Luminous Performances Characterization of YAG: Ce^{3+} Phosphor/Silicone Composites Using Both Reflective and Transmissive Laser Excitations

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***Abstract*—YAG: Ce^{3+} phosphor/silicone composites are widely used in solid-state lighting as a light converter to achieve white lighting. However, because of high thermal resistance and low thermal stability, the luminous performance of YAG: Ce^{3+} phosphor/silicone composite deteriorates rapidly when excited by high-power-density blue-laser. To explore the potential of blue laser-excited YAG: Ce^{3+} phosphor/silicone composites, the luminous performances under different blue laser power conditions were characterized by both the reflective and transmissive excitations using a self-built three-integrating-sphere system. Furthermore, the Monte-Carlo Ray-tracing simulation was used to illustrate the light-transmission and energy conversion mechanism in the phosphor/silicone composites. The results showed that: (1) The YAG: Ce^{3+} phosphor/silicone composite could be excited by the 0.292W laser light with the peak wavelength of 445nm, excessive laser power will cause phosphor thermal quenching and silicone carbonization. (2) The luminous flux of the composite under both the reflective and transmissive excitations gradually increased with the increase of phosphor concentration; correspondingly, the color coordinate moved to the yellow region, and the Correlated Color Temperature (CCT) gradually decreased. (3) The simulation results indicated that under the same phosphor concentration, the luminous flux obtained by reflection excitation was largely higher than that by the transmission excitation, as the light re-conversion and strong back-scattering were occurred in the reflective and transmissive laser excitation respectively.**

***Index Terms*—Light-emitting diode, YAG: Ce^{3+}, phosphor/silicone composites, laser excitation, Monte-Carlo ray-tracing.**

I. Introduction

SOLID-STATE lighting (SSL) was highly integrated into our daily life [1]. Among the solid-state light sources, the phosphor-converted white light-emitting diode (pc-wLED) is the most widely used in several applications. However, a defect exists in pc-wLED named "efficiency droop" [2]. In contrast, laser diodes (LDs) can achieve higher efficiency at high current densities, and they also have the advantages of higher power density and long irradiation distance [3]. Therefore, the phosphor-converted white light laser diode (pc-wLD) shows huge promise in next-generation high-performance SSL applications [4].

The color converter is an important component for white LEDs and LDs. Currently, phosphor/silicone composites are commonly used as the color converter in pc-wLED packaging owing to their low cost and easy preparation. Extensive researches have been conducted on the optical properties of phosphor/silicone composites used in pc-wLEDs. Liu *et al.* [5] studied the packaging method of phosphor/silicone composites in LED and found that the phosphor layer location and thickness decided the uniform of LED light output. Shen *et al.* [6] investigated the interaction of YAG yellow powder and phosphor/silicone film. The results showed that with the increase in the YAG concentration, the transmittance of phosphor/silicone decreased and the luminous efficiency increased, while the peak wavelength and full width at half maximum (FWHM) of blue and yellow lights were basically unchanged. Through TracePro simulation, Tan *et al.* [7] revealed that with the increase of phosphor particles in the phosphor/silicone composite, the luminous flux increased first, and then decrease. Moreover, through ASAP simulation, Sommer *et al.* [8] found that the phosphor concentration in the phosphor/silicone composite will affect its spatial color distribution.

Because the traditional LED packaging usually adopts the dispensing method, i.e., the phosphor/silicone composite is directly coated on the LED chip surface, this method will reduce the light extraction efficiency and is not conducive to heat

