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Improving light output by micro-TiO₂ scatters in pc-WLED encapsulants

TSUNG-HSUN YANG,¹  BENOÎT GLORIEUX,^{2,*}  KAI-WEI SHIAU,¹
YU-YU CHANG,¹ YEH-WEI YU,¹  AND CHING-CHERNG SUN¹

¹Department of Optics and Photonics, National Central University, Chung-Li, 32001, Taiwan

²CNRS, Université de Bordeaux, ICMCB, UMR 5026, 87 Avenue du Dr. Albert Schweitzer, 33608 F Pessac Cedex, France

*benoit.glorieux@icmcb.cnrs.fr

Abstract: Phosphor-converted white light emitting diodes (pc-WLEDs) are used worldwide for an extensive amount of applications. The device is a complex combination of various components that introduce various technical issues: materials, electrical, chemical, thermal, and so on. All of these combined to obtain a targeted optical characteristic. While most of the pc-WLEDs are sufficient for basic illumination performance, there are still many issues to improve the pc-WLED performance. In this work, we deal with the incorporation of micron size particles of titanium oxide (TiO₂) in silicone encapsulant that contains yttrium aluminum garnet (YAG) phosphor in remote phosphor pc-WLED. Based on the light output and the scattering spatial distribution measurements of the phosphor plates, we have found that several essential performance indices, like the color uniformity, the efficiency, and the amount of phosphor for the pc-WLEDs, can be adjusted by tuning the amount of TiO₂ particles and thus be optimized. With a comprehensive model using a Monte-Carlo ray tracing process combined with the Mie scattering theory, two TiO₂ loading conditions are revealed. The first one is the sparse condition that the TiO₂ particles act as the scattering particles such as to increase the output flux to improve the efficiency of YAG. The second one is the dense condition that the TiO₂ particles act more as barrier particles such so to decrease the output flux.

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The white-light source produced by phosphor-converted White Light Emitting Diode (pc-WLEDs) is nowadays widely developed to replace progressively most of the solid-state light sources [1]. Its process is based on the combination of a blue emitting die (usually InGaN) and phosphor particles (usually Yttrium Aluminum Garnet, YAG) than are incorporated in a specific packaging on the top of the die. The phosphor particles absorb partially the blue light coming from the die and down-convert it into yellow emission [2]. While the use of pc-WLEDs is well established, there is still many lacks regarding the optimization of the combination of all the components inside the lighting devices, especially about the color quality [3]. Lot of improvements can still be performed to optimize perfectly the pc-WLEDs [4]. In this context, the request of adapted modeling tools is a key issue [5].

The properties of pc-WLEDs are various and depend on the targeted use of the light. It can be the luminous intensity, the colorimetry, the color homogeneity, the spatial light distribution, the color rendering ability [6]. In order to optimize a specific property, various characteristics of the pc-WLEDs device can be the adapted like the die materials [7], the phosphor component [8], the electrical source [9], and the combination of each component. Many solutions were proposed, by adjusting the coating [10], using specific lens [11], using an adapted sapphire substrate [12], a graded refractive index multilayer phosphor package [13], or a total internal reflection collimator [14]. The use of organic-inorganic composite in the encapsulation materials is reviewed by Shen et al. [15] In all the cases, improving one property induce a drawback in another property.

Our work is dedicated to the study and optimization of the diffuser particles loading on the top of the die. These phenomena were investigated in previous publications. Two kinds of influence were analyzed as a function of the particle size. Indeed, the particle size get a direct on the Rayleigh and the Mie scattering properties [16,17].

