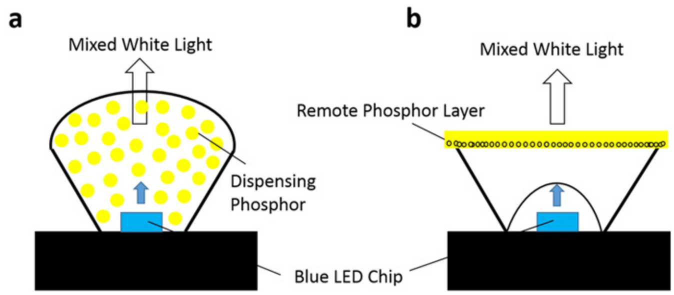

Fig. 1. Schematic diagram of LED packaging. (a) Package of dispensing phosphor, (b) Package of remote phosphor.

by melt-extrusion and rolled into a fluorescent PC film by open mill [15]. Ying et al. concluded that the convex remote phosphor structure had higher luminous efficiency with higher color uniformity than conventional flat remote phosphor structure [16].

Silicone is commonly used for packaging materials with high transparency, good stability, flexibility and good gas-permeability, whereas studies of the remote phosphor layer prepared directly from slurry of phosphor powder and silicone are less. In this work, we demonstrated a simple and efficient process to fabricate large-area and uniform flexible yellow fluorescent film that is applicable for the PC-WLED package such as Flip chip and Chip on Board (COB). Moreover, the optical properties, thermal aging and ultraviolet aging tests of the flexible phosphor film used in LED packaging samples were described in detail.

2. Experiments

2.1. Materials

A Ce-doped yttrium aluminum garnet (YAG-04, Intematix) with excitation wavelength ranging from 430 nm to 490 nm was used in this experiment. As shown in Fig. 2a, the YAG phosphor powders are approximate micro-spherical particles, the mean size (D50) is about 17 μm [Fig. 2b]. Furthermore, adopting polydimethylsiloxane (PDMS, OE-6550, DOW CORNING), tetrahydrofuran (THF, AR), *n*-hexane (*n*-C_6H_{14}, AR) and teflon (PTFE) coagulating mold with a size of 28 × 28 × 1 mm.

2.2. Processing procedure

First, the silicone PDMS glue A with glue B were blended at the ratio of 1:1 in weight and trace THF acting as solvent was injected. Then YAG:Ce phosphor was mixed, which accounted for 12.5% of total quality of silicone, and a little *n*-C_6H_{14} as dispersant was dripped so as to form a uniform phosphor suspension. Subsequently, the uniform mixture was filled into PTFE mold and the small air bubbles inside the phosphor suspension were removed through vacuum pumping for 2 min. Finally, phosphor film was baked and cured at 100 °C for 1 h. For ultraviolet aging with UV wavelength of 272 nm and thermal aging tests, prepared film samples were aged at room temperature and 200 °C for one week, respectively.

2.3. Apparatus

The morphologies of raw YAG phosphor and remote phosphor film were characterized using field emission scanning electron microscope (FE-SEM, JSM-6700F). The surface topography of the phosphor film was measured by atomic force microscope (AFM, SPA-300 HV). UV–vis absorption spectrum was taken using Hitachi-3900 spectrometer. Fluorescence spectra were measured by Horiba Fluoromax4 Spectrometer (the slit width was 1 nm). Electroluminescence (EL) spectra were measured using a computer controlled PMS-50 plus UV–vis-near IR spectrometer with an integrating sphere. The thermal aging and ultraviolet aging tests of remote phosphor film were carried out in heat oven and ultraviolet irradiation lightbox, respectively. The tensile testing was conducted at an electronic universal testing machine with the dumbbell-shaped specimen that had a gauge length of 10 mm, a gauge width of 10 mm and a thickness of 1 mm. All measurements were made at room temperature unless otherwise stated.

3. Results and discussion

3.1. Micro-morphology of the YAG phosphor film

Fig. 3a shows the appearance of the YAG phosphor film prepared by the process proposed in this work. YAG phosphor particles gradually subside into one side of PDMS during the solidification because of gravity and immiscibility. Fig. 3b exhibits the surface topography of the sediment side of phosphor film, which is relatively flat with a surface roughness RMS of 1.35 nm. As can be seen from Fig. 3c and d, phosphor particles are in orderly lateral rows in PDMS and separated with obvious spacing among each other. Compared with the agglomerated raw YAG particles with D50 of 17 μm, the YAG phosphors in prepared phosphor film is just 10 μm or so in average size, confirming that *n*-hexane acts effectively in dispersing YAG phosphor particles. However, the particles accumulate compactly with the layer thickness of about 70 μm in the longitudinal direction. This structure can allow sufficient blue light pass through the prepared phosphor film to produce white light by mixing with converted yellow light since PDMS has good transmittance. On the other hand, it can also scatter the incident blue light efficiently.

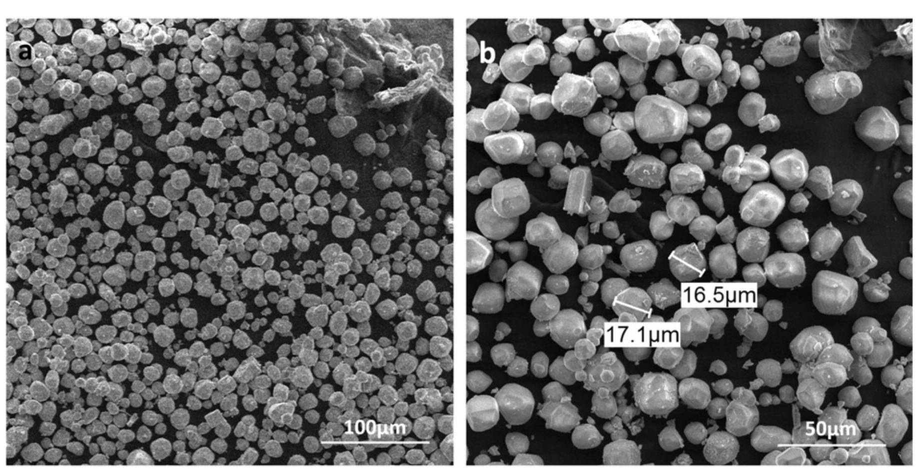

Fig. 2. SEM images of YAG:Ce phosphor.