Multi-wavelength phosphor model based on fluorescent radiative transfer equation considering re-absorption effect

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A B S T R A C T

The light propagation properties within the phosphor layer in phosphor-converted light-emitting diodes (pc-LEDs) have been extensively described by the two-wavelength model, in which two individual excitation and emission wavelengths are selected. The ignorance of the wavelength-dependence may cause deviation and cannot return the spectrum. In this paper, we extended the two-wavelength model to a multi-wavelength model based on fluorescent radiative transfer equation (FRTE) to characterize the overall light propagation behavior in the phosphor layer. In addition to the light absorption, forward scattering, and fluorescence characteristics, the re-absorption effect was further considered. The spectral radiance at any spatial location and angular direction for each wavelength was iteratively solved using the spectral element method (SEM). The model was validated by comparing the calculated spectrum and angular correlated color temperature (CCT) with experiments and good agreement was achieved with the corresponding maximum deviations of 7.6% and 3.6%, respectively. We also conducted a comparison between the multi-wavelength model with the two-wavelength model. We found that the two-wavelength model may underestimate or overestimate the optical power in the blue region and yellow region, respectively. It was attributed to assuming the peak absorption coefficient and quantum efficiency for all excitation wavelengths in the two-wavelength model.

1. Introduction

Light propagating within the phosphor layer exhibits absorption, strong forward scattering, and fluorescence characteristics [1–4]. Numerical and analytical methods have been already proposed to model the phosphor in the last decade. Monto-Carlo ray-tracing simulation has been a very accurate numerical method in modeling the optical-related objects, making it the most widely applied method for phosphor modeling [5–7]. However, due to the probability-based computing method, usually a million rays or even more need to be traced to obtain accurate results [8]. To get a deep understanding of the light propagation properties inside the phosphor, researchers have proposed some analytical methods, including the diffusion-approximation (DA) method [9] and the extended Kubelka-Munk (KM) theory for fluorescence [2,10–12]. These methods have been demonstrated to be very efficient compared to the Monto-Carlo method. Recently, we have developed a general one-dimensional phosphor model based on the fluorescent radiative transfer equation (FRTE) [13], which is the governing equation to describe the overall light propagating properties of the fluorescent media. By calculating the radiance at any location and direction, we can evaluate the spatial and angular optical properties efficiently.

However, most of the mentioned methods are based on the two-wavelength method [5–7,9–12]. In other words, only peak excitation and emission wavelengths are selected to be solved. Typically for the cerium-doped yttrium aluminum garnet (YAG: Ce) phosphor excited by a blue light-emitting diode (LED), the individual blue and yellow lights are solved. In this case, the spectrum of the phosphor-converted lighting emitting diode (pc-LED) cannot be obtained. In addition, the ignorance of the wavelength-dependence of the phosphor parameters may also cause deviation of the calculated results. To solve this problem, many studies have further considered the wavelength-dependence using the Monto-Carlo simulation [14–16]. By inputting the excitation and emission spectrum and the wavelength-dependent phosphor parameters, the spectrum can be estimated accurately.

Alternatively, Leyre et al. have developed an analytical extended adding-doubling method considering the wavelength-dependence [17]. This method is demonstrated to be very efficient to predict the angular and spectral distribution of the fluorescent media. In their model, the excitation and emission spectrum does not overlap with each other. However, for most commercial phosphor materials, there is an overlap between the excitation and emission spectrum [1]. In this case, some of the converted light will be absorbed again by the phosphor particle and

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re-emitted as light with the longer wavelength, which is known as the re-absorption effect [18]. For phosphor with large spectral overlap, the re-absorption effect cannot be neglected. To include the re-absorption effect, Leyre et al. have also extended the analytical adding-doubling method to take the spectral overlap into account [19].

In this work, we extended the previous two-wavelength model to a multi-wavelength model based on FRTE considering the re-absorption effect. The wavelength domain was divided into many discrete wavelength nodes. For each wavelength, the FRTE and the boundary conditions were established. Then, this series of complicated partial differential-integral equations (FRTEs) was solved using the spectral element method (SEM) [20], which has been successfully applied to solve RTE [21], and two-wavelength FRTEs [13]. The spectral radiance at any spatial location and angular direction for each wavelength was calculated by using this method. Next, the output spectral radiant flux distribution and then the optical power, luminous flux, average correlated color temperature (CCT) and color rendering index (CRI) were calculated. In addition, the angular CCT and spectral radiant intensity distribution were also obtained. In the following, the model was validated by comparing these calculated properties with experiments. Finally, the presented multi-wavelength model was compared with the two-wavelength model.