Titanium oxide (TiO_2) is known to improve the light extraction of a pc-WLEDs [18,19]. With a specific TiO_2 microstructure on the surface of the die, Désières et al. [20] improve the power of the LED, due to the design and the high refractive index of the compound. Mont et al. [18] loaded surfactant-coated nanoparticles of TiO_2 inside an epoxy encapsulant positioned on the top of a pc-WLED. By adjusting the sizes and the number of particles, they optimize the effective refractive index of the encapsulant, so they improve the extraction of light through the device by reducing optical losses from scattering and Fresnel reflection. Huang et al. [19] found similar results. But, in addition, they showed that loading TiO_2 particles increase the thermal conductivity, so improve the thermal stability and the reliability of the device. In both cases, they did not improve the color homogeneity. It is due to the fact that for low size particles, there is no scattering effect; the particle is almost transparent to the blue and yellow light. Chou et al. [21] in addition of the luminous flux, enhanced slightly the color uniformity of their LED device, by loading 50nm size of TiO_2 . Gwang et al. [22] modified the surface of nanometric TiO_2 particles before loading into the encapsulant of pc-WLED. Their process improved the dispersibility of TiO_2 , inducing an enhancement of luminous flux and color uniformity, but lower drastically the CCT. In all these cases, any model were proposed to understand and predict the optical properties of pc-WLED.

Other influences were investigated by adding micron-size particles. In this case, the scattering characteristics of the loaded particles get a drastic influence on the optical properties of the lighting device. The most common self-influence on adding diffuser particles is the improvement on the color uniformity and the drop of the output flux. However, these effects are not the same has a function of the CCT (Correlated Color Temperature) [23]. Various kind of particle were investigated as scattering particles in order to enhance the properties of the pc-WLEDs, like silica [24,25], quantum dots [26], yttrium oxide [27], calcium carbonate [28]. It is shown that adding particles modify the color spatial homogeneity, the efficiency and the CCT [29]. Modifying the CCT is an issue, because modifying pc-WLEDs emission color change the field of application of the device. So, an efficient way of analyzing the influence of the particles loading is the compare the properties for an identical CCT. Huang et al. [30] show that adding anatase TiO_2 , rutile TiO_2 and zirconia ZrO_2 reduced considerably the CCT deviation and, in some case, enhanced the luminous flux. TiO_2 rutile get the better contribution due to its scattering property [31]. But their analysis does not consider the CCT modification.

Nguyen et al. [29] mentioned that for an 8,500K device, increase the amount of diffuser particles induce the decrease of YAG phosphor to maintain an 8,500 K device. Among various diffuser (SiO_2 , CaCO_3 , CaF_2 , TiO_2), TiO_2 is the most appropriate to increase the color uniformity. It is confirmed by Lee et al. [31] who estimated the optimization in CCT distribution and the drop of luminous efficacy for various position of TiO_2 particles.

All these previous publications correspond to experimental works explained by basic theoretical point of view. Yang et al. [32] did developed a powerful model using a Monte-Carlo ray tracing process combined with the Mie scattering theory to simulate the optical properties (Light scattering, efficiency, CCT, and luminous flux). This model was used in our work.

The aim of this work is to perform systematic analyses of experiments related to the color uniformity, CCT, the efficiency, the amount of phosphor as a function of TiO_2 loading, in order to propose a model predicting and optimizing these properties using a Monte-Carlo ray tracing process combined with the Mie scattering theory.

1. Preparation of experimental samples

The techniques used in this work were already described in previous publications [32,33]. The materials used are: yellow YAG phosphor (Cool White YAG 00902 from Hung Ta Co.), TiO₂ (anatase phase, Aldrich 99.9%), silicone (KER-2600 (A/B) from ShinEtsu Chemical Co).

The YAG particles size is of 8.5 μm for D₅₀, with a refractive index of 1.83 and a density of 4.55 g/cm³. The TiO₂ particles are composed of a huge number of small particles (D₅₀ = 1.6 μm), but particles reach a large range of size and can be up to 25 μm. TiO₂ particles get a refractive index of 2.609 and a density of 4.17 g/cm³. Silicone is formed by a mixture of polymer and curing agent. It gets a refractive index of 1.41 and a density of 1 g/cm³.

Samples are composed of plates are elaborated by mixing defined amounts of YAG, TiO₂ particles and a silicone on a specific mold after removing the air inside the mixture with the use of a vacuum chamber. The mold is then heated at 120°C on a hot plate during 5 min to cure the silicone. Plates get thickness from 0.6 mm to 1.5 mm, and weight concentration of YAG and TiO₂ ranging respectively from 3.0% to 18.0% and from 0.00% to 0.20%. The pc-WLED configuration is in Remote Phosphor configuration [34].

