

A nearly ideal phosphor-converted white light-emitting diode

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A phosphor-converted light-emitting diode was obtained with nearly ideal blue-to-white conversion loss of only 1%. This is achieved using internal reflection to steer phosphor emission away from lossy surfaces, a reflector material with high reflectivity, and a remotely located organic phosphor having (1) unity quantum efficiency (η_q), (2) homogeneous refractive index to minimize scattering, and (3) refractive index-matched to the encapsulation to eliminate total internal reflection. An inorganic composite phosphor is also reported with a nearly homogeneous refractive index to minimize diffuse scattering of emitted light, thereby maximizing the effective phosphor η_q and light extraction. © 2008 American Institute of Physics. [DOI: 10.1063/1.2901378]

In the United States, the goal of solid-state lighting has been defined by the Department of Energy (DOE) and the Optoelectronics Industry Development Association (OIDA). The DOE plans¹ to reproduce the spectrum of sunlight at 50% system efficiency, while the OIDA goal² is to achieve 200 lm/W efficacy with good color rendering by 2020.

The phosphor-converted light-emitting diode (pcLED) configurations shown in Fig. 1 utilize a blue LED chip partially converted by the phosphor to obtain white emission. A conventional pcLED [Fig. 1(a)] has a broadband yellow-emitting yttrium aluminum garnet (YAG):Ce powder phosphor coating on the LED die surface. This configuration is least efficient because the diffuse phosphor directs 60% of total white light emission (consisting of reflected blue and emitted yellow) back toward the chip, where high loss occurs. A scattered photon extraction (SPE) pcLED [Fig. 1(b)] was shown³ to be 61% more efficient than the conventional pcLED because of the separation of the die and extraction of backward-emitted rays from the sides of the optic. Significant losses still occur inside the phosphor layer due to quantum conversion (QC) loss and trapping by total internal reflection (TIR). The pcLED configuration of Fig. 1(c) introduced by Luo *et al.*⁴ uses a remote phosphor, diffuse reflector cup, and hemispherical optic to minimize trapped light. Losses still occur at reflector surfaces as rays tend to be trapped between the phosphor layer and reflector cup. The enhanced light extraction by internal reflection (ELiXIR) pcLED (Ref. 5) [Fig. 1(d)] utilizes a semitransparent rather than diffuse phosphor layer that is separated from the chip by an air gap. Internal reflection at the phosphor/air interface redirects much of the backward phosphor emission away from the die and reflective surfaces *without loss*. The semitransparency of the phosphor layer allows light to pass without deflection and escape the device more easily than diffuse phosphor layers.

Once photons exit the blue LED chip, there are two sources of loss in a pcLED: internal loss in the phosphor during QC of LED light (phosphor quantum efficiency, η_q) and absorption of both LED and phosphor-emitted photons inside the device package (package efficiency, η_p). The $\eta_q\eta_p$ product is the figure of merit for phosphor conversion efficiency (CE). An ideal pcLED will have 100% utilization of photons emitted by the LED chip (i.e., $\eta_q\eta_p=1$). The ulti-

