ORIGINAL ARTICLE

Journal
the American Ceramic Society

A low sintering temperature glass based on SiO_2–P_2O_5–ZnO–B_2O_3–R_2O system for white LEDs with high color rendering index

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Abstract
A low sintering temperature glass based on the SiO_2–P_2O_5–ZnO–B_2O_3–R_2O (R=K and Na) system was studied as a matrix for embedding phosphors to fabricate color tunable white LEDs. The proposed system, which uses no heavy-metal elements and can be sintered at 500°C, incorporates thermally weak commercial phosphors such as $CaAlSiN_3$:Eu^{2+} to produce phosphor-in-glasses (PiGs). Changing the mixing ratio of glass to phosphors affected the photo-luminescence spectra and color coordinates of the PiGs when mounted on a blue LED. The color rendering index (CRI) and color correlated temperature (CCT) of the LEDs were also varied with the mixing ratio, providing color tunable white LEDs. A high CRI, up to 93, as well as highly improved thermal stability were obtained, along with a low sintering temperature compared to other glass systems, suggesting the practical feasibility of the proposed system.

KEYWORDS
glass, low temperature, optical materials, phosphors, sintering

1 | INTRODUCTION

Inorganic color converting materials are currently being used to replace organic resin materials in white LEDs (wLEDs), because conventional organic resins have lower thermal and chemical stability, which shortens LED lifetime.[1-4] Among the inorganic alternatives, phosphor ceramics sintered with phosphor powders have been developed and commercialized, but they suffer from high production costs because their fabrication requires a high sintering temperature of over 1600°C, as well as high pressure.[5,6] To produce wLEDs, however, phosphor-in-glass (PiG) can be easily prepared by sintering a mixture of phosphor and glass powders at temperatures below 800°C.[7-19] PiG plate has been reported to have enhanced thermal stability compared with conventional

silicone resin in high power LEDs[7-9,14,17] and thus has recently been commercialized for high power applications such as automobile headlamps.[17]

Although the color coordination of wLEDs can be easily controlled by the content of the yellow $Y_3Al_5O_{12}$:Ce^{3+} (YAG:Ce^{3+}) phosphor and the thickness of the plate, their color rendering index (CRI) and color correlated temperature (CCT) have been limited because they lack red emission. Demand has recently been increasing for a high power white light source with a high CRI value and a warm white with a CCT lower than 4100 K, to provide natural color reproduction and various white sources for high power applications such as automobile headlamps and indoor/outdoor lighting.

Various efforts to improve CRI and reduce CCT have been proposed, for example, by adding transition metals or rare earth ions to the glass matrix to produce the red emission.[12,13,20] Lee et al. proposed an assembled PiG with green and red phosphors as a high CRI color converter, but the device requires multiple fabrication steps and has limited sizes available.[10] The practical feasibility of PiGs simultaneously embedding with YAG:Ce^{3+} and the CaAlSiN$_3$:Eu^{2+} (CASN:Eu^{2+}) red phosphor was also demonstrated.[11,18] Since embedding CASN:Eu^{2+} is critically important for high CRI and it starts to significantly lose its emission intensity after sintering above 600°C, due to oxidation and thermal decomposition,[8,11,20] it has been a great concern to find a proper glass matrix with a low sintering temperature. Silicate glasses based on the SiO_2–Na_2O–RO (R=Ba, Zn; SNR)[11] and SiO_2–B_2O_3–ZnO–Na_2O (SBZN)[18] systems can be sintered at 550°C and they have exhibited CRI values higher than 90 successfully embedding CASN:Eu^{2+}. Glasses with heavy-metal oxides such as tellurite and bismuth based glasses have low glass transition temperatures and have been also studied to compose PiG plates.[13,15,21-24] The heavy-metal oxide based glasses, for example Bi_2O_3–ZnO–B_2O_3 glass, can be sintered even at 400°C.[24] However, the glass performance was seriously degraded by the inherent visible absorption of the heavy-metal ions such as Te and Bi-ions, which restricted conversion efficiency as well as their practical application.

