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Phosphor-in-glasses composites containing light diffusers for high color uniformity of white-light-emitting diodes

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Abstract

ZrO_2 light diffuser layers was placed on phosphor-in-glass composites (PiGs) to improve color uniformity of white-light-emitting diodes (white LEDs). Color coordinates of central part and periphery of white LEDs were (0.34, 0.38) and (0.36, 0.43), respectively; difference between two area decreases with the introduction of a diffuser layer. Color temperature difference also decreased from 770 to 464 K. The addition of diffuser layers reduced the luminous efficacy of the white LEDs from 90.3 to 83.4 lm/W.

Background

Phosphor-based white-light-emitting diodes (white LEDs) provide alternative ways of lighting with high luminous efficacy and stability [1]. They use mixtures of cerium-doped yttrium aluminum garnet (Ce^{3+}:YAG) phosphor powders and epoxy resins that are pasted on blue LED chips. However, these combinations encounter problems such as color properties deviation and degradation of resins, especially in high-power applications [2–4]. Glass materials have been proposed as alternative binders for phosphor powders to alleviate the thermal degradation of polymer resins. Phosphor-in-glass composites (PiGs) have higher efficiencies than do polymer-based mixtures [5–7].

Optical path lengths of the rays through are different depending on the angles between the rays and surface of PiGs; therefore, blue rays that pass through the edge of an LED device have longer path length and experience higher absorption that leads to higher intensity of yellow light than do rays that pass through in the perpendicular direction. As a result, white light rays from one LED chip are close to white at the center, but increasingly yellowish toward the periphery; the result is the 'yellow-ring effect' [8, 9]. This can be reduced with adjusted shape of color converting elements and LED package structure [10–13]. Diffuser layers were used to diffuse blue rays so that optical path lengths in all direction become similar. This adverse effect was alleviated in polymer-based white LEDs by using light diffuser layer coating [14–16].

We used ZrO_2 powders mixed into PiG composites to realize white LEDs with high color uniformity. Commercial Ce^{3+}:YAG phosphors and high refractive-index glass powders were used to prepare PiGs, and ZrO_2 powders were used as light diffusers. This ZrO_2 layer considerably decreased the difference in color temperatures between the center and periphery of the PiG composites.

Methods

Commercial Ce^{3+}:YAG phosphor powders (diameter <40 μm) were mixed in a 1:9 weight proportion with 50PbO-35B_2O_3-15SiO_2 (mol %) glass frits with diameter <5 μm and refractive index of 1.77. These mixtures were further ground using a mortar, and pellets with diameters of 15 mm were prepared by applying 150 MPa uniaxial pressure. For PiG composites containing diffuser powders, an additional layer of glass frits containing ~10 wt.% of ZrO_2 powders was placed underneath the PiG pellets (Fig. 1). They were then sintered together at temperature of 650 °C for 5 h on a glass plate with a thickness of 0.7 mm. Afterwards, specimens were polished to a final thickness of 1.3 mm including the glass substrate of 0.7 mm thickness using abrasive paper and suspension liquid with 1 μm polishing particles, to control transmittance of blue light.

Field emission scanning electron microscope (FE-SEM; XL30S FEG, Philips Electron Optics B.V.) images and energy dispersive spectroscopy (EDS) were used to investigate the distribution of diffuser layers. Total fluxes from blue LEDs and white LEDs were collected by an integrating sphere to examine luminous efficacies and color coordinates. In cases of white LEDs, we first attached color converting elements on blue LEDs to prevent detection of the backward lights. Total transmittance of specimens was measured by comparing flux from LEDs with and without specimens. A UV–Vis-NIR spectrometer (Lambda-750S, Perkin-Elmer) was used to measure the parallel transmittance. Haze parameters were calculated using these two transmittances to obtain the quantity of rays diffracted by the diffuser powder. A hemispherical angle-splitting jig was placed on white LEDs to measure the color properties at different angles. Apertures were controlled at intervals of 10° to detect light that passed through the specific aperture. The size of apertures could not be smaller than 10°, since the intensity of lights from periphery is too low to be detected. However the distributions of color temperatures are continuous so the deviation of measurement from real values is negligible.