mate OIDA roadmap goal for pcLED CE is $\eta_q\eta_p=0.855$. Conventional white pcLEDs typically have low (<0.40) CE as a consequence of low² η_p (0.40–0.60) and low⁶ YAG:Ce η_q (~ 0.77). In order of decreasing significance, low CE is due to the following factors:⁶ (1) only 40% of white light-emitted/transmitted at the phosphor layer is emitted in the forward direction, while the 60% which is emitted/reflected at the phosphor layer in the backward direction experiences high loss as it must reflect from the LED chip or submount, then traverse the entire diffuse phosphor layer before exiting the device, (2) 0.23 QC loss ($1-\eta_q$) when YAG:Ce converts blue to a balanced white, and (3) 0.13 TIR loss inside the planar phosphor layer. Both the previously white pcLEDs of Narendran *et al.*³ and Luo *et al.*⁴ addressed the primary loss factor by using a remote phosphor. The Narendran device³ resulted in a 61% increase in efficiency and flux. The Luo device⁴ added a hemispherical cap optic and diffuse reflector cup, resulting in $\eta_p\approx 0.91$, an improvement of 82% over the conventional white pcLED. Ray trace simulations by Luo *et al.* showed that the addition of the hemispherical optic reduced the fraction of trapped modes to 0.019. Using a remote phosphor, however, results in increased loss at reflecting surfaces. The remaining 0.07 package loss in the Luo pcLED was probably due to absorption of phosphor-scattered light at the LED and reflectors. Both the Narendran and the Luo pcLEDs used a planar powder phosphor layer, but differed in phosphor identity. The SPE used conventional YAG:Ce powder phosphor while Luo *et al.* used an unspecified broadband yellow phosphor. Therefore, the Narendran pcLED still suffers a 0.23 phosphor QC loss and likely 0.13 TIR absorption loss. The Luo device reduced TIR absorption

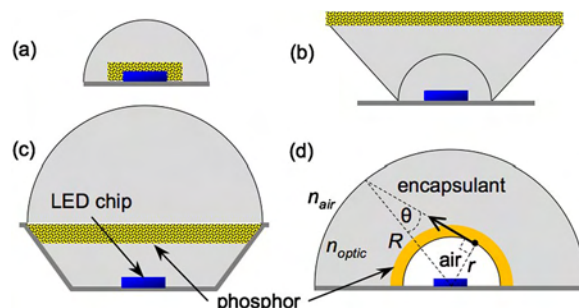


FIG. 1. (Color online) Cross-sectional views of several pcLED packages: (a) conventional phosphor on chip, (b) scattered photon extraction³ remote phosphor, (c) remote phosphor with hemispherical dome,⁴ and (d) ELiXIR (Ref. 5) remote hemispherical shell semitransparent phosphor (this work).

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losses to 0.019 by using a diffuse reflector cup⁷ to scatter whispering gallery modes, but had an unknown QC loss.

When the phosphor is in direct contact with the die, the package loss is dominated by absorption of the 60% of phosphor emission directed backward toward the chip. When the phosphor is separated from the die, loss is dominated by absorption at reflective surfaces and by trapping of light inside the diffuse phosphor. The ELiXIR luminaire addresses these loss mechanisms by reducing phosphor emission back into the die to <0.1%, combining internal reflection to redirect emission heading toward the reflector and the use of a high reflectivity specular reflector⁸ with $R > 98.5\%$, and eliminating trapping of light inside the phosphor.

The intermediate phosphor location, in both the ELiXIR and Luo devices is significant from an efficiency point of view. To minimize the amount of backward emission into the chip, the phosphor should be located as far from the chip as possible, preferably on the exterior surface of the encapsulating optic. However, for maximum transmission of phosphor-converted light at the exterior optic/air interface, the phosphor should be positioned at the center of the sphere, so that all rays will strike at normal incidence and have maximum Fresnel transmission. At minimum, TIR of phosphor-emitted rays at the exterior optic/air interface should be avoided because TIR rays have long path lengths inside the device and consequently encounter increased loss. Consider the phosphor-emitted ray from the point at the right angle vertex of the triangle in Fig. 1(d). The ray is emitted at an angle tangent to the phosphor/air interface. Of all possible ray directions, this ray strikes the exterior optic/air interface at the maximum angle θ with respect to normal incidence. Note also that $\sin \theta = r/R$, where r is the phosphor distance from the device center and R is the exterior optic radius. From Snell's law and using $n_{\text{air}} = 1$, the condition to avoid TIR is $\sin \theta \leq 1/n_{\text{optic}}$ or $r/R < 1/n_{\text{optic}}$. To minimize total optical loss, the phosphor should be located far enough from the exterior optic/air interface to satisfy $r/R < 1/n_{\text{optic}}$, yet far enough from the LED chip to reduce phosphor flux incident on the chip and the associated loss.

