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Study on phosphor sedimentation effect in white light-emitting diode packages by modeling multi-layer phosphors with the modified Kubelka-Munk theory

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In this study, we studied the phosphor sedimentation effect in white phosphor-converted light-emitting diode packages by modeling the multi-layer phosphors with gradient concentrations. The essence of phosphor sedimentation can attribute to the variation of phosphor concentrations. By modifying the Kubelka-Munk theory, we built a multi-layer phosphor model with considering the light scattering, light absorption, and light conversion process simultaneously. With a brief review of Kubelka-Munk theory, multi-layer phosphors were modeled on the basis of single-layer phosphor model. The phosphor sedimentation effect was characterized by modeling multi-layer phosphors with gradient concentrations, whereas keeping the total amount of phosphors at the same level. It is found from the five calculation cases that phosphor sedimentation will cause the drop of light extraction efficiency (LEE) by 13.04%. Furthermore, the phosphor layer with inverse-gradient concentrations will enhance the LEE 16.56%. To figure out the reasons, the light losses were calculated, and it is proved that the light loss is enhanced when phosphor sedimentation happens. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4792051]

I. INTRODUCTION

High brightness GaN-based light-emitting diode (LED) packages have been extensively applied in our daily life for their extraordinary characteristics.[1–5] Among the current state-of-the-art white LEDs, phosphor-converted scheme is most widely adopted one, in which the phosphors absorb the incident short-wavelength light from the LED chips and convert part of the absorbed light into long-wavelength emission. The lights of two selected wavelengths are usually color complementary, and the combination of the transmitted wavelengths gives the appearance of white light.

In the common LED packaging, the phosphor particles are usually embedded in silicone matrix, and then directly or indirectly coated onto the LED chips.[6] The typical phosphor particle diameter is from 2 to 20 μm, and the specific gravity is about 4.5 g/cm^3 (relatively heavy compared to silicone). The most desirable situation is to remaining the phosphor particle in suspension homogeneously during the dispensing process and curing process. However, due to the gravity effect, the phosphor particles may sedimentate in the non-Newtonian silicone matrix, especially when the matrix is heated during the curing process. The viscosity of the silicone decreases with the increase of temperature, thus phosphor sedimentation happens at high temperature more easily. The instance of obvious phosphor sedimentation phenomenon can be seen in Fig. 1. It is seen that most particles accumulate at the bottom of the matrix due to the sedimentation.

In the LED packaging industry, phosphor sedimentation is a serious problem, which may cause decay of luminous efficiency, color non-homogeneity, etc.[7–9] As far as we know, there are few studies referring to phosphor sedimentation. Lee *et al.*[7] investigated the influence of phosphor sedimentation on the luminous efficacy of white LEDs by changing the chip structures. Sommer *et al.*[8] simulated the phosphor sedimentation in LEDs by the commercial software package ASAP and found that the color temperature and flux-output are highly sensitive to the variations of phosphor distribution. In our previous study,[9] we studied the sedimentation phenomenon by observing the change of phosphor concentrations of a layer of phosphors with time elapses. Monte Carlo simulations were conducted using the software package Tracepro based on the experimental phosphor concentrations. From our previous study, we concluded that the essence of phosphor sedimentation effect on the optical performance lies in the variations of phosphor concentration distribution in the matrix.

However, there are no systematic modeling studies of the phosphor sedimentation effect so far. Some[10] of the existing studies about phosphor modeling simplified the complicated light conversion processes by considering the light scattering and light absorption while neglecting the light conversion from blue excitation to yellow emission. Some[11,12] assumed the phosphor layer as homogeneous, which could not consider the sedimentation effect. The main obstacles to model the phosphor sedimentation lie in the complex light propagation inside the phosphor layer with gradient concentrations, and the inapplicability of homogeneous medium hypothesis.

