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Innovative Thin PiG Plates Boost the Luminous Efficacy and Reliability of WLEDs for Vehicles

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Abstract: In this study, we demonstrate the high luminous efficacy of 118 lm/W and the high reliability of white LEDs (WLEDs) through 450 °C thermal aging, utilizing four-inch YAG: Ce³⁺ phosphor-in-glass (PiG) plates designed for vehicle headlights. The sintering process of mixing glass and phosphor typically generates pores, which can scatter light and reduce the luminous efficacy of the fabricated PiG. In this study, we produced four-inch PiG plates under four different fabrication conditions to evaluate their luminous efficacy. Our results revealed that the PiG plate with a thin thickness of 0.08 mm exhibited a 16.83% increase in luminous efficacy compared to the 0.15 mm plate, attributed to reduced light interaction with the pores. Unlike silicone-based phosphor WLEDs, which offer high performance but lower reliability due to the silicone resin's low transition temperature (150 °C), our novel thin PiG plate achieves high performance and reliability. This advancement suggests that the proposed thin PiG plate could replace traditional silicone-based phosphors, enabling the development of high-quality WLEDs for vehicle headlights in automotive applications.

Keywords: phosphor-in-glass; white LED; thin plate

Citation: Huang, H.-W.; Huang, C.-W.; Chen, Y.-C.; Cheng, W.-C.; Liu, C.-N.; Chiang, C.-C. Innovative Thin PiG Plates Boost the Luminous Efficacy and Reliability of WLEDs for Vehicles. *Ceramics* **2024**, *7*, 1147–1158. <https://doi.org/10.3390/ceramics7030075>

Academic Editor: Francesco Baino

Received: 18 July 2024

Revised: 18 August 2024

Accepted: 20 August 2024

Published: 26 August 2024



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1. Introduction

High-power phosphor-converted white LEDs (pc-WLEDs) are increasingly in demand due to their low cost, reliability, and high luminous efficacy. These remarkable properties enable pc-WLEDs to quickly replace traditional light sources in interior lighting, exterior lighting, and automotive headlights [1–3]. In particular, the reliability of the high-power pc-WLEDs is crucial for automotive headlight applications due to driving safety considerations [4,5]. However, the traditional method of encapsulating pc-WLEDs is to embed phosphor in a silicone resin. Silicone resin has poor thermal stability and is prone to yellowing and cracking, which will affect the optical characteristics and service life of the pc-WLEDs [6,7]. Currently, there are many studies using ceramic, single crystal, and glass-based inorganic materials instead of the silicone resin to solve the thermal stability problem [1,8,9]. According to studies, these high-temperature fabrications are difficult to apply to commercial production because the fabrication temperatures of the ceramic-based phosphor and single crystal-based phosphor exceed 1500 °C and 1900 °C, respectively [9]. Therefore, the development of the phosphor-in-glass (PiG)-based pc-WLED for automotive headlight applications is necessary. However, due to the voids present between the powder particles, pores are always generated when the mixture of the glass and phosphor is sintered, and the formed pores cause light scattering in the obtained PiG, resulting in the luminous efficacy of the PiG being lower than that of the silicone-

based phosphor. It is essential to study the possibility using the PiG to replace silicone-based phosphor [10–12]. Recently, the high-performance and high-reliability of the two-inch PiG for pc-WLED, employing a novel wet-type cold isostatic pressing (CIP), have been reported [12]. In a previous report, a novel wet-type CIP was used to reduce the pores in the PiG. However, there is still a certain degree of pores in the PiG, which deteriorates the characteristics of the PiG. Also, a two-inch diameter PiG for automotive headlight applications may not meet economical size requirements for commercial use. Therefore, there is a need to conduct research on a large-size PiG and reduce the interaction between pores and light to develop high-performance and high-reliability four-inch $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ (YAG: Ce^{3+}) PiG to further increase the adoption of the PiG in the solid-state lighting market.

In this study, high-performance and high-reliability WLEDs employing four-inch YAG: Ce^{3+} PiG plates for vehicle headlights are presented and demonstrated. A glass matrix ($\text{SiO}_2\text{-Na}_2\text{O-Al}_2\text{O}_3\text{-CaO}$) has high transparency and excellent thermal stability, so that the obtained PiG exhibits satisfactory optical properties and thermal performance durability [13]. Through precisely adjusting the thickness of the four golden formula four-inch PiG plates to 0.08, 0.10, 0.12, and 0.15 mm, the four golden formula four-inch PiG plates have similar chromaticity and correlated-color temperature (CCT) when comparing the differences in their luminous efficacy. This study found that the luminous efficacy of the PiG plate with a thickness of 0.08 mm was 16.83% higher than that of 0.15 mm due to the reduced interaction between the pores and light. Finally, the four-inch PiG plates with a thickness of 0.08 mm were verified by an accelerated aging test at a stress temperature of 450 °C and 1008 h. As a result, the International Commission on Illumination (CIE) shift was 4.9×10^{-3} , and the lumen loss was 1.7%. This indicates that the PiG plates also have high thermal stability due to the high transition temperature of the glass matrix up to 550 °C. At present, high-performance silicone-based phosphor WLEDs have been commercially used in vehicle headlights, but due to the low transition temperature (150 °C) of silicone resin, its reliability is low. In contrast to silicone-based phosphor WLEDs, this study presents a novel thin PiG plate with both high-performance and high-reliability WLEDs for vehicle headlights. With the benefits of the proposed thin PiG plate, an opportunity to replace traditional silicone-based phosphors may be provided, to achieve high-quality WLEDs for vehicle headlights in automotive applications.