As sintering at lower temperature can further reduce production costs and also prevent thermal degradation of phosphors more effectively, it is highly important to find a proper and practical transparent host glass which can further reduce the sintering temperature. Thus, in this study, we investigated a new glass system based on the SiO_2–P_2O_5–ZnO–B_2O_3–R_2O (R=K and Na) (SPZ) system which has no inclusion of heavy-metal oxides for PiG fabrication and can be sintered even at 500°C. Glass composition and sintering conditions were studied. PiG plates were fabricated by varying the mixing ratio of glass and phosphors to compose color tunable wLEDs. CRI, CCT as well as thermal stability were examined to confirm the practical feasibility of the proposed system.

2 | EXPERIMENTAL PROCEDURE

The nominal composition of the SPZ glass was $5SiO_2$–$(80–x)P_2O_5$–$xZnO$–$15(B_2O_3+R_2O)$ (R=K and Na). High purity (>99.9%) raw materials were weighed and melted at 1300°C for 1 hour using an alumina crucible. The melt was quenched in a brass mold and then ground into a powder with particles below 100 μm in size. The average particle size of YAG:Ce^{3+} and CASN:Eu^{2+} phosphors was 17 μm (D_{50}) and 9 μm (D_{50}), respectively as shown in Figure S1. The glass power was thoroughly mixed with phosphors and 0.4 g of the mixed powder was formed into a 10 mm disk shaped green body by a uniaxial press. The glass to YAG:Ce^{3+} phosphor (GtP) ratio was varied from 9:1 to 8:2 in weight and CASN:Eu^{2+} was additionally added, up to 3 wt%. The green body then was sintered at varying temperatures and then polished to 250 μm in thickness for further examination.

The thermal properties of the glass were inspected by differential scanning calorimeter analyzer (DSC; DSC-60, Shimadzu, Tokyo, Japan) while an X-ray diffractometer (XRD; MiniFlex 600, Rigaku, Tokyo, Japan) was employed to monitor possible crystalline phases within the glass matrix. Visible transmission of the PiG plate was examined with a UV/VIS spectrophotometer (Evolution 201, Thermo Fisher Scientific, Waltham, MA).

To compose a white LED the PiG plate was mounted on a commercial blue LED chip with a center wavelength of 450 nm, supplied by CTL Inc. A visible spectrometer (DARSA PRO 5200, PSI, Suwon, Korea) equipped with an integrating sphere (C9920-02, Hamamatsu, Hamamastu, Japan) recorded the electro-luminescence (EL) spectra of the blue LED chip and the photo-luminescence (PL) spectra of the PiG plate, along with the CRI and CCT of the mounted LEDs. Thermal quenching property was monitored with a fluorescence spectrometer (F-4500, Hitachi, Tokyo, Japan) while the LFA 447 Nanoflash[TM] system (NETZSCH, Selb, Germany) measured the thermal conductivity of the plate. The morphology of the PiG plate was observed using a field-emission scanning electron microscope (FE-SEM; MIRA LMH2, TESCAN, Brno-Kohoutovice, Czech Republic).

3 | RESULTS AND DISCUSSION

When glasses were prepared with varying ZnO content, the glasses were clear except the glass with x=45 (SPZ45), which showed devitrification due to phase separation. The

glass transition temperature (T_g) was estimated to be ~419, 412 and 410°C for x=30, 35 and 40 (SPZ30, SPZ35 and SPZ40), respectively, when inspected with DSC at a heating rate of 10°C/min using an aluminum pan. The decrease in T_g can be attributed to the destruction of the structural connectivity of [PO$_4$] units with ZnO content, as previously reported.[25,26] The T_g determined for the present glasses was lower than that of SNR (~457°C)[11] and SBZN ~ 425°C) systems[18] which have a sintering temperature of 550°C, and thus a lower sintering temperature can be anticipated for PiG fabrication.