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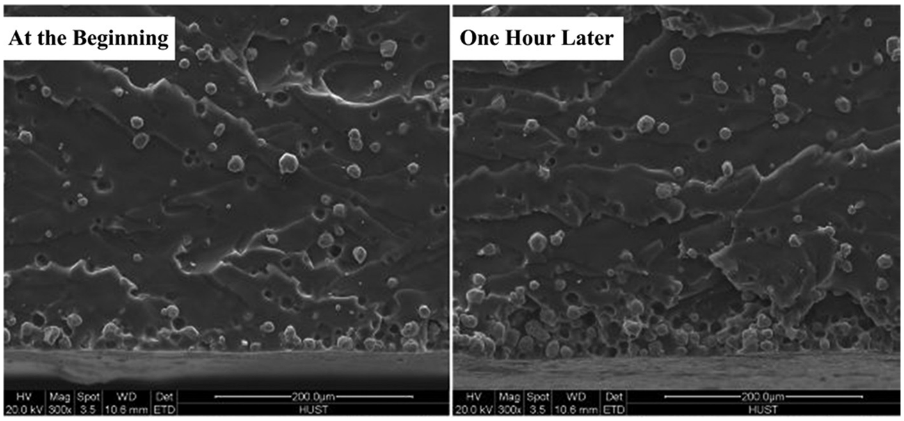

FIG. 1. Fractographs of phosphor layer after sedimentation.

In this paper, we modified that Kubelka-Munk theory by taking account of the light scattering, light absorption, and light conversion simultaneously, and modeled the multi-layer phosphors. The phosphor sedimentation consequence can be modeled by assigning gradient concentrations to the multi-layer phosphor models. The detailed modeling processes were introduced. The phosphor sedimentation effect on the light extraction efficiency (LEE) was presented, and the reasons behind the phenomena were analyzed.

This paper is organized as follows. In Sec. II, we would like to give a brief review of Kubelka-Munk theory, and then the modeling of single-layer phosphors was followed. On the basis of single-layer model, we built the multi-layer phosphors model in the end. In Sec. III, we applied the multi-layer model to investigate the phosphor sedimentation effect. The detailed discussions were also presented. Some conclusions were drawn in Sec. IV.

II. MODELING METHODOLOGY

A. Kubelka-Munk theory

Within a phosphor layer, the light propagates along with the light conversion processes, in which light absorption and light conversion take place every time light is scattered by individual particles. The Kubelka-Munk theory[13] is based on the solution of a set of simultaneous differential equations with some simplifications for the complicated light conversion processes. The simplifications include: (1) the phosphor layer is assumed as continuous optical medium; (2) the optical properties are determined by two phenomenological constants, namely, the absorption coefficient and scattering coefficient.

As shown in Fig. 2(a), the light intensity in medium at the invasion depth of z is divided into the forward scattering component $I(z)$ and the backscattering component $J(z)$. They are coupled with each other by the so called Kubelka-Munk equations as follows:

$$\begin{aligned} \frac{dI(z)}{dz} &= -(a+s)I(z) + sJ(z), \\ \frac{dJ(z)}{dz} &= (a+s)J(z) - sI(z), \end{aligned} \tag{1}$$

where a and s denote the absorption and scattering coefficients, respectively. With proper boundary conditions, Eq. (1) can be solved. As illustrated in Fig. 2(b), we can obtain the forward scattering light intensity $I_0(d)$, $I_d(d)$ and backscattering light intensity $J_0(d)$, $J_d(d)$ at the boundaries $z=0$ and $z=d$, respectively.

It is seen that the medium is considered as homogeneous because there are no variations in the absorption and scattering coefficients when the phosphor thickness changes or the invasion depth z changes. Moreover, the light conversion process is neglected from Eq. (1). At a time, only one wavelength can be considered. When the wavelength changes, the method is similar except to change the absorption and scattering coefficients according to the corresponding wavelengths.

