The realization of a whole palette of colors in a green gap by monochromatic phosphor-converted light-emitting diodes

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Abstract: A variety of efficient green, yellow, and amber monochromatic phosphor-converted light-emitting diodes (pc-LEDs) were fabricated by simply capping a long-wave pass filter (LWPF) on top of LED packing associated with each corresponding powder phosphor. In this paper, the luminous efficacy and color purity of two green, three yellow, and two amber pc-LEDs were reviewed by comparing the optical properties and current/temperature stability of each LWPF-capped pc-LED. The simple combination of LWPFs and phosphor materials in the pc-LEDs provide a simple means of addressing the low luminous efficacy problem of III-V monochromatic semiconductor LEDs in the various colors of the wavelength range between green and amber (known as the "green gap"). This technique also represents a simple approach to mitigate the sub-linearity problem of the efficacy versus the driving current occurring at a relatively low current in III-V green LEDs (known as "green droop") to the level of a blue LED. This nano-multilayered filter-capped pc-LED can open further research into developing new color-converting materials (such as powder phosphors, and/or quantum dots) to extend the color palette in the wavelength region of the "green gap" and to improve the efficacy and color purity of color pc-LEDs.

OCIS codes: (220.0220) Optical design and fabrication; (230.0230) Optical devices; (230.1480) Bragg reflectors; (230.3670) Light-emitting diodes.

References and links

1. J. M. Phillips, M. E. Coltrin, M. H. Crawford, A. J. Fischer, M. R. Krames, R. Mueller-Mach, G. O. Mueller, Y. Ohno, L. E. S. Rohwer, J. A. Simmons, and J. Y. Tsao, "Research challenges to ultra-efficient inorganic solid-state lighting," Laser Photon. Rev. **1**(4), 307–333 (2007).
2. MM. R. Krames, O. B. Shchekin, R. Mueller-Mach, G. O. Mueller, L. Zhou, G. Harbers, and M. G. Craford, "Status and Future of High-Power Light-Emitting Diodes for Solid-State Lighting," J. Disp. Technol. **3**(2), 160–175 (2007).
3. W. Schnick, "Shine a light with nitrides," Phys. Status Solidi RRL **3**(7-8), A113–A114 (2009).
4. D. A. Steigerwald, J. C. Bhat, D. Collins, R. M. Fletcher, M. O. Holcomb, M. J. Ludowise, P. S. Martin, and S. L. Rudaz, "Illumination With Solid State Lighting Technology," IEEE J. Sel. Top. Quantum Electron. **8**(2), 310–320 (2002).
5. N. Kimura, K. Sakuma, S. Hirafune, K. Asano, N. Hirosaki, and R.-J. Xie, "Extra high color rendering white light-emitting diode lamps using oxynitride and nitride phosphors excited by blue light-emitting diode," Appl. Phys. Lett. **90**(5), 051109 (2007).
6. K. Fujiwara, H. Jimi, and K. Kaneda, "Temperature-dependent droop of electroluminescence efficiency in blue (In,Ga)N quantum-well diodes," Phys. Status Solidi C **6**(S2), S814–S817 (2009).
7. M. Funato, M. Ueda, Y. Kawakami, Y. Narukawa, T. Kosugi, M. Takahashi, and T. Mukai, "Blue, Green, and Amber InGaN/GaN Light-Emitting Diodes on Semipolar {1122} GaN Bulk Substrates," Jpn. J. Appl. Phys. **45**(26), L659–L662 (2006).