2. Model definition

Fig. 1 illustrates the schematic of the one-dimensional phosphor model and boundary conditions. For a planar phosphor plate with a large ratio of the diameter to the thickness, the one-dimensional assumption can be feasible, i.e., the optical variables only vary along the thickness (z-) direction [13]. We can see that in the middle filled area, when excitation light (usually blue light) hits the phosphor particle, part of the light is absorbed and the other is scattered without being absorbed. Some of the absorbed light is then down-converted to green-yellow light by the Stokes shift. The converted light may be re-absorbed by the neighboring phosphor particles and then again down-converted to light with a longer wavelength. This is also called the re-absorbing effect, which may pose an effect on the optical performance of phosphor. In the following, the method of evaluating the re-absorption effect, as well as the mentioned light propagating properties, will be presented.

Fig. 2 shows the excitation and emission spectrum (the normalized intensity are respectively denoted as $f_{ex}$ and $f_{em}$) of a typical YAG:Ce phosphor applied in PC-LEDs. There is a large spectral overlap between them, which ranges from $\lambda_{ex} (f_{em} = 0)$ to $\lambda_{em} (f_{ex} = 0)$. These two critical values divide the full visible spectrum ranging from 380 nm to 780 nm into three regions, i.e., the excitation region ($\Omega_{ex}$), the overlapped region ($\Lambda_{ol}$), and the emission region ($\Omega_{em}$), corresponding to the marked three types of spectral radiance ($I(\Omega, \lambda_{ex})$, $I(\Omega, \lambda_{ex})$, and $I(\Omega, \lambda_{em})$) to be solved in Fig. 1. Then, these three regions are uniformly discretized into $N_{ex}$, $N_{ol}$, and $N_{em}$ wavelength nodes with the corresponding interval of $\Delta_{ex}$, $\Delta_{ol}$, and $\Delta_{em}$ respectively. According to the previous literatures [13, 22], a series of extended governing FRTEs in one dimension for the excitation, overlapped, and emission regions can be respectively expressed as (1)-(3).

For each wavelength $\lambda_{ex}$ (i = 1, 2, ..., $N_{ex}$) in the excitation region, only light absorption and scattering occur:

$$\frac{\partial I(\Omega, \lambda_{ex})}{\partial z} = -[\kappa_{a}(\lambda_{ex}) + \kappa_{s}(\lambda_{ex})]I(\Omega, \lambda_{ex}) + \frac{1}{4\pi} \sum_{j=1}^{N_{em}} I(\Omega', \lambda_{em}) p(\Omega, \Omega') d\Omega'$$ \hspace{1cm} (1)

For each wavelength $\lambda_{ol}$ (k = 1, 2, ..., $N_{ol}$) in the overlapped region, besides light absorption and scattering, the fluorescence and re-absorption occur simultaneously:

$$\frac{\partial I(\Omega, \lambda_{ol})}{\partial z} = -[\kappa_{a}(\lambda_{ol}) + \kappa_{s}(\lambda_{ol})]I(\Omega, \lambda_{ol}) + \frac{1}{4\pi} \sum_{j=1}^{N_{em}} I(\Omega', \lambda_{em}) p(\Omega, \Omega') d\Omega'$$ \hspace{1cm} (2)

For each wavelength $\lambda_{em}$ (i = 1, 2, ..., $N_{em}$) in the emission region, there is no re-absorption. And light absorption, scattering, and fluorescence occur simultaneously:
The re-absorption that the integration over the entire emission wavelength region is 1. excitation wavelength in the overlapped region (our model. The expression of (3) is very similar to (2) except that all the angular direction $\Omega$.

$1$) region to the selected emission wavelength in the overlapped region ($\lambda_{ol}$). For each wavelength, the solution procedure is similar to the previous method of solving two-wavelength FRTEs except for the calculation of emission weight. The spectral radiance is iteratively calculated by solving the corresponding discrete-ordinates equation. If the stop criterion, which means that the maximum relative error of the spectral radiance is below $10^{-4}$, is satisfied, the iteration process terminates. The spectral radiance in the excitation, overlapped and emission regions can be all obtained in sequence by using the same method.

Finally, the overall optical performance can be extracted from the spectral radiance $I(z, \Omega, \lambda)$. The internal spectral radiant flux $S(z, \lambda)$ can be derived as the integration of $I(z, \Omega, \lambda)$ over the total solid angle:

$$S(z, \lambda) = \int I(z, \Omega, \lambda) d\Omega \quad (6)$$

Then, the radiant flux $\Phi(z)$ can be calculated as the integration of $S(z, \lambda)$ over the full wavelength region at each solution node:

$$\Phi(z) = \int_{0}^{780} S(z, \lambda) d\lambda \quad (7)$$

For a fluorescent material, the output optical properties are always important. Based on the definition of the Fresnel reflection of boundary 3, when the incident angle $\theta$ is below the critical angle $\theta_c$, the transmitted spectral radiant intensity $I_{\text{out}}(\theta, \varphi, \lambda)$ with a transmitted angle of $\theta, \varphi$ at $z = d$ can be expressed as:

$$I_{\text{out}}(\theta, \varphi, \lambda) = [1 - R(\cos \theta)] I(\theta, \varphi, \lambda), \quad \theta < \theta_c \quad (8)$$

where $R$ denotes the specular reflectivity and can be obtained referred to [13]. It can be seen that the obtained $I_{\text{out}}(\theta, \varphi, \lambda)$ contains both the angular and spectral distribution and can be used to calculate the angular CCT distribution according to [23]. Also, other output optical properties, including the spectral radiant flux, optical power, luminous flux, average CCT, and CRI (represented by Ra), can all be directly calculated using $I_{\text{out}}(\theta, \varphi, \lambda)$ [23].

In addition to the optical performance, the thermal performance can also be evaluated using this model. Considering the light-to-heat process, the heat generation density versus invasion depth $q(z)$ can be calculated as the sum of the unconverted part of the absorbed spectral radiant flux at each wavelength:

$$q(z) = \int_{0}^{780} \left( \kappa_e(\lambda_{ex}) + \kappa_s(\lambda_{ex}) \right) \eta_{\text{qe}}(\lambda_{ex}) \int_{0}^{780} \eta_{\text{qe}}(\lambda_{ex}) \omega(\lambda_{ex}, \lambda_{em}) d\lambda_{em} \right) \quad (9)$$

By inputting the $q(z)$ as a distributed heat source into the heat diffusion equation, we can further obtain the temperature distribution within the phosphor layer [24–27]. In this process, the optical parameters are assumed to be independent on temperature.

3. Experiments

To verify the multi-wavelength model, a remote pc-LED module same with [13] was fabricated. A commercial YAG:Ce yellow phosphor (YAG-04, Intematix Inc.) was used in our experiment. To prepare the phosphor film, the phosphor powder was mixed with the transparent silicone matrix (Dow Corning GE-6550 A and B in 1:1 mix ratio) with boundary conditions are applied. The details of the boundary conditions can be found in the previous work [13].

To calculate the spectral radiance $I(z, \Omega, \lambda)$, the governing Eqs. (1)–(3) together with the boundary conditions are solved using SEM. First, the input parameters, appearing in the governing equations and boundary conditions, need to be obtained. Then, the wavelength, spatial and angular domains must be discretized. The wavelength domain discretization has been illustrated above. The method for discretizing the spatial and angular domains is same with [13]. Next, the multi-wavelength FRTEs are solved from the shorter wavelength to the longer wavelength by step. For each wavelength, the solution procedure is similar to the previous method of solving two-wavelength FRTEs except for the calculation of emission weight. The spectral radiance is iteratively calculated by solving the corresponding discrete-ordinates equation. If the stop criterion, which means that the maximum relative error of the spectral radiance is below $10^{-4}$, is satisfied, the iteration process terminates. The spectral radiance in the excitation, overlapped and emission regions can be all obtained in sequence by using the same method.

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4. Results and discussions

To implement the model, some input parameters need to be obtained at first. The excitation, emission spectrum and the quantum efficiency of the used YAG: Ce phosphor were adopted from the experimental data published in [28], as shown in Fig. 2. We can see that a large spectral overlap occurs between 450 nm and 550 nm. The excitation region ranges from 380 nm to 450 nm and the emission wavelength ranges from 550 nm to 780 nm. Fig. 3 shows the emission weight calculated from the emission spectrum using (4). We can see that there are three critical lines dividing the figure into two parts. The emission weight is greater than zero on the condition that the excitation wavelength is lower than \( \lambda_{c,1} \) (550 nm), the emission wavelength is higher than the excitation wavelength, and the emission wavelength is higher than \( \lambda_{c,1} \) (450 nm). The maximum weight is located at the peak emission wavelength. By inputting the wavelength-dependent absorption coefficient of the YAG: Ce crystal phosphor adopted from [29], we calculated the wavelength-dependent absorption and scattering coefficients of the phosphor/silicone mixture using Mie theory [30]. In this work, we use the Henyey-Greenstein (HG) phase function and the anisotropy parameter is calculated to be 0.82 [13]. The refractive index of the phosphor plate is set to be 1.53 [1]. The LED emission spectrum is also plotted in Fig. 2 with a peak wavelength of 450 nm and the total output optical power is measured to be 0.41 W under 0.35 A. The following results are all based on these conditions. The multi-wavelength output optical power is measured to be 0.41 W under 0.35 A. The multi-wavelength optical power, luminous flux, average CCT, and CRI under driving current of 0.35 A and different phosphor concentrations were measured using an integrating sphere (ATA-1000, Everfine Inc.). In addition, the angular CCT and spectral radiant intensity distribution were measured using the same experimental setup with [13].