Results and discussion

Secondary electron SEM images (Fig. 2a) of PiGs with 10 wt. % ZrO_2 diffusers did not show a clear distinction between the diffuser layers and normal PiG region. However, the diffuser layer with ~200-μm thickness was clearly distinguished in the elemental analysis using EDS (Fig. 2b). Concentration of Pb decreased at the interface between the glass substrates and PiG layers where we added an additional layer containing ZrO_2 powders. Because the molar mass of Ce^{3+}:YAG is approximately five times that of ZrO_2, we could observe a large decrease in Pb concentration even though the frits were mixed with same

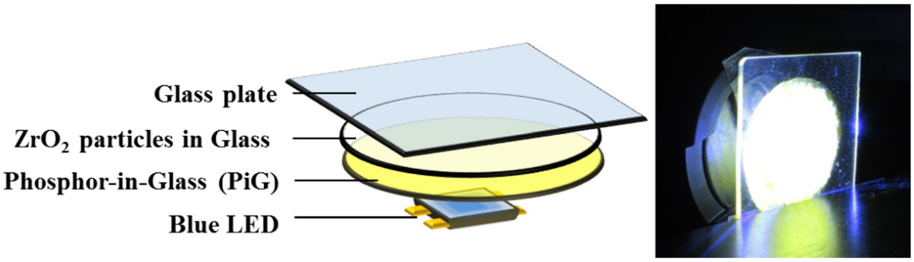

Fig. 1 Configuration of a PiG specimen with a diffuser layer, and a photograph of white light generated from the specimen

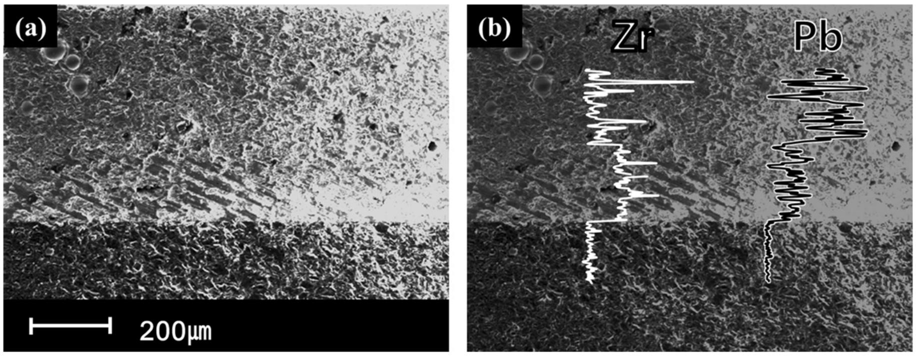

Fig. 2 **a** Secondary electron SEM image of cross section of PiG with a ZrO_2 diffuser layer. **b** Line profiles of Pb and Zr concentration analyzed by EDS

weight percentage in the PiG and diffuser regions; this difference suggests that ZrO_2 diffuser particles remained between the PiGs and glass substrate during sintering.

Specimens containing ZrO_2 diffuser layer and frits without phosphors were prepared to investigate the effect of diffuser layers. Intensities of light passing through specimens at different angles were recorded using the blue LED as a light source. Intensity of light at the periphery of the LED-diffuser system increased when ZrO_2 diffuser layer was included, whereas intensity at the central portion decreased (Fig. 3). These changes reduced the intensity difference between central region and periphery of the white LED device.