2. Experimental Methodology

2.1. Manufacturing the Phosphor-in-Glass

A glass matrix (D263, Schott AG, Jena, Germany) composed of 60 mol% SiO_2 , 25 mol% Na_2CO_3 , 9 mol% Al_2O_3 and 6 mol% CaO was mixed uniformly, then heated and melted at 1300 °C, and then gradually cooled to room temperature. The resulting cullet was ground into glass powder and then sieved to a particle size of about 10 μm . The yellow luminescence center was YAG: Ce^{3+} phosphor (NYAG4454, Intematrix Corp., Fremont, CA, USA), and its particle size was about 15 μm . Then, YAG: Ce^{3+} phosphor with four different weight ratios were uniformly spread out into the glass powder under atmospheric pressure. A wet-type CIP technique was performed to form a precursor with a high-uniform bulk density, to be sintered at a temperature of 700 °C for 30 min. Finally, the four fabricated conditions of the four-inch YAG: Ce^{3+} PiG bulks were sliced to the thicknesses of 0.08, 0.10, 0.12, and 0.15 mm. In this study, a total of four four-inch PiG fabrication conditions were generated. The YAG: Ce^{3+} phosphor weight ratio and thickness of PiG-0.08 mm, PiG-0.10 mm, PiG-0.12 mm, PiG-0.15 mm are 30 wt% and 0.08 mm, 24 wt% and 0.10 mm, 20 wt% and 0.12 mm, and 16 wt % and 0.15 mm, respectively. The uniformly mixed phosphor–glass powders were subjected to a wet-type cold isostatic pressing (CIP) technique under atmospheric pressure. This process formed a precursor with high uniform bulk density, ensuring homogeneity and compactness. The formed precursor was sintered at 700 °C for 30 min, allowing the glass matrix to encapsulate the phosphor

particles and form a dense and uniform PiG composite. The PiG composites were sliced into wafers with different thicknesses. These wafers were subjected to a polishing process to achieve the desired thicknesses. During polishing, the material hardness of the PiG was carefully considered, and the powder ratio design in the fabrication process was optimized to ensure uniformity and mechanical integrity across the different thicknesses. Table 1 lists the four fabricated conditions of the four-inch PiG. Figures 1a–d show a four-inch PiG bulk, a PiG plate, PiG chips on a blue tape for placement, and a WLED package with PiG chips. Figure 1a shows the PiG powder that has been sintered into a bulk crystal. As depicted in Figure 1b, the bulk crystal was sliced using wire cutting techniques and placed into a cutting mold. The crystal was cut at equal intervals to achieve a thickness of approximately 160 μm . The plates are polished to achieve the desired thicknesses, ensuring uniformity across the four different thicknesses of the mixed materials. In Figure 1c, the polished PiG wafers were mounted onto a support film. The wafers were used to laser into individual dies, which were prepared for subsequent packaging processes. Figure 1d illustrates a designed PCB (Printed Circuit Board) with both thermal management and circuitry. The PiG chips of different thicknesses were selected and packaged onto the PCB equipped with blue LEDs. The chips were encapsulated onto the light-emitting components using a white adhesive. This encapsulating material is designed to withstand high temperatures up to 320 $^{\circ}\text{C}$, ensuring the durability of the package.

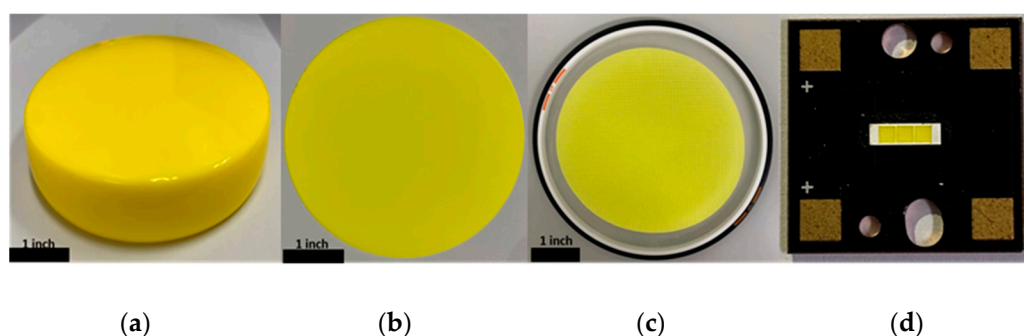


Figure 1. The related photos of a diameter of four-inch PiG. (a) Bulk; (b) Plate; (c) Die cutting, and (d) Package.

Table 1. Four fabricated conditions of the four-inch PiG.

Formula	PiG-0.08 mm	PiG-0.10 mm	PiG-0.12 mm	PiG-0.15 mm
Amount of mixture (g)	500	500	500	500
Amount of glass matrix (g)	350	380	400	420
Amount of YAG: Ce^{3+} (g)	150	120	100	80
Sintering temperature ($^{\circ}\text{C}$)	700	700	700	700
Diameter (inch)	4	4	4	4
Thickness of PiG bulk/plate (mm)	25/0.08	25/0.10	25/0.12	25/0.15

This study investigates the effects of varying phosphor-to-glass ratios across different thicknesses, aimed at exploring the intrinsic photoluminescent properties and emission intensity of the phosphor sheets, both in their own right and when excited by a blue LED, under consistent CIE and CCT conditions.

