Performance Enhancement of White LEDs Through Phosphor/Silicone Composite Particles

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***Abstract*—Settling of the heavy phosphor particles in the silicone encapsulation is a root-cause for variation of the correlated color temperature causing yield reduction in the fabrication of white light-emitting diodes (LEDs). When prefabricated particles consisting of the phosphor and the silicone are used, settling can be significantly reduced and a dramatic enhancement of the consistency of LED performance can be achieved. A reduction of the change in correlated color temperature after 60 min of settling on the chip from >900 K for traditional phosphor particles to only 8 K for particles with a weight ratio of phosphor and silicone of 1:1 in an otherwise identical coating procedure is demonstrated.**

***Index Terms*—Light-converting materials, settling-free, phosphor, silicone, high-power white light-emitting diodes (LEDs).**

I. Introduction

HIGH-POWER white light-emitting diodes (LEDs) are highly popular in our daily life due to their high light efficiency, low power consumption, environmental friendliness and high reliability [1], [2]. There is a continuous effort to further improve their optical performance in order to make them the dominant light source for illumination [3]–[5]. Apart from optical performances, the high price of LED products still sets the main obstacle for further development. Thus, reducing the manufacturing cost has become a research hotspot in the LED packaging community [6].

Most white LEDs feature a combination of blue LED chips and yellow phosphor [7]. For such white LEDs, phosphor coating, in which phosphor particles and silicone gel are mixed and coated around LED chips, is a necessary packaging process step [8]. In this process, the phosphor particle settling phenomenon inevitably occurs because of the density difference between the phosphor and silicone [9]. Such phosphor settling results in a large variation among LED modules in mass packaging in terms of the coating phosphor amount and phosphor particle distribution. Consequently, optical performance including the light efficiency and color cannot be well controlled and a big fluctuation between LED products results [10]–[12]. Those LEDs whose optical performance is out of the specification range must be discarded as waste products. Therefore, decreasing the optical performance variation caused by phosphor settling is a key issue.

In order to suppress phosphor settling, two main technical solutions have been developed, i.e. increasing the encapsulant viscosity and using small phosphor particles [13]. However, using high-viscosity encapsulation makes the process difficult and time-consuming resulting in low manufacturing efficiency. Small phosphor particles enhance scattering and thus can reduce the device efficiency [14]. Thus, more ideal settling suppressing methods are still in demand.

In this letter, phosphor/silicone composite particles, which reduce toward settling are shown to enhance packaging optical performance consistency for high-power white LEDs. The preparation and enhancement in optical performance consistency are described in detail.

II. Toward Settling-Free Principle

The phosphor particle's settling velocity under gravity can be given by [15],

$$u_s = \left[\frac{3}{4} \cdot \frac{g d_p (\rho_p - \rho)}{C_D \rho}\right]^{\frac{1}{2}} \tag{1}$$

where u_s is the settling velocity. d_p and g are the phosphor particle diameter and gravity constant, respectively. ρ_p and ρ are the densities of the phosphor and silicone, with values of 4.8 g/cm^3 and 1.2 g/cm^3, respectively C_D is the drag force coefficient. From Eq. 1, it can be seen that smaller particle size, lower density difference between particle and silicone, and a larger drag force coefficient can reduce the phosphor settling velocity. Based on these principles, we propose the use of phosphor/silicone composite particles.

Fig. 1 illustrates two possible types of composite particles. The phosphor particle is wrapped by the silicone or attached to the silicone. The particle shape ranges from nearly spherical to irregular. The effective density of the composite particle can be calculated by

$$\rho'_p = V_p \times \rho_p + (1 - V_p) \times \rho \tag{2}$$

where ρ'_p is the composite particle density, V_p is the phosphor volume fraction within such a particle. Due to the lower

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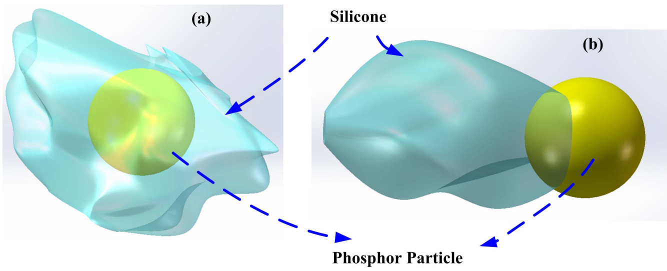

Fig. 1. Schematic illustration of the structures of phosphor/silicone composite particles. (a) Silicone coating around phosphor particle. (b) Silicone attaching to phosphor particle.