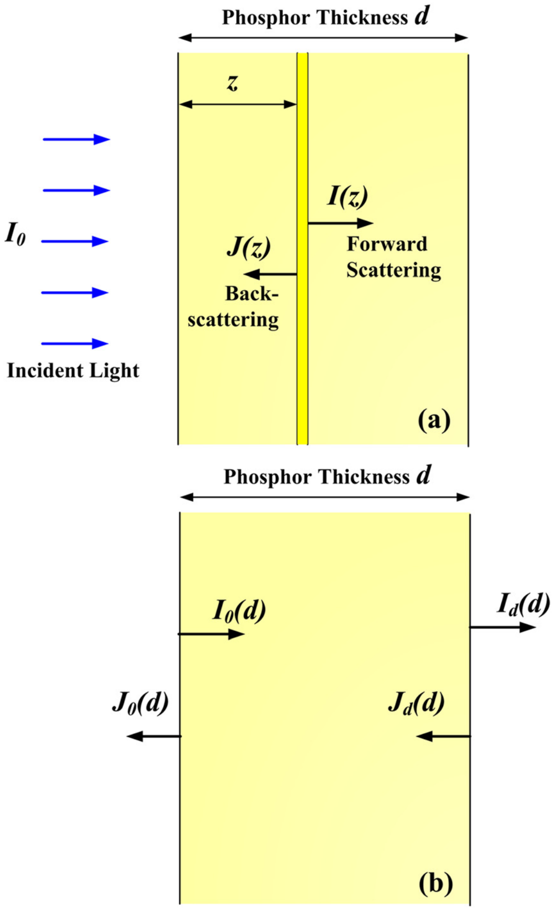

FIG. 2. (a) Forward scattering and backscattering functions with invasion depth z in a phosphor layer and (b) forward scattering and backscattering at the boundaries for a thickness of d phosphor layer.

Nevertheless, many studies[14–17] about phosphor optical properties were conducted based on this theory.

B. Modeling single-layer phosphors

In our previous study,[18,19] we modified the Kubelka-Munk theory by taking the light conversion process into consideration to make the modeling closer to the real situations. By introducing the energy conversion efficiency of phosphors from blue light to yellow emission, we can re-write Eq. (1) as

$$\begin{cases} \dfrac{dI^B(z)}{dz} = -2(a^B+s^B)I^B(z)+2s^B J^B(z) \\ \dfrac{dJ^B(z)}{dz} = 2(a^B+s^B)J^B(z)-2s^B I^B(z) \\ \dfrac{dI^Y(z)}{dz} = -2(a^Y+s^Y)I^Y(z)+2s^Y J^Y(z)+\eta a^B[I^B(z)+J^B(z)] \\ \dfrac{dJ^Y(z)}{dz} = 2(a^Y+s^Y)J^Y(z)-2s^Y I^Y(z)-\eta a^B[I^B(z)+J^B(z)], \end{cases} \tag{2}$$

where η is the energy conversion coefficient and the corresponding superscripts B and Y denote the blue light and yellow light, respectively. The appearance of coefficient 2 is because the mean path of light inside the isotropic phosphor layer is double of the thickness.[17] By solving Eq. (2), we could obtain the general forms of forward scattering function $I(z)$ and backscattering function $J(z)$ as Eqs. (3) and (4) for blue light and yellow light, respectively. For blue light,

$$\begin{aligned} I^B(z) &= A(1-\beta)e^{\alpha z}+B(1+\beta)e^{-\alpha z}, \\ J^B(z) &= A(1+\beta)e^{\alpha z}+B(1-\beta)e^{-\alpha z}. \end{aligned} \tag{3}$$

For yellow light,

$$\begin{aligned} I^Y(z) &= C(1-\nu)e^{\mu z}+D(1+\nu)e^{-\mu z} \\ &\quad + \frac{2\eta a^B}{\nu(\mu^2-\alpha^2)}[A(\mu-\nu\alpha)e^{\alpha z}+B(\mu+\nu\alpha)e^{-\alpha z}], \\ J^Y(z) &= C(1+\nu)e^{\mu z}+D(1-\nu)e^{-\mu z} \\ &\quad + \frac{2\eta a^B}{\nu(\mu^2-\alpha^2)}[A(\mu+\nu\alpha)e^{\alpha z}+B(\mu-\nu\alpha)e^{-\alpha z}], \end{aligned} \tag{4}$$

with

$$\begin{aligned} \alpha &= 2\sqrt{a^B(a^B+2s^B)}, \quad & \beta &= \sqrt{a^B/(a^B+2s^B)}, \\ \mu &= 2\sqrt{a^Y(a^Y+2s^Y)}, \quad & \nu &= \sqrt{a^Y/(a^Y+2s^Y)}, \end{aligned} \tag{5}$$

where A, B, C, and D are the undetermined constant coefficients, which can be solved with proper boundary conditions for blue light and yellow light. Similarly, we can obtain the forward scattering and backscattering light intensities at the boundaries $z=0$ and $z=d$ for blue light and yellow light, respectively. More details can be referred to our previous works.[18,19]

It is seen that this method takes account of the light conversion process, but still assumes the phosphor layer as homogeneity medium. The phosphor concentration variations along the invasion depth z into the phosphor layer are still unable to be considered.