2.2. Measurements of Phosphor-in-Glass

The glass transition temperature and melting temperature of the glass powder were measured by a differential thermal analysis (DTA, PerkinElmer STA 6000). Use a fluorescence spectrometer (HITACHI F-4500) to evaluate the photoluminescence excitation spectroscopy and light emission spectrum of the yellow YAG: Ce^{3+} phosphor. The crystalline

phase of the four-inch PiG powder was evaluated by an X-ray diffraction analyzer (XRD, BRUKER D8 SSS). An integrating sphere measurement system (Isuzu OPTICS ISM-360 series) was used to measure the light emission spectrum, the chromaticity coordinates of the International Commission on Illumination (CIE), the luminous efficacy, and the CCT of the four-inch PiG plates. A spectrophotometer (Hitachi U3900-H) was used to measure the transmittance of the four-inch PiG plates with the thicknesses of 0.08 and 0.15 mm, respectively. A FE-SEM (JEOL JSM-7800F) was used to determine the porosity on the surface of the four-inch PiG plate.

2.3. Thermal Stress Aging Tests

The thermal stability of the four-inch PiG plates with a thickness of 0.08 mm was evaluated by thermal aging test. The four-inch PiG plates were aged at 150 °C, 250 °C, 350 °C, and 450 °C for 1008 h. The optical properties, including chromaticity coordinates and luminous efficacy, were characterized to assess the thermal stability of the four-inch PiG plates before and after the thermal aging test. To further analyze the relationship between LED driving energy and the lifetime of the phosphors, the PiG plates with varying thicknesses were subjected to cyclic LED driving conditions. The LED driving energy was varied to observe its effect on the degradation rate and lifespan of the phosphors. The cyclic lifetime was determined by monitoring the optical properties over repeated cycles of LED operation. The relationship between LED driving energy and phosphor degradation was analyzed, providing insights into the durability and performance of the phosphor materials under different operating conditions.

3. Results and Discussion

As shown in Figure 2, the DTA curve of the glass powder has been measured by a differential thermal analysis to show the glass transition temperature (T_g) and melting temperature (T_m) of the glass matrix. Figure 2 shows that the T_g and T_m are 557 °C and 603 °C, respectively. Therefore, a sintering temperature of 700 °C in this study may be suitable for the fabrication of the four-inch PiG. The photoluminescence excitation (PLE) spectrum and emission spectrum of the YAG: Ce³⁺ phosphor have been measured by a fluorescence spectrometer, as shown in Figure 3. Figure 3 shows the excitation and emission spectrum of the YAG: Ce³⁺ phosphor with peaks at 450 nm and 563 nm, respectively. The mixture of the two will present white light due to the yellow light and blue light being complementary colors in chromatics. The XRD patterns of the four-inch PiG powder at a sintering temperature of 700 °C was determined by an X-ray diffraction analyzer. As shown in Figure 4, all the diffraction peaks were in accordance with the JCPDs card no. 33-0040 pattern, indicating no crystallization of the glass phase was found. The presence of peaks with broader full width at half maximum (FWHM) or lower intensity can be primarily attributed to interactions between the glass particles and the phosphor, which may alter the diffraction pattern.

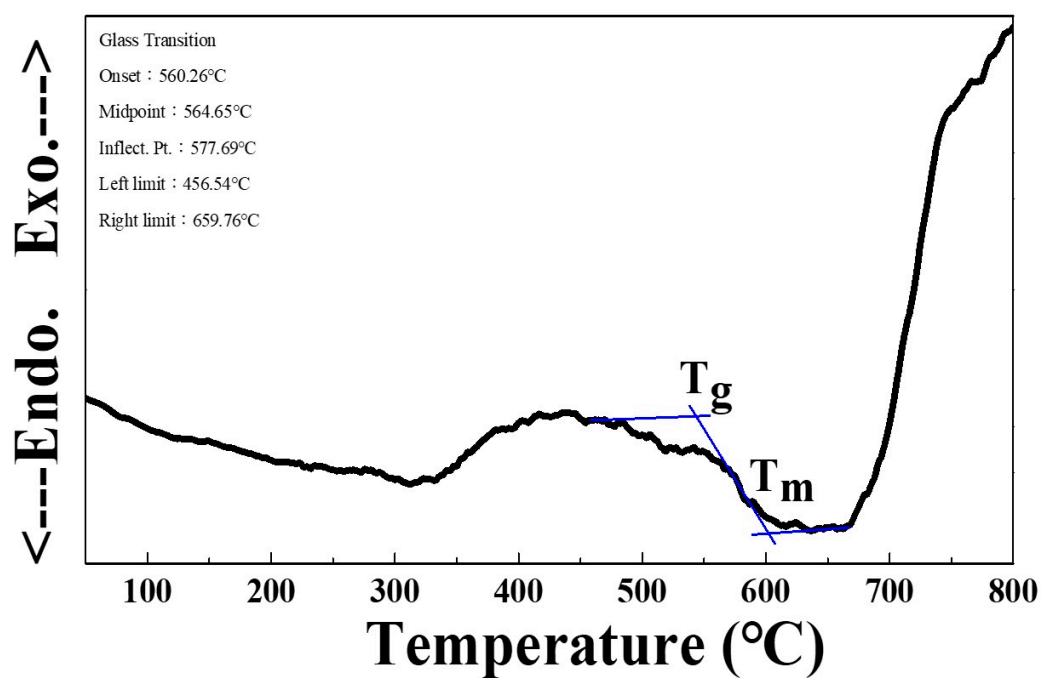


Figure 2. DTA thermogram of the glass powder; The data for the glass transition temperature (T_g) and melting temperature (T_m) were obtained through the tangents of the slopes of the curves.