Two set-ups are used to performed the experiments. The first one is used to measure the scattering light distribution of the plates [33]. A He-Ne 632.8 nm laser is used as light source. The phosphor plate is positioned in the center of the rotational stage where the power meter is measuring the scattering light. These measurements are performed for all the plates. The second set-up is used to measure the emission of the phosphor plate in a pc-WLED device as a function of the wavelength. A blue chip (EZ-700 from Cree; 449 nm) is connected on a MCPCB board, and a black acrylic tube allows positioning the plate on the top of the device and to remote the plate from blue die to reduce the thermal effect (Fig. 1). The MCPCB board is connected to an efficient home-made heat dissipation to provide a stabilization of the blue light output. Measurements are performed by placing the device on an integrating sphere (Sphere Optics, Ilumnia, injection current 50 mA, voltage from 2.91 to 2.95 V) to characterize the total spectral emission from 350 to 750 nm. The integrating sphere allows measuring the incident blue light (device without plate), then the output blue light passing through the plate and the yellow light emitted by the phosphor.

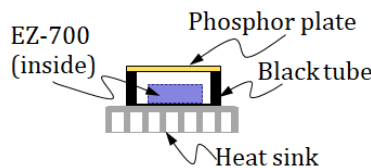


Fig. 1. Configuration of the experimental pc-WLED set-up.

2. Modelling

The first goal is to assess the key parameters involved in the mathematical model used to fit the measured data. Indeed, the light scattering is monitored by the size particles, the concentration and the refractive index of the plate components.

The light scattering is simulated by a model [35] using a Monte-Carlo ray tracing process combined with the Mie scattering theory. The goal is to compare the experimental data with the theoretical data, as a function of particles concentrations, plate thickness and phosphor/TiO₂ ratio.

The weight percentage w_t of the particles is defined by $w_t(\%) \equiv \frac{w_p}{w_p + w_s} \times 100\%$, which w_p is the weight of the particles, and w_s is the weight of the silicone. The total volume of the phosphor

particles is equal to

$$V_{ph} = \frac{V_t}{1 + \frac{D_p}{D_s} \left(\frac{1}{w[\%]} - 1 \right)} \quad (1)$$

with D_p is the density of the phosphor particles, D_s is the density of silicone, and V_t is the total volume of the phosphor particles and the silicone. Then, the equivalent phosphor particles number N is:

$$N = \frac{V_{ph}}{\int \frac{4}{3}\pi r^3 p(r) dr} = \frac{V_t}{\left[\int \frac{4}{3}\pi r^3 p(r) dr \right] \left(1 + \frac{D_p}{D_s} \left(\frac{1}{w[\%]} - 1 \right) \right)} \quad (2)$$

with r is the radius of phosphor particles, and $p(r)$ is the corresponding amount fraction for the phosphor particle of radius r . Various experiments were performed on samples containing YAG and TiO_2 . Photo of samples and examples of comparison between simulated and experimental light scattering are shown both in the Figs. 2 and 3.

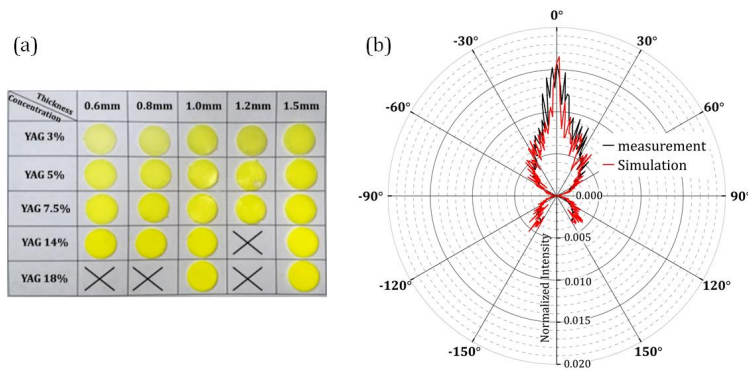


Fig. 2. (a) Photo of the investigated plates containing YAG and (b) the comparison between the simulated and the measured light scattering for plate of 1.5mm thickness and 3% of YAG