In order to find the proper sintering temperature, the ground glass powders with varying ZnO content were packed into a disk shape and sintered at various temperatures for 30 minutes. As presented in Figure 1, the glasses could even be sintered at 500°C, and showed changes in shape due to viscous flow at 525°C, implying the developed glasses had good flow properties at low temperature. The SPZ40 glass was inspected by XRD for possible crystallization following heat treatment, and as shown in Figure 1, no apparent crystalline phases were detected up to a sintering temperature of 550°C, implying the glass was highly stable during the sintering process. However, a ZnP$_2$O$_7$ phase can be formed above 575°C in the SiO$_2$–P$_2$O$_5$–ZnO system, which restricts transmittance and sintering temperature.[27]
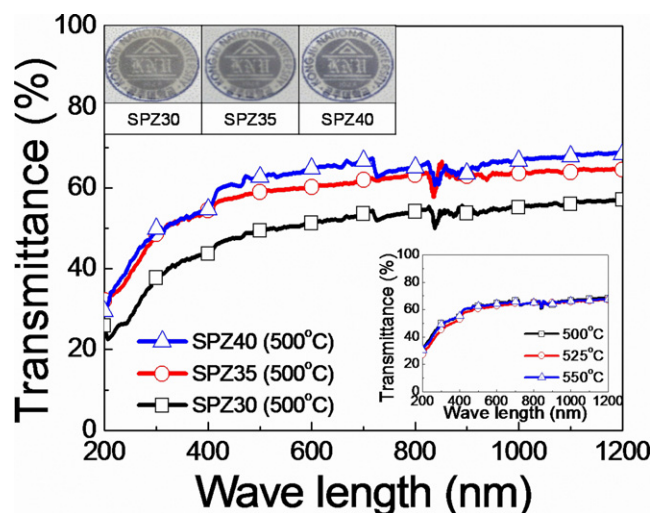
When the UV/VIS transmittance of the glasses sintered at 500°C for 30 minutes was monitored after polishing to 250 μm in thickness (Figure 2), transmittance reached up to 65%, which is higher than that has been reported for other SNR and SBZN glasses sintered at 550°C.[11,18] It should be noted that the transmittance increased with ZnO content.
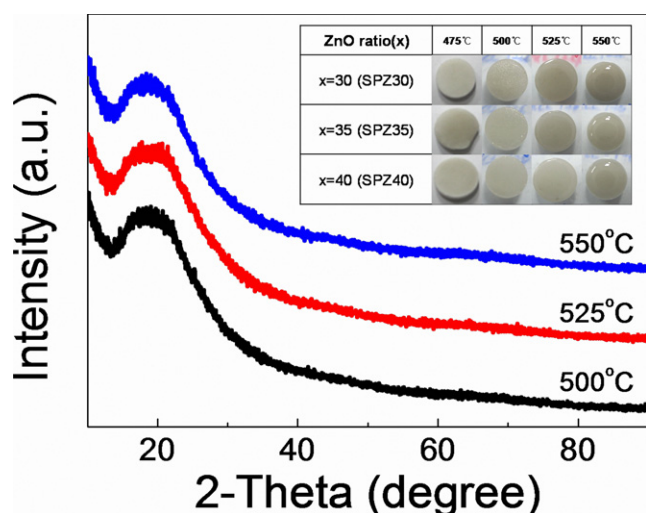
This can be attributed to the improved viscous flow of the proposed glass with increasing ZnO content, as predicted by the decreasing T_g. Scattering due to trapped pores and the viscous flow of the glass during the sintering process are mainly responsible for producing haze and transmittance.[17,18] When the sintering temperature was increased to 550°C, as displayed in the inset figure, the transmittance of the sintered plate with x=40 (SPZ40) was not significantly improved, suggesting that the glass can be sintered at 500°C, and still produce sufficient viscous flow and transmittance. Further enhancement of transmittance can be expected by properly controlling processing conditions, for example, by reducing contamination during powder processing, and increasing pressure during packing to remove trapped pores.[17] Chemical durability is also important for practical application and the glass showed no apparent chemical reaction on its surface under normal atmospheric condition even for 30 months as exhibited in Figure S2.

