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Investigation of Remote-Phosphor White Light-Emitting Diodes with Multi-Phosphor Layers

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To understand how multiple phosphors in a mixture or stacked layers affect the performance of the remote-phosphor “scattered photon extraction (SPE)” white light-emitting diode (LED), a laboratory study was conducted with commercial yellow [quantum efficiency (QE) = 0.91] and red (QE = 0.59) phosphors in equal amounts. The highest light output was obtained when the longer-wavelength red phosphor was placed as the second layer. Experiments showed that when using two phosphors in an SPE package, several factors influence the performance: mixture or stacked layers; specific layer order; phosphor densities; phosphor external QE; overall spectral power distribution (SPD); phosphor excitation and emission spectra and efficiencies. © 2010 The Japan Society of Applied Physics

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Phosphor-converted, InGaN-based white light-emitting diodes (LEDs) are now competing with traditional light sources in a number of applications. However, to achieve the industry-set efficacy goal of 200 lm per watt, improvements are needed at several stages, including phosphor-conversion efficiency and extraction efficiency of phosphor-converted photons. Past studies have addressed synthesis methods to improve the phosphor quantum efficiency^{1,2)} and placement of phosphor away from the chip to improve light extraction efficiency.^{3–5)} The scattered photon extraction (SPE) method, in which the optic between the chip and the remote-phosphor layer is tailored to extract the backscattered photons, has shown 60% improvement in phosphor-converted photon extraction.³⁾ While light output and luminous efficacy improvements are essential, improvements are also needed in the color properties. Mixing a red phosphor, like SrS:Eu²⁺ or Sr₂Si₅N₈:Eu²⁺, with a green or yellow phosphor, like YAG:Ce³⁺, SrGa₂S₄:Eu²⁺, or SrSi₂O₂N₂:Eu²⁺, is one way to improve color.^{6–8)} However, mixing several phosphors randomly can reduce efficiency through emission and reabsorption of light by different phosphor types.^{6,9–13)} As a solution, past studies have investigated stacked phosphor layers placed adjacent to the LED chip and concluded that placing the phosphor with “the lowest energy of excitation edge”¹¹⁾—the longer-wavelength red phosphor—as the first layer above the chip results in higher efficiency due to reduced reabsorption.^{11–14)} This is because the overlap is avoided between the phosphor emission (“red”) from the first layer and the phosphor excitation (“green” or “blue”) from the second layer. It is not clear, however, whether the same rule of phosphor order applies to remote-phosphor configurations. The objective of the study presented here was to understand how multiple phosphors in a mixture or stacked layers affect the final performance of a remote-phosphor white LED, namely the SPE package, in terms of light output and color properties.

Commercial YAG:Ce³⁺ (yellow) and SrS:Eu²⁺ (red) phosphors were applied to three SPE lenses in different configurations and mounted on three high-power blue LEDs (Fig. 1). The SPE LED consists of transparent optics in the shape of an inverted pyramid with a layer of phosphor placed on the optics’ wide base. The SPE optics direct the blue LED radiation toward the phosphor layer, and this radiation is either absorbed, converted and emitted, or

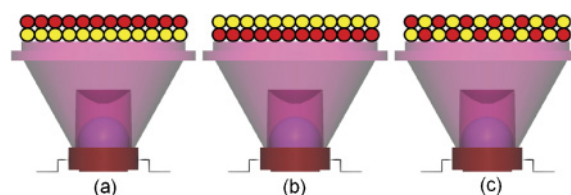


Fig. 1. (Color online) SPE LED packages with multi-phosphor layer configurations: (a) Y-R (yellow first, red second); (b) R-Y (red first, yellow second); (c) YR random mixture.