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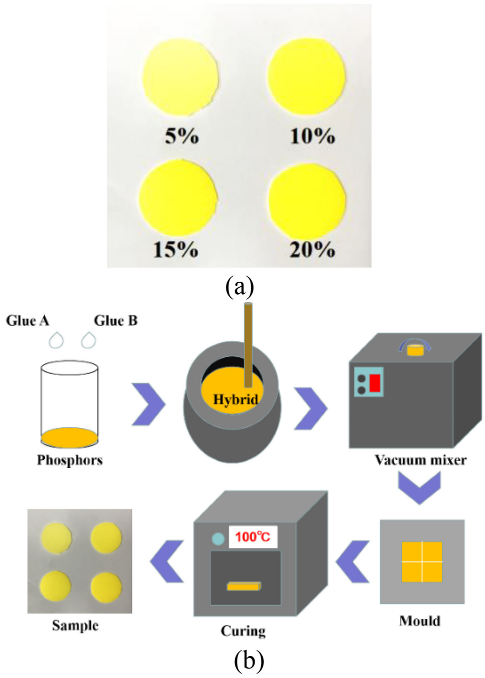


Fig. 1. (a) YAG: Ce^{3+} phosphor/silicone composite samples; and (b) its preparation process.

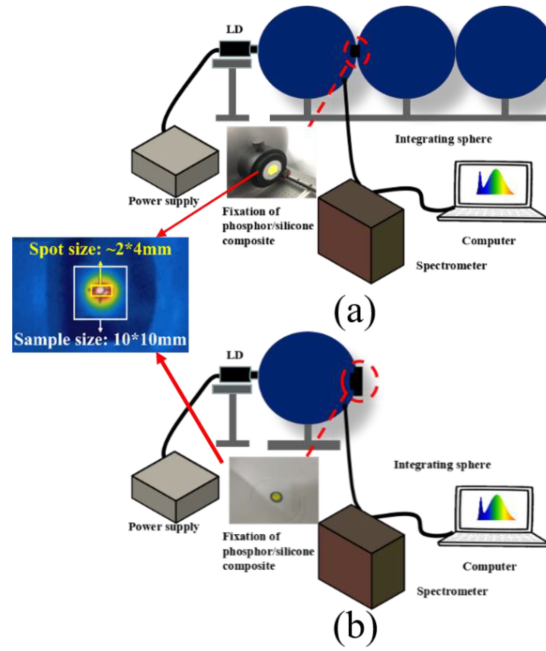


Fig. 2. Schematic diagram of three-integrating-sphere test system: (a) T-mode; (b) R-mode.

dissipation. Therefore, the remote phosphor concept has been proposed [9], that means an suitable distance will exist between blue chip and yellow phosphor convertor. Compared with LED, laser diode has a higher focusing power density [10], which will cause more serious thermal challenges. Therefore, a certain distance between phosphors and LD chip should be maintained, in order to finish independent thermal management for LD chip and phosphor/silicone composites. According to the relative direction of the input and output lights, the laser-excited remote phosphor (LERP) package can be divided into the transmissive mode (T-mode) and the reflective mode (R-mode). In the T-mode, the blue light emitted by the laser chip and yellow light converted by phosphor directly transmit through the phosphor layer. The direction of the output white light is the same as the direction of the input blue laser light. In the R-mode, a mirror layer or reflective substrate with high reflectivity is added behind the phosphor layer [11], so that the direction of the output white light is opposite to that of the input blue laser light. Previous studies have found that it is easily for T-mode to accomplish a compact and small-sized laser white light source [12], [13], while the R-mode has a better heat dissipation and luminous efficacy [14], [15]. Ma *et al.* [16] produced transmissive and reflective pc-wLD modules and compared analyzed their optical and thermal properties, the results showed that the R-mode had better luminous efficiency and color stability.

Since the light power density of the LD far exceeds that of the LED, many researchers have proposed and developed inorganic color converters in pc-wLD, such as single-crystal phosphor [17]–[19], ceramic phosphors [20]–[22], and phosphor-in-glass [23]–[27]. Only a few literatures are focused on improving the thermal stability of laser-excited phosphor/silicone composite. For example, Akvilė *et al.* [28] prepared a laser-excited phosphor/silicone composite with high thermal conductivity (~0.95W/m•K) by adding hexagonal boron nitride. To dissipate the heat accumulating in the surface, Ding *et al.* [29] proposed a converter with porous copper framework/paraffin embedded into the phosphor/silicone composite. The maximum surface temperature of proposed converter is 139 °C, when excited by blue laser with the optical power of 7.74W. Yan *et al.* [30] presented another phosphor/silicone composite converter which was mounted on the copper foam coated by boron nitride. Under high-power laser excitation of 8.13W, this converter is still able to maintain thermal stability. However, the abovementioned color converters are complex preparation and high cost at present, and the researches on phosphor/silicone composite is still not enough.