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8. R. Mueller-Mach, G. O. Mueller, M. R. Krames, and T. Trottier, "High-Power Phosphor-Converted Light-Emitting Diodes Based on III-Nitrides," IEEE J. Sel. Top. Quantum Electron. **8**(2), 339–345 (2002).
9. R. Mueller-Mach, G. O. Mueller, T. A. Trottier, M. R. Krames, A. Kim, and D. A. Steigerwald, "Green phosphor-converted LED," Proc. SPIE **4776**, 131–136 (2002).
10. R. Mueller-Mach, G. O. Mueller, M. R. Krames, O. B. Shchekin, P. J. Schmidt, H. Bechtel, C.-H. Chen, and O. Steigelmann, "All-nitride monochromatic amber-emitting phosphor-converted light-emitting diodes," Phys. Status Solidi RRL. **3**(7-8), 215–217 (2009).
11. J. R. Oh, S.-H. Cho, H. K. Park, J. H. Oh, Y.-H. Lee, and Y. Rag, "Full down-conversion of amber-emitting phosphor-converted light-emitting diodes with powder phosphors and a long-wave pass filter," Opt. Express **18**(11), 11063–11072 (2010).
12. J. S. Kim, P. E. Jeon, J. C. Choi, and H. L. Park, "Emission color variation of $M_2SiO_4:Eu^{2+}$ (M = Ba, Sr, Ca) phosphors for light-emitting diode," Solid State Commun. **133**(3), 187–190 (2005).
13. Y. R. Do, K.-Y. Ko, S.-H. Na, and Y.-D. Huh, "Luminescence Properties of Potential $Sr_{1-x}Ca_xGa_2S_4$:Eu Green- and Greenish-Yellow-Emitting Phosphors for White LED," J. Electrochem. Soc. **153**(7), H142–H146 (2006).
14. Y. Q. Li, A. C. A. Delsing, G. de With, and H. T. Hintzen, "Luminescence Properties of Eu^{2+}-Activated Alkaline-Earth Silicon-Oxynitride $MSi_2O_{2-\delta}N_{2+2/3\delta}$ (M = Ca, Sr, Ba): A Promising Class of Novel LED Conversion Phosphors," Chem. Mater. **17**(12), 3242–3248 (2005).
15. J. L. Wu, G. Gundiah, and A. K. Cheetham, "Structure–property correlations in Ce-doped garnet phosphors for use in solid state lighting," Chem. Phys. Lett. **441**(4-6), 250–254 (2007).
16. J. K. Park, C. H. Kim, S. H. Park, H. D. Park, and S. Y. Choi, "Application of strontium silicate yellow phosphor for white light-emitting diodes," Appl. Phys. Lett. **84**(10), 1647–1649 (2004).
17. H. S. Jang, H. Yang, S. W. Kim, J. Y. Han, S.-G. Lee, and D. Y. Jeon, "White Light-Emitting Diodes with Excellent Color Rendering Based on Organically Capped CdSe Quantum Dots and $Sr_3SiO_5:Ce^{3+},Li^+$ Phosphors," Adv. Mater. (Deerfield Beach Fla.) **20**(14), 2696–2702 (2008).
18. L. Li, T. J. Daou, I. Texier, T. T. Kim Chi, N. Q. Liem, and P. Reiss, "Highly Luminescent $CuInS_2$/ZnS Core/Shell Nanocrystals: Cadmium-Free Quantum Dots for In Vivo Imaging," Chem. Mater. **21**(12), 2422–2429 (2009).
19. M. Peter, A. Laubsch, W. Bergbauer, T. Meyer, M. Sabathil, J. Baur, and B. Hahn, "New developments in green LEDs," Phys. Status Solidi A **206**(6), 1125–1129 (2009).
20. A. Laubsch, M. Sabathil, W. Bergbauer, M. Strassburg, H. Lugauer, M. Peter, S. Lutgen, N. Linder, K. Streubel, J. Hader, J. V. Moloney, B. Pasenow, and S. W. Koch, "On the origin of IQE-'droop' in InGaN LEDs," Phys. Status Solidi C **6**(S2), S913–S916 (2009).
21. J. R. Oh, S.-H. Cho, Y.-H. Lee, and Y. R. Do, "Enhanced forward efficiency of $Y_3Al_5O_{12}:Ce^{3+}$ phosphor from white light-emitting diodes using blue-pass yellow-reflection filter," Opt. Express **17**(9), 7450–7457 (2009).
22. J. R. Oh, S.-H. Cho, Y.-H. Lee, and Y. R. Do, "Lowering Color Temperature of $Y_3Al_5O_{12}:Ce^{3+}$ White Light Emitting Diodes Using Reddish Light-Recycling Filter," Electrochem. Solid-State Lett. **13**(1), J5–J7 (2010).
23. A. F. Turner, and P. W. Baumeister, "Multilayer mirrors with high reflectance over an extended spectral region," Appl. Opt. **5**(1), 69–76 (1966).
24. L. Wang, P.-F. Gu, and S.-Z. Jin, "Enhancement of flip-chip white light-emitting diodes with a one-dimensional photonic crystal," Opt. Lett. **34**(3), 301–303 (2009).
25. A. Žukauskas, R. Vaicekauskas, F. Ivanauskas, H. Vaitkevičius, and M. S. Shur, "Spectral optimization of phosphor-conversion light-emitting diodes for ultimate color rendering," Appl. Phys. Lett. **93**(5), 051115 (2008).
26. R.-J. Xie, N. Hirosaki, M. Mitomo, K. Sakuma, and N. Kimura, "Wavelength-tunable and thermally stable Li-α-sialon:Eu^{2+} oxynitride phosphors for white light-emitting diodes," Appl. Phys. Lett. **89**(24), 241103 (2006).
27. C.-C. Chung, and J.-H. Jean, "Synthesis of Ca-α-SiAlON:Eu_x phosphor powder by carbothermal-reduction-nitridation process," Mater. Chem. Phys. **123**(1), 13–15 (2010).
28. S. C. Allen, and A. J. Steckl, "ELiXIR—Solid-State Luminaire With Enhanced Light Extraction by Internal Reflection," J. Disp. Technol. **3**(2), 155–159 (2007).
29. S. C. Allen, and A. J. Steckl, "A nearly ideal phosphor-converted white light-emitting diode," Appl. Phys. Lett. **92**(14), 143309 (2008).
30. N. Narendran, Y. Gu, J. P. Freyssinier-Nova, and Y. Zhu, "Extracting phosphor-scattered photons to improve white LED efficiency," Phys. Status Solidi A **202**(6), R60–R62 (2005).