PiG plates were thus fabricated using SPZ40 at 500°C with various ratios of glass to YAG:Ce^{3+} yellow phosphor (GtP), from 9:1 to 8:2. The sintered PiGs were optically polished to a thickness of ~250 μm and showed reasonable transparency, as exhibited in Figure 3. The color converting properties of the PiG plates were examined by mounting them on a blue LED operated at 20 mA. As can be seen in the actual photos of the PiG mounted LEDs in Figure 3, the mounted LEDs produced white emission under blue excitation, and their color coordination was linearly shifted to yellow as the phosphor content was increased. The yellow shift can be further identified from the emission spectra (the inset figure), which shows the development of


FIGURE 1 X-ray diffraction results of SPZ40 glass with varying sintering temperatures. In the inset figure, glasses with varying ZnO content which were sintered at different temperatures for 30 min are shown [Color figure can be viewed at wileyonlinelibrary.com]

FIGURE 2 Transmittance of polished glass plates without phosphors and with varying ZnO content after sintering at 500°C. Thickness was maintained at 250 μm. The inset figure represents the transmittance of the SPZ40 glass for varying sintering temperatures [Color figure can be viewed at wileyonlinelibrary.com]
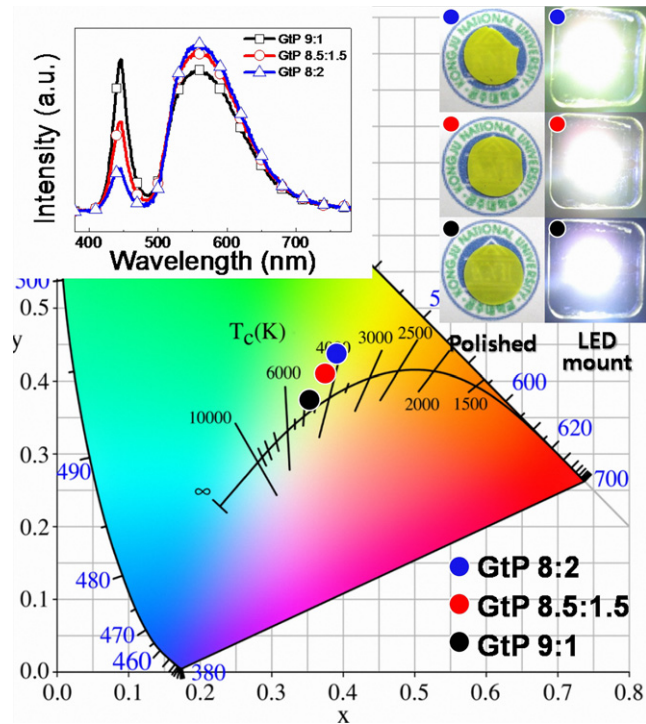
FIGURE 3 CIE color coordinates of LEDs mounted with PiGs having various GtP ratios from 9:1 to 8:2. The inset figure shows their EL+PL spectra. Images of the sintered PiGs and the mounted LEDs are also displayed on the right side of the figure [Color figure can be viewed at wileyonlinelibrary.com]

a yellow PL emission in lieu of the blue EL emission with the increase in phosphor content.