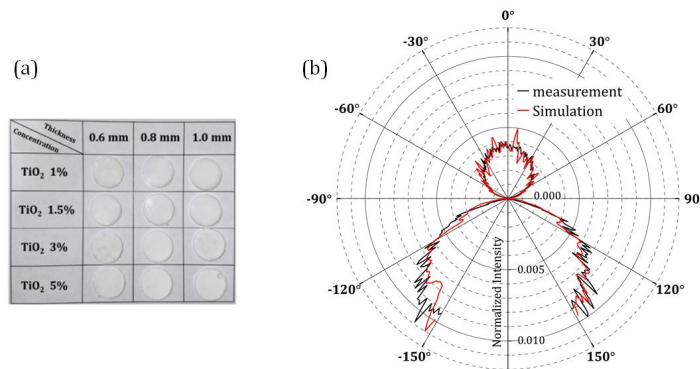


Fig. 3. (a) Photo of the investigated plates containing TiO_2 and (b) the comparison between the simulated and the measured light scattering for plate of 1.0 mm thickness and 1.5% of TiO_2 .

The modelling process is described in previous publications [32,33,35]. In order to fit the measurement on plates containing YAG, an effective radius of 2.2 μm and an effective refractive index of 1.83 was used [35]. To fit the measurement on plates containing TiO_2 , an effective radius of 0.365 μm and an effective refractive index of 2.49 was used. The difference between

the calculated value and data from the provider is due to the limit of the Mie theory and the Monte-Carlo ray tracing process used for modelling. Indeed, real powders contain a combination of YAG and TiO_2 particles, having non-spherical shapes, with a broad size distribution. So, in order for the model to fit perfectly the experimental scattering, the refractive index and the radius are adjusted. Adjusting these parameters induce that these parameters shift from a real value to an effective value.

As seen in the Fig. 2, YAG particles induce a narrow scattering, meaning that the light scattered by the YAG particle is focus in one direction. The backscattering is minor. The result is different for the TiO_2 particles as shown in Fig. 3 (a). The scattering is spatially uniform, but with an important backscattering. This phenomenon implies a larger recycling effect of the light. The light hitting the TiO_2 particles is then ore dispersed in the plate and get more chance to interact with YAG particles.

As for YAG and zirconium particles ZrO_2 [36], measurement and experimental show identical scattering (experimental and modelling) for configurations having identical number of particles, but with different concentration of particles and thickness of plate are performed. Examples are shown in the Fig. 4.

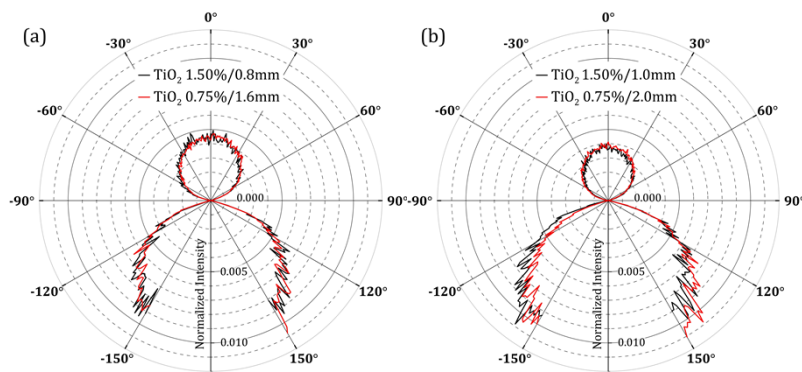


Fig. 4. Comparison between simulated and experimental light scattering for plates (a) of 0.8 mm thickness and 1.5% of TiO_2 , plates of 1.6 mm thickness and 0.75% of TiO_2 , and plates (b) of 1.0 mm thickness and 1.5% of TiO_2 , plates of 2.0 mm thickness and 0.75% of TiO_2 .

3. Light output dependent on TiO_2 concentration

The next step is to analyze the luminous properties of the samples in the integrating sphere to measure their total light outputs. As the particle number parameter defined in Eq. (2) is the key parameter to simulate the scattering. An important amount of measurements was performed in Fig. 5. The particle number replaces both the phosphor particles concentration and plates thickness to effectively analyze the optical properties.

