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Optical properties of phosphor-in-glass through modification of pore properties for LED packaging

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ABSTRACT

The volume and size of the voids present between the frit and the phosphor particles used before sintering determine the pore properties of the resulting phosphor-in-glass (PIG). The pores formed from the voids influence the path of the incident light, thus changing the optical properties of the PIG. Therefore, the trends observed for the shrinkage and the green and sintered densities of the PIG were investigated using SiO_2-B_2O_3-ZnO-K_2O glass frit of four sizes to understand the tendency for the pore size, porosity, and optical properties of PIG. It has been demonstrated that variation in the pore properties according to the particle size influences parameters defining the light scattering phenomenon, such as the scattering angle of the light and the scattering coefficient, as well as the color rendering index, correlated color temperature, and package efficacy. The results obtained for the variation in the optical properties with the frit size can be used as a reference to select the appropriate glass frit size to achieve the required optical properties for a light-emitting diode (LED) package.

1. Introduction

White light-emitting diodes (WLEDs) have attracted increasing attention because of their advantages, such as high efficacy, environmental friendliness, and low electricity consumption [1–3]. LEDs are being applied not only for interior lighting but also in medical equipment, automotive headlamps, and outdoor lamps [4–6]. Glass encapsulants with good chemical, mechanical, and thermal properties have been replacing conventional encapsulant materials such as resins and silicone to protect the LED chips from the external environment and to ensure their long life and reliability [7,8]. In particular, phosphor-in-glass (PIG) is advantageous as the desired color can be easily achieved by mixing different phosphors and the efficacy can be improved by applying the PIG to the remote type packages [2,4,6].

Several studies have been conducted to utilize the advantages of PIG and to improve the efficacy of LED packages [4,6–9]. Various processes, such as screen printing [6], spin coating [8], and tape casting [9], have been used for the preparation of PIGs, and the structure of PIG has been modified to prevent reabsorption between the phosphors [10]. Studies have also been conducted to improve the color uniformity of the emitted light using additives as light diffusers [11,12]. In particular, the importance of the relationship between the microstructure and the optical properties was confirmed by observing the effect of the pore characteristics inside the plate on the transmittance and luminous efficacy of the PIG [13].

However, a fundamental study of the trends in the pore properties and optical properties of the PIG based on the particle size of the glass frit has not been carried out so far. The size of the pores in the glass plate is affected by the presence of an additional void volume between the particles after sintering [14]. The changes in the pore size affect the scattering and traveling direction of light, thereby changing the transmittance and light absorption of the phosphor [13,15]. On the other hand, it has been confirmed that even if the emission intensity of the phosphor itself changes with the size of the phosphor particle [16], the change in the particle size of the phosphor affects the morphology of the PIG when the same glass frit is used, thereby causing leveling-off of the luminous efficacy [17]. Therefore, it is necessary to study the relationship between the changes in the glass frit size and the pore properties as it significantly affects the optical properties of the PIG [13,15].

In order to understand the variations in the microstructure and the optical properties with the frit size, the relationship between the internal pore properties and the optical properties of the PIG was investigated using glass frit of four sizes based on the D_{50}

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particle size of the YAG phosphor. It was confirmed that the scattering angle of the incident blue light and the scattering coefficient based on the pore properties are important factors in improving the luminous efficacy. Furthermore, the pore properties of the plate affect not only the emission intensity of the light but also the color rendering index (CRI) and correlated color temperature (CCT) of the PIG. The purpose of this study is to investigate the effect of the particle size on the microstructure and optical properties of the PIG in order to achieve a higher efficacy and desired chromaticity using the same glass composition and phosphor.

2. Experimental procedure

The $18SiO_2$-$35B_2O_3$-$42ZnO$-$5K_2O$ glass frit (mol%) was selected as the PIG matrix because of its low glass transition temperature (T_g) of 450 °C and low glass softening temperature (T_s) of 580 °C. The particle size distribution of the glass frit was varied using a stainless steel sieve with four mesh sizes (20, 25, 38, and 46 μm; Nonaka Rikaki Co. Ltd., Japan). The specific surface area of each screened glass frit was determined by Brunauer–Emmett–Teller (BET) analysis (ASAP 2020; Micromeritics, USA). YAG phosphor ($Y_3Al_5O_{12}$:Ce^{3+}; FORCE4 Co. Ltd., Korea) with a peak wavelength of 553 nm was used to generate a white light with blue light from the LED chip. The particle size of each screened glass frit and phosphor was measured using a particle size analyzer (Mastersizer 2000; Malvern, UK) (Table 1).