The ELiXIR luminaire consists of a blue LED die located at the center of a hemispherical poly(methyl methacrylate) PMMA shell with an interior phosphor coating composed of a fluorescent dye dispersed in a modified PMMA matrix ($n=1.5$). TIR of phosphor emission at the exterior optic/air interface was avoided by placing the phosphor at a position $r/R \sim 0.5$. The general structure and fabrication details have been described previously.⁵

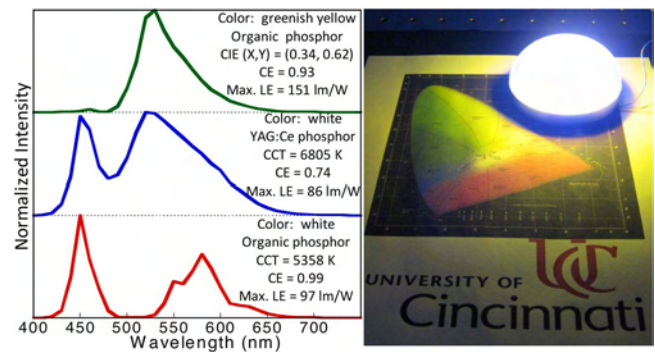


FIG. 2. (Color online) ELiXIR pcLED spectra: green, organic white, YAG:Ce white (left). Photograph of organic white ELiXIR pcLED in operation (right).

The reflector material consisted of 3M Vikuiti enhanced specular reflector film,⁸ a multilayer polymer reflector having reflectivity $>98.5\%$ across the visible spectrum. High efficiency Cree EZR1000 460 nm LED die⁹ were mounted onto an Al base. The spectra of three ELiXIR luminaires are shown in Fig. 2(a). The green luminaire has an organic phosphor layer consisting of Lumogen Yellow¹⁰ fluorescent dye in a modified PMMA host material. Blue LED emission is almost extinguished, resulting in CIE coordinates of (0.34, 0.62). The two white luminaires have different phosphor layers: YAG:Ce in a transparent silicone and Lumogen Orange¹¹ fluorescent dye in a modified PMMA material. The YAG:Ce phosphor, commonly used in commercial white pcLEDs because of its broad yellow emission spectrum, is not optimal because of its relatively low η_q and diffuse scattering characteristics. The organic phosphor is semitransparent and results in little scattering, making maximum efficiency possible, but features a narrower emission spectrum. A photograph showing a CIE color chart illuminated by the white luminaire with organic phosphor is shown in Fig. 2(b).

Devices were tested at drive currents ranging from 1 to 350 mA. At 30 mA, the blue LED chip achieves a wall plug efficiency (η_{LED}) of 0.360 at an output power of 30.3 mW. A maximum flux of 313 mW is achieved at 350 mA, where η_{LED} decreases to 0.255. The green luminaire achieves a maximum luminous efficacy (LE) of 151 lm/W at 30 mA and a maximum flux of 131 lm at 350 mA. The white luminaire with YAG:Ce phosphor achieved 86 lm/W at 30 mA and 75 lm at 61 lm/W at 350 mA. The white luminaire with organic phosphor yielded a LE of 97 lm/W at 30 mA and 84 lm at 69 lm/W at

TABLE I. Comparison of ELiXIR luminaire efficiency and flux values with other pcLEDs at 400 mA dc drive current.

pcLED	η_{pcL}	η_{LED}	η_s	η_q	η_p	LE (lm/W)	flux (lm)
Conventional SrGa_2S_4 green ^a	b	b	~ 0.9	~ 0.9	~ 0.6	37	50
ELiXIR greenish yellow ^c	0.08	0.13	0.83	0.97	0.78	40	56
Conventional YAG:Ce white	~ 0.85	0.77	$\sim 0.5^d$	$\sim 30^e$	$\sim 43^e$
SPE YAG:Ce white ^e	~ 0.85	0.77	~ 0.81	~ 47	~ 63
Luo <i>et al.</i> YAG:Ce white ^f	$\sim 0.91^g$
ELiXIR greenish yellow	0.188	0.241	0.841	0.97	0.96	101	145
ELiXIR YAG:Ce white	0.153	0.241	0.860	0.77	0.96	58	77
ELiXIR organic white	0.204	0.241	0.857	1.00	0.99	65	94

^aReference 13.