C. Modeling multi-layer phosphors

As aforesaid, the consequence of the phosphor sedimentation is the variations of phosphor concentration distribution in the matrix. For a phosphor layer with gradient concentrations, the above models are helpless for this problem. Therefore, we have to model the multi-layer phosphors with gradient concentrations.

Considering a phosphor layer with gradient concentrations, as shown in Fig. 3, we can divide it into N sublayers, in which the thickness of each sublayer is small enough that its concentration can be considered as homogeneous. Therefore, each sublayer can be modeled using aforementioned single-layer model. The general forms of the forward scattering and backscattering light intensities of each sublayer at the boundaries for both blue light and yellow light are obtained as follows: For blue light at boundaries,

$$\begin{cases} I_0^B(d_i) = A_i(1-\beta_i)+B_i(1+\beta_i) \\ J_0^B(d_i) = A_i(1+\beta_i)+B_i(1-\beta_i) \\ I_d^B(d_i) = A_i(1-\beta_i)e^{\alpha_i d_i}+B_i(1+\beta_i)e^{-\alpha_i d_i} \\ J_d^B(d_i) = A_i(1+\beta_i)e^{\alpha_i d_i}+B_i(1-\beta_i)e^{-\alpha_i d_i}. \end{cases} \tag{6}$$

For yellow light at boundaries,

$$\begin{cases} I_0^Y(d_i) = C_i(1-\nu_i)+D_i(1+\nu_i) \\ \qquad + \dfrac{2\eta_i a_i^B}{\nu_i(\mu_i^2-\alpha_i^2)}[A_i(\mu_i-\nu_i\alpha_i)+B_i(\mu_i+\nu_i\alpha_i)] \\ J_0^Y(d_i) = C_i(1+\nu_i)+D_i(1-\nu_i) \\ \qquad + \dfrac{2\eta_i a_i^B}{\nu_i(\mu_i^2-\alpha_i^2)}[A_i(\mu_i+\nu_i\alpha_i)+B_i(\mu_i-\nu_i\alpha_i)] \\ I_d^Y(d_i) = C_i(1-\nu_i)e^{\mu_i d_i}+D_i(1+\nu_i)e^{-\mu_i d_i}+\dfrac{2\eta_i a_i^B}{\nu_i(\mu_i^2-\alpha_i^2)} \\ \qquad \times[A_i(\mu_i-\nu_i\alpha_i)e^{\alpha_i d_i}+B_i(\mu_i+\nu_i\alpha_i)e^{-\alpha_i d_i}] \\ J_d^Y(d_i) = C_i(1+\nu_i)e^{\mu_i d_i}+D_i(1-\nu_i)e^{-\mu_i d_i}+\dfrac{2\eta_i a_i^B}{\nu_i(\mu_i^2-\alpha_i^2)} \\ \qquad \times[A_i(\mu_i+\nu_i\alpha_i)e^{\alpha_i d_i}+B_i(\mu_i-\nu_i\alpha_i)e^{-\alpha_i d_i}], \end{cases} \tag{7}$$

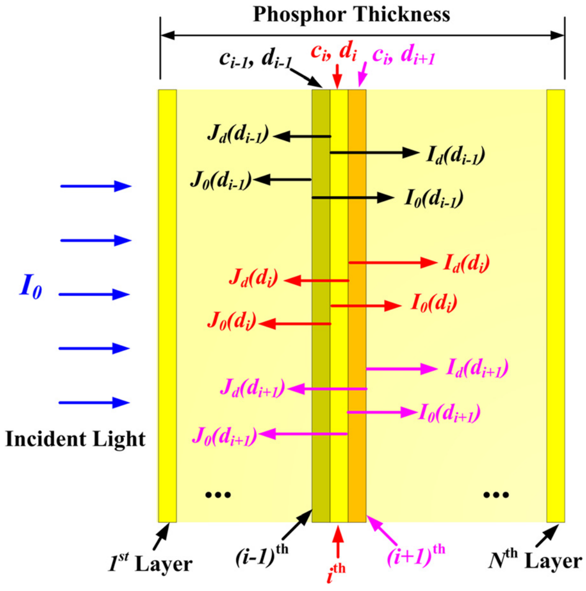

FIG. 3. Forward scattering and backscattering in multi-layer phosphors.