Therefore, to explore the application potential of YAG: Ce^{3+} phosphor/silicone composites in pc-wLD, this paper characterizes the luminous performance of laser-excited remote YAG: Ce^{3+} phosphor/silicone composites by both the reflective and transmissive excitations under a self-built three-integrating-sphere system.

II. Sample Preparation, Experiment and Simulation Setups

A. Sample Preparation

As shown in Fig. 1(a), four YAG: Ce^{3+} phosphor/silicone composite samples with different phosphor concentrations (the concentrations are 5%, 10%, 15%, 20%, respectively.) were prepared in this study. The selected yellow phosphor (YAG-04) and silicone (KJC-1200A/B) were purchased from Intematix Co.Ltd and Shin-Etsu Co.Ltd, respectively. Fig. 1(b) shows the sample preparation process: 1), weighing the silicone A/B and phosphor; 2) mixing phosphor/silicone; 3) stirring and defoaming phosphor/silicone mixture using vacuum mixer; 4) injecting the mixture into a mould; 5) heating the mould with100 °C for

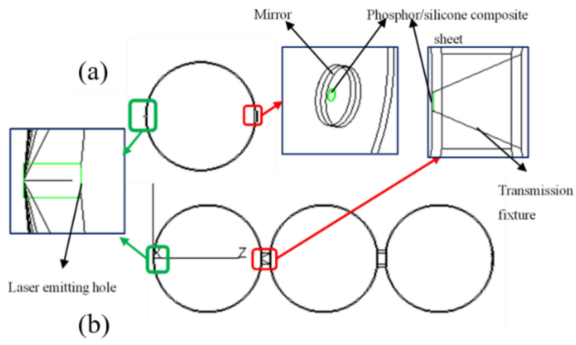


Fig. 3. The simulation model of three-integrating-sphere system in TracePro7.0 software: (a) R-mode; (b) T-mode.

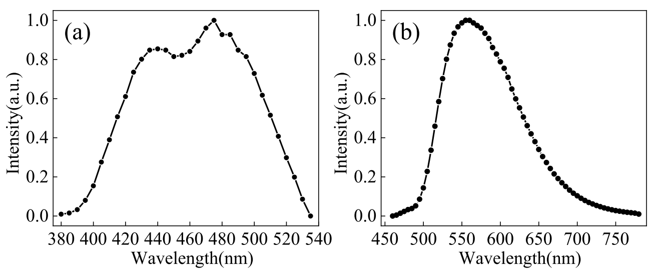


Fig. 4. (a) The excitation spectrum and (b) The emission spectrum of YAG: Ce^{3+} phosphor used in this study.

3 hours; 6) finishing the composite samples with a thickness of 1 mm.

B. Experiment Setups

As illustrated in Fig. 2, the experimental test equipment was mainly composed of a power-adjustable laser diode with the peak wavelength of 445 nm and a self-built three-integrating-sphere system. The output power of this laser diode was range from 0.08 W to 0.68W, the spot size of the blue laser on the phosphor samples is about 2*4 mm. In this study, the luminous performances of YAG: Ce^{3+} phosphor/silicone composites were characterized by transmission (T-mode) and reflection (R-mode) excitations. During the T-mode testing, the laser was placed at the light inlet of the first integrating sphere, and the test sample was placed between the first and second integrating spheres by a transmission fixture (Fig. 2a). The first, second, and third integrating spheres collected reflected light, transmitted light, and collimated light, respectively. In the R-mode testing, the laser position was remained. The test sample was placed at the light outlet of the first sphere by a reflection fixture. as depicted in Fig. 2(b), only the first integrating sphere was used in R-mode testing.

