

Highly circularly polarized white light using a combination of white polymer light-emitting diode and wideband cholesteric liquid crystal reflector

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Abstract: We present a simple and intriguing device that generates highly circularly polarized white light. It comprises white polymer light-emitting diodes (WPLEDs) attached to a wideband cholesteric liquid crystal (CLC) reflector with a wide photonic bandgap (PBG) covering the visible range. The degree of circular polarization realized is very high over the visible range. The wide PBG was realized by introducing a gradient in pitch of the cholesteric helix by controlling the twisting power within the CLC medium. WPLEDs fabricated using a ternary (red, green, and blue) fluorescent polymer blend with the same moiety showed a low turn-on voltage, high brightness, high efficiency, and good color stability.

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1. Introduction

White polymer light-emitting diodes (WPLEDs) have attracted considerable attention because of their several advantages in a wide range of potential applications such as solid-state lighting [1,2], flat-panel display backlights [3], and flexible optoelectronic devices. WPLEDs could be crucial for next-generation color displays, and thus, a diverse range of

active materials have been investigated, such as a host light-emitting polymer doped with fluorescent polymers [4–7] or phosphorescent dyes [8–10] and polymers with specially engineered structures obtained, for example, by the introduction of side-chain chromophores [11] or copolymers [12,13]. In addition, device design and device-making processes are also important factors determining device performance for WPLEDs.

Furthermore, circularly polarized (CP) light sources based on WPLEDs are of considerable interest in applications such as three-dimensional displays, optical data storage systems, and optical communication devices. Circular polarizers are also widely used to improve the viewability of emissive devices and can act as excellent glare reduction filters for displays in bright light environments [14]. Recently, several approaches have been reported to produce CP electroluminescence (EL) from organic light-emitting diodes (OLEDs) [15–18], and a simple method to generate CP green light emission by attaching a stack of multiple cholesteric liquid crystal (CLC) films to OLEDs were reported [19]. However, the aforementioned CPEL exhibited several demerits such as limited wavelength range, low degree of circular polarization, and low device performances, etc.

CLCs exhibit wavelength- and polarization-selective reflection due to the helical arrangement of the molecules; that is, CLCs selectively reflect CP light with the same handedness as the helix. This feature makes CLCs one of the most interesting one-dimensional self-assembled photonic crystals (PCs) [20,21]. For normal incidence of light along the helical axis, selective reflection occurs if wavelength λ_R of the light is on the order of pitch p of the helix:

$$\lambda_R = np \quad (1)$$

where n is the average refractive index calculated as $n = ((n_o^2 + n_e^2) / 2)^{1/2}$ (n_o and n_e are the ordinary and extraordinary refractive indices of the CLCs, respectively). Selective reflection occurs within a bandwidth $\Delta\lambda = \lambda_R \cdot \Delta n / n$, where $\Delta n = n_e - n_o$ is the birefringence. Thus, the reflection bandwidth $\Delta\lambda$ is proportional to Δn . However, because the value of Δn is limited ($\Delta n \sim 0.2$), the reflection bandwidth is too limited for the abovementioned application [22,23]. This limited $\Delta\lambda$ can be overcome in practice by employing a multilayered stack of several different polymeric CLCs (PCLCs) [19,24–26] with different reflection bands or by implementing a gradual pitch gradient across the CLC film [27,28].

Herein, we present a simple and intriguing device structure that generates highly CP white light from WPLEDs incorporating a single PCLC reflector with a wide photonic bandgap (PBG). The WPLEDs were fabricated using a blend of red, green, and blue (RGB) fluorescent polymers with the same moiety. The wide PBG covering the visible range for the single PCLC reflector was realized via the pitch gradient method. Our fabricated WPLED showed low turn-on voltage, high brightness, high efficiency, and good color stability.

2. Experimental

2.1 Preparation of white light-emitting polymer blend solution

As the host polymer, soluble poly-spirobifluorene copolymer (Merck Co., SPB-02T), which is referred to as “M-blue” was used. Soluble poly(*p*-phenylene vinylene) (PPV) copolymer known as “Super Yellow” (SY; Merck Co.) was used as the fluorescent green-emitting dopant and another soluble poly-spirobifluorene copolymer (purchased from Merck Co., SPR-001), which is referred to as “M-red,” was also used as the fluorescent red-emitting dopant. Detailed chemical structures are shown in Fig. 1(a). All three fluorescent polymers were dissolved in toluene (11 mg/ml), and the concentration of the dopants was controlled by adding an appropriate amount of a diluted toluene solution of the SY and M-red dopants. First, the concentration of SY was set to the optimum value (0.1 wt%) by adding the required

amount of SY to the M-blue matrix. Then, the concentration of M-red was varied from 0 to 0.5 wt%.