Using the spectrum presented in Fig. 5, we also calculated the overall optical performances including optical power, luminous flux, average CCT, and CRI (Ra). Fig. 6 shows the comparison of these properties between the measurement and calculation. We can see that the optical power rises constantly with concentration due to the increased light absorption. The luminous flux rises instead with concentration because the optical power in the green-yellow range increases and this part contributes more to the luminous flux [23]. As the concentration increases, both the average CCT and CRI decrease. For the four properties, overall good agreements are achieved with similar developing trends and relative close values. And the maximum deviation among those properties is 4.1%, which is occurred at 0.05 g/cm³ for the luminous flux.

In addition, the angular optical performance is also calculated and compared. Fig. 7(a) plots the measured and calculated angular CCT distribution. In low view angle, the calculated CCT agrees well with the measured CCT. There exists deviation when the view angle increases with the maximum deviation of 3.6%. The main source of the deviation may lie in the estimates of the input parameters and the smooth surface assumption. We further compare the measured and calculated normalized spectral radiant intensity at different angles. Fig. 7(b) to (d) show the comparisons of three representative view angles of 1.4°, 20.3°, and 58.3° with corresponding simulated CCT of 7658 K, 6868 K, and 5848 K, respectively. The values of these view angles are determined by the angular discretization and Fresnel boundary condition. As the view angle rises, the relative power in yellow region increases, corresponding to a decrease in CCT. For three angles, the spectral radiant intensity obtained by the experiment and model shows good agreement.

![Fig. 3. The emission factor calculated from the emission spectrum in Fig. 2 using (4).](image)

![Fig. 4. The radiant flux \( \Phi(z) \) and heat generation density \( q(z) \) versus normalized invasion depth \( z/d \).](image)
with similar trend and close values. As a whole, the presented model is accurate enough to model the light propagation properties and evaluate the spectral and angular performance of phosphor material.

Finally, we compare the previous two-wavelength model and the presented multi-wavelength model. Fig. 8 plots the total optical power $P_{\text{out}}$, blue light power $P_{\text{out},B}$, and yellow light power $P_{\text{out},Y}$ calculated by these two models. For the convenience of comparison, $P_{\text{out},B}$ and $P_{\text{out},Y}$ are defined as the integration of the spectral flux over the wavelength of

![Fig. 5. Comparisons of the output spectral radiant flux distribution between the experiment and model at different phosphor concentration of (a) 0.05 g/cm$^3$, (b) 0.15 g/cm$^3$, and (c) 0.30 g/cm$^3$, respectively.](image)

![Fig. 6. Comparisons of optical performance: (a) optical power, (b) luminous flux, (c) average CCT, and (d) color rendering index (Ra) between experiment and model versus varying phosphor concentration from 0.05 g/cm$^3$ to 0.30 g/cm$^3$ with an interval of 0.05 g/cm$^3$.](image)
blue light range and yellow light range, respectively. According to [30], light with spectra from 380 nm to 490 nm is defined as blue light, and light with spectra from 490 nm to 780 nm is defined as yellow light. We can see that $P_{\text{out,B}}$ obtained by the two-wavelength model is lower than that by the multi-wavelength model, whereas the case of $P_{\text{out,Y}}$ is just in the opposite. It is due to that for all the excitation wavelengths, the absorption coefficient and quantum efficiency are both assumed to be identical to the peak absorption coefficient and the peak quantum efficiency at peak excitation wavelength in the two-wavelength model. In this case, more blue light will be absorbed and more yellow light will be converted, which may further cause deviation in predicting the chromatic properties. As for the total optical power, it is interesting to see that $P_{\text{out}}$ calculated by these two models matches with each other, indicating that both two models are feasible in predicting $P_{\text{out}}$. This is also consistent with our previous literature [13].

5. Conclusions

In this work, we presented an extended multi-wavelength model based on the fluorescent radiative transfer equation to describe the simultaneous light absorption, scattering, fluorescence, and re-absorption processes. The re-absorption effect was included by introducing an emission weight in this model. The spectral radiance at any spatial location and angular direction for each wavelength was calculated by solving the multi-wavelength FRTEs using SEM. The presented model was validated by comparing the predicted spectrum and angular CCT with the experiments, with the corresponding maximum deviation of 7.6% and 3.6%, respectively. We also compared the two-wavelength model with the presented model and found that the two-wavelength model may underestimate or overestimate the optical power in the blue and yellow region, respectively. Moreover, the presented model can be easily coupled into the heat diffusion equation to evaluate the thermal and further the optical-thermal interacted performances. In summary, the efficient and accurate multi-wavelength model can be very useful in predicting the angular, spectral and even thermal properties of phosphor and other fluorescent media.

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