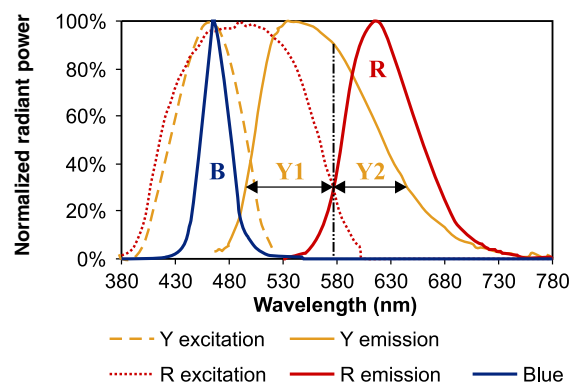


Fig. 2. (Color online) Excitation and emission spectra of the yellow and red phosphors; emission spectrum of the “blue” LED excitation light source.

scattered by the phosphor particles. Figure 1 illustrates three phosphor configurations: Y-R (yellow first, red second); R-Y (red first, yellow second); and YR-mixture (yellow and red randomly mixed into a single layer). Figure 2 illustrates the excitation and emission spectra of these two phosphors. The spectral power distributions (SPD) of the three SPE LED packages were measured using an integrating sphere with a spectroradiometer. In all cases, the amount of phosphor was kept the same, 10 mg for each lens. The amount of phosphor determines both the total amount of downconverted light and the ratio of forward to backward extracted light.¹⁴⁾

To compare results, in addition to the experiment, light output and chromaticity values were calculated based on the theory proposed by Fran *et al.*,¹⁵⁾ and optical ray-tracing analyses were conducted using LightTools™ software and a method similar to that used in an earlier study.¹⁶⁾

The transmitted, T_r , and reflected, R_e , radiant power from a single yellow phosphor layer, incident with a blue excitation light source, can be written as shown in Fig. 3,¹⁵⁾

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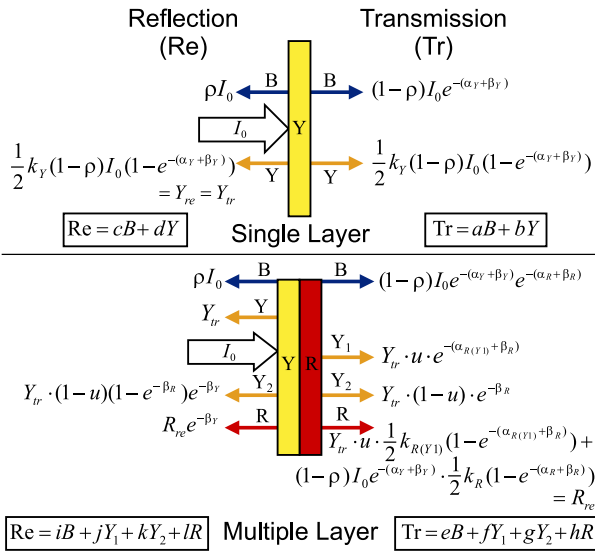


Fig. 3. (Color online) Transmitted and reflected radiant power passing through a single phosphor layer and multiple phosphor (yellow and red) layers.

where I_0 is the incident blue radiant power, α and β are phosphor absorption and scattering coefficients, respectively, k is the phosphor conversion efficiency, and ρ is the Fresnel reflection coefficient. α_Y or α_R and k_Y or k_R denote either a yellow (“Y”) or red (“R”) phosphor absorption coefficient and conversion efficiency when excited by “blue” photons, while β_Y or β_R is the scattering coefficient. Light transmitted and reflected from a single phosphor layer will include blue (“B”) and yellow (“Y”) radiant power as shown in Fig. 3, where the coefficients a through d represent the amount of B or Y radiant power. For multiple phosphor layers (yellow and red), converted yellow radiant power from the first layer is divided into Y_1 and Y_2 , where Y_1 can be absorbed by the red phosphor because it lies within the absorption (excitation) spectral range of red phosphor, while Y_2 cannot be absorbed by the red phosphor because it lies outside this range (Fig. 2). The ratio of Y_1 to Y is denoted as u . The parentheses in the subscript (i.e., $\alpha_{R(Y_1)}$ and $k_{R(Y_1)}$) denote the red phosphor absorption coefficient and conversion efficiency when excited by Y_1 photons (Fig. 2). For blue radiant power incident on multiple phosphor layers, reflected and transmitted light will include some portion of the blue, red, and yellow (Y_1 and Y_2) radiations. These radiant powers can be written as shown in Fig. 3, where the coefficients e through l represent the amount of radiant power in each component of the spectrum (defined in Fig. 2).