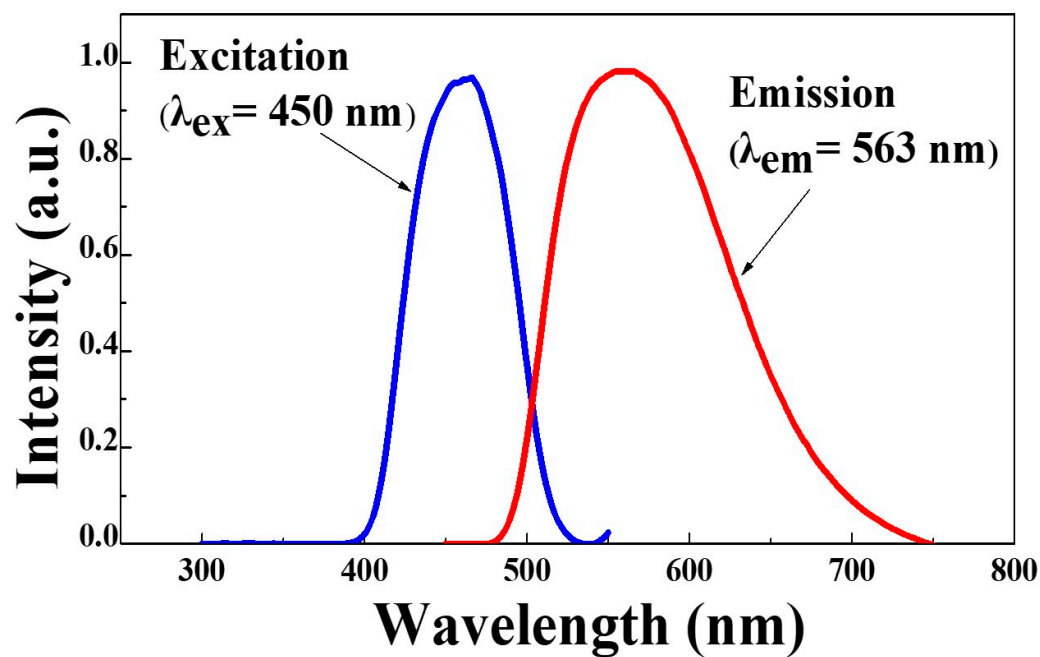


Figure 3. PLE and emission spectrum of the YAG: Ce^{3+} phosphor.

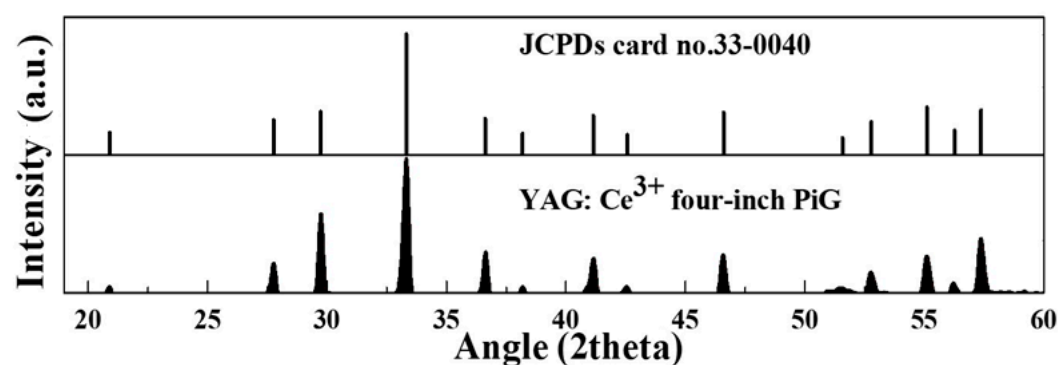


Figure 4. XRD patterns of the four-inch PiG powder.

In this study, a total of four four-inch PiG fabrication conditions were generated to develop high-performance and high-reliability four-inch YAG: Ce³⁺ PiG for automotive headlight applications. Through precisely adjusting the thickness of the four golden formula four-inch PiG plates to 0.08, 0.10, 0.12, and 0.15 mm, the four golden formula four-inch PiG plates have similar chromaticity and CCT, as shown in Tables 1 and 2 and Figure 5. Table 2 lists the optical properties of the four fabricated condition four-inch PiG plates, which were excited by a 450 nm blue LED chip. The high luminous efficacy was 118 lm/W for the PiG plates with a thickness of 0.08 mm. Table 2 shows that the luminous efficacy of the four-inch PiG plates decreases as thickness increases. The luminous efficacy of the PiG plate with a thickness of 0.08 mm was about 16.83% higher than that of the PiG plate with a thickness of 0.15 mm.