Results were analyzed in term of luminous flux (lumen) as a function of YAG particle number. In principle, only the YAG particles function of the wavelength conversion, while the TiO_2 particles do not. So, here we consider the YAG particle numbers only rather than the total particle numbers of YAG and TiO_2 both. All the results of the luminous fluxes from the samples with various YAG weight concentrations of 3.0%, 5.0%, 7.5%, 14.0%, and 18.0% combining with several different TiO_2 weight concentrations of 0.00%, 0.03%, 0.05%, 0.07%, 0.12%, 0.17%, and 0.20% are shown in Fig. 5.

The luminous flux is the combination of two curves phenomena, that are relevant of cases of few and high amount of particle number. This curve reveals a maximum of luminous flux for an optimal particle number. This optimal particle number corresponds to the most proper

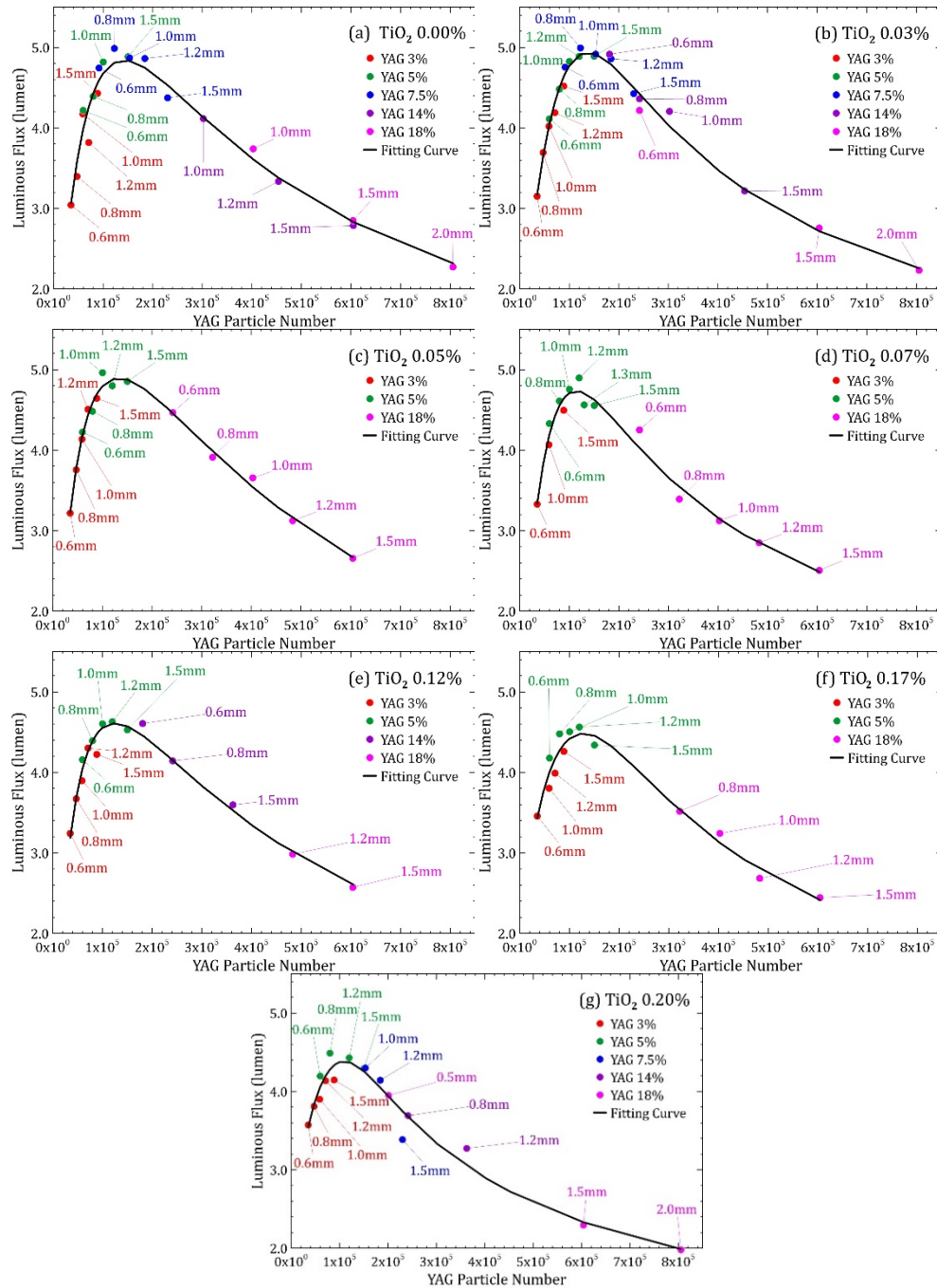


Fig. 5. Luminous fluxes depend on the YAG particles numbers from the samples with various YAG weight concentrations of 3.0%, 5.0%, 7.5%, 14.0%, and 18.0% combining with TiO₂ weight concentrations of (a) 0.00%, (b) 0.03%, (c) 0.05%, (d) 0.07%, (e) 0.12%, (f) 0.17%, and (g) 0.20%.

condition for the phosphor particles uniformly suspending in the silicone resins. In case of less particles number, the phosphor particles are considered as sparse in the silicone and does not have self-influence. The light beam is scattered by few amounts of phosphor particles. In this case, increasing the particles number increase the probability to convert blue light from the die into yellow light by the phosphor, and its correlated color temperature (CCT) decreases. In case of high amount of particles number, the density of particles increases in the silicone. The phosphor particles are considered as dense in the silicone and get self-influence. The light is scattered by many particles. The probability for the light to escape out of the plate decrease as a function of particles number.