1. Introduction

The beautiful mono-color generation of phosphor-converted light-emitting diodes (pc-LEDs) is relevant when it is realized that the entire palette of colors of phosphors is available in the Commission Internationale d'Eclairage (CIE) chromaticity diagram. Of course, the full spectrum of colors is possible from III-V LEDs and related compounds without phosphors. However, there are a number of problems associated with several of the monochromatic LEDs from this source: First, it has proven to be difficult to achieve reasonable efficiency for emission in the deep green to amber region of spectrum between 510 – 610 nm. This is well known as the "green gap" or "yellow gap" problem, and it impedes the use of efficient deep green, yellow, or amber color from a direct emissive LED [1–3]. Second, the poor working lifetime of some LEDs with green or amber emission wavelengths leads to different aging

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characteristics of different colors while white light or mixed color is created through a combination of the light of several colored LEDs [4,5]. Third, there is a large temperature or current dependence of the emission wavelength of some deep green or amber LEDs [6,7]. As is well known, these problems can be easily addressed through the use of pc-LEDs by combining a blue InGaN LED and blue excited phosphors [8,9]. InGaN-based blue LEDs showed greater external quantum efficiency and better thermal or current dependence compared to both InGaN-based deep green and AlGaInP-based amber LED. Phosphors also show high quantum efficiency, even at elevated temperature on the surface of LEDs, as well as high chemical and thermal stability. Even if the blue InGaN LED emission undergoes a wavelength shift with the current or temperature, the wide excitation band of various LED phosphors can span the change. Hence, the emission band of LED phosphors is not wavelength-sensitive or at least much less so than that of the LED.

Very recently, two types of 'full' down-conversion approaches were suggested by Muller-Mach et al. [10] and by the authors [11] in an effort to address problems with low LED performance and temperature and current dependence at wavelengths in the amber region. Muller-Mach et al. introduced an innovative idea to close the "green gap" problem using a densely sintered translucent ceramic of $(Ba,Sr)_2Si_5N_8$:Eu amber phosphor. They used new morphology of sintered ceramic phosphors to block the transmitted blue LED light passing through phosphor layers and reduce the scattering loss of powders. However, there are only a few available sintered ceramic phosphors that can be excited efficiently by blue LEDs in the present. Meanwhile, we suggested amber light pc-LEDs using Eu-doped silicate powder phosphors in association with a blue-mirror-yellow-pass filter (long-wave pass filter, LWPF). A modified quarter-wave type of LWPF consisting of nano-multilayered film was simply introduced on top of an amber phosphor-coated InGaN-based LED die to block and recycle unabsorbed transmitted blue emission to create a highly efficient amber monochromatic LED [11]. This concept can be easily enlarged to create a variety of monochromatic color LEDs because there are a large number of powder phosphors or quantum dots in the present market or in research labs use well-established technology.

In a conventional approach for obtaining full down-conversion color using blue-excited pc-LEDs coated with powder-based phosphors, the phosphor layer should be thick and highly concentrated to block the unabsorbed blue emission from the pc-LED [11]. This approach is hampered by low phosphor conversion efficiency due to the additional scattering associated with the high concentration of the phosphor content in the paste. As previously reported, the simple capping of a pc-LED by a LWPF enhances the emission output and color purity from the pc-LED due to the recycling and blocking of blue light. In addition, the low concentration phosphor of optimum LWPF-assisted pc-LED results in reducing scattering loss from the phosphor layer. Therefore, an efficient LWPF-assisted monochromatic pc-LED displays a color point which is dependent on the material type of the phosphors even with a low concentration of the phosphors in paste. Given the present state of technology and the current publications, there are many possible phosphor or quantum dot candidates that cover the colors in the wavelength range between green and amber [12–18]. Among them, several potential green/yellow/amber phosphors were selected and tested as to whether they could be applied into various monochromatic pc-LEDs capped with LWPFs.

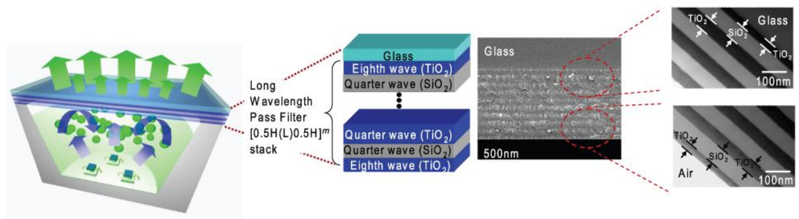

Fig. 1. Schematic diagram of the phosphor-coated LED device structure with the embedded color (green, yellow or amber) light passing through and blue light reflected from the dielectric multilayer coated glass substrate. The enlarged side view shows the basic sequence of the modified quarter-wave stacks ($[0.5H(L)0.5H]^9$ (L: low index layer, SiO_2, H: high index layer, TiO_2)) of the LWPF and SEM/TEM images.

As shown in the schematic diagram of the LWPF-capped pc-LEDs in Fig. 1, various colored pc-LEDs were fabricated by dropping the same amounts of each phosphor pastes at an optimum concentration onto a cup-type blue LED. To display the realization of monochromatic pc-LEDs, the luminous efficacy, CIE color coordinates and color purities of various monochromatic pc-LEDs were reviewed using various powder phosphors associated with LWPFs. Furthermore, the current and temperature-dependence of the luminous efficacy of green/yellow/amber pc-LEDs capped with LWPFs were compared. In so doing, the different sub-linearity problem of the efficacy versus the driving current between III-V blue, green, yellow and amber LEDs was addressed (known as the "different current droop") [19,20]. Finally, further research into attaining the requirements of powder-based phosphors that can be utilized with high-quality monochromatic pc-LEDs was discussed. This straightforward concept of monochromatic pc-LEDs can open research and development areas related to the production of new color-converting materials to be used in green/yellow/amber phosphors and new types of omnidirectional reflective (ODR) filters or 1D/3D photonic crystal filters which function as blue-mirror-yellow-pass filters.