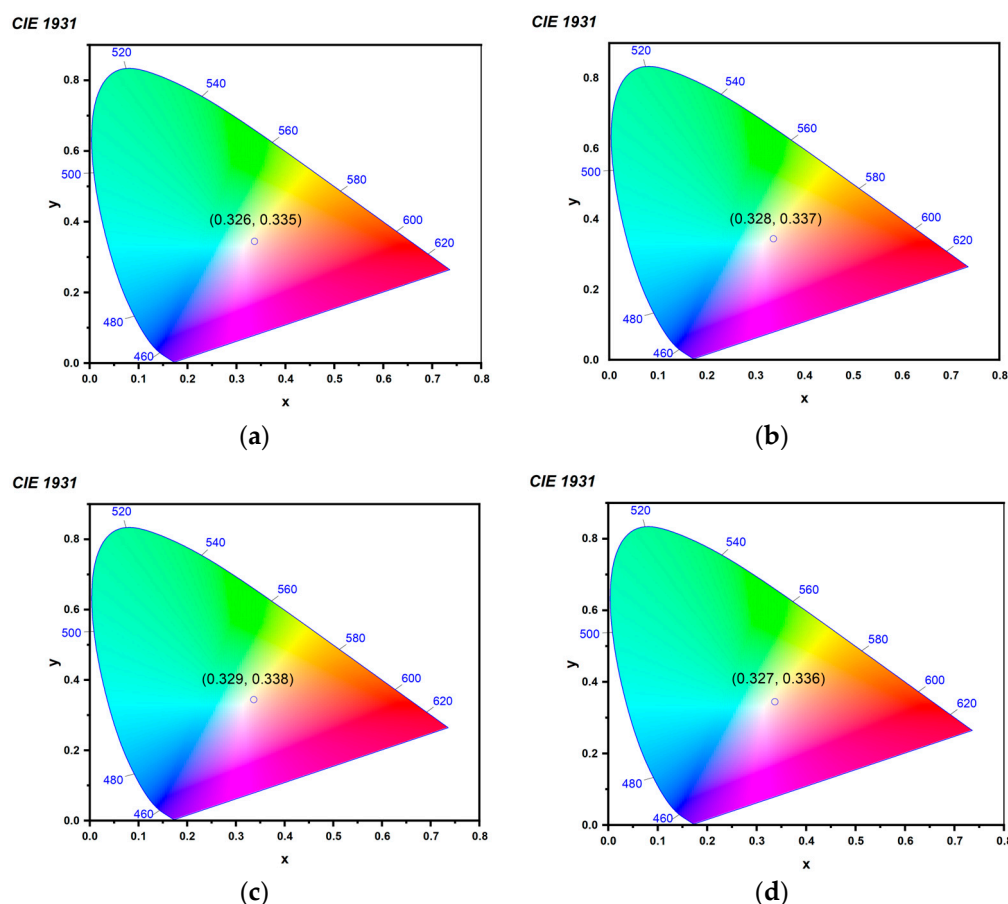


Figure 5. The CCT image of different thickness in the YAG:Ce³⁺; (a) 0.15 mm, (b) 0.12 mm, (c) 0.10 mm and (d) 0.08 mm. By adjusting the phosphor-to-glass powder ratio in fluorescent plates of

varying thicknesses, the emitted light color was confirmed to remain consistent.

Table 2. Optical properties of the four fabricated condition four-inch PiG plates.

Formula	Thickness (mm)	Light up	CIE (x, y)	CCT (K)	Luminous Efficacy (lm/W)	EQE (%)
PiG-0.15 mm	0.15		(0.326, 0.335)	5700 K $\pm$ 300	101	20.57
PiG-0.12 mm	0.12		(0.328, 0.337)	5700 K $\pm$ 300	105	20.62
PiG-0.10 mm	0.10		(0.329, 0.338)	5700 K $\pm$ 300	111	21.39
PiG-0.08 mm	0.08		(0.327, 0.336)	5700 K $\pm$ 300	118	21.47

A comparison of External Quantum Efficiency (EQE) revealed that a PiG plate with a thickness of 0.08 mm exhibited approximately 1% higher efficiency than one with a thickness of 0.15 mm. This study investigates the optical properties of PiG plates of varying thicknesses, maintaining the same CCT and CIE values by altering the phosphor ratio to glass powders. Regarding EQE and luminous efficiency, thicker phosphor layers facilitate multiple absorption and scattering events as photons pass through the material. This increased interaction may reduce quantum efficiency, as some photons might be reabsorbed instead of successfully escaping the material. Furthermore, increased thickness can lead to more significant optical losses, reducing luminous efficiency. In contrast, thinner phosphor layers with a higher proportion of phosphor can potentially enhance quantum efficiency, as the probability of photon absorption and subsequent luminescence is higher. The reduced proportion of glass powder minimizes scattering and absorption losses, potentially improving luminous efficiency. However, if the glass powder proportion is too low, it might compromise the material's stability and other optical properties, adversely affecting overall light output efficiency. This issue can be further explored through aging or environmental testing.

Therefore, the thickness of the four-inch PiG plate was significantly affecting the optical characteristics and needs further study. Based on the results in Table 2, we performed further analysis to determine the cause of the low luminous efficacy for four-inch PiG plates with a thickness of 0.15 mm. The results are shown in Figures 6–8 and Table 3.