C. Simulation Model

A simplified three-integrating-sphere system was built in TracePro7.0 software, and then the Ray-tracing simulation based on the Monte Carlo algorithm was conducted. The R-mode and T-mode simulation models are shown in Fig. 3(a) and (b), respectively.

Fig. 4 displays the excitation and emission spectrum of the YAG: Ce^{3+} phosphors used in the simulation. According to reference [31], the phosphor's scattering coefficients for blue and yellow lights were set as 1 mm^{-1} and 16 mm^{-1}, respectively; the phosphor's absorption coefficients for blue and yellow lights were set as 4 mm^{-1} and 0.0233 mm^{-1}, respectively. The quantum efficiency of phosphor was 0.92 and its refractive index was 1.82. The refractive index of silicone was 1.4. The refractive index of the phosphor/silicone composite n_{mix} was calculated using the following equation:

$$n_{mix} = \sigma_{phos}{}^{*} n_{phos} + \sigma_{sil}{}^{*} n_{sil} \tag{1}$$

where σ_{phos} is the concentration of phosphor, n_{phos} is the refractive index of phosphor, σ_{sil} is the silicone's concentration, and n_{sil} was the refractive index of silicone.

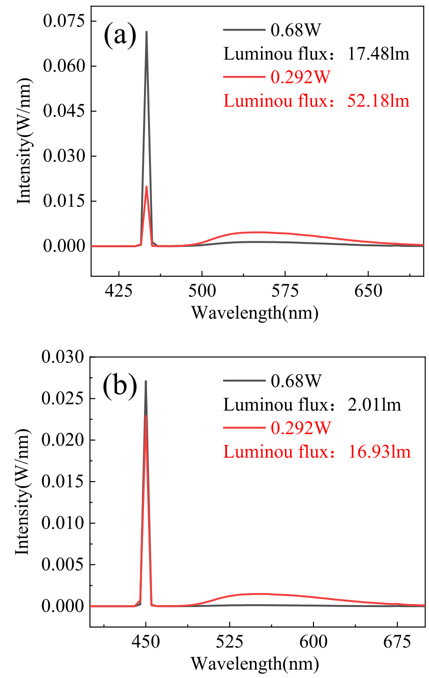
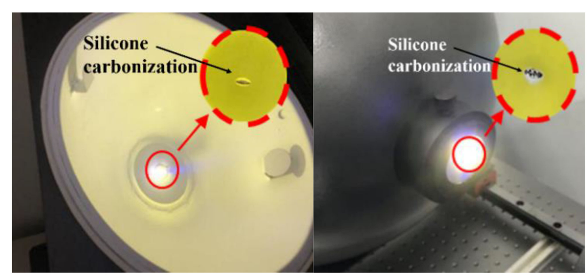


Fig. 5. The SPDs of 10% YAG: Ce^{3+} phosphor/silicone composite measured under (a) R-mode; and (b) T-mode integrating sphere 2.



(a) (b)

Fig. 6. The carbonization of YAG: Ce^{3+} phosphor/silicone composites excited by 0.68 W blue laser: (a) R-mode; (b) T-mode.

III. Results and Discussions

A. Influence of Laser Power

Fig. 5(a) show the Spectral Power Distributions (SPDs) of 10% YAG: Ce^{3+} phosphor/silicone composite sample measured under the R-mode with two laser power excitations. Transmitted

