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Effect of phosphor settling on the optical performance of phosphor-converted white light-emitting diode

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ABSTRACT

Phosphor settling phenomena exists during the phosphor coating process in the light emitting diode (LED) packaging industry. It is perceived that phosphor settling will affect the concentration of the phosphor, and consequently the concentration will influence the optical performance of phosphor-converted white LED light source. In this paper, an experiment based on the real packaging process was conducted to investigate the phosphor settling phenomena. It was found that the concentration variation of the phosphor embedded in the silicone matrix was very small (less than 1%). Based on the observation of the experiments, the effect of the phosphor settling in the silicone matrix on light extraction efficiency (LEE), correlated color temperature (CCT), angular color uniformity (ACU) and light intensity distribution curve (LIDC) was investigated and discussed by the three dimensional Monte Carlo ray-tracing simulations. It was discovered that the effect of the phosphor settling on the optical performance could be neglected when using the present packaging process.

1. Introduction

As being considered as the next-generation light source, white light-emitting diode (LED) has been developed rapidly in recent years. It has a lot of advantages over traditional light sources, such as relatively cool operating temperature, high luminous efficiency, long lifetime and low power consumption [1,2]. With the improvement of our daily life, not only the light efficiency but also the light quality is increasingly requested, which are crucial for a widespread penetration of white LEDs into more applications and markets. The correlated color temperature (CCT), angular color uniformity (ACU) and light intensity distribution curve (LIDC) are the three significant parameters for the light quality [3,4].

The most commonly used white LED is the phosphor-converted white LED, which is generated by combining a short wavelength blue emitter and a phosphor wavelength converter. One of the common phosphor converters is cerium-doped yttrium aluminum garnet (YAG:Ce) phosphor particles mixed with silicone matrix material, which absorbs blue light from the LED die and reemits yellow light. Since the thickness [5–7], concentration [5,6], location [8], geometry [9,10] and packaging methods [11–13] of the phosphor layer play important roles in determining the output performance and the light quality of white LEDs, the phosphor coating is a key step in the white LED packaging industry and we should try to make the thickness and concentration as uniform as possible to realize the color homogeneity. However, no matter coated by manual operation or the phosphor dispensing machine, the phosphor particles may settle inside the silicone matrix because of the gravity. In fact, there are two steps in the LED packaging process where the phosphor settling may occur. One is that the phosphor in the syringe may settle when the workers use the syringe to disperse the phosphor silicone matrix onto the top surface of the LED die. The other one happens during the phosphor silicone curing process in the vacuum oven after the coating process. The former process results in the concentration variation and impacts the consistency of the same batch of LEDs. As far as we are concerned, there are few literatures involved with phosphor settling. Sommer et al. [14] pioneered the phosphor settling work in terms of the impact of the phosphor distribution on the performance of white LEDs. They studied the phosphor settling in the former process and assumed that the phosphor converter was divided into two volumes with identical geometry and the concentration difference ratio was as large as 1:24. Being different from Sommer's work, we will deal with the phosphor settling after the phosphor silicone matrix was dispersed onto the LED dice.

In this paper, the phosphor settling was investigated by both experiments and simulations. We conducted the experiments and

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examined the concentration variation of phosphor silicone matrix at first. Based on the observation in the experiments, three dimensional ray-tracing simulations were performed to identify the light extraction efficiency (LEE), correlated color temperature (CCT), angular color uniformity (ACU) and light intensity distribution curve (LIDC) of phosphor-converted white LEDs.

2. Experiments

Since the volume of phosphor silicone matrix dispersed onto the top surface of the single LED die is very small, it is difficult to observe the settling phenomena. In our experiments, we mixed the YAG:Ce phosphor particles with Dow Corning OE6550 A/B glue at the mass ratio of 0.3:1:1 to form the phosphor silicone matrix. The phosphor particles and the silicone matrix were stirred uniformly and then the air inside the matrix was drawn out by means of vacuum pump. The phosphor silicone matrix was filled into a quartz cuvette whose thickness was only 1 mm. As shown in Fig. 1(a), according to the Beer–Lambert law, there is a logarithmic relation between the light transmittance and the product of the extinction coefficient (or called as molar absorptivity), the concentration and the path length of the light. The formula is given by Eq. (1) [15]:

$$T=\frac{I_1}{I_0}=10^{-\varepsilon lc} \tag{1}$$

where T is the light transmittance, I_0 and I_1 are the incident and transmitted light intensity, respectively, ε and c are the extinction coefficient and concentration of phosphor silicone matrix, respectively and l is the path length of the light.