TABLE I
RATIO OF THE SETTLING DRIVING FORCE OF COMPOSITE PARTICLES TO THAT OF TRADITIONAL PHOSPHOR PARTICLES

Phosphor volume fraction(V_p)	1	0.7	0.4	0.1
d'_p / d_p	1	1.13	1.36	2.15
$[d'_p(\rho'_p - \rho) / d_p(\rho_p - \rho)]$	1	0.79	0.54	0.22

density of the silicone ρ, the density of the composite particle ρ'_P is lower than the density of the phosphor ρ_P, so there is less density difference between composite particles and the liquid silicone. Besides, composite particles increase C_D by changing the particle shape from spherical to irregular resulting in a higher surface to volume ratio [16]. These two factors reduce the settling effect. However, the particle size gets larger by adding solid silicone, which favours settling. For convenience, the composite particle is assumed as a spherical shape. Thus the product of the particle size and density difference in the right term of Eq. 1 is the key item for the settling velocity and represents the settling driving force. To evaluate the comprehensive effect on phosphor settling, the ratio of the settling driving force of composite particles to that of traditional phosphor particles for different phosphor volume fraction V_P is calculated and shown in table I. d'_p is the composite particle diameter. When V_P decreases from 1 to 0.1, the ratio of settling driving force decreases from 1 to 0.22. Therefore, the settling can be effectively suppressed by the composite particles. This effect is bigger for bigger composite particles whose density approaches that of the silicone matrix. However, if the particles are too big they can cause problems in dispensing the composite particle/silicone mixture. Thus the phosphor volume fraction of the composite particles is selected to be 0.2 in our experiments, that is, the weight ratio of phosphor to silicone is 1:1.

III. PREPARATION OF PHOSPHOR/SILICONE COMPOSITE

The phosphor particles with an average diameter of 13 μm and silicone (OE 6550, Dow Corning, USA) were used for the preparation of phosphor/silicone composite particles. Fig. 2 shows the fabrication process flow which is carried out in a super-clean laboratory. Before using, all the vessels were cleaned ultrasonically in acetone, isopropanol and deionized water and then dried in a stream of nitrogen gas. First, the solid phosphor-silicone mixture plate was fabricated by mixing phosphor with silicone at a weight ratio of 1:1, removal of air by vacuum, and curing at 150 °C. Then, the mixture plate was grinded to gain particles. After the grinding process, the particle size wasn't in homogeneous, so a filtration process was introduced to remove large particles (i.e. with a diameter above 40 μm) which might cause equipment blocking or other problems in the following packaging. These large particles were recycled, grinded and filtered to gain proper size particles to keep the phosphor concentration and particle size as constant as possible.

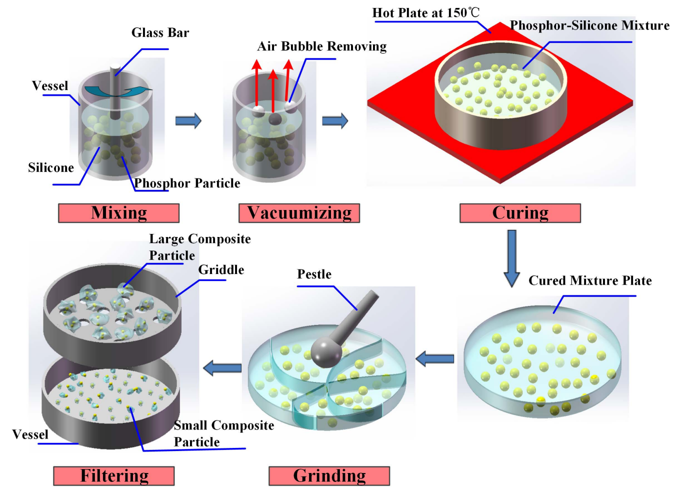

Fig. 2. Schematic diagram of overall fabrication process flow for the composite particles.

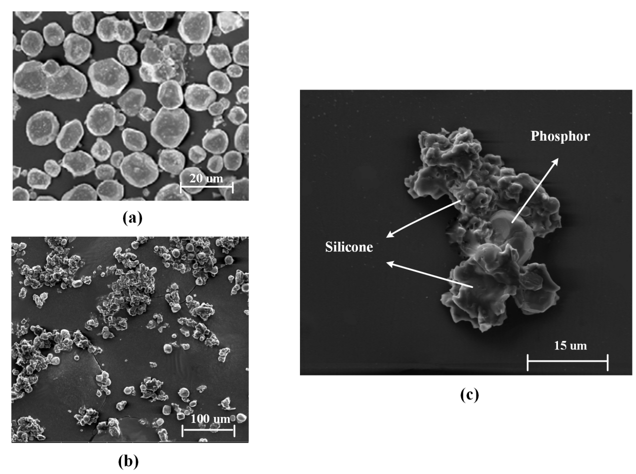

Fig. 3. SEM images of traditional and composite light-converting particles. (a) Traditional phosphor particles. (b) Composite particles. (c) A single composite particle.