Each phenomenon can be empirically described by $P = e^{b_x} \cdot N^{m_x}$, wherein P is the output flux in unit of lumen, N is the YAG particles number, and b_x and m_x (x can be either s or d) are empirical coefficients. Moreover, b_x and m_x are different in the case of the sparse condition (b_s and m_s) or o the dense condition (b_d and m_d). From the observations on the practical cases, b_s is negative and m_s is positive for the sparse condition, while b_d is positive and m_d is negative for the dense condition. A factor k is implemented to determine the coupling between the sparse condition and the dense condition. A large k value is relevant of a sharp shifting between the sparse condition and the dense condition, while a low value of k is relevant of a broad range of particle number where the sparse and the dense condition are mixed. So, all the samples in Fig. 5(a)~5(g) can be fitted to Eq. (3),

$$[\ln P - (m_s \cdot \ln N + b_s)] \times [\ln P - (m_d \cdot \ln N + b_d)] = k. \quad (3)$$

With no TiO₂ particles in the plate, a genetic algorithm allows to determine the fitting parameters are: $k = 0.39121$; $b_s = -3.26385$; $m_s = 0.87840$; $b_d = 6.64564$; $m_d = -1.01908$. The same experiments were performed on all the samples, containing various amount of YAG and TiO₂. Results are shown in the Fig. 6, as well as the fitting parameters presented in Table 1.

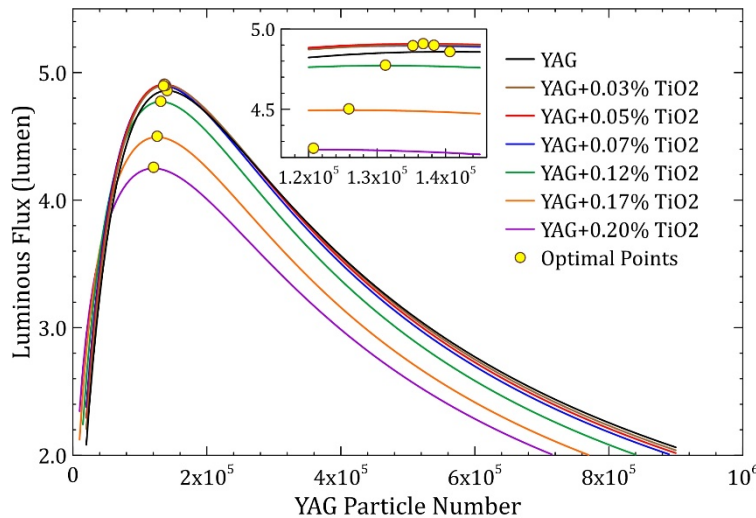


Fig. 6. Luminous flux measured on all the phosphor plates. All the parameters are determined by fitting to Eq. (3). All the optimal points are evaluated according to Eq. (9) and enlarged in the inset.