Therefore, the concentration could be calculated as

$$c=-\frac{1}{\varepsilon l}\log_{10}T \tag{2}$$

Since the variation of concentration was not easy to observe, a Hitachi U-3310 ultraviolet/visible spectrophotometer was employed to observe the changes of the light transmittance instead. The wavelength range of the spectrophotometer was from 400 nm to 800 nm. As shown in Fig. 1(b), in order to fix the observation position and make sure the accuracy of the transmittance, a black shade board with a hole of 1 mm in diameter was used. The light could transmit from the hole but not the shade. We moved the positions of the cuvette and the shade board to observe the changes of the transmittance in the top layer and the center region of the phosphor silicone matrix. We recorded the transmittance data every 3 min, and for the sake of observation of obvious phosphor settling phenomena, the whole experimental process lasted as long as half an hour.

The phosphor particle size is around 13 μm, therefore the light would be scattered when reaching the phosphor particles. The diameter of the light beam was the same with the small hole, which is much larger than the phosphor particle size. In the experiments, we recorded the intensity of the incident and transmitted rays to calculate the transmittance according to Eq. (1). The scattered light is diffused to random directions, therefore the scattered light will not do much to the transmitted light intensity. Although the scattering effect on a single ray is obvious, the recorded result is the statistic one and the scattering effect on the whole light beam is small. Thus the path length of light could be regarded as constant. Based on these, we think the phosphor concentration was dependent on the transmittance. We could calculate the variation of phosphor concentration based on the transmittance obtained from the experiments.

The experimental results were shown and analyzed as follows. Fig. 2 shows the variation of transmittance in the whole wavelength range of the spectrophotometer. It could be observed that there existed a “valley” at the wavelength range of 460 nm. In fact, the main wavelength of blue light was exactly 460 nm, which was just the main excited wavelength of the YAG:Ce phosphor. So, the phosphor layer absorbed the blue light and reemitted yellow light, but the two lights did not counteract each other, which resulted in a “valley” in the transmittance curve. It was also observed that the transmittance increased after half an hour later in the whole wavelength range. From Eq. (2), the increase of transmittance meant that the concentration decreased and the settling occurred. Since the wavelength of blue light emitted by the LED die was about 460 nm, the changes of the transmittance at 460 nm was our primary concern. At the beginning, the transmittance at 460 nm was 0.49, while after half an hour the transmittance at 460 nm became 0.53. The change rate of transmittance was $(0.53-0.49)/0.49$, there was a logarithmic dependence between the concentration and transmittance, therefore the change rate of concentration after half an hour was less than 1%.

As shown in Fig. 3, during the same period of time, the transmittance at the wavelength of 460 nm varied at the top layer and center region. It is obvious that the transmittance at the top layer changed larger than that at the center region and the transmittance at the center region almost kept the same, which indicated that the phosphor settling mainly occurred at the top layer. Obviously, the concentration at the top layer decreased while the concentration at the center region kept the same, so the concentration of the bottom layer increased according to the conservation of mass. Therefore in the real LED packaging industries, we should get rid of the bottom layer and the top

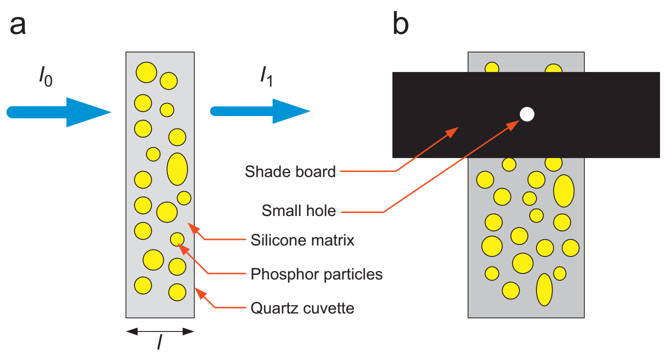

Fig. 1. Experimental diagram based on the Beer–Lambert absorption of a beam of light.