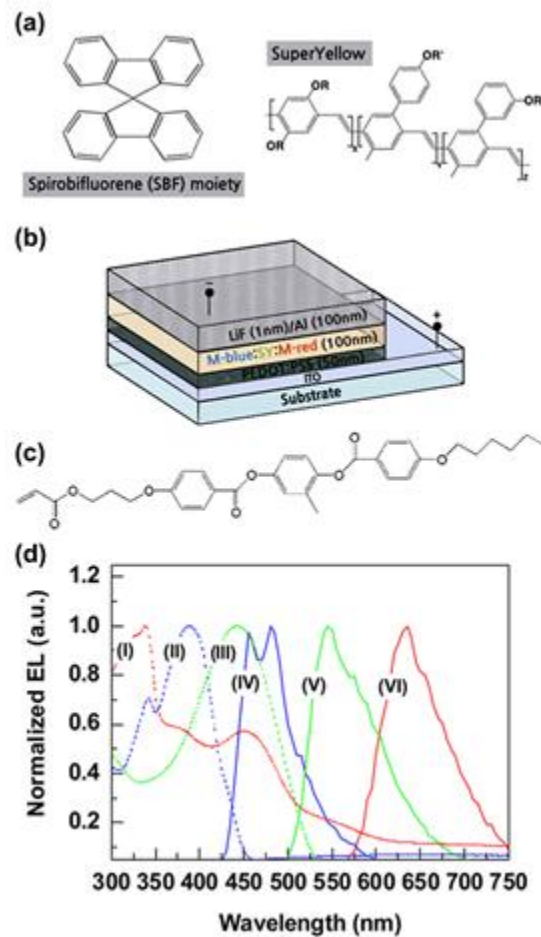


Fig. 1. (a) Detailed chemical structure of spirobifluorene (SBF) moiety contained in M-blue and M-red polymers and Super Yellow (SY). (b) Device configuration used in this study. The device structure was ITO/PEDOT:PSS/M-blue:SY:M-red/LiF/Al. (c) Chemical structure of host LC synthesized by our group. (d) Normalized UV-Vis absorption (dashed line) and photoluminescence (PL) (solid line) spectra of M-blue, SY, and M-red in the film: (I) absorption of the M-red film, (II) absorption of the M-blue film, (III) absorption of the SY film, (IV) PL of the M-blue film, (V) PL of the SY film, (VI) PL of the M-red film.

2.2 PLED fabrication

Glass substrates with patterned indium-tin-oxide (ITO) with a sheet resistance of $20 \Omega/\text{square}$ were carefully cleaned with detergent, followed by ultrasonication in DI water, acetone, and IPA for 10 min each. The substrates were dried in a vacuum oven and treated with UV/ozone for 20 min. Then, a PEDOT:PSS layer was spin-coated onto the ITO-patterned glass at a speed of 4000 rpm and baked at 140°C for 10 min. The ternary blend solutions were successively deposited onto the PEDOT:PSS layer at a speed of 2000 rpm to form a 100-nm-thick emissive layer. Thermal annealing was performed at 150°C for 1 h in a glove box. The LiF (1 nm)/Al (100 nm) cathodes were deposited by thermal evaporation under high vacuum ($<10^{-6}$ Torr). The device architecture is shown in Fig. 1(b).

2.3 Absorption and photoluminescence measurement

Absorption spectra were obtained with a Cary 5000 UV-Vis spectrophotometer (Varian, USA), and photoluminescence (PL) spectra were recorded using an FLS920 spectrophotometer (Edinburgh Instrument Ltd.) equipped with a 150-W xenon lamp at room temperature.

2.4 PLED device characterization

The current density (J)–voltage (V)–luminance (L) characteristics of the devices were measured with a computer-controlled Keithley 2400 source meter. A spectroradiometer (CS-2000, Konica Minolta Sensing, Inc.) was used to measure the EL spectra of the devices.

2.5 Wideband CLC reflector

The CLC reflectors were fabricated from a photo-reactive host LC with a mono-acrylate functional group synthesized by our group. The host LC exhibits nematic phase range from 76 to 149°C [29]. The chemical structure is depicted in Fig. 1(c). The CLC mixture comprised 95 wt% host LC and 5 wt% chiral dopant (CM223, BASF) with a small amount of photo-initiator (Igacure 907, Ciba-Geigy). CM223 has di-acrylate functional groups, which can also photo-react with the host LC. The prepared mixture was introduced into cells sandwiched between two glass substrates by capillary action at 80°C; separation between the glass substrates was maintained by a glass bead (5 μm). The LC cells were then annealed for 60 m to align the LC molecules homogeneously. Then, a planar structure in which the helical axis of the CLC molecules is perpendicular to the glass substrates was realized. The CLC films were kept at 80°C to induce massive displacement between the constituent during UV-light exposure (365 nm, 40~50 μWcm^{-2} , 70~90 s). Then, cross-linked CLC films with a spatial helical pitch gradient were obtained. We succeeded in fabricating broadband CLC reflector covering the visible range [27,28]. The merits of our reflector fabricated here in comparison with conventional multi-layered reflector are as follows; wide wavelength range covering the visible range, high degree of circular polarization, and simple making-process. These merits are attributed to the spontaneous pitch gradient process.