2. Experimental methods

Two types (L1 and L2) of dielectric LWPFs were fabricated on glass substrates. For the fabrication of the LWPF stacks, terminal eighth-wave thick TiO_2 (L1: 25 nm, L2: 26 nm) and quarter-wave thick SiO_2/TiO_2 (L1: 73/50 nm, L2: 73/52 nm) nano-multilayered films were coated onto a glass substrate by e-beam evaporation at 250°C. The base pressure in the e-beam chamber was fixed at 4.0×10^{-5} torr. The deposition was performed at an acceleration voltage of 7 kV with an oxygen partial pressure of 1.9×10^{-4} torr. The refractive indices (n) and extinction coefficients (k) of the e-beam evaporated SiO_2 and TiO_2 films were measured using a spectroscopic ellipsometer (Sentech, SE800). The detailed wavelength dispersion characteristics of the n and k values of the as-grown SiO_2 and TiO_2 films were reported previously by the authors [21,22]. These measured n and k values were used to simulate the reflectance (R), transmittance (T), and absorption (A) in the design of the various types of LWPFs. For the design of the LWPF multilayer films for the blue-excited pc-LEDs, the characteristic matrix method was used to simulate the reflectance (R), transmittance (T), and absorption (A) of the optical structure of LRF stacks [23,24]. In the simulation, the thicknesses of the high-index (TiO_2) and low-index (SiO_2) films were varied to tune the spectral position of the reflectance band. In this publication, two different types of LWPFs (L1 = 510 and L2 = 530 nm at the band-edge of the long-wavelength) were fabricated as capping filters to analyze the effect of LWPF films on the forward emission of pc-LEDs with various green, yellow and amber phosphors. The basic structure and SEM/TEM image of modified quarter-wave stacks of nano-multilayered film ($[0.5TiO_2/SiO_2/0.5TiO_2]^m$ with m = 9, eighth-wave high-index TiO_2 (0.5H), and quarter-wave low-index SiO_2 (L) and high-index TiO_2 (H)) were used for the LWPF in this study. As shown in Fig. 1. TEM images clearly indicated that this sequence simply entails the addition of a pair of eighth-wave layers of high-index TiO_2 layers to the

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quarter wave stack, one at each end. The TEM images also confirm that the slight difference of the long-wavelength edge of the reflectance band was tuned by controlling the thicknesses of the high- and low-index multilayer.

To fabricate the pc-LEDs, a blue chip (λ_{max} = 445 nm) was used simultaneously as a blue light source and an excitation source for the various color phosphors of pc-LEDs. Blue, green and amber monochromatic LED chips were purchased from Alti-semiconductor Co. Ltd. Two green, three yellow, and two amber powder phosphors were also used in this experiment. $SrGa_2S_4$:Eu (G2) green and $(Sr,Ba,Ca)_3SiO_5$:Eu (A1) amber phosphors were synthesized in our lab through a solid-state reaction. The synthetic procedures followed the procedures detailed in a previous publication by the authors [13] and by Park et al [16]. The other powder phosphors were obtained from several phosphor companies. Those commercialized phosphors were not optimized phosphors for the LWPF-capped monochromatic pc-LEDs investigated here, as the seven powder phosphors used in this experiment were optimized for application into white pc-LEDs. Optimum amounts of each color phosphor were dispersed in a silicone binder, and the same amounts of the resulting phosphor pastes were dropped onto a cup-type blue LED to create the different color pc-LEDs. On top of the various color pc-LEDs, a LWPF-coated glass substrate was attached with an air gap.

The forward emissions of the emission spectra from seven different powder phosphors, III-V blue, green, amber LEDs, blue-excited pc-LEDs and blue-excited LWPF-capped monochromatic pc-LEDs were measured in a normal direction or an integrated sphere using a spectrophotometer (PSI Co. Ltd., Darsar). The luminous efficacy and quantum efficiency were defined as the brightness and the integrated emission spectra of both the phosphor-coated conventional and LWPF-assisted pc-LEDs, respectively, at a constant current or power. The external efficiency and color purity of the various LWPF-coated color pc-LEDs were compared with the current at optimum phosphor concentrations. The transmission and diffusive reflectance spectra of the LWPFs were measured using a UV/Visible/Near IR spectrophotometer (Varian, Carry 5000). The transmission spectra were made at normal incidence to the surface of LWPFs on glass substrates and the diffusive reflectance spectra were measured at normal incidence with an integrating sphere. The thicknesses and cross-sectional images of the LWPFs on glass substrates were determined by field emission-type scanning electron microscopy (FE-SEM) (JSM 7401F, JEOL) operated at 10 kV and by transmission electron microscopy (TEM) (JEM2100F, JEOL).