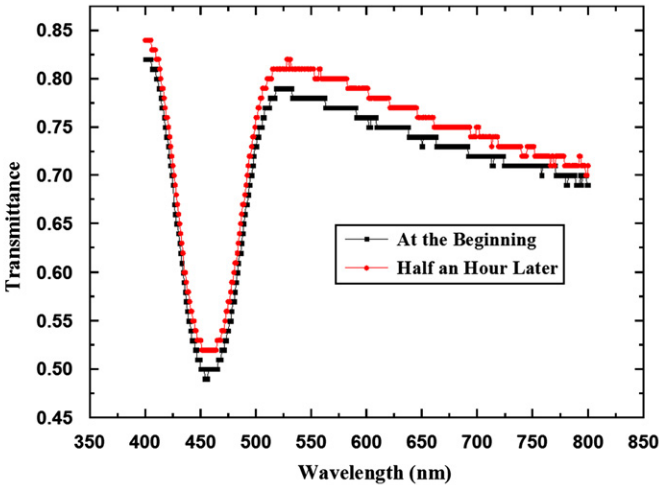

Fig. 2. Variation of transmittance at the beginning and half an hour later with the increase of wavelength from 400 nm to 800 nm.

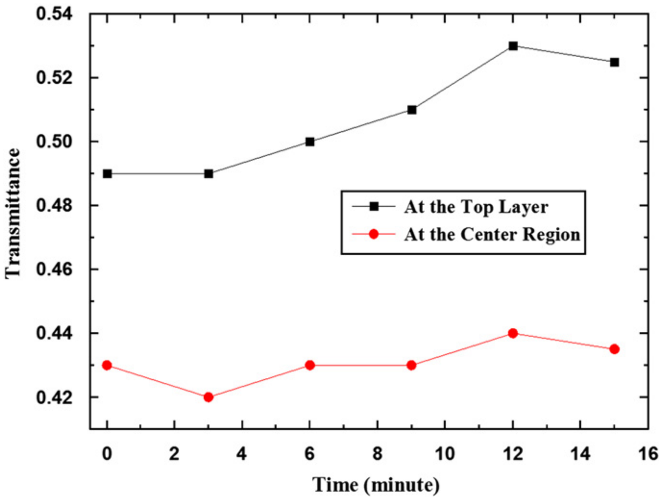

Fig. 3. Transmittance comparison at the wavelength of 460 nm at the top layer and the center region by moving the shade board with the increase of time.

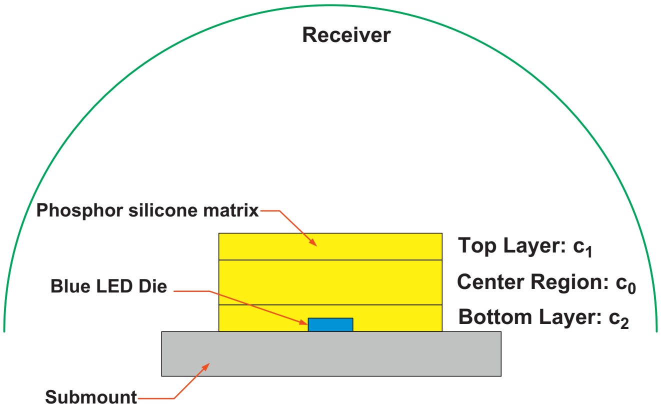

Fig. 4. Sketch of the simulation model.

layer of the phosphor silicone matrix in the syringe before the coating process to guarantee the concentration homogeneity.

In our experiments, it was found that the phosphor concentration difference was less than 1% and the phosphor settling mainly occurred at the bottom and the top layers of the phosphor silicone matrix. Usually, the larger the viscosity is, the less amount the phosphor will settle. Since the matrix viscosity is 4 Pa s, the experimental results also have significance on the manufacturing industry. Based on these observations, we will build the optical models and conduct Monte Carlo simulation to investigate the phosphor settling effect.