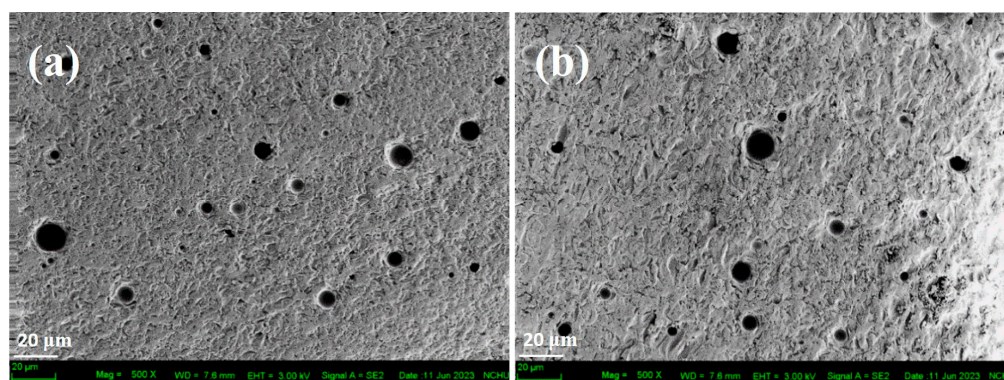


Figure 6. FE-SEM micrographs of the PiG plates with the thickness of (a) 0.08 mm and (b) 0.15 mm.

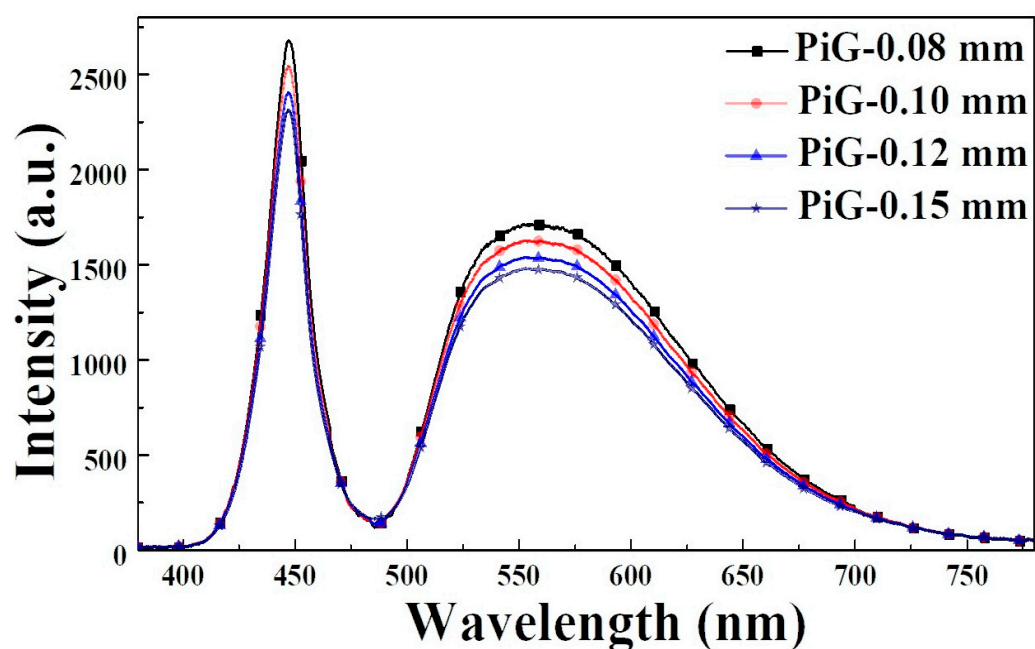


Figure 7. Emission spectrum of the four fabricated condition four-inch PiG plates.

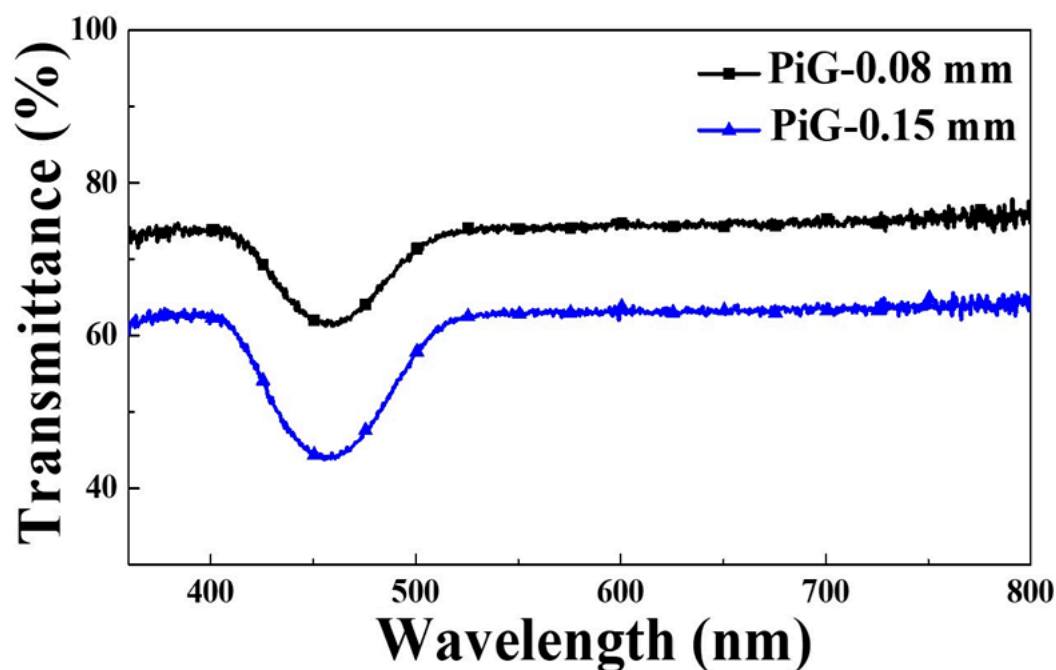


Figure 8. Measured transmittance of the PiG plates with the thickness of 0.08 mm and 0.15 mm.

