Wavelength Conversion Material "Phosphor-Glass Composites" for High Power Solid-State Lighting

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Keywords: Glass composite, Wavelength conversion, Phosphor, Solid-state lighting, weather resistance

ABSTRACT

Phosphor-Glass Composites, in which phosphors are dispersed, have been developed. The phosphor-glass composites are expected as the excellent wavelength conversion material for high power solid-state lighting, which have high-humidity resistance, long lifetime and extremely small deviation in emission color.

1. INTRODUCTION

Phosphor converted-Light Emitting Diodes / Laser Diodes (pc-LEDs/LDs) have been attracting much attention as next generation solid-state lighting devices [1, 2]. In comparison with the conventional illumination lamps, pc-LEDs/LDs devices have a lot of advantages such as high energy efficiency, long lifetime and small environmental burdens. Recently, with the having improved performance of both LED light sources and phosphors, pc-LEDs have been used for various applications (display backlighting, automotive lights, data-projectors and general illumination lights).

Currently, most common pc-LEDs consist of Ce^{3+} doped YAG phosphor sealed in resin matrix and a blue-LED. However, the resin matrix tends to be deteriorated by heat from the high power blue-LED light source [3]. This deterioration results in undesirable color changes and deformations of the resin matrix, therefore lifetime of the pc-LEDs is short.

To solve these problems, we have developed a wavelength conversion material, 'Phosphor-Glass Composites' [4] (named Lumiphous™), in which phosphors are dispersed homogeneously.

As the glass matrix of the phosphor-glass composites, for example, SiO_2-B_2O_3-RO(R=Mg, Ca, Sr or Ba), B_2O_3-ZnO-RO and SnO-P_2O_5 glass systems are applied. These glass systems and their glass transition temperature (Tg) are shown in Table 1. Tg is one parameter which shows thermal resistance of the phosphor-glass composites. Glass system is selected by considering refractive index differences and reactive property between the glass and phosphor powder. For the phosphors with low heat resistance, the glass systems of low Tg are usually adopted, for instance, B_2O_3-ZnO-RO or SnO-P_2O_5 glass systems. The difference of refractive index between glass and phosphor makes a large influence on light scattering of blue light. Luminous efficiency can be enhanced by introducing the proper light scattering into the phosphor-glass composite.

Phosphor-glass composites entirely composed of inorganic materials have a number of advantages in the use of wavelength conversion devices, especially in terms of high-humidity resistance and small deviation in emission color. In this research, we show superior performance of the phosphor-glass composites.

Table 1 Glass compositions and their glass transition temperature.

Glass composition system	SiO_2-B_2O_3-RO	B_2O_3-ZnO-RO	SnO-P_2O_5
Tg (°C)	500 - 650	400 - 500	300 - 350

2. EXPERIMENTAL PROCEDURES

In this research, a SiO_2-B_2O_3-RO glass system was used. Powder of the glass and phosphors were well mixed under the prescribed condition. The mixture was pressed into each shape and heat-treated at a temperature higher than Tg. The phosphor-glass composite samples were processed into the forms of 10 x 10 x 0.2 mm and 1.2 x 1.2 x 0.2 mm to evaluate the humidity resistance and the emission color. To investigate the humidity resistance, the samples were subject to an unbiased highly accelerated stress test (HAST).The test condition was 121°C, 95 % RH and 2 atm for

300 hours. The total luminous flux and the emission color were calculated from spectral power distribution measured in an integrating sphere under 460 nm and 445 nm blue-LED excitation at room temperature.

3. RESULTS AND DISCUSSION

Figure 1 shows the changes of relative total luminous flux during the HAST for the phosphor-glass composite and a phosphor-resin composite. $La_3Si_6N_{11}:Ce^{3+}$ (LSN) [5] yellow phosphor was dispersed in both samples. The content of the phosphor in the phosphor-glass composite and the phosphor-resin composite are 13.5 wt% and 10.0 wt%, respectively. The phosphor-resin composite was made by a commercially available heat-curable silicone resin (cure condition; 120 °C-1 hour). As seen in Fig.1, the relative total luminous flux of the glass composite sample did not change at all even if the sample was exposed in HAST for 300 hours. The appearance of the sample also did not change. On the other hand, the relative total luminous flux of the resin composite sample decreased with increasing test time. This decrease in total luminous flux may be caused by a deterioration of silicone resin in high temperature and high humidity environment [6].