3. Monte Carlo ray-tracing simulation

The widely used three-dimensional Monte Carlo ray-tracing method was adopted to simulate the effect of the phosphor settling on the optical performance of phosphor-converted white LEDs. As shown in Fig. 4, the conventional blue LED die was mounted on the top surface of the submount and had the dimensions of 1 mm × 1 mm. For further details, fine structure of the LED die was also considered. The luminescent Multi-Quantum Well (MQW) were sandwiched by an n-GaN layer and heterostructures of p-GaN layer and p-AlGaN layer. A current spreading layer fabricated by Indium Tin Oxide (ITO) and a sapphire substrate were also taken into account. The top and bottom surfaces of the MQW were set as luminescent surfaces with Lambertian light distribution. The electrode pattern was not taken into consideration for simplicity. The absorption coefficients and for p-GaN, MQW and n-GaN were 5 mm^{-1}, 8 mm^{-1} and 5 mm^{-1}, respectively; their refractive indices were 2.45, 2.54 and 2.42, respectively [6]. By setting the absorption coefficients and refractive indices of the materials, the optical model of the conventional blue LED die was successfully obtained.

As shown in Fig. 4, the simulation model consisted of a blue LED die, conformal coating phosphor layer and a hemispherical receiver. The current conformal coating phosphor layer was about 100 μm thick [16,17], so did the phosphor layer in our model. In order to simulate the phosphor settling, the volume of the phosphor layer was divided into three parts with different concentrations, namely the top layer, the center region and the bottom layer. The volumes and concentrations satisfied the conservation of mass as Eq. (3):

$$V_0c_0 = V_1c_1 + V_2c_2 + V_3c_0 \tag{3}$$

where V_1, V_2 and V_3 were the volumes of the top layer, the bottom layer and the center region, respectively. V_0 was the total volume of phosphor layer. c_1, c_2 and c_0 were the concentrations of the top layer, bottom layer and center region, respectively. c_0 was also the reference concentration.

The volumes of the top layer and the bottom layer were the same and both were half of the volume of the center region (we assumed), namely

$$V_1 = V_2 = \tfrac{1}{2}V_3 = \tfrac{1}{4}V_0 \tag{4}$$

Substituting Eq. (4) into Eq. (3), Eq. (3) was simplified into

$$2c_0 = c_1 + c_2 \tag{5}$$

In the simulation Case 1, all the concentrations were the same with $c_0 = 0.23\ \text{g/cm}^3$ as a reference with no phosphor settling. Based on the experimental results, we assumed that the phosphor settling phenomena just took place in the top and the bottom layer, which resulted in that the concentration of the top layer decreased while the concentration of the bottom layer increased according to Eq. (5). We changed the concentrations as $c_1 = 0.22\ \text{g/cm}^3$ and $c_2 = 0.24\ \text{g/cm}^3$ to consider that the phosphor settling occurred in Case 2. In Case 3, the concentrations c_1 and c_2 were changed to 0.21 g/cm^3 and 0.25 g/cm^3, respectively, to assume the phosphor settling occurred further. Both in Cases 2 and 3, the concentration of the center layer was fixed as c_0, which meant that the settling did not occur in the center region.

The optical simulations for the color conversion process took into consideration of the absorption of the blue light by the yellow phosphor particles, the reemission of yellow light as well as the scattering of both blue light and yellow light within the phosphor silicone matrix and the LED die. For the success of the simulations, the blue and yellow light were separately simulated by the Monte Carlo ray-tracing method, and specific wavelengths of 465 nm and 555 nm were used in the calculation to represent blue and yellow light, respectively [11]. The wavelength-dependent refractive indices and scattering indices of the phosphor silicone matrix determined the optical performance and therefore were very important, which varied with the concentration. For the blue light, as mentioned before, the top and bottom surfaces of the MQW layer were the luminescent surfaces. For yellow light, the phosphor layer first absorbed the blue light and then re-emitted yellow light from the phosphor silicone matrix. The optical model has been validated and utilized to do simulations by our previous studies [3,6,8,11]. All the rays out of the phosphor silicone matrix were received and used to calculate the LEE, CCT, ACU and LIDC.