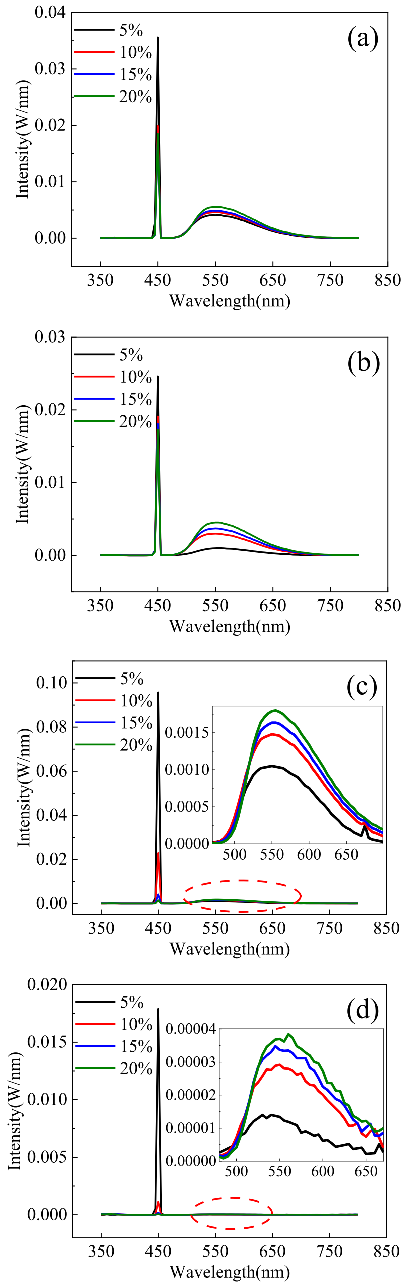


Fig. 7. The SPDs *v.s.* phosphor concentrations: (a) R-mode. (b) T-mode integrating sphere 1. (c) T-mode integrating sphere 2. (d) T-mode integrating sphere 3.

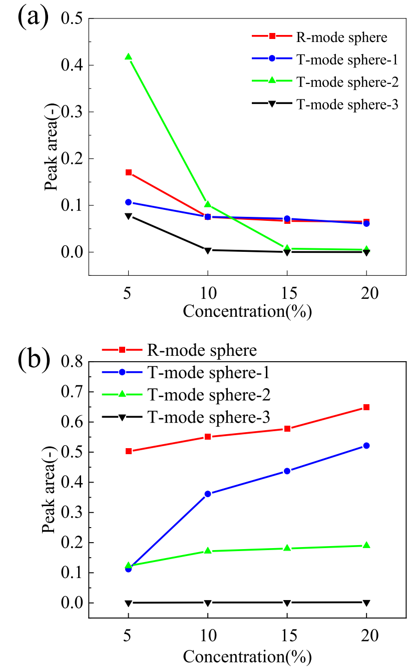


Fig. 8. The disassembled SPD peak area *v.s.* phosphor concentration: (a) $A_{\rm B}$. (b) $A_{\rm Y}$.

light of this sample in T-mode was measured in second integrating sphere, and the SPDs is shown in Fig. 5(b).

When the laser optical power increased from 0.292W to 0.68W, the spectral intensity of the blue laser increased, but the spectral intensity of the converted yellow light decreased. For both T-mode and R-mode, the luminous fluxes measured under laser excitation of 0.292W are higher than that of 0.68W. This might be explained by the occurrence of thermal quenching and silicone carbonization (see Fig. 6). The phosphor/silicone composite material was directly attached to the aluminum reflective fixture in the R-mode, which results in better heat dissipation effect. Therefore, compared with the T-mode, the degree of silicone carbonization of R-mode was relatively weak.

B. Influence of Phosphor Concentration

According to the above analysis, the laser optical power was fixed as 0.292W in this section. Fig. 7(a)–(d) show the SPDs collected by R-mode, T-mode integrating sphere 1 (reflected light), T-mode integrating sphere 2 (transmitted light) and T-mode integrating sphere 3 (collimated light), respectively. when the phosphor concentration increased, for each integrating sphere, the spectral intensity of the blue light part gradually decreased, while those of the yellow light part gradually increased. With the increase of phosphor concentration, more blue light was absorbed by phosphor particles and then was converted to yellow light. Comparison of Fig. 7(b) and (c), the converted yellow light region spectral intensity of reflected light is much larger than that of transmitted light, which proves that backside-scattering occurs more readily in phosphor/silicone composite, when excited by blue laser. With the concentration of 5%, some collimated light can be observed in integrating sphere 3, that means several blue laser lights directly penetrate the sample without conversion by phosphor. When the concentration was more than 10%, this phenomenon was greatly reduced.