IV. RESULTS AND DISCUSSION

Fig. 3 shows scanning electron microscopy (SEM) pictures of the traditional phosphor particles and the composite particles. The traditional phosphor particles are nearly spherical. In the composite particles, however, the phosphor particle is surrounded by solid silicone and

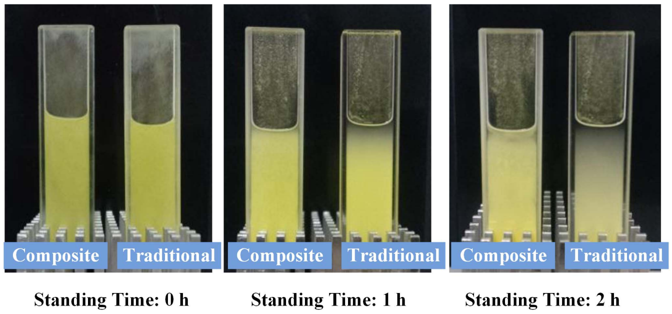

Fig. 4. Settling effect comparison between mixtures with traditional and composite light-converting particles after different standing times of 0 h, 1 h, 2 h.

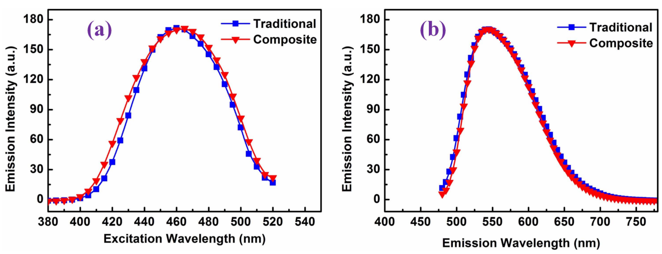

Fig. 5. Comparison of a) excitation spectra and b) emission spectra (under excitation with 464 nm) of traditional and composite light-converting particles.

exhibits an irregular shape. More than 120 individual composite particles have been measured. Their diameters range from 18 μm to 40 μm, and the average diameter is around 30 μm.

A common phosphor concentration of 0.15 g/cm^3 is chosen for all following experiments. As the phosphor concentration of the composite particles is known, the weight of the required liquid silicone can be calculated. The weight ratio of composite particles to liquid silicone is 0.3:1, Comparably the ratio of the traditional phosphor to liquid silicone is 0.13:1. The two kinds of mixtures were prepared and filled to cuvettes for testing the settling effect. Fig. 4 shows the settling results after standing times of 1 h and 2 h. It can be seen that the upper layer of the mixture with traditional phosphor particles is more transparent after 1 h which becomes more obvious after 2 h. This is due to the strong settling effect leading to less phosphor particles staying in the upper layer. No settling can be seen for the composite particles from these pictures after 2 h. This effectively demonstrates the strongly reduced settling of such composite particles.

To evaluate the optical properties of the composite particles, the excitation and emission spectra of the phosphor/silicone mixture and the composite particle/silicone mixture were measured by a spectrometer (Jasco FP-6500, Japan). Fig. 5 shows that the composite particles almost yield the same spectrum as traditional phosphor particles. Their peak excitation and emission wavelengths are both 464 nm and 544 nm. This indicates that the composite particle fabrication processes do not change the light-conversion properties.

The fabricated composite particles were applied in LED packaging to examine their effect on the optical performance consistency. There are two periods of phosphor settling,

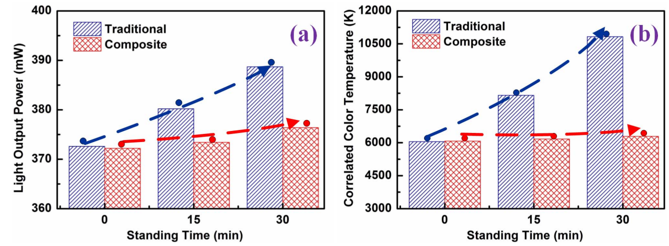

Fig. 6. Evaluating optical performance consistency caused by settling in coating equipments. (a) LOP variation, (b) CCT variation with different standing times.

settling in coating equipments and settling after phosphor coating in every single LED module. Two kinds of experiments were designed in terms of these two types of phosphor settling. More than four samples were fabricated under the same conditions and their average results were taken.