with

$$\alpha_i = 2\sqrt{a_i^B(a_i^B + 2s_i^B)}, \quad \beta_i = \sqrt{a_i^B/(a_i^B + 2s_i^B)}, \\ \mu_i = 2\sqrt{a_i^Y(a_i^Y + 2s_i^Y)}, \quad \nu_i = \sqrt{a_i^Y/(a_i^Y + 2s_i^Y)}, \tag{8}$$

where the subscript i denotes the ith sublayer, d_i is the thickness, and c_i is the concentration of the ith sublayer. The subscript 0 and d denote the boundaries when z equals 0 or d_i of each sublayer, respectively. From Fig. 3, we can see the forward scattering and backscattering light intensities of each sublayer are coupled with its adjacent sublayers as

$$\begin{cases} I_d(d_i) = I_0(d_{i+1}) \\ J_d(d_i) = J_0(d_{i+1}) \end{cases}, \; i = 1, 2, ..., N-1. \tag{9}$$

Since the phosphors are coated on LED chips and the substrate, the backscattered blue and yellow light projected at the surfaces of chips or substrate will be reflected back to the phosphor layer. Therefore, the boundary conditions of the whole phosphor layer for the blue light and yellow light are expressed as, respectively,

$$I_0^B(d_1) = I_0 + \gamma^B J_0^B(d_1), \quad J_d^B(d_N) = 0, \tag{10}$$

$$I_0^Y(d_1) = \gamma^Y J_0^Y(d_1), \quad J_d^Y(d_N) = 0, \tag{11}$$

where γ^B and γ^Y are the reflection coefficients on the surfaces of chips or substrate. With Eqs. (9)–(11), we can balance the number of equations and undetermined coefficients in Eqs. (6) and (7), thus we can obtain all the light intensities at all the interfaces between every two sublayers. It is seen that the forward-scattering and backscattering functions for both blue light and yellow light of each sublayer cannot be totally solved unless the necessary absorption and scattering coefficients of phosphor particles are obtained in advance. These coefficients are changed with phosphor concentrations and difficult to measure actually. In this study, we adopted the Mie-Lorenz theory to calculate the necessary coefficients since the theory is valid for all possible ratios of particle radius to the wavelength.[20] The detailed calculations of these coefficients can be referred to the Appendix.

III. STUDY ON PHOSPHOR SEDIMENTATION BASED ON MULTI-LAYER MODEL

As shown in Fig. 1, we can see that with time elapses, the phosphor particles would sedimentate and larger concentration gradient would form in the volume of matrix due to the gravity effect of phosphor particles. Based the multi-layer phosphors models, we can assign the multiple layers with gradient concentrations to indicate the phosphor sedimentation processes. In the meantime, the total amount of phosphor particles in the sedimentation processes should be constant, thus

$$\sum_{i=1}^{i=N} c_i d_i = const. \tag{12}$$

For the phosphor layer with gradient concentrations, the transmitted light through the whole phosphor layer is the forward scattering light at the boundary of the Nth sublayer. Thus, the LEE can be calculated as the sum of the transmitted light intensities of both blue and yellow light

$$LEE = [I_d^B(d_N) + I_d^Y(d_N)]/I_0, \tag{13}$$

where I_0 is the incident light intensity to the phosphor layer, as shown in Figs. 2 and 3.

TABLE I. Five cases in the calculations.

	Case 1	Case 2	Case 3	Case 4	Case 5
Layer number	1	3	2	4	2
Thickness (μm)	300	100	150	75	150
Concentrations (g/cm^3)	0.2	0.15	0.15	0.1	0.1
				0.15	
		0.2		0.25	
		0.25	0.25	0.3	0.3

In this study, we calculated five cases and their concentrations and thickness are listed in Table I. According to the real LED packaging, we assumed that the initial concentration for the whole phosphor layer was 0.2 g/cm^3 in case 1. With time elapses, the phosphor sedimentation happens and the concentration gradient forms. Due to the gravity effect, the phosphor sedimentation will result in that the concentrations of upper layers decreases while those of the lower layers increases. Thereinafter, we used the gradient concentration to refer to the gradient caused by phosphor sedimentation in particular. We used the concentration variations from case 1 to case 5 to indicate the phosphor sedimentation process.