4. Results and analyses

The light extraction efficiency (LEE) is a key factor to influence LED luminous efficiency. Owing to the total internal reflection optical loss and Fresnel loss, the LEE is usually less than 1. In our simulations, the blue LED die was set as 1 W and by calculating the light energy incident on the receiver, the LEE could be calculated as the following equation:

$$\eta_{LEE} = \frac{\Phi_{received}}{\Phi_{emitted}} \tag{6}$$

where $\Phi_{emitted}$ and $\Phi_{received}$ were the light flux emitted by the blue LED die and the light flux including the blue light and yellow light received by the semispherical receiver, respectively.

As shown in Fig. 5, it is found that the LEEs in the three cases were almost the same. Here, we introduced a yellow to blue ratio (YBR) to illustrate the variation of correlated color temperature (CCT). The bigger the YBR is, the lower the CCT is [9]. Literally, the YBR was defined as,

$$\mathrm{YBR} = \frac{\Phi_{yellow}}{\Phi_{blue}} \tag{7}$$

where Φ_{yellow} and Φ_{blue} were the light flux of yellow light and blue light incident on the receiver, respectively.

The angular color uniformity (ACU) represents the spatial homogeneity in the whole radiation angles, which is an important parameter affecting the light quality. As shown in Eq. (8), the ACU is defined as the ratio of minimum YBR to maximum YBR in the whole radiation angle from 0° to 180°.

$$\mathrm{ACU} = \frac{\mathrm{YBR}_{\mathrm{min}}}{\mathrm{YBR}_{\mathrm{max}}} \tag{8}$$

where $\mathrm{YBR}_{\mathrm{min}}$ and $\mathrm{YBR}_{\mathrm{max}}$ were the minimum and maximum YBR in the whole radiation angles, respectively.

As shown in Fig. 6, the YBR in the three cases had the same trend and the three YBR curves almost overlapped each other. The increases of YBR at the edge of the radiation angle indicated the yellow rings in the edge of the radiation patterns. Although we could not identify the differences of these YBR curves by eyes, the decrease of ACU indicated that the phosphor settling deteriorated the optical performance. In Case 1, the ACU was 0.793, which was larger than those in Cases 2 and 3. The ACU in Case 3 was the least, which implied that the more phosphor particles settled, the worse the light quality was. However, taking the ACU in the three cases into consideration, the ACU did not decrease too much. Compared with the referenced Case 1, the ACU in Case 3 changed less than 0.03%.

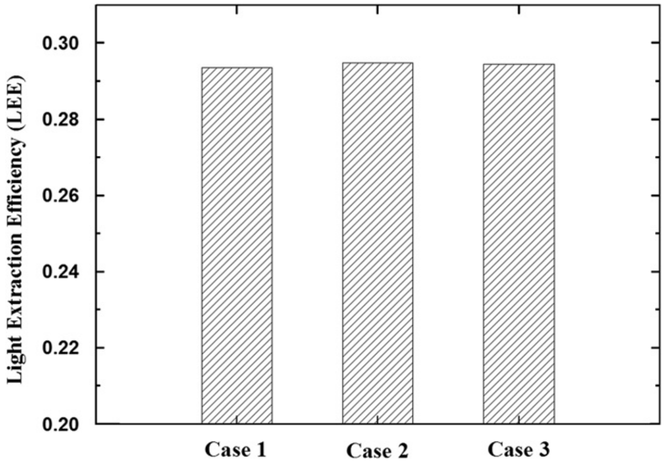

Fig. 5. Comparison of LEE among the three cases.

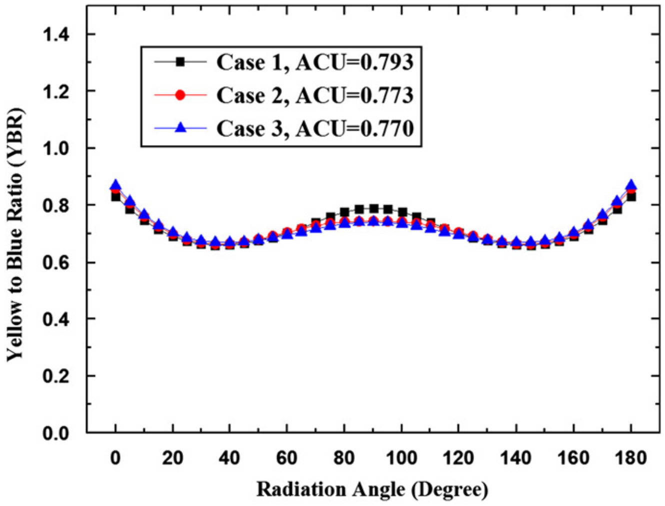

Fig. 6. Variations of YBR and ACU in the whole radiation angles under the three cases.