^b $\eta_{\text{pcL}}/\eta_{\text{LED}}$ was given as ~ 0.5 .

^cReference 5.

^dReference 2.

^eReference 3.

^fReference 4.

^gRay trace simulation result.

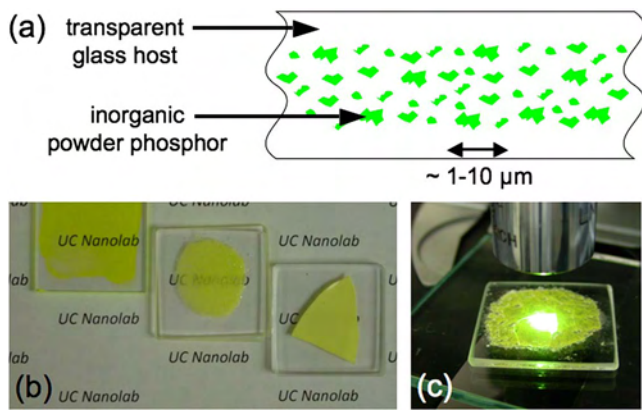


FIG. 3. (Color online) Material composite approach for high performance phosphors: (a) schematic of composite material structure. (b) Photographs comparing transparency (from left to right): yellow organic in modified PMMA, YAG:Ce in SF57 glass, and YAG:Ce in silicone. (c) Photograph of PL from YAG:Ce/SF57 glass composite phosphor pumped at 325 nm.

350 mA, a significant improvement over the YAG:Ce device. Table I contains efficiency values for pcLEDs presented in this work and in the literature. The electrical to optical efficiency of the entire pcLED device η_{pcL} is the product of η_{LED} , η_s , Stokes CE, η_q , and η_p .

Previously reported conventional and ELiXIR green-yellow region pcLEDs have conversion efficiencies (CE = $\eta_q \eta_p$) of 0.54 and 0.76, respectively. The green-yellow ELiXIR pcLED reported here achieved a η_p of 0.96 and CE of 0.93, with the increase mainly due to decreased reflector absorption. The η_p for the conventional white YAG:Ce pcLED was assumed to be 0.50, the midpoint of typical values² to allow estimation of η_p values from other reports. The η_q for YAG:Ce conversion of blue-to-white was reported⁶ as 0.77. This yields CE=0.385 for the conventional white pcLED. The SPE pcLED showed a 61% increase in flux and efficiency over the conventional pcLED with identical phosphor, resulting in an improved $\eta_p=0.81$ (1.61×0.50). The ELiXIR pcLED with YAG:Ce phosphor achieved CE=0.74, which yields a $\eta_p=0.96$ if η_q is assumed to be 0.77. This calculated η_p is equal to that of the green ELiXIR with a semitransparent phosphor. Finally, the white ELiXIR pcLED with an organic orange-emitting phosphor achieved a η_p of 0.99. Combining η_p with $\eta_q=1.00$ for this phosphor, the device CE is a nearly ideal 0.99, the highest value reported for a pcLED. The η_p exceeds that of the green-yellow device because the phosphor layer is optically thinner, so a smaller fraction of total photons is emitted back toward the lossy reflector.

The ELiXIR pcLED demonstrated here has achieved nearly ideal blue-to-white CE using a semitransparent organic phosphor and index-matched encapsulating optic. Organic materials, however, lack sufficient photostability for a practical solid-state lighting device emitting watts of optical power for thousands of hours. We have investigated an inorganic composite phosphor using an inorganic powder phosphor suspended in a transparent host with a similar refractive index. This phosphor satisfies the maximum efficiency requirements of low scattering and existence of a transparent optic material with $n_{\text{optic}} \geq n_{\text{phosphor}}$. The structure of the reduced-scatter phosphor composite is shown in Fig. 3. A demonstration of this phosphor composite material incorpo-

rated YAG:Ce phosphor in a Schott SF57 glass matrix. A side-by-side comparison with YAG:Ce in a transparent silicone matrix [Fig. 3(b)] illustrates the decreased optical scatter the inorganic composite material. In Fig. 3(c), strong photoluminescence (PL) is observed from a YAG:Ce/SF57 composite sample pumped at 325 nm.