The CIE color coordination, color co-related temperature (CCT), CRI values and luminous efficacy (lm/W) of the PiG-mounted LEDs with varying GtP ratios are summarized in Table 1. It should be noted that the PiG with a GtP ratio of 9:1 produced a white emission with a high CRI value of 84, which is comparable to that of commercial wLEDs. The wLEDs also exhibited reasonable luminous efficacy around 90 lm/W which was increased with phosphor content due to the improved yellow emission. However, as previously reported,[7,11,18] their color coordination as well as CRI remained limited due to the weak red-emission of the yellow phosphor, even with various GtP.
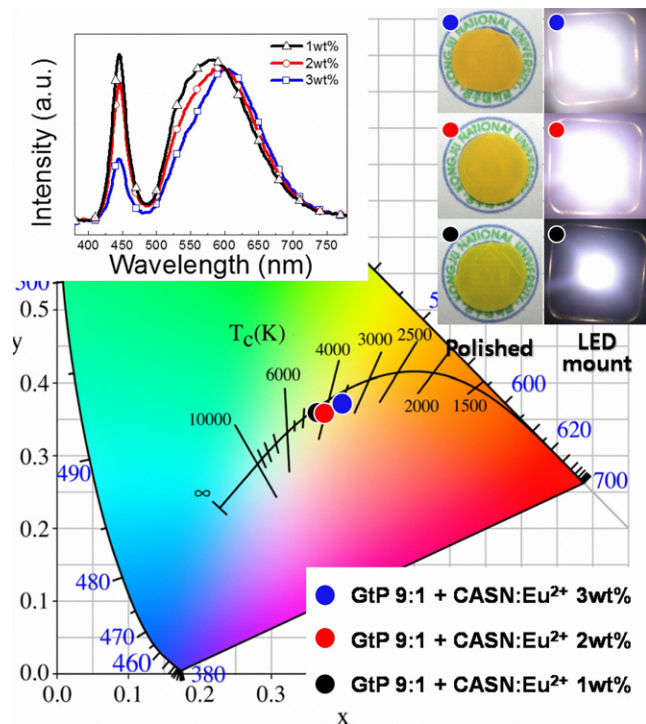
To improve CRI and color tunability, CASN:Eu^{2+} red phosphor was then incorporated into the PiG. Up to 3 wt% CASN:Eu^{2+} was additionally added to the PiG at a GtP ratio of 9:1. The PiG plates prepared with ~250 μm in thickness also showed reasonable transparency and changed their color with increasing CASN:Eu^{2+} content, as displayed in Figure 4. When the plates were mounted on a blue LED operated at 20 mA, they also produced white emission, as shown in the figure.

The EL+PL spectra of the mounted LEDs clearly shows the red shift of the color converted emission resulting from the evolution of the red-emission peak at the expense of blue EL emission, with increasing CASN:Eu^{2+} content. The red shift may be also elucidated by possible reabsorption of yellow emission by red phosphor along with the reduced chance of blue absorption for yellow phosphor by the additional introduction of red phosphors within the glass matrix. As seen in Figure 4, the color coordination has also been moved well into the warm white range following the Planckian locus. The color coordination, CCT, CRI, and luminous efficacy of the PiG mounted LEDs prepared with varying CASN:Eu^{2+} content are summarized in Table 1. It should be noted that CCT has been effectively reduced down to ~3500 K with the addition of the red phosphor, which further assisted the realization of a warm white LED. Luminous efficacy has been decreased with the introduction of red phosphor since overall PL emission peak shifts to red as shown in Figure 4. Moreover, the CRI has been highly enhanced, even up to 93, which is higher than SNR glass[11] and comparable to the SBNR glass system.[18] In light of the fact that the SNR and SBZN glass systems have a higher sintering temperature (550°C), these results clearly demonstrate that color tunability as well as high CRI can be obtained even at a low sintering temperature of 500°C, indicating the practical feasibility of the present glass system for color tunable wLED applications. The color stability of wLED with increasing operation current has been also examined (Figure S3) and showed insignificant change in color coordination as reported previously.[15]

Figure 5 shows an FE-SEM image of the PiG plate containing the YAG:Ce^{3+} and CASN:Eu^{2+} phosphors sintered at 500°C. Relatively large YAG:Ce^{3+} and small CASN:

TABLE 1 CIE color coordinates (CIE), color co-related temperature (CCT), color rendering index (CRI) and luminous efficacy (lm/W) of PiG mounted LEDs, with varying GtP ratio and CASN:Eu^{2+} content at the operation current of 20 mA

|	PiG with YAG:Ce^{3+}					PiG with YAG:Ce^{3+} and CASN:Eu^{2+} [a]			
GtP ratio	CIE (x, y)	CCT (K)	CRI	Efficacy(lm/W)	CASN:Eu^{2+} (wt%)	CIE (x, y)	CCT (K)	CRI	Efficacy(lm/W)
8:2	(0.3909, 0.4368)	4067	75	103.7	1	(0.3648, 0.3599)	4719	88	92.4
8.5:1.5	(0.3742, 0.4105)	4330	78	95.8	2	(0.3753, 0.3586)	4258	91	86.9
9:1	(0.3525, 0.3743)	5292	84	89.4	3	(0.3998, 0.3721)	3534	93	77.6

[a]GtP was fixed at 9:1.