The LEE of each case was calculated according to Eq. (13), and the calculation results were shown in Fig. 4. It is seen that the LEE decreases gradually from 0.3304 (case 1) to 0.2873 (case 5) by 13.04%, which indicates that phosphor sedimentation will cause the drop of LEE. The larger the concentration gradient is, the lower the LEE is. This trend was also reported by Lee *et al.*[7] by experiments.

We also calculated the phosphor layers with inverse-gradient concentrations, in which the concentration gradient was upside down. In other words, the concentration of upper layers was larger than that of the lower layers. One example is illustrated in Fig. 5. Similarly, the LEE calculation results were plotted in Fig. 6. It is seen that the LEE increases from

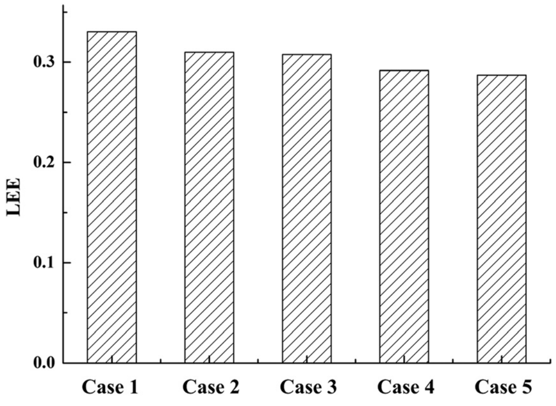

FIG. 4. Variation of LEE in five cases.

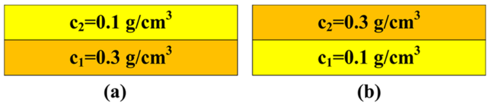

FIG. 5. Comparison of case 5 with (a) gradient concentration and (b) inverse-gradient concentration.

0.3304 (case 1) to 0.3851 (case 5) by 16.56%. The larger the concentration inverse-gradient is, the higher LEE is. This phenomenon implies a potential way to enhance the LEE that we can realize phosphor layer with gradient concentrations, and then put the cured phosphor layer upside down in the LED packaging. In this way, the phosphor layer with inverse-gradient concentrations was realized and the LEE could be enhanced.

To figure out the reasons behind these phenomena, we calculated the blue light and yellow light losses caused by the absorption of the chips or substrates, expressed as

$$Light\ Loss = [(1-\gamma^B)J_0^B(d_1) + (1-\gamma^Y)J_0^Y(d_1)]/I_0. \quad (14)$$

The light losses in the five cases with gradient concentrations and inverse-gradient concentrations were calculated and plotted in Fig. 7. It is seen that the light loss with gradient concentrations increases by 22.96% from case 1 to case 5, whereas the light loss with inverse-gradient concentrations decreases by 14.13% accordingly. The trends imply that when the phosphor layer with gradient concentrations (referring to phosphor sedimentation), more light would be absorbed by the chips or substrates. These phenomena may be explained as follows. When the concentration of the lower phosphor layer is larger than that of the upper phosphor layer (referring to phosphor sedimentation), on the one hand, more light would be backscattered on the bottom of the phosphor layer; on the other hand, the refraction index of the lower layer is larger than that of the upper layer, thus more total internal reflection events happen and more light would be trapped inside the phosphor layer due to the difference of the refraction indices. Therefore, more light would be backscattered and absorbed by the chips and substrates in the end. As a result, light loss increases but LEE decreases when the phosphor layer has gradient concentrations; light loss decreases but LEE increases when the phosphor layer has inverse-gradient concentrations.

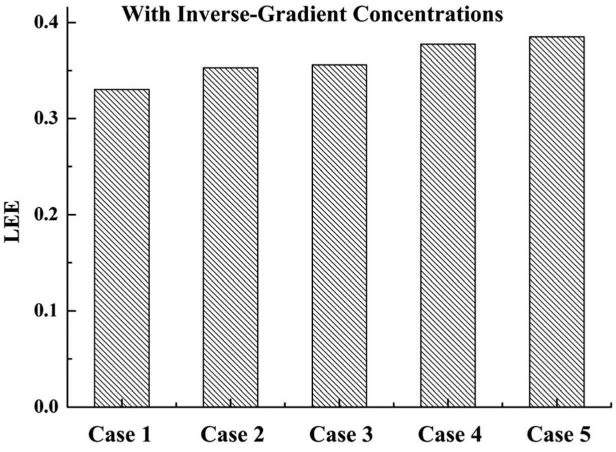

FIG. 6. Variation of LEE with inverse-gradient concentrations.