PiGs containing both Ce^{3+}:YAG phosphors and ZrO_2 diffuser powders were mounted in front of the blue LED light source. Color coordinates of white light generated from these arrangements were measured at angles θ up to 70° from the perpendicular direction (Fig. 4). White light emerging from the central part of white LEDs ($\theta < 10°$) had color coordinates of (0.34, 0.38) and (0.33, 0.37) for PiGs with and without a diffuser layer, respectively; this difference means that the diffuser layer caused light from the central portion of white LED to became yellowish. Scattering causes greater decrease in the flux of blue light than in the flux of yellow light; therefore the blue spectrum in the middle section of LEDs decreased in intensity and color coordinates moved to the yellow side. In contrast, color coordinates measured at the edge of the system at $\theta = 70°$ were (0.37, 0.45) without a diffuser layer and moved to (0.36, 0.43) when a diffuser layer

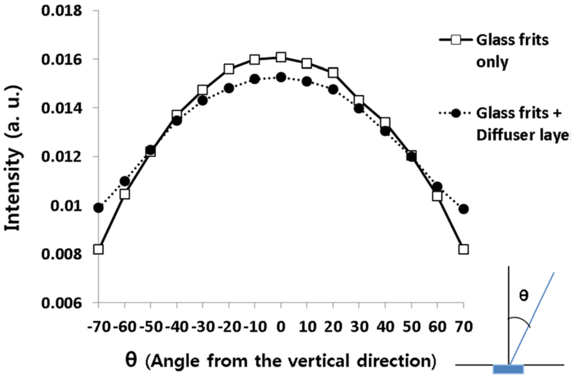

Fig. 3 Angular distribution of intensity of blue light passed through sintered glass frits with and without ZrO_2 diffuser layer. Neither specimen contained Ce^{3+}:YAG phosphors

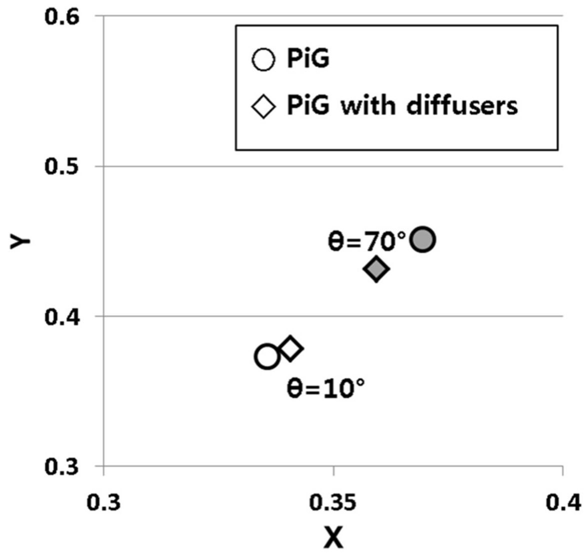

Fig. 4 Color coordinates of white light from white LEDs using normal PiGs and PiGs with diffuser layers measured from the central part ($\theta = 10°$ from perpendicular direction) and at the periphery ($\theta = 70°$)

was added; the result was reduction in the intensities of the yellow rings at the periphery of the white light. Color temperatures of the central part of white light were higher than those at the edge in PiGs both with and without a diffuser layer (Fig. 5). However, the temperature difference between center and periphery decreased from 770 to 464 K while average color temperatures decreased from 5351 to 5221 K, when the diffuser layer was introduced. The haze parameters were measured to investigate relationships between scattering capability of diffuser layer and angular distribution of color. Haze parameters were calculated using following equation:

$$T_{tot} = T_p + T_{diff}, \tag{1}$$

where T_{tot}, T_p and T_{diff} are total transmittance, parallel transmittance and diffusive transmittance, respectively. T_p represents rays that maintain their direction, whereas