Regarding the Luminous flux shown in Fig. 6, it can be seen that except for low amount of YAG particles, increasing the amount of TiO₂ decrease the luminous flux. As for other diffusing particles [19,36], added into the phosphors packaging of LED, these particles increase the scattering probability to the light beam, so decrease its probability to escape out of the device.

Table 1. All the fitting parameters in Eq. (3) with the corresponding TiO₂ concentrations. Uncertainties of these value is 0.00002 (*m_d*, *m_s*); 0.00004 (*b_d*), 0.00003 (*b_s*) and 0.00001 (*k*)

TiO ₂ Concentration	Fitting Parameters				
	<i>m_d</i>	<i>m_s</i>	<i>b_d</i>	<i>b_s</i>	<i>k</i>
0.00%	-1.01908	0.87840	6.64564	-3.26385	0.39121
0.03%	-1.01907	0.89137	6.65958	-3.27005	0.44011
0.05%	-1.01906	0.90046	6.66936	-3.27439	0.47803
0.07%	-1.01905	0.90995	6.67956	-3.27893	0.52106
0.12%	-1.01903	0.93555	6.70710	-3.29117	0.65755
0.17%	-1.01900	0.96424	6.73796	-3.30488	0.85338
0.20%	-1.01898	0.98317	6.75832	-3.31393	1.01359

Comparing these results to the zirconia influence on the output flux [36], the influence of TiO₂ seems similar, but with a range of TiO₂ amount about 50-fold less compared to the ZrO₂. It is due to the high refractive index of TiO₂ (2.6) compared to the refractive index of zirconia (2.13).

By fitting the parameters (*b_x* and *m_x*) of the Fig. 6, it appears that these parameters get a linear relation to the proportion of the TiO₂: $w_t/(1 - w_t)$. On the other hand, the *k* parameter gets an exponential influence to the proportion of TiO₂.

$$b_s = -3.26385 - 0.20032 \times \frac{w_t}{1 - w_t} \quad (4)$$

$$m_s = 0.87840 + 0.41907 \times \frac{w_t}{1 - w_t} \quad (5)$$

$$b_d = 6.64564 + 0.45073 \times \frac{w_t}{1 - w_t} \quad (6)$$

$$m_d = -1.01908 + 0.00041 \times \frac{w_t}{1 - w_t} \quad (7)$$

$$k = 0.39121 \times e^{(3.80806 \times \frac{w_t}{1 - w_t})} \quad (8)$$

b_s increases as the weight percentage of TiO₂ increases. This parameter is related to the light recycling effect, which is enhanced by TiO₂ particles as mentioned earlier. *m_s* decrease as the weight percentage of TiO₂ increases. This parameter is related to the Stokes Shift loss (difference of energy between the incident light and the emitted light). Indeed, enhancing the recycling effect induce an increase of the Stokes Shift loss. *b_d* decreases as the weight percentage of TiO₂ increases. This parameter is relevant of the extraction of the light from the plate. As the particle number increases, the light gets more chance to be trapped in the device. *m_d* increases slightly as the weight percentage of TiO₂ increases. This parameter would be relevant of the light recycling effect. But the impact of TiO₂ particles is quite low on this parameter. *k* decreases as the weight percentage of TiO₂ increases. As previously mentioned, this parameter is relevant of the transition between the sparse condition and the dense condition. Adding TiO₂ particles, certainly due to the recycling effect, induces a smooth transition between each condition.

Moreover, the optimal YAG particles number N_{opt} , inducing a maximal luminous flux, can be determined by checking the condition of $\partial \ln P / \partial \ln N = 0$. Based on Eq. (3), the optimal YAG particles number N_{opt} can be found as:

$$\ln N_{opt} = \frac{m_s m_d (b_d - b_s) - (m_s - m_d) \sqrt{-m_s m_d k}}{(m_s - m_d) m_s m_d} \quad (9)$$