Table 3. Porosity of the four fabricated condition four-inch PiG plates.

Formula	PiG-0.08 mm	PiG-0.10 mm	PiG-0.12 mm	PiG-0.15 mm
Porosity of four-inch PiG (%)	1.34	1.37	1.38	1.33

The microstructure measurements of the four-inch PiG plates are shown in Figure 6 and Table 3. Figure 6 shows FE-SEM micrographs of the four-inch PiG plates with the thicknesses of (a) 0.08 mm and (b) 0.15 mm, respectively. As shown in Table 3, the porosity of the four-inch PiG plates with the thicknesses of 0.08 mm, 0.10 mm, 0.12 mm, and 0.15 mm, estimated from the micrographs, are in the range of 1.34%, 1.37%, 1.38%, and 1.33%,

respectively. From the measurement results, there was no significant difference in the porosity of the four-inch PiG plates under the four fabrication conditions. However, the number of pores increases as the thickness increases. This indicates that in the PiG plate, the interaction between the pores and light increase, resulting in more light scattering, which in turn affects the optical performance of the PiG plate [11]. This may be one of the reasons why the luminous efficacy of the PiG plate with a thickness of 0.15 mm is lower than that of the PiG plate with a thickness of 0.08 mm. Figure 6 depicts the light emission spectrum of the four-inch PiG plates. The peak intensities of the emission spectrum at 450 nm and 563 nm decrease as the thickness of the PiG plate increases. It can be clearly seen that the luminous efficacy of the PiG plate will also decrease with the increase in the thickness of the PiG plate. As a result, the PiG plate with a thickness of 0.08 mm for vehicle headlight applications obtained significantly improved luminous efficacy due to the reduced interaction between the pores and light. This was consistent with the result of Table 2. Figure 8 shows the measured transmittance of the PiG plates with the thicknesses of 0.08 mm and 0.15 mm. This was due to the number of pores, which increased as the thickness of the PiG plate increased. These induced an interaction between the pores and light, resulting in more light scattering. Moreover, several factors influence the glass powder's optical properties. Firstly, the refractive index mismatch between the glass and the phosphor particles leads to reflection and refraction at the interface, causing partial absorption or scattering of light. Secondly, interface defects or irregularities introduced during the bonding process can act as scattering centers, further increasing light absorption. Additionally, suppose the absorption spectrum of the glass overlaps with the emission spectrum of the phosphor. In that case, the glass may absorb some of the emitted light, thereby reducing the overall light output efficiency. Photon reabsorption phenomena may also occur, where specific wavelengths of photons emitted by the phosphor are reabsorbed as they pass through the glass material, leading to a decrease in photon yield. Finally, the thickness of the glass powder layer can exacerbate light absorption due to the increased optical path length, which heightens the probability of absorption. These factors collectively contribute to a potential reduction in overall light transmittance. Therefore, the transmittance value of PiG plate with a thickness of 0.08 mm was higher than that of PiG plate with a thickness of 0.15 mm. The result was consistent with the light emission spectrum of the four-inch PiG plates in Figure 7. As shown in Figure 9, we further measured the uniformity of the five four-inch PiG plates with a thickness of 0.08 mm using an integrating sphere measurement system, at points (A), (B), (C), (D), (E), (F), (G), (H), and (I). The measured results of a total of 45 points were shown in Figure 10. The differences in CIE (x, y) of a total of 45 points were less than 3.9×10^{-3} . From the nine-point measurement results, the precise fabricating process produced the four-inch PiG plates with a thickness of 0.08 mm with good uniformity and excellent performance. To study the thermal stability of the PiG plate with a thickness of 0.08 mm, a thermal aging test at stress temperatures of 150 °C, 250 °C, 350 °C, and 450 °C for 1008 h were performed. Figure 11 shows the results of the PiG plate with a thickness of 0.08 mm before and after thermal aging tests at stress temperatures of 150 °C, 250 °C, 350 °C, and 450 °C for 1008 h. The optical performance of the CIE and luminous efficacy did not significantly change before and after thermal aging. This indicates that the PiG plate with a thickness of 0.08 mm also had high thermal stability.

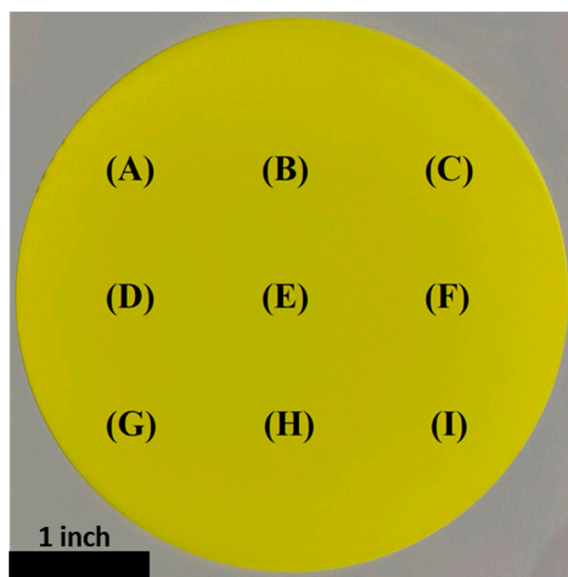


Figure 9. Nine-point measurement for four-inch PiG plate with a thickness of 0.08 mm. The measurement points are denoted as (A) to (I).