FIGURE 4 CIE color coordinates of LEDs mounted with PiGs having various CASN:Eu^{2+} content at a fixed GtP ratio of 9:1. The inset figure represents their EL+PL spectra. Images of the sintered PiGs and the mounted LEDs are also displayed on the right side of the figure [Color figure can be viewed at wileyonlinelibrary.com]
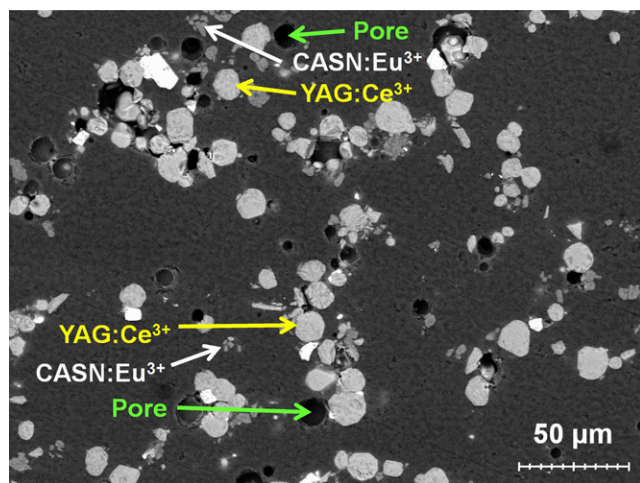
FIGURE 5 FE-SEM image of the PiG with a GtP ratio of 9:1 and 3 swt% of CASN:Eu^{2+} [Color figure can be viewed at wileyonlinelibrary.com]

Eu^{2+} phosphor powders were well dispersed within the glass matrix, without noticeable interaction between the glass and phosphors. Elemental mapping of the characteristic elements for glass (P), YAG:Ce^{3+} (Y) and CASN:Eu^{2+} (Ca) confirmed their distribution within the glass matrix as displayed in Figure S4. It should also be noted that trapped pores of less than ~10 μm in size can be observed within
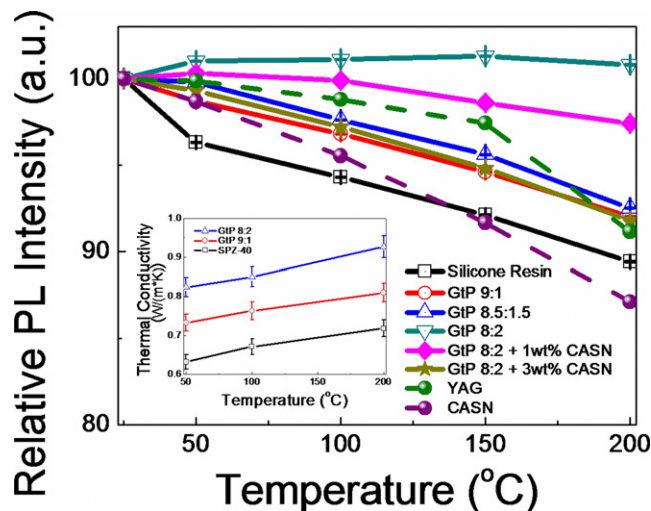
FIGURE 6 Thermal quenching (TQ) property of PiGs with various GtP ratios (open symbols with solid line) and CASN:Eu^{2+} content (solid symbols with solid line). TQ property of YAG: Ce^{3+}(YAG) and CASN:Eu^{2+}(CASN) phosphors were also shown for comparison (solid symbols with dashed line). The inset figure shows the thermal conductivity of the glass plate without phosphor, and PiG plates with various GtP ratios [Color figure can be viewed at wileyonlinelibrary.com]

the plate. Those pores can act as additional scattering centers, since their size corresponds to the Mie scattering range, and thus will restrict the maximum transmittance of the plate. Further adjustment of the mixing conditions, such as glass powder size and sintering condition, can effectively remove the trapped pores and improve the optical properties of the plate. For example, the complete elimination of trapped pores was achieved by gas pressure sintering (GPS) under 2000 kPa N$_2$ using glass powder with particles less than ~50 μm in size.[17]