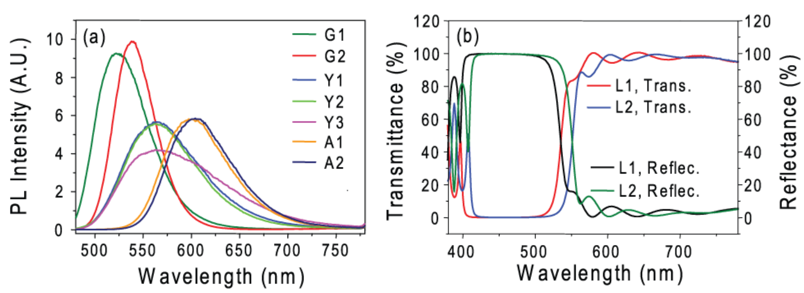

Fig. 2. (a) The measured photoluminescent (PL) spectra of seven different green, yellow, and amber powder phosphors. (b) Measured transmittance and reflectance spectra of the two different LWPFs (L1 and L2) on glass substrates.

3. Results and discussion

Figure 2(a) shows the photoluminescence (PL) spectra excited by blue light and Table 1 summarizes the optical properties of various phosphor candidates for application into monochromatic pc-LEDs. Although they are not the best phosphor candidates for this purpose, they are well known for their application to white pc-LEDs. Most widely used phosphors in white pc-LEDs for lighting applications were developed to have a broad spectrum (Fig. 2(a)) to enhance the color rendering index (CRI) [25]. The narrow spectrum of

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phosphors is more apt to reach a high level of color purity for application into monochromatic pc-LEDs. Nonetheless, various monochromatic pc-LEDs were fabricated using two green, three yellow and two amber phosphors of the type available in market or that could be synthesized in a lab to compare their optical properties when applied to monochromatic pc-LED with a LWPF. As previously reported in the optimization process of one amber powder phosphor (A1 phosphor in this experiment) for application into an amber pc-LED [11], different optimum concentrations of each phosphor were determined, as shown in Table 1.

Table 1. The optical properties of various phosphors

	Phosphors	CIE color coordinates		Relative quantum efficiency (%)[a]	Relative brightness (%)[a]	Color Purity	Peak wavelength (nm)	Optimum phosphor concentration in paste (%)
		x	y					
G1	$(Sr,Ba)_2SiO_4$:Eu	0.26	0.66	100	100	79.8	521	40
G2	$(Sr,Ca)Ga_2S_4$:Eu	0.30	0.67	83.6	97.7	85.4	537	10
Y1	$(Sr,Ba)_2SiO_4$:Eu	0.45	0.54	88.4	84.6	94.1	563	40
Y2	$(Sr,Ca)Si_2O_2N_2$:Eu	0.45	0.54	82.9	81.4	97.4	561	30
Y3	$(Y,Gd)_3(Al,Ga)_5O_{12}$:Ce	0.47	0.52	84.8	70.0	97.7	562	10
A1	$(Sr,Ba,Ca)_3SiO_5$:Eu	0.58	0.42	79.4	55.0	99.6	597	20
A2	$(Sr,Ba,Ca)_3SiO_5$:Eu	0.59	0.40	78.9	50.5	98.8	603	20

[a]The relative quantum efficiency and relative brightness were compared with G1 phosphor.

The reflectance spectra of the two LWPFs showed that the emission band of the blue InGaN LED at a shorter wavelength can be overlapped with the reflection bands of the LWPFs at a shorter wavelength (Fig. 2 (b)). The transmittance of the various blue-excited phosphors at the wavelength region between green and amber continuously exceeded 90%. In this publication, two different types of LWPFs (L1 = 510 and L2 = 530 nm at the band-edge of the long-wavelength) were fabricated as capping filters to be applied into various colored monochromatic pc-LEDs with the appropriate combination of each LWPF and corresponding phosphor. The long-wavelength edges of the high-reflectance band of the selected LWPFs for each color are displayed in Table 2.

Table 2. The optical properties of various LWPF-capped pc-LEDs

	Band edge of LWPF	CIE color coordinates		Relative external quantum efficiency (%)[a]	Relative luminous efficacy (%)[a]	Luminous efficacy (lm/w)[b]	Color purity	Peak wavelength (nm)
		x	y					
	Blue LED	0.16	0.02	100	100	5.9	99.2	445
G1	L1 (510nm)	0.27	0.64	71.6	1260	74.7	75.6	530
G2	L1 (510nm)	0.30	0.66	66.6	1430	82.7	91.0	538
Y1	L2 (530nm)	0.46	0.53	71.8	1190	70.9	96.7	564
Y2	L2 (530nm)	0.46	0.53	60.5	1010	61.2	96.2	564
Y3	L2 (530nm)	0.48	0.51	65.6	951	57.8	97.8	568
A1	L2 (530nm)	0.57	0.43	66.6	886	52.7	97.7	596
A2	L2 (530nm)	0.58	0.41	63.1	816	48.5	97.5	602

[a]The relative external quantum efficiency and relative luminous efficacy were compared with Blue LED.
[b]The luminous efficacy was measured at equal power (at 100mA).