Each screened glass frit was mixed with 5 wt% of YAG phosphor using a tubular shaker mixer (T2F; Willy A. Bachofen, Switzerland) for 48 h at 300 rpm. Glass and PIG samples were classified from G1 to G4 and P1 to P4, respectively, based on the glass frit size (Table 1). The shrinkages of the glass frit and the phosphor-containing glass frit were analyzed by hot stage microscopy (HSM, Misura HSM; Expert System Solutions Inc., Italy). The green body density of the glass frit and the phosphor-containing glass frit was measured using a pellet pressed with a metal mold (Hantech Inc., Korea) of diameter 25 mm, and then the pellets of the glass frit and the phosphor-containing glass frit were sintered in an electric furnace at 580 °C for 10 min at a heating rate of 10 °C/min. The sintered density of both the glass plate and the PIG were measured using a pycnometer (AccuPyc II 1340; Micromeritics, USA), and then they were polished to a thickness of 500 μm for analyzing the optical properties.

The total transmittance of the glass plate and the PIG was measured using a UV–visible spectrophotometer (UV 2450; Shimadzu Corp., Japan), and the CRI, CCT, electroluminescence (EL) spectra, and luminous efficacy (lm/W) were measured using a spectroradiometer with an integrating sphere with a diameter of 50 cm (GS-1290-3 Spectroradiometer; Gamma Scientific, USA). The PIG was placed at a certain distance from the blue LED chip using a reflector holder placed between the LED chip and the PIG. The middle of the PIG plates was cut to observe the cross-section, and cross-sectional images of the glass plate and PIG were obtained using scanning electron microscopy (SEM, S-4300; Hitachi, Japan). Pore properties such as the pore size and porosity were analyzed using the images obtained (analysis area of about 6400 × 500 μm) using the image analysis software Image-Pro Plus (Ver. 6.0, Media Cybernetics Inc., USA). The scattering coefficient of the glass plate and PIG was calculated by the effective scattering coefficient (C_{sca}) using the analyzed pore properties, and the scattering length was defined as the inverse of the scattering coefficient (C_{sca}^{-1}) [18–20].

3. Results and discussion

A smaller particle size of the glass frit, G1 and P1, showed a higher shrinkage rate at a sintering temperature of 580 °C (Fig. 1). Glass frit and phosphor-containing glass frit contain voids of different sizes between the particles depending on the frit size, and the voids exist as pores after sintering [21,22]. Therefore, the properties of the pores existing inside the sintered plate vary according to the particle size of the glass frit [21]. Additionally, a reduction in the pore size can be expected when using a small size of the glass frit, and increase in the pore size can be expected with decrease in the shrinkage rate when phosphor is added to the glass frit to form a PIG.

Both the glass frit and the phosphor-containing glass frit tended to have a lower green density as the particle size decreased, but an improved sintered density was observed after sintering approximately at the softening temperature of 580 °C for 10 min (Fig. 2). The increase in the shrinkage rate due to the decrease in the particle size of the glass frit affected the improvement in the sintered density. The green density of the phosphor-containing glass frit was similar to that of the glass frit; however, the sintered density of the

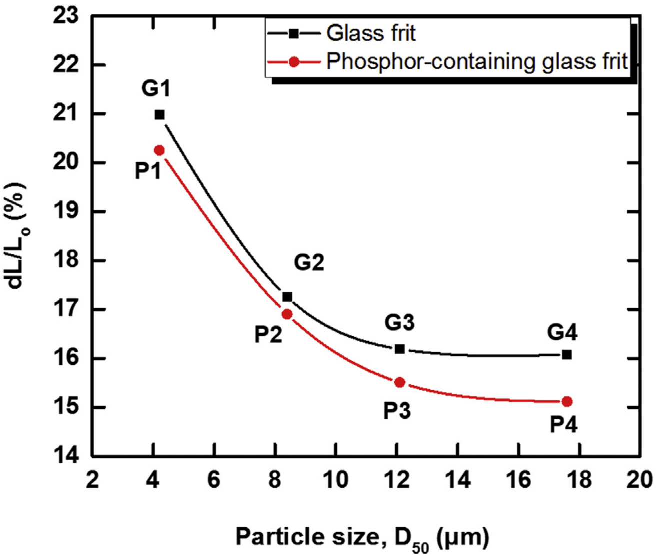

Fig. 1. Linear shrinkages of the glass frit and the phosphor-containing glass frit based on the particle size.

Table 1
Particle sizes of the glass frit and phosphor used for the glass plates and PIGs.

Samples		Particle size (μm)				PIG samples (5wt% of YAG)
		D (10)	D (50)	D (90)	D (max)	
Glass frit	G1	1.6	4.2	7.5	11.6	P1
	G2	1.6	8.4	23.3	45.1	P2
	G3	1.7	12.1	32.8	61.0	P3
	G4	2.0	17.6	51.5	96.3	P4
Phosphor	YAG:Ce^{3+}	10.0	15.4	23.6	38.7	–