Conventional powder phosphors in pcLEDs are typically encapsulated in a matrix of epoxy or silicone. Glass encapsulated phosphors have also been reported,¹² showing an increase in stability over epoxy encapsulation, though still having similar scattering properties due to a large index difference. The encapsulation materials have a maximum refractive index of ~ 1.6 while still maintaining good transparency. The YAG:Ce phosphor, however, has a refractive index of 1.85 in the visible region. The large difference in refractive indices combined with small particle size and weak absorption results in diffuse scattering of incident and emitted light. This phosphor scatter reduces efficiency due to (1) increased path length for light inside the phosphor, leading to reabsorption losses and decreasing the effective η_Q of the phosphor and (2) randomizing of light directionality passing through the phosphor, leading to longer path lengths and increased contact with high loss areas such as reflectors, phosphor layer, and LED chip. Diffuse reflector surfaces have been used to increase efficiency by scattering modes trapped by TIR. When seeking near perfect extraction efficiency, however, diffuse surfaces are best avoided because of additional loss due to ray path randomization.

In summary, the combination of a semitransparent, high efficiency phosphor and a device structure engineered to minimize internal loss has enabled blue-to-white conversion with nearly ideal efficiency. The YAG:Ce composite material is a practical implementation of these principles for increased efficiency solid-state lighting. Luminaires incorporating phosphor composite materials can exceed the OIDA efficiency targets for the year 2020.

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¹Navigant Consulting Inc. for the U. S. Department of Energy, 2006, see <http://www.netl.doe.gov/ssl/PDFs/SSLMultiYearPlan.pdf> for full text.

²J. Y. Tsao, Ed., 2002, see <http://lighting.sandia.gov/lightingdocs/> for full text.

³N. Narendran, Y. Gu, J. P. Freyssonier-Nova, and Y. Zhu, *Phys. Status Solidi A* **202**, R60 (2005).

⁴H. Luo, J. K. Kim, E. F. Schubert, J. Cho, C. Sone, and Y. Park, *Appl. Phys. Lett.* **86**, 243505 (2005).

⁵S. C. Allen and A. J. Steckl, *J. Disp. Technol.* **3**, 155 (2007).

⁶Y. Zhu, N. Narendran, and Y. Gu, *Proc. SPIE* **6337**, 63370S (2006).

⁷H. Luo, J. K. Kim, Y. A. Xi, E. F. Schubert, J. Cho, C. Sone, and Y. Park, *Appl. Phys. Lett.* **89**, 041125 (2006).

⁸Vikuiti Enhanced Specular Reflector (ESR) 3M Corp., 2003, see http://solutions.3m.com/wps/portal/3M/en_US/Vikuiti1/ for data sheet.

⁹EZ1000 LEDs, Cree Inc., 2006, see <http://www.cree.com/products/pdf/CPR3CR.pdf> for data sheet.

¹⁰Lumogen F Yellow 083, BASF Corp, 2005, see <http://www.basf.com/additives.pdfs/lumye083.pdf> for data sheet.

¹¹Lumogen F Orange 240, BASF Corp, 2005, see <http://www.basf.com/additives.pdfs/lumorg240.pdf> for data sheet.

¹²S. Fujita, S. Yoshihara, A. Sakamoto, S. Yamamoto, and S. Tanabe, *Proc. SPIE* **5941** 594111 (2005).

¹³R. Mueller-Mach, G. O. Mueller, T. Trottier, M. R. Krames, A. Kim, and D. Steigerwald, *Proc. SPIE* **4776**, 131 (2002).