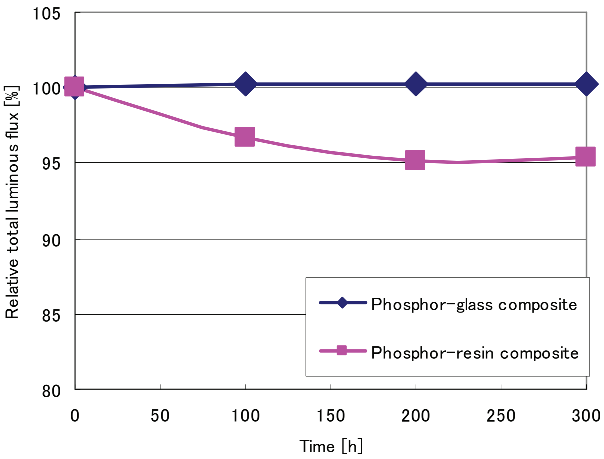

Fig.1 The change of relative total luminous flux in the phosphor-glass composite and the phosphor-resin composite during the highly accelerated stress test under the conditions of 121°C, 95% RH and 2 atm. (Phosphor: LSN)

Figure 2 shows the emission colors of the phosphor-glass composites with the appropriate mix of LSN and α-SiAlON:Eu^{2+} orange phosphor[7, 8] in the CIE chromaticity diagram. In this case, the emission color of the phosphor-glass composites can be freely controlled within a region surrounded by the chromaticity point of LSN, α-SiAlON and 460 nm blue-LED excitation. The emission colors can be easily fitted on the locus of the blackbody radiation by changing a ratio of LSN and α-SiAlON shown in Fig. 2.

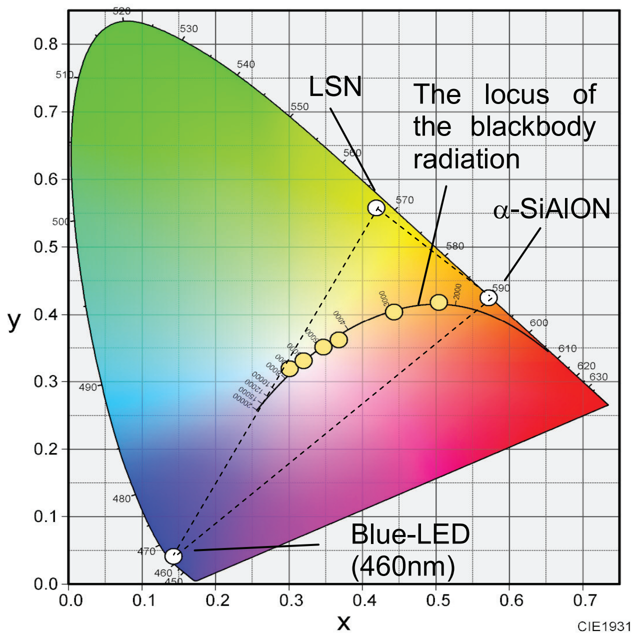

Fig.2 The emission colors of the phosphor-glass composites with the appropriate mix of LSN and α-SiAlON in the CIE chromaticity diagram. (excitation: 460 nm)

The deviation of emission color in the 80 samples was evaluated. LSN and α-SiAlON were used. A content of phosphor in the samples was 13.5 wt% and the weight ratio of LSN to α-SiAlON was 85 to 15. The distribution of emission color for the samples is shown in Fig. 3. The deviation of chromaticity x is within a small range of 0.006. This deviation falls inside 3-step MacAdam ellipse used as a chromaticity control standard. This is attributed to the homogeneous dispersion of the phosphor in glass matrix and precise dimensional control in process.