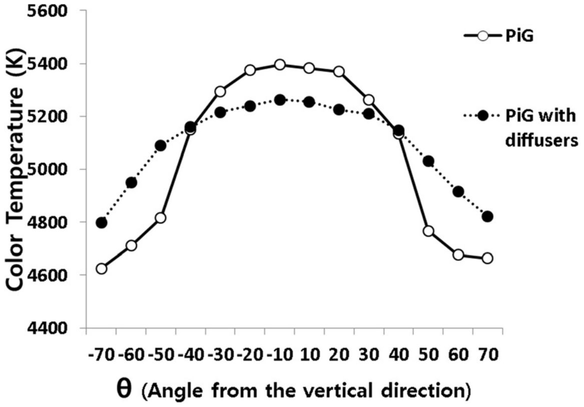

Fig. 5 Angular distribution of color temperatures of white light generated from LED/PiG combinations with diffuser layers

T_{diff} represents rays diffracted from their original direction while passing through the specimens. Haze parameters $\left(H = \frac{T_{diff}}{T_{tot}} \times 100\,\%\right)$ were measured with blue and white LEDs to compare diffusion of blue and yellow light. The specimen that contained 10 wt. % ZrO_2 had $H = 57.2$ % when a blue LED was used as a light source, but $H = 51.7$ % when a white LED was used. Because white light generated from white LED is mixture of yellow and blue light, this result confirms that the amount of scattering is less for the yellow light compared to blue light.

A portion of the blue light from LEDs is absorbed by phosphor particles and converted into yellow light as they pass through the PiG layers. Some of it may be scattered by phosphor particles but remaining portion simply passes through the PiG layer without interaction. Therefore, the light in the central part of white LEDs is bluish compared to the periphery. In contrast, when the white light generated from PiG layer passes through diffuser layer, blue light is scattered more than is yellow light. The scattered blue light are either absorbed by phosphor particles or scattered away from the system. In this way, blue light is distributed throughout the system that led to the light with an improved uniformity.

Luminous efficacies of LED-PiG systems with various diffuser concentrations were measured (Table 1). Luminous efficacy of normal PiGs sintered on glass plates was 90.3 lm/W. As concentrations of ZrO_2 diffuser particles increased to 20 wt. %, luminous efficacy decreased to 78.6 lm/W. Transmittance of specimens that contained a large amount of ZrO_2 was small, but lights scattered by the diffuser layer still seemed to contribute to the total flux. Color temperature differences between center and periphery decreased from 717 to 392 K when the concentration of ZrO_2 increased to 20 wt. %. However, when concentration of ZrO_2 particles exceeds 10 wt. %, overall color temperature begins to decrease and the light becomes yellowish. Therefore 10 wt. % of ZrO_2 particles which was considered as maximum for white light generation was used. Diffuser layers containing 10 wt. % of ZrO_2 used in this work showed a luminous efficacy of 83.4 lm/W.

Conclusions

Phosphor-in-glass composites were synthesized by mixing Ce^{3+}:YAG phosphors and commercial glass frits; 10 wt. % of ZrO_2 diffuser particles were mixed with glass frits and co-sintered to yield a diffuser layer thickness of ~200 μm. Color coordinates of white LEDs using PiGs with diffuser layers were (0.34, 0.38) when measured at the vertical angle and (0.36, 0.43) at the periphery. Since the normal PiG-based white LEDs without ZrO_2 diffuser layer had color coordinates of (0.33, 0.37) and (0.37, 0.45); the difference between center and periphery decreased when the diffuser was used. Color temperature differences between these two areas decreased from 770 to 464 K. Addition of 10 wt. % of ZrO_2 diffuser decreased the luminous efficacy of white LEDs from 90.3 to 83.4 lm/W.

Table 1 Luminous efficacies of white LEDs using PiGs containing various concentrations of diffuser layers

ZrO_2 concentration (wt. %)	0	5	10	15	20
Luminous efficacy (lm/W)	90.3	86.0	83.4	82.7	78.6

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

SY carried out the preparation of PiG specimens, measurements and analysis of optical properties of PiGs and white LEDs, and drafted the manuscript. WJC participated in the design of the study and interpretation of data. JH participated in the design of the study, revised the manuscript and gave final approval of this version. All authors read and approved the final manuscript.

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