Then, the optimal YAG particles number N_{opt} as a function of the weight concentration of TiO₂ w_t is shown as the curve I in Fig. 7 based on Eqs. (4) ~ (9). In the same time, the optimal light

output P_{opt} corresponding to the optimal YAG particles number N_{opt} for a certain w_t of TiO₂ is described in Eq. (3), and all the results of various concentrations of TiO₂ are shown in the curve II in Fig. 7. Finally, the optimal output flux P_{opt} for various TiO₂ concentrations w_t is shown as the curve III in Fig. 7. The optimal YAG particles number N_{opt} decreases as the concentration of TiO₂ increases. Indeed, for the configuration where the output flux is maximum, by increasing the amount of TiO₂, the probability for an incident blue beam to be transfer into yellow light increase, because of the TiO₂ scattering. The TiO₂ particles improve the efficiency of YAG. Therefore, the overall maximal condition comes out as $w_{t,max}$ is 0.048%, N_{max} is 136,829, and P_{max} is 4.908 lm for this experimental pc-WLED system.

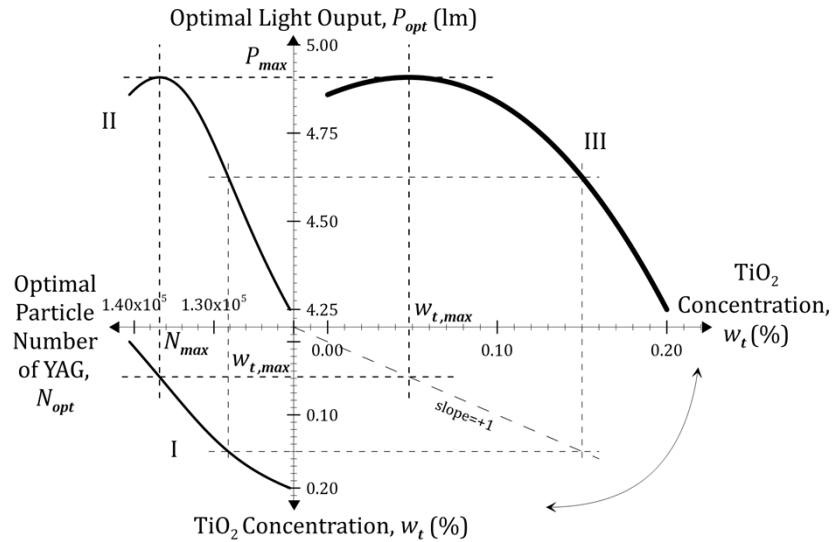


Fig. 7. Optimal YAG particles number N_{opt} (curve I, Eqs. (4)~9) and the light output P_{opt} (curve II, Eq. (3)) as a function of the weight concentration of TiO₂. Then, the maximal light output P_{max} and the corresponding maximal YAG particle number N_{max} can be found and finally the corresponding TiO₂ concentration $w_{t,max}$.

4. Conclusion

Various experiments were performed to investigate the influence of TiO₂ particles on the optical properties of remote phosphor pc-WLEDs. Experiments consist on measuring the light scattering distribution and the emission of the phosphor plate. Modelling was performed in order to simulate the scattering and the efficiency. The particle number replaces both the phosphor particles concentration and plates thickness. Optical properties were analyzed by using the particle number as a key parameter. So two well defined models were established, the first one related to the output flux P (Eq. (3)), the second one related to the optimal particles number N_{opt} (Eq. (9))

Micron size TiO₂ particles can be used to adjust the targeted properties of a pc-WLEDs to effectively increase the total light output and hence to enhance their energy conversion efficiency. TiO₂ particle get various influence because the TiO₂ particles increase the pathway of the incident blue light inside the package, so increase the possibility for the blue beam to be absorbed by a phosphor particle. In other words, it increases the efficiency of the YAG phosphor. But adding too much TiO₂ particles increase too much the pathway and the scattering, that implies a decrease in the probability of the light to escape out of the device. Empirically, at low amount of TiO₂, the condition of the particles inside the plate can be considered as sparse condition, i.e. particles do not have self-influence. The light beam is scattered by few numbers of particles. So, the output

flux increases slightly. But adding more TiO₂ shift the condition into dense condition, so induce a drastic decrease of the efficiency. Finally, the TiO₂ particles improve the efficiency of YAG. Therefore, the overall maximal condition comes out as $w_{l,max}$ is 0.048%, N_{max} is 136,829, and P_{max} is 4.908 lm for this experimental pc-WLED system.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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