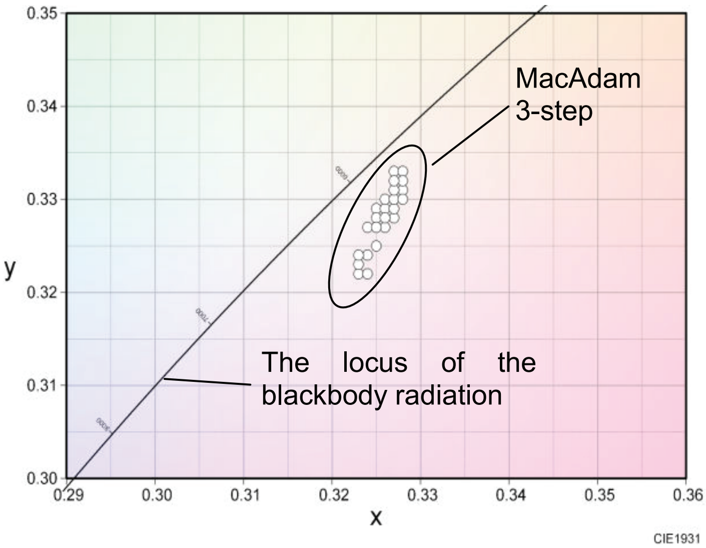

Fig.3 The distribution of emission color for the phosphor-glass composites. The content of the phosphors (LSN and α-SiAlON) is 13.5 wt%, the weight ratio of LSN to α-SiAlON is 85 to 15.

Finally, a formability of the phosphor-glass composites is shown. For example, plate, disk, rod, dome and ball-shaped samples are shown in Fig. 4. The phosphor-glass composites can be processed by a variety of methods, such as cutting, grinding, polishing, pressing, bending and so on.

Additionally, a variety of emission colors of phosphor-glass composites is shown in Fig. 5. The various emission colors can be produced by selecting a suitable combination of glass and phosphor.

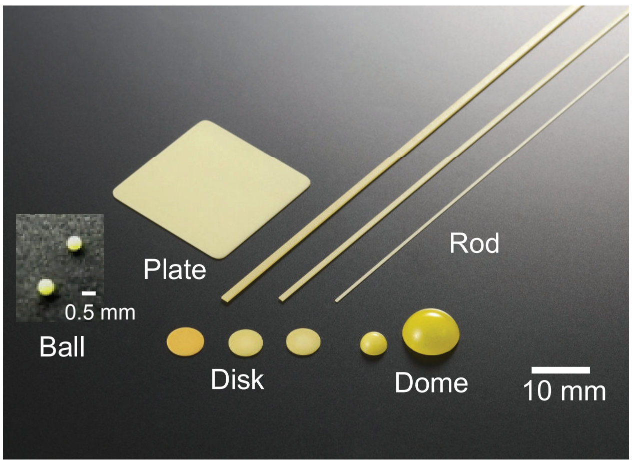

Fig.4 The examples of formability of phosphor-glass composite.

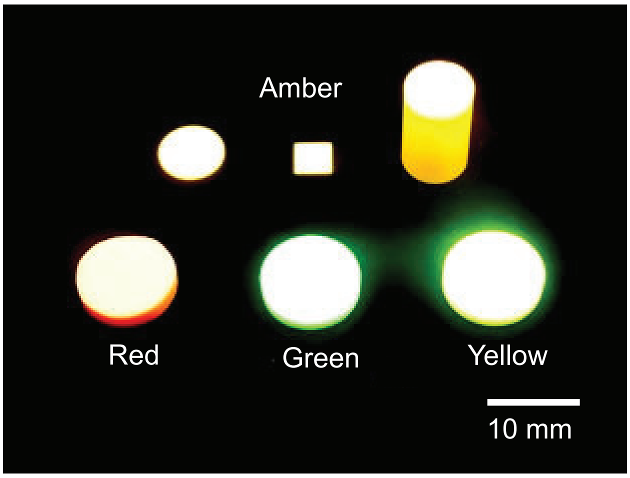

Fig.5 A variety of color coordination in phosphor-glass composites when they are excited by UV-lamp.

4. SUMMARY

The humidity resistance and the deviation of emission color of the phosphor-glass composites were evaluated. The phosphor-glass composites showed excellent humidity resistance in HAST compared with the resin-phosphor composite. The deviation of emission color was so small even if two kinds of phosphors were dispersed in the glass matrix, it would be difficult to control the emission color precisely. These results indicate that the phosphor-glass composites are suitable for the wavelength conversion materials for high power solid-state lighting.

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