The experiments for checking phosphor settling in coating equipments were conducted with the dip-transfer coating method as described in Ref [17]. The phosphor/silicone mixture and the composite particle/silicone mixture were filled into different coating equipments. The upper-layer of both kinds of mixtures with the same volume in coating equipments after standing times of 0 min, 15min and 30 min were transferred and coated onto different LED modules. After phosphor coating, optical characteristics were measured immediately to avoid the influence of phosphor settling after coating. The results are shown in Fig. 6. We can find that the average light output power (LOP) with traditional phosphor increases by 16 mW and the average correlated color temperature (CCT) increases by 4774K with standing time. LED modules coated with the composite show an average LOP increase of 4 mW and CCT increase of around 200K. Thus, the LOP and CCT variations caused by phosphor settling in coating equipments were strongly reduced. These differences in optical performance consistency can be explained as follows. In the mixture with traditional phosphor, due to phosphor settling, the phosphor concentration in the upper layer decreases with standing time. As a result, the actual phosphor amount involved in blue-to-yellow light conversion reduces along with standing time, even though the mixture volume keeps the same. Less phosphor usually leads to higher CCT and LOP [6]. However, the mixture with the composite particles keeps the same phosphor concentration, consequently maintaining stable optical performance.

The other experiment for testing optical variation due to phosphor settling in LED modules was conducted by measuring each LED module at different times after phosphor coating. As shown in Fig. 7, after 60 min, the average LOP increases from 346 mW to 372 mW in LED modules coated with traditional phosphor, and the average CCT increases by 906K from 5965K to 6859K. However, the phosphor/silicone composite results in stable performance with the average LOP increasing by only 6 mW and an average CCT variation of only 8K. Therefore, the LOP and CCT consistency are dramatically enhanced. The above optical performance variation in the same module results from the change of phosphor particle distribution due to phosphor settling. The standing times of

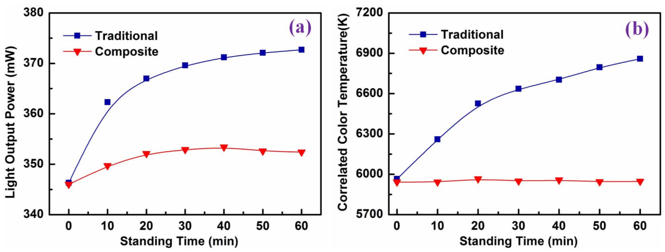

Fig. 7. Experimental results on optical performance consistency caused by phosphor settling in LED modules. (a) LOP variation, (b) CCT variation with different standing times.

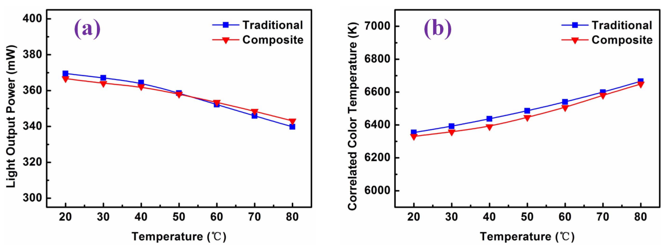

Fig. 8. Thermal stability comparison between traditional and composite light-converting materials. (a) LOP variation, (b) CCT variation with different operating temperatures.

LED modules in real production are different and hard to control, so the severe settling by traditional phosphor particles leads to low optical performance consistency.

It should be noted that the LOPs increase more in all packaging experiments with traditional phosphor as the light-converting material. However, this comes together with a strong increase in CCT and thus is no advantage.

To evaluate the thermal stability, LED modules coated with both kinds of light-converting materials were measured at different working temperatures ranging from 20 °C to 80 °C (Fig. 8). Over the whole temperature range up to 80 °C there is no significant difference in performance. The LOP drops by 29 mW for the traditional phosphor and by 24 mW for the composite particles while the increase in CCT is around 315K. This shows that the composite particles do not change the temperature characteristics of the devices.

V. Conclusion

In summary, phosphor/silicone composite particles were shown to improve the optical performance consistency in LED packaging. This is due to reduced settling of the particles that have a smaller difference in density to the silicone matrix than the traditional phosphor particles. The composite particles show almost the same material optical properties and thermal stability as the commercial phosphor particles. The average total LOP variation caused by settling decreases from 42 mW to 10 mW. Meanwhile, the total CCT variation reduces from 5680K to 208K when composite particles with a weight ratio of phosphor to silicone of 1:1 are used at the same average phosphor concentration in the coating solution.

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