Furthermore, to analyze the influence of phosphor various concentration on the SPD, an extended Gaussian peak function was used to fit the measured SPDs. The extended Gaussian model is expressed as follows:

$$y = \mathrm{y}_0 + \frac{A_B}{\mathrm{w}_1\sqrt{\pi/2}}\mathrm{e}^{-2\frac{(\mathrm{x}-\mathrm{x}_{c1})^2}{\mathrm{w}_1^2}} + \frac{A_Y}{\mathrm{w}_2\sqrt{\pi/2}}\mathrm{e}^{-2\frac{(\mathrm{x}-\mathrm{x}_{c2})^2}{\mathrm{w}_2^2}} \quad (2)$$

where y_0 is the initial value of the ordinate of the function, x is the wavelength, $A_{\rm B}$ is the peak area of the laser blue light, $x_{\rm c1}$ is the peak wavelength of the laser blue light peak, w_1 is the full width at half maximum (FWHM) of the laser blue peak, $A_{\rm Y}$ is the phosphor-emitted yellow light peak area, $x_{\rm c2}$ is the peak wavelength of the phosphor-emitted yellow light, and w_2 is the FWHM of the phosphor-emitted yellow light.

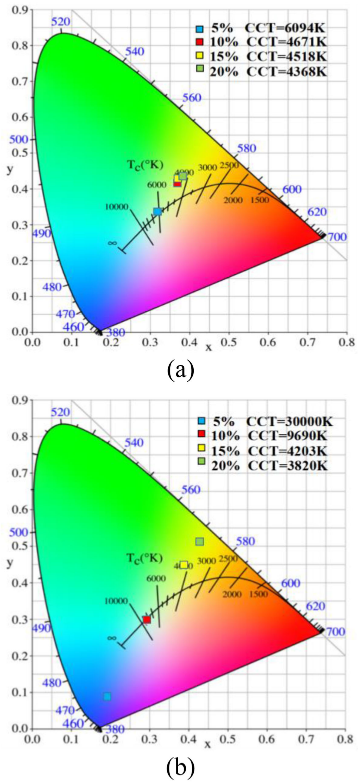


Fig. 9. The color coordinates *v.s.* phosphor concentration: (a) R-mode. (b) T-mode integrating sphere 2.

TABLE I
THE LUMINOUS FLUX DATA OF TEST SAMPLES WITH DIFFERENT CONCENTRATIONS COLLECTED AT EACH INTEGRATING SPHERES

Phosphor concentration	R-mode /lm	T-mode integrating sphere-2 /lm	T-mode integrating sphere-3 /lm	Sum of integrating sphere 2 and 3 /lm
5%	47.58	14.18	0.62	14.8
10%	52.18	16.93	0.34	17.27
15%	54.87	18.51	0.37	18.88
20%	61.87	19.26	0.4	19.66

Fig. 8 illustrates the changes of fitted SPD peak area at the R-mode and T-mode integrating spheres. For the R-mode, when the phosphor concentration increases from 5% to 10%, A_B/A_Y dropped greatly, then kept slightly decrease with increasing concentration from 10% to 20%, which results in the CCTs of test samples rapidly decreased first, and then kept relatively close, as displayed in Fig. 9(a). The cold white light close to the Planck curve could be obtained when the phosphor concentration was 5%.

For the sphere 2 of T-mode, as the phosphor concentration increased from 5% to 20%, the A_B gradually decreased, and the A_Y gradually increased, so that the A_B/A_Y decreased. Therefore, as shown in Fig. 9(b), the color temperature gradually decreased, and the color coordinates moved to the yellow area. Finally, a bluish cold white light near the Planck curve could be obtained when the phosphor concentration was 10%.