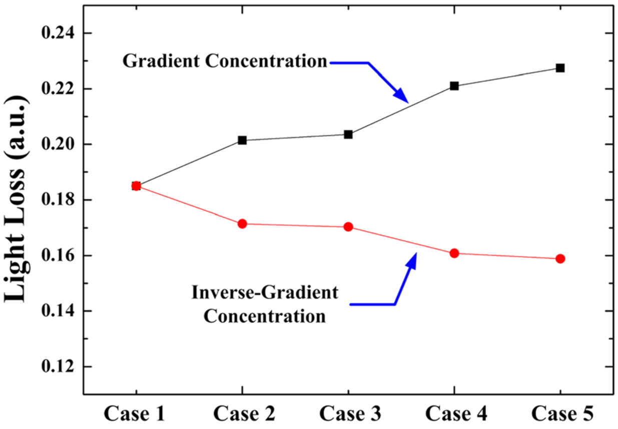

FIG. 7. Comparisons of light loss with gradient concentrations and inverse-gradient concentrations.

IV. CONCLUSIONS

In this paper, we studied the phosphor sedimentation effect in white LED packages by modeling the multi-layer phosphors. In our model, we modified the Kubelka-Munk theory, and took account of the light scattering, light absorption, and light conversion process simultaneously. The modeling of multi-layer phosphors was presented step by step in detail. It is found that phosphor sedimentation will cause the drop of LEE by 13.04% in the five calculation cases. On the contrary, the phosphor layer with inverse-gradient concentrations will enhance the LEE 16.56%. With the calculations of the light loss, the reasons why phosphor sedimentation decreases the LEE may attribute to the enhancement of light loss by the absorption of LED chips or substrates consequently.

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APPENDIX: ABSORPTION AND SCATTERING COEFFICIENTS CALCULATION

According to Mie-Lorenz theory, the extinction efficiency Q_{ext}, the scattering efficiency Q_{sca}, and the absorption efficiency Q_{abs} are normally calculated by following equations:

$$Q_{ext} = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)\mathrm{Re}(a_n + b_n), \quad (A1)$$

$$Q_{sca} = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2), \quad (A2)$$

$$Q_{abs} = Q_{ext} - Q_{sca}, \quad (A3)$$

where a_n and b_n are the expansion coefficients with even symmetry and odd symmetry, respectively, which can be determined by

$$a_n = \frac{m^2 j_n(mx)[xj_n(x)]' - j_n(x)[mxj_n(mx)]'}{m^2 j_n(mx)[xh_n^{(1)}(x)]' - h_n^{(1)}(x)[mxj_n(mx)]'}, \tag{A4}$$

$$b_n = \frac{j_n(mx)[xj_n(x)]' - j_n(x)[mxj_n(mx)]'}{j_n(mx)[xh_n^{(1)}(x)]' - h_n^{(1)}(x)[mxj_n(mx)]'}, \tag{A5}$$

where $j_n(x)$ and $h_n^{(1)}(x)$ are the Bessel function and first kind of Hankel function, respectively. x is the size parameter, which is calculated by Eq. (A6). m is the complex refractive index of the particle relative to the ambient medium. In the LED packaging, the ambient medium of phosphor particles is silicone gel, thus the complex refractive index of phosphor particle is calculated by Eq. (A7);

$$x = krn_{sil} = \frac{2\pi n_{sil}}{\lambda} r, \tag{A6}$$

$$m = n_{\mathrm{phos}}/n_{sil}, \tag{A7}$$

where k is the wave number, r is the equivalent sphere radius, λ is the wavelength. n_{phos} and n_{sil} are the refractive index of phosphor and silicone gel, respectively. Then the light scattering and absorption coefficients can be calculated as follows:

$$\begin{aligned} \mu_{sca} &= Q_{sca} A V_d, \\ \mu_{abs} &= Q_{abs} A V_d, \end{aligned} \tag{A8}$$

where A is the geometrical cross area of the particle ($=\pi r^2$) and V_d is the volume density of phosphor particles. In addition, the infinite summations in Eqs. (A1) and (A2) for the extinction and scattering efficiencies calculations converge after a certain number $N_{\max} = x + 4x^{1/3} + 2$ depending on x. Usually, the recursion formulas are used up to $N_{\max}$-th order.

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