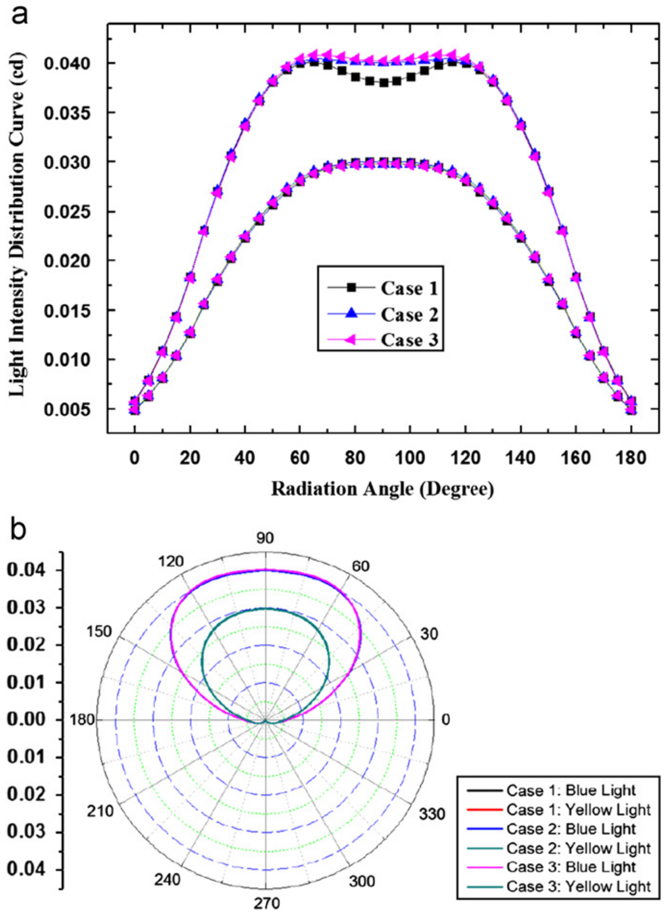

Fig. 7. Rectangular (a) and polar (b) light intensity distribution curve in the whole radiation angles.

The light intensity distribution curve (LIDC) is another key optical parameter. Different LIDCs of LED have different applications. Fig. 7 shows the rectangular LIDC and polar LIDC both for blue light and yellow light. All the LIDCs were close to the Lambertian, which just agreed with our original assumptions. In comparisons among the three cases, the LIDCs lapped over each other both for blue light and yellow light.

From the simulation results, it was found that these four parameters, LEE, CCT, ACU and LIDC degraded by the phosphor settling, but the influence was little when the concentration had small change. Taking all the parameters into consideration, in the present packaging process, the phosphor settling effect during the curing process on the optical performance of LEDs could be neglected.

5. Conclusions

In order to investigate the effect of phosphor settling on the optical performance of phosphor-converted white LED light source, both the experimental study and the three dimensional Monte Carlo ray-tracing simulations were conducted. In our experiments, it was found that the concentration of the phosphor silicone matrix during the curing of the real packaging process changed less than 1%. The phosphor settling occurred at the top layer and the concentration of the majority volume kept the same. Based on these observations, an accurate model including a blue conventional LED die and conformal coating phosphor silicone matrix was set up. The simulations indicated that although the phosphor settling occurred, the effect on optical performance was very small and could be neglected.

To completely get rid of the settling effect, the process engineers of LED packaging should not use the bottom layer and the top layer of phosphor silicone matrix in the syringe before the coating process since the phosphor settling occurred in these two layers. Also, they can increase the phosphor dispensing velocity or enlarge the volume of the syringe to alleviate the settling and improve the homogeneity of the optical performance.

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