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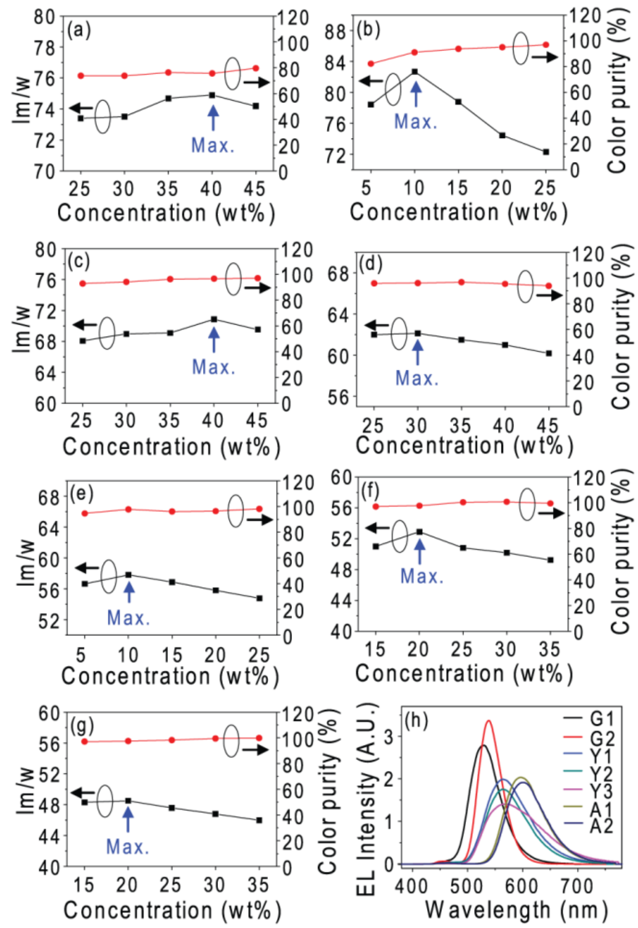

Fig. 3. The optimum concentration of each LWPF-capped pc-LEDs (a) G1, (b) G2, (C) Y1, (d) Y2, (e) Y3, (f) A1, (g) A2, (h) The measured EL spectra of a pc-LED with seven powder phosphors excited by a blue LED (with optimum phosphor concentrations for each color) capped with a LWPF.

As shown in Fig. 3(a)–3(g), the concentration of the phosphors in each LWPF-capped pc-LED was carefully selected to guarantee the maximum level of luminous efficacy and color purity similar to that of the corresponding phosphor. The electroluminescence (EL) spectra of seven different colors of LWPF-capped pc-LEDs are also shown in Fig. 3(h). The peak maximum EL wavelengths from various LWPF-capped pc-LEDs nearly coincide with the PL wavelengths from seven powder phosphors under excitation at 450 nm (Fig. 2(a)). There were very few peaks observed in the blue region of each spectrum of all LWPF-capped pc-LEDs in this study due to the blocking and recycling of the pumping blue light by a capped LWPF. A variety of pure colors between green and amber can be obtained by varying the type of phosphor material in the LWPF-capped pc-LEDs.

To investigate the suitability of fabricating monochromatic pc-LEDs from corresponding phosphors, the optical properties of full down-converted LWPF-capped pc-LEDs were also summarized, as shown in Table 2. Except for bluish green color (G1), the peak wavelength, color coordinates and color purities of other monochromatic pc-LEDs are close to those of the corresponding phosphors summarized in Table 1. The color deviation of the G1 green pc-LED from Eu-doped orthosilicate-based green phosphor results from the partial filtering of the transmitted blue passing through the phosphor layer due to the closeness between the blue emission peak of the InGaN LED and the green emission of the orthosilicate-based phosphors.

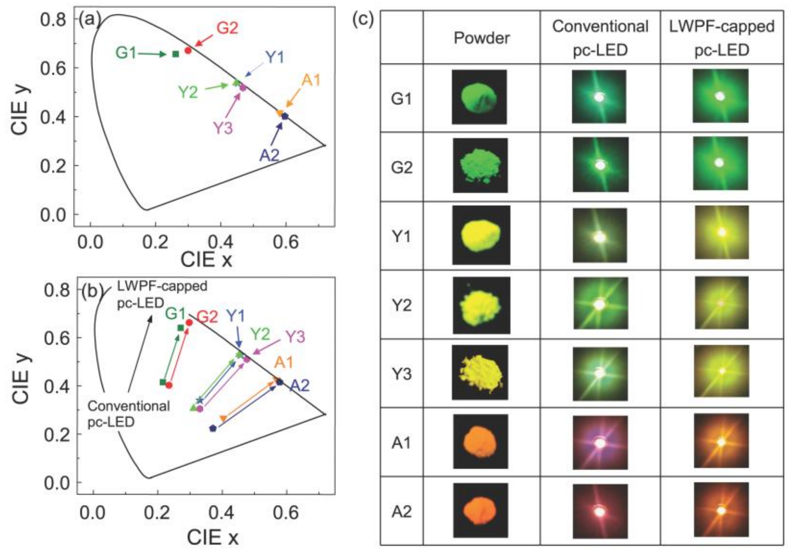

Fig. 4. (a) 1931 CIE color coordinates of two green, three yellow, and two amber powder phosphors. (b) 1931 CIE color coordinates of seven pc-LEDs without a LWPF and seven pc-LEDs with LWPFs. (c) Images of the seven powder phosphors (left), seven pc-LEDs without a LWPF (middle) and those with a LWPF (right). All measurements were performed under an equal amount of current (100 mA).