The total radiant power, including transmitted and reflected, can be written in the spectral form as

$$S(\lambda) = (e + i)B(\lambda) + (f + j)Y_1(\lambda) + (g + k)Y_2(\lambda) + (h + l)R(\lambda). \quad (1)$$

From eq. (1), the luminous flux (lumen) can be calculated from¹⁷⁾

$$\Phi = K_m \sum S(\lambda)V_\lambda(\lambda)\Delta\lambda. \quad (2)$$

where K_m is a constant, V_λ is the relative photopic luminous efficiency function, and $\Delta\lambda$ is the wavelength interval.¹⁷⁾ The CIE 1931 chromaticity coordinates, x , y , and z , can be calculated using the three imaginary primary colors,

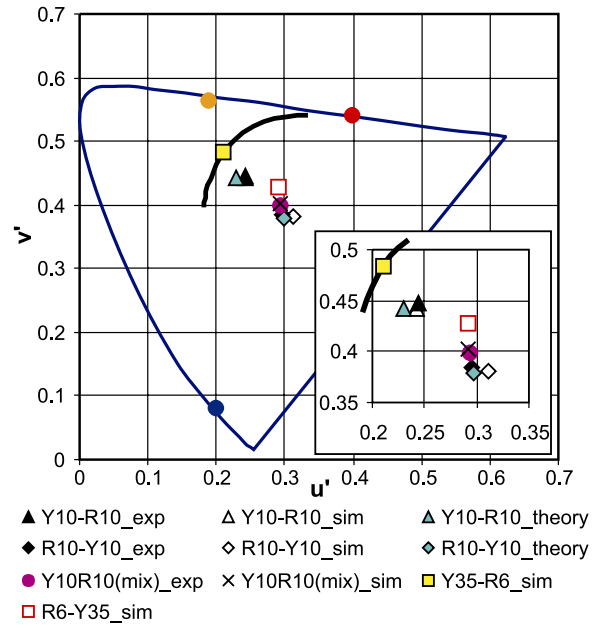


Fig. 4. (Color online) Chromaticity results on CIE 1976 UCS diagram for phosphor configurations of Y10-R10, R10-Y10, and Y10-R10 mixture from experiment, optical ray-tracing (simulation), and theory calculation, along with Y35-R6 and R6-Y35 from optical ray-tracing.

Table I. Light output results in radiant energy and luminous flux for three phosphor configurations from experiment, optical ray-tracing (simulation), and theory calculation.

Phosphor	Experiment		Simulation		Theory	
	P (W)	Φ (lm)	P (W)	Φ (lm)	P (W)	Φ (lm)
Y10-R10	0.102	27.8	0.099	26.6	0.108	29.6
R10-Y10	0.089	17.5	0.082	15.7	0.092	17.4
R10-Y10 mix	0.088	18.5	0.088	18.6	—	—

given by $X = h \sum S(\lambda)x(\lambda)\Delta\lambda$; $Y = h \sum S(\lambda)y(\lambda)\Delta\lambda$; $Z = h \sum S(\lambda)z(\lambda)\Delta\lambda$; where h is an arbitrary constant, and $x(\lambda)$, $y(\lambda)$, and $z(\lambda)$ are the spectral tristimulus values from the appropriate color matching function.¹⁷⁾ Finally, the (x, y) chromaticity coordinates can be converted to CIE 1976 (u', v') chromaticity coordinates using the following relationships:¹⁷⁾

$$u' = \frac{4x}{-2x + 12y + 3}; \quad v' = \frac{9y}{-2x + 12y + 3}. \quad (3)$$