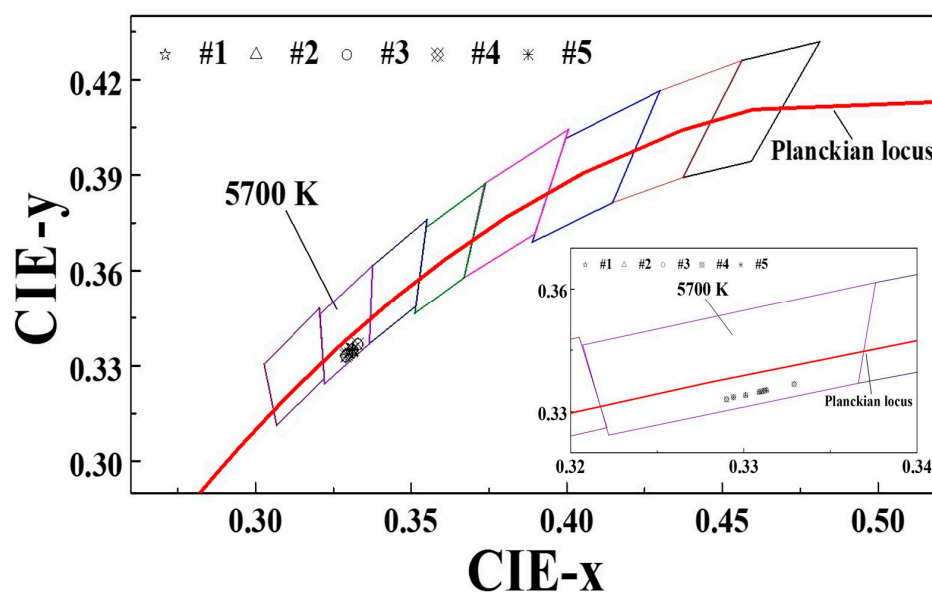


Figure 10. CIE 1931 (x, y) chromaticity coordinates of the PiG plate with a thickness of 0.08 mm. The measurements, denoted as 1 to 5, were conducted sequentially. As evident from the magnified view, the values remained highly consistent across all five measurements.

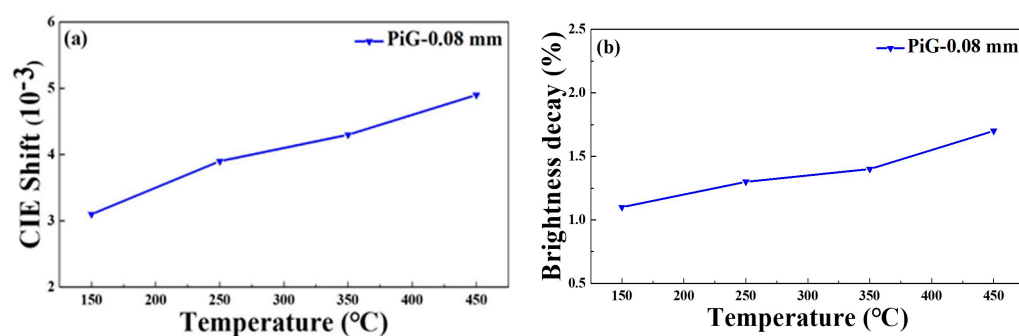


Figure 11. Thermal aging tests of the PiG plate with a thickness of 0.08 mm for the (a) CIE shift, and (b) lumen loss.

4. Discussion and Conclusions

In summary, we have demonstrated a novel thin PiG plate showing both high-performance and high-reliability WLEDs employing four-inch YAG: Ce³⁺ PiG plates for vehicle headlight applications. The luminous efficacy of the PiG plate with a thin thickness of 0.08 mm was increased by 16.83% compared to the plate with a thickness of 0.15 mm, due to the reduction in the interaction between the pores and light. From the nine-point measurement results, the precise fabricating process produced the four-inch PiG plates with a thickness of 0.08 mm with good uniformity and an excellent luminous efficacy of 118 lm/W. The four-inch PiG plates with a thickness of 0.08 mm were verified by an accelerated aging test at a stress temperature of 450 °C and 1008 h. The results showed that there was no significant change in CIE and luminous efficacy before and after thermal aging. This indicated that the PiG plate had high thermal stability. In this study, given the benefits of the proposed thin PiG plate, an opportunity to replace traditional silicone-based phosphors may be provided, to achieve high-quality WLEDs for vehicle headlights in automotive applications.

Author Contributions: Conceptualization, H.-W.H.; Methodology, W.-C.C.; Validation, Y.-C.C.; Formal analysis, C.-C.C.; Writing—original draft, C.-W.H.; Writing—review & editing, C.-N.L. All authors have read and agreed to the published version of the manuscript.

Funding: Ministry of Science and Technology, Taiwan (MOST) (108-2218-E-005-018 to fund the research for U.S. 363,261 and 107-2218-E-005-025 to fund the research for U.S. 152,738).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

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