Furthermore, Figs. 4(a) and 4(b) compare the CIE color coordinates of the PL emissions of phosphors and the EL emissions of the pc-LEDs with or without a LWPF in a chromaticity diagram. Here, the variations between the phosphor powders and the pc-LEDs with a LWPF are very small in comparison to those between the powders and the pc-LEDs without a LWPF. The similarities in the CIE color coordinates indicate that the LWPF-capped pc-LEDs can reproduce any colors from different phosphors in the green and amber region in the chromaticity diagram. The emitting images and color purities of the various fabricated pc-LEDs with and without a LWPF and each phosphor powder under UV excitation are also compared in Fig. 4(c) and in Tables 1 and 2. The introduction of a LWPF converts the mixed whitish colors of all types of pc-LEDs to monochromatic colors at an optimum concentration of phosphors. These images and color purities also confirm that various monochromatic colors of LWPF-capped pc-LEDs are well matched with those of the corresponding powder phosphors. In the green regions, the relatively low color purity of the phosphors creates the low color purity of the color LWPF-capped pc-LEDs. For further advancement of the color purity of green pc-LEDs, it is necessary to develop color-conversion materials with a narrow bandwidth of the emission spectrum. From a scientific perspective, semiconductor quantum dots having a narrow bandwidth may be good candidates for color-converting materials in monochromatic LWPF-capped pc-LEDs to obtain high purity green color [17,18].

As previously reported, the highest efficacy can be attained in a LWPF-capped pc-LED with an optimum concentration of each phosphor owing to the reduced scattering loss of powder phosphors at a relatively low powder concentration and due to the recycling of the reflected blue light by the LWPF [11]. Moreover, the superior performance of the monochromatic the LWPF-capped pc-LED was reported to relative to that of a direct emitting pc-LED with a highly concentrated phosphor. Figure 5(a) compares the efficacy stability of different colors LWPF-capped pc-LED with the temperature. The silicate-based green (G1) and yellow (Y1) pc-LEDs show a bad temperature dependence of color and luminous efficacy. Among them, the thiogallate-based green (G2) LWPF-capped pc-LED showed moderate dependence and the silicate-based amber (A1, A2) and oxynitride-based yellow (Y2) LWPF-capped pc-LEDs show a good dependence. The variation of the temperature stability among the different colored pc-LEDs is due to the different temperature stability

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values of each corresponding phosphor and the phosphor concentration. As previously reported [26,27], the different phosphor materials show different thermal quenching phenomena (See the Fig. 5(b)) and the highly concentrated phosphor paste is more rapidly quenched due to the increased ambient temperature compared to the less concentrated paste (as shown in the inset of Fig. 5(a)). Therefore, the different and complicated stability trend of monochromatic LWPF-capped pc-LEDs depends mainly on both the material type of the phosphor in the pc-LED and the phosphor concentration the in paste.

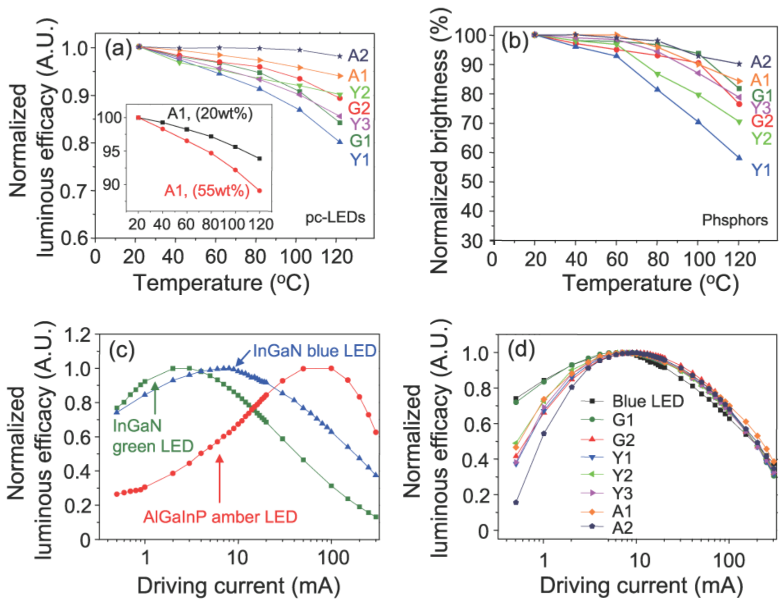

Fig. 5. (a) Comparison of the luminous efficacy of the seven different pc-LEDs with a LWPF (with the optimum concentration of phosphor for each color) as a function of the ambient temperature under an equal amount of current (100 mA). The inset shows the luminous efficacy of two amber LWPF-capped pc-LEDs at concentrations of 20 wt % and 55 wt% as a function of the ambient temperature. (b) Comparison of the brightness of the seven different colored corresponding powder phosphors as a function of the ambient temperature. (c) The measured and normalized luminous efficacy of blue/green/amber III-V semiconductor LEDs as a function of current density. (d) The measured and normalized luminous efficacy of the seven different pc-LEDs with a LWPF (with the optimum concentration of phosphor for each color) as a function of current density.