Figure 4 plots the chromaticity values (u', v') of the output light on the CIE 1976 chromaticity diagram for the three phosphor configurations. The results from experiment, theory-based calculation, and optical ray-tracing (simulation) agree well. The chromaticity values are quite different for the three phosphor configurations, even though the amount of yellow and red phosphors is the same in all cases. Table I shows the radiant power and luminous flux. Here again the results from experiment, calculation, and simulation agree well. The Y10-R10 package, where the longer-wavelength red phosphor is the second layer, yielded the highest light output among all three configurations: 15% more in radiant power and 59% more in luminous flux than the R10-Y10 package.

This result is opposite of that found in a past study¹¹⁾ for an LED package with the phosphor layer in contact with the

Table II. Optical ray-tracing (simulation) results of light output, in radiant watts and lumens, for three phosphor configurations with similar and dissimilar external quantum efficiencies.

Phosphor	Simulation		Simulation	
	QE(Y) = 0.91	QE(R) = 0.59	QE(Y) = 0.91	QE(R) = 0.91
	P (W)	Φ (lm)	P (W)	Φ (lm)
Y10-R10	0.099	26.6	0.108	28.8
R10-Y10	0.082	15.7	0.105	21.3
R10-Y10 mix	0.088	18.6	0.105	22.7

chip, where a longer-wavelength red phosphor was used as the first layer and a shorter-wavelength blue phosphor was used as the second layer (R-B), yielding 21% more radiant power than the opposite phosphor configuration (B-R).¹¹⁾ The light output and the color properties depend on the SPD, which in this case depends on the composition of the relative radiant power of the phosphor-converted photons and the remaining pump (“blue”) photons. The likely reason for the difference in results is that in a conventional LED package where the phosphor is in contact with the chip, a significant portion of the converted photons from the first layer that are emitted backward are absorbed by the chip. However, in the SPE package, these backward-emitted photons are extracted out of the LED package, contributing to the total light output. In both types of package, the forward-emitted converted photons from the first layer will enter the second phosphor layer and undergo scattering losses and absorption–emission if the energy falls within the excitation energy range of the second phosphor. However, the SPE package contributes a greater proportion of photons from the first phosphor layer to the total light output, and therefore the first phosphor layer becomes the dominant layer. The red phosphor used in this study had lower external quantum efficiency (59%) than the yellow phosphor (91%). This could possibly explain the lower radiant power output of the SPE package with the R10-Y10 configuration.

Because the optical ray-tracing results matched well with the experiment, subsequent studies included only ray-tracing. A second study investigated the effect of each phosphor’s external quantum efficiency on the results. Here the yellow and red phosphors were assumed to have equal external quantum efficiencies. The results showed that the radiant power output is very close for all three configurations, while the luminous flux is different (Table II). This difference is a result of the different SPDs of light from the different phosphor configurations (Fig. 5). Different SPDs, when weighted by the luminous efficiency function, $V(\lambda)$, will yield different lumen values (Table II).

Because many applications require white light with chromaticity values on or very close to the blackbody locus, a third study was carried out with different amounts of yellow (35 mg) and red (6 mg) phosphor to achieve ideal chromaticity values. Comparing Y35-R6 with R6-Y35, the results showed Y35-R6 was on the blackbody locus and had 26% more radiant watts and 80% more lumens than R6-Y35,

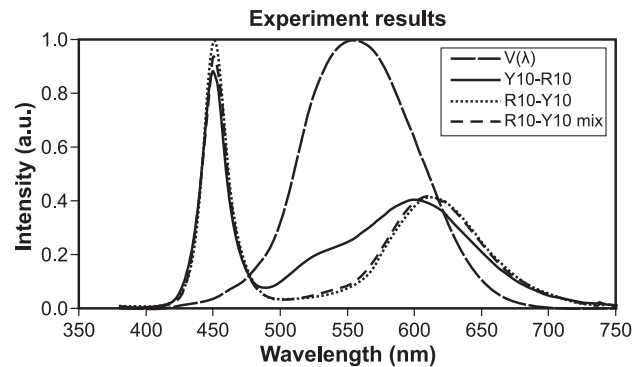


Fig. 5. $V(\lambda)$ and SPDs of the three phosphor configurations with dissimilar external quantum efficiencies (from simulation).

which had chromaticity values far below the blackbody locus (Fig. 4).