The thermal stability of the present glass system was examined by investigating the thermal quenching property (TQ) of the PiG plates. The relative change in the PL intensity of plates with varying GtP ratios at their peak emission wavelength was monitored under a 450 nm LED pump, up to 200°C. TQ properties of phosphor powders used for this study were also examined for comparison. The increased non-radiative transition rate of the active ions was primarily responsible for the TQ behavior of the phosphors.[28] Passivation of the phosphors using glass materials can effectively shield the phosphors from the thermal source and thus improve the TQ property, unlike phosphors with silicone resin.[8,9,11]

Figure 6 depicts the TQ properties of various PiG plates and clearly shows the improvement in TQ property with the use of the present glass, as compared with the phosphor in silicone resin. It should be noted that the TQ property was improved by the YAG:Ce^{3+} phosphor content, and the PiG with a GtP ratio of 8:2 showed almost no thermal

degradation up to 200°C. The same enhancement of TQ property with increasing YAG:Ce^{3+} phosphor content has also been observed with SBZN glass.[18] The YAG:Ce^{3+} phosphor possesses high thermal conductivity (11.2 $Wm^{-1}K^{-1}$) and thermal stability. As shown in the inset figure of Figure 6, thermal conductivity of the PiG plates was increased with YAG:Ce^{3+} content. The increase in TQ property with increasing YAG:Ce^{3+} content can be attributed to the increased thermal conductivity of the PiG plate, which dissipate the thermal energy of the plate. Although it is not clearly understood yet, it seems that there exist compromise between glass and YAG:Ce^{3+} content for the resultant TQ property. Further study is required to completely understand the TQ behavior of PiG plates. The TQ property was then reduced with the introduction of CASN:Eu^{2+}. It can be elucidated by the poorer thermal stability of CASN:Eu^{2+} [11,16] than YAG:Ce^{3+} as shown in Figure 6. As discussed earlier, the PL emission peak shifts to red due to the evolution of red emission as CASN:Eu^{2+} content increases. Since the TQ property was measured at the peak emission wavelength of PL spectra, as CASN content increases, contribution of CASN to the emission increases and thus it will be more affected by CASN decreasing the TQ property.

Different porosity of each sample may also give an effect on the thermal properties. When we estimated the porosity of PiG samples varying phosphor content by inspecting SEM images, they showed similar porosity showing little relationship of the porosity to the thermal quenching property (Figure S5). However, it should be mentioned that the present glass clearly demonstrated improved thermal stability and feasibility for the color tunable high power white LED applications, even with the low sintering temperature of 500°C.

4 | CONCLUSION

Glasses based on the 5SiO$_2$–(80–x)P$_2$O$_5$–xZnO–15 (B$_2$O$_3$+R$_2$O) (R=K and Na) system were studied to develop a glass with a low sintering temperature, without heavy-metal oxides, for PiG fabrication. A glass with x=40 (SPZ40) showed reasonable transparency and could be sintered even at 500°C without crystallization. PiG plates with varying phosphor content were successfully manufactured at 500°C without noticeable degradation of the phosphors. Color tunable wLEDs were obtained when PiG plates with various YAG:Ce^{3+} contents were mounted on a blue LED. The additional introduction of CASN:Eu^{2+} further improved the CRI, up to 93. The developed glass with a GtP ratio of 8:2 showed also almost no thermal quenching property, and with the addition of CASN:Eu^{2+} also exhibited a higher thermal quenching property than silicone resin, clearly demonstrating the feasibility of the present glass system for high power wLED applications. However, further studies are required to reduce trapped pores and to elucidate the TQ property of PiG plates in detail.

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