Figures 5(c) and 5(d) show the normalized dependence of the efficacy vs. the current of blue/green/amber III-V semiconductor LEDs and LWPF-capped pc-LEDs. The figures clearly show that the different current stability among blue/green/amber III-V semiconductor LEDs results from the different compositional origin of the InGaN-based green and blue and AlGaInP-based amber LEDs. Otherwise, Fig. 5(d) shows that the current stability values of LWPF-capped pc-LEDs are not better than those of InGaN-based pump LEDs; however, they continue to display a trend similar to that of the blue pump LED. This indicates that LWPF-capped pc-LEDs may have nearly similar current stability compared to the wide variation of current stability among blue/green/amber monochromatic III-V LEDs that do not contain phosphors. In particular, it is a well-known phenomena that sub-linearity of the efficacy versus the driving current occurs at a lower level for a green InGaN LED (530 nm; above ~2 mA) compared to a blue InGaN LED (450 nm; above ~10mA). This is known as the “green droop” [19,20]. The similar current stability of a green/yellow/amber pc-LED with a LWPF can makes it possible to address the different current-dependence problems of various colored semiconductor LEDs as well as the green droop issue at a low driving current.

We also compared the equal-power luminous efficacy of full down-conversion green (G2)/amber (A1) LWPF-capped pc-LEDs to that of green/amber monochromatic III-V LEDs

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as a function of the driving current to address the superior optical properties of an LWPF capped pc-LED compared to those of a direct monochromatic LED that does not incorporate phosphors. Figures 6(a) and 6(b) show that the luminous efficacy ratings of the green and amber LWPF-capped pc-LED are higher than the efficacy of a corresponding green and amber monochromatic LED over the actual range of current used in practical applications (green: 5 ~300 mA, amber: 0.5 ~300 mA). The luminous efficacy of the full down-conversion green/amber LWPF-capped pc-LED and the green/amber monochromatic LED is 82.7/36.8 and 52.7/34.7 lm/W at 100 mA, respectively. The measured luminous efficacy ratings of the green and LWPF-capped pc-LED are 2.25 and 1.52 times higher than that of a direct-emitting green and amber LED without the use of phosphors.

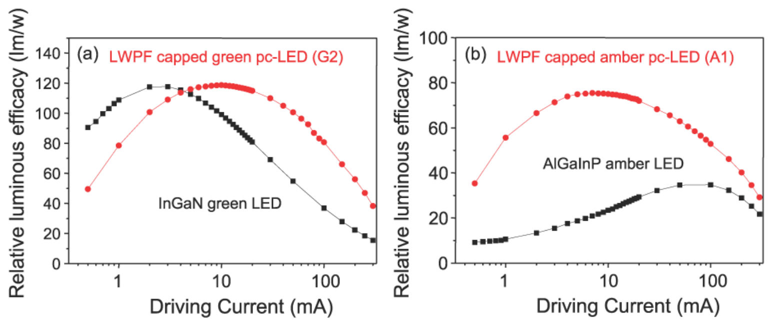

Fig. 6. Comparison of the luminous efficacy of a LWPF-capped pc-LED and a direct-emitting monochromatic semiconductor LED without phosphors as a function of the current density; (a) green color, (b) amber color.

As shown in Fig. 6 and Table 2, seven different colored LWPF-capped pc-LEDs show efficient performance compared to green and amber III-V LEDs without the use of phosphors. There are critical factors for determining the luminous efficacy of monochromatic LWPF-capped pc-LEDs. Of course, the performance of full down-converted LWPF-capped pc-LEDs increase with an increase in the performance of the InGaN-based pump LED. In addition, the performance of pc-LEDs depends directly on the quantum efficiency and packaging efficiency of each phosphor used in the pc-LEDs. Despite the fact that both efficient pumping blue LED and color-converting phosphor are combined, the possible loss of emission light from the phosphor layers should be reduced to produce efficient color pc-LEDs. For the purpose of decreasing the scattering loss and backward emission loss of the phosphor layers, many optical structures and phosphor morphologies in pc-LED coated paste have been suggested for use with white pc-LEDs [28–30]. Therefore, the effective combination of the efficient architecture of a blue pump LED, efficient phosphor materials and an optical structure of phosphor layers can be selected in future works to maximize the luminous efficacy and current/temperature stability of color LWPF-capped pc-LEDs.

4. Conclusions

Although the best architecture of pump LED, the highest efficient phosphor, or the best optical structure of the phosphor layer were not used in this experiment, this simple combination of a pc-LED and a LWPF nano-multilayered film represents a simple means of realizing the entire palette of colors in the CIE chromaticity diagram from efficient full down-converted, mono-chromatic LEDs given the availability of color-converting materials such as powder phosphors or quantum dots. The colors, performance levels, current-stability and temperature-stability data of the green, yellow and amber pc-LEDs as assessed here confirm that the concept of monochromatic LWPF-capped pc-LEDs can be enlarged to develop any color of efficient LED in the wavelength ranges in the green gap or the yellow gap of LEDs using powder phosphor or quantum dots. This paper simply reviewed the basic concepts for the realization of green, yellow, and amber colors using facile LWPF-capped pc-LEDs. Much more elaborate work remains to be done to determine the optimum phosphor material and morphology for a highly efficient monochromatic LWPF-capped pc-LED in the green gap, taking into account the development of new types of highly efficient color-converting materials and including an efficient combination of a blue pump LED, a phosphor layer and a LWPF coating in an achievable technique.

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