In conclusion, when using two types of phosphor in an SPE LED package, several factors will influence the final performance, including: mixture or stacked layers; specific order of the layers; densities of the phosphor medium; external quantum efficiencies of the different phosphors; overall SPD; phosphor excitation and emission spectra and efficiencies.

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- 1) Y. D. Jiang, G. Villalobos, J. C. Souriau, H. Paris, C. J. Summers, and Z. L. Wang: *Solid State Commun.* **113** (2000) 475.
- 2) H. Chang, I. W. Lenggoro, T. Ogi, and K. Okuyama: *Mater. Lett.* **59** (2005) 1183.
- 3) N. Narendran, Y. Gu, J. P. Freyssinier-Nova, and Y. Zhu: *Phys. Status Solidi A* **202** (2005) R60.
- 4) H. Luo, J. K. Kim, E. F. Schubert, J. Cho, C. Sone, and Y. Park: *Appl. Phys. Lett.* **86** (2005) 243505.
- 5) H. Masui, S. Nakamura, and S. P. DenBaars: *Jpn. J. Appl. Phys.* **45** (2006) L910.
- 6) R. Mueller-Mach, G. O. Mueller, M. R. Krames, and T. Trottier: *IEEE J. Sel. Top. Quantum Electron.* **8** (2002) 339.
- 7) M. Yamada, T. Naitou, K. Izuno, H. Tamaki, Y. Murazaki, M. Kameshima, and T. Mukai: *Jpn. J. Appl. Phys.* **42** (2003) L20.
- 8) R. Mueller-Mach, G. Mueller, M. R. Krames, H. A. Höpfe, F. Stadler, W. Schnick, T. Juestel, and P. Schmidt: *Phys. Status Solidi A* **202** (2005) 1727.
- 9) K. Sakuma, K. Omichi, N. Kimura, M. Ohashi, D. Tanaka, N. Hirotsaki, Y. Yamamoto, R. Xie, and T. Suehiro: *Opt. Lett.* **29** (2004) 2001.
- 10) K. Ishida, I. Mitsuishi, Y. Hattori, and S. Nunoue: *Appl. Phys. Express* **1** (2008) 082201.
- 11) T. Fukui, H. Sakuta, K. Mishihiro, T. Miyachi, K. Kamon, H. Hayashi, N. Nakamura, Y. Uchida, S. Kurai, and T. Taguchi: *J. Light Visual Environ.* **32** (2008) 43.
- 12) T. Taguchi: *Proc. SPIE* **7422** (2009) 74220B.
- 13) T. Fukui, K. Kamon, J. Takeshita, H. Hayashi, T. Miyachi, Y. Uchida, S. Kurai, and T. Taguchi: *Jpn. J. Appl. Phys.* **48** (2009) 112101.
- 14) Y. Won, H. S. Jang, K. W. Cho, Y. S. Song, D. Y. Jeon, and H. K. Kwon: *Opt. Lett.* **34** (2009) 1.
- 15) Y. Fran and T. Tseng: *J. Phys. D* **32** (1999) 513.
- 16) Y. Zhu and N. Narendran: *J. Light Visual Environ.* **32** (2008) 115.
- 17) G. Wyszecki and W. S. Stiles: *Color Science—Concepts and Methods, Quantitative Data and Formulae* (Wiley, New York, 1982) 2nd ed., p. 156.