Collected at each integrating sphere under the two excitation modes, the luminous flux data of test samples with different concentrations are shown in Table I. Under the same phosphor

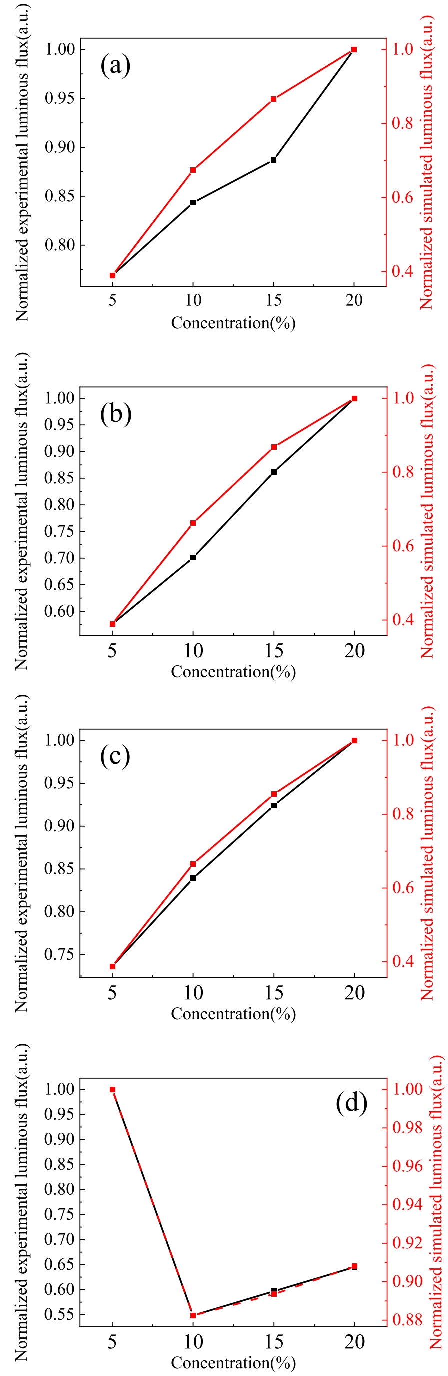


Fig. 10. The normalized luminous flux *v.s.* phosphor concentration: (a) R-mode. (b) T-mode integrating sphere 1. (c) T-mode integrating sphere 2. (d) T-mode integrating sphere 3.

concentration, the luminous flux measured under the R-mode was largely higher than the sum those in both T-mode integrating sphere 2 and 3. Fig. 10 plotted the normalized luminous flux results of both experiment and simulation, which showed the similar trends *v.s.* phosphor concentration. The simulation results were obtained by the Monte Carlo ray-tracing simulation in TracePro7.0. Fig. 11 shows the simulations results of the sample with phosphor concentration of 10% under both reflection and transmission excitation. The luminous flux obtained

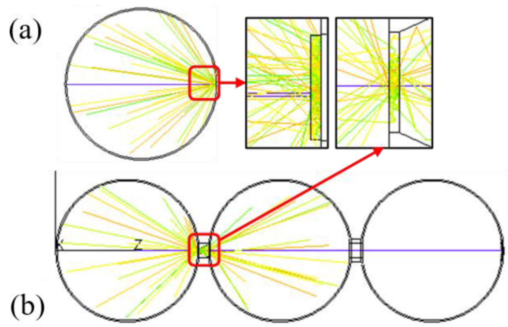


Fig. 11. The Monte Carlo ray-tracing simulation result: (a) R-mode. (b) T-mode.

by reflection excitation was largely higher than that obtained by the transmission excitation, as the light re-conversion and strong back-scattering were occurred in the reflective and transmissive laser excitation, respectively.

IV. Conclusion

In this study, the luminous performance of YAG: Ce^{3+} phosphor/silicone composites used in white LED packaging was characterized by both reflective and transmissive laser excitations in a self-built three-integrating-sphere system. The results indicated that: (1) the prepared YAG: Ce^{3+} phosphor/silicone composites could be excited by a blue laser under the optical power of 0.292 W and excessive power laser will lead to the thermal quenching and silicone carbonization; (2) the luminous flux of the YAG: Ce^{3+} phosphor/silicone composites under both the reflective and transmissive excitations gradually increased with the increase of phosphor concentration and the CCT gradually decreased; (3) the reflection excitation with the light re-conversion produced more luminous flux than the transmission excitation with a strong back-scattering.

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