3. Results and discussion

As shown in Fig. 1(b), the WPLEDs were fabricated by sequential deposition of indium tin oxide (ITO; 150 nm) as the transparent anode, poly(styrenesulphonate)-doped poly(3,4-ethylenedioxythiophene) (PEDOT:PSS) as the hole injection/transport layer (HIL/HTL), a polymer blend (100 nm) as the emissive layer, LiF (1 nm) as the electron injection/transport layer (EIL/ETL), and Al (100 nm) as the metallic cathode. The emissive polymer blend consisted of three polymers, namely, M-blue (Merck Co.) as the blue emitter, super yellow (SY) (Merck Co.) as the green emitter, and M-red (Merck Co.) as the red emitter; the proportions of the red, green, and blue (RGB) fluorescent polymers were adjusted to realize white emission, i.e., Commission Internationale de L'Eclairage (CIE) coordinates (x , y) of (0.33, 0.33).

It should be noted that it is difficult to achieve white light emission with a simple ternary blend because significant color tuning of luminescence occurred simply by the addition of a small amount of the red and green dopants to the host blue matrix. Thus, precise control of the amounts of dopants added to the fluorescent host is required to suppress complete Förster energy transfer between the components. In addition, compatibility among the polymers in the blend is another key issue that determines the stability of white light emission. Most polymer blends are not highly miscible owing to their low entropy of mixing [30]. This can lead to phase separation and, as a result, variation in color with the applied voltage and low device efficiency. Thus, a homogeneous polymer blend of highly miscible polymers with analogous moieties is critical for improved device performance and stability [29,31]. Here,

the chemical structures of SY and the spirobifluorene (SBF) moiety in M-blue and M-red, as shown in Fig. 1(a), suggest that good compatibility can be expected because the polymers have the same moiety.

The UV-absorption and photoluminescence (PL) spectra of the host and dopant fluorescent polymers are shown in Fig. 1(d). The emission spectrum of M-blue ($\lambda_{\text{max}} = 460$ and 490 nm) shows spectral overlap with the absorption spectrum of SY ($\lambda_{\text{max}} = 445$ nm) and the broad absorption spectrum of M-red. Therefore, Förster energy transfer from the host M-blue to acceptors SY and M-red can be expected. However, the energy transfer efficiency is very sensitive to the distance between the donor and acceptor, and white emission can be obtained by partial energy transfer via dilution of the SY and M-red polymers in the M-blue polymer.

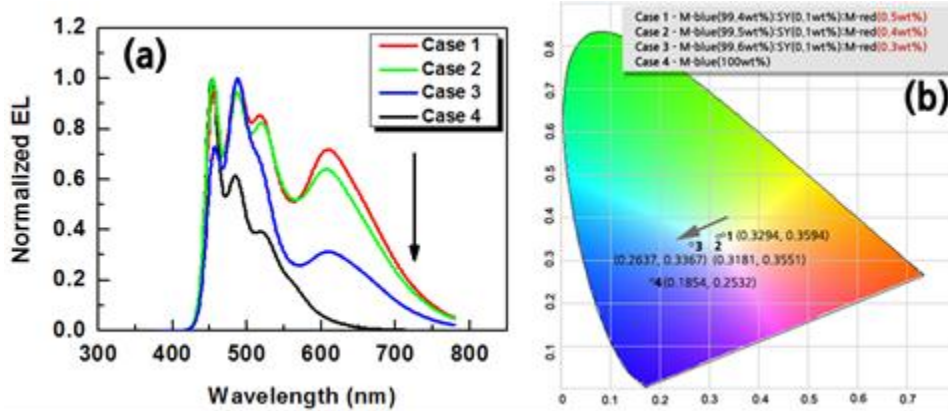


Fig. 2. (a) Normalized EL spectra of white emission from PLEDs with 0.1 wt% SY and varying amounts of red dopant M-red at luminance of 500 cd/m^2 : 0.5 (case 1), 0.4 (case 2), 0.3 (case 3), and 0 wt% M-red (100% M-blue). (b) CIE color diagram of white emission from PLEDs with varying amounts of M-red.

The amount of green-emitting dopant (SY) incorporated into the M-blue matrix was fixed at M-blue:SY = 99.4:0.1 by weight because the EL intensity of the green emission was equal to that of the blue emission for the optimized WPLEDs. Then, the amount of red-emitting dye was carefully adjusted to obtain white emission. As seen in the normalized electroluminescence (EL) spectra in Fig. 2(a), the device with the composition of M-blue:SY:M-red = 99.4:0.1:0.5 wt% exhibited three balanced emission peaks for the three primary colors, covering the visible range from 400 to 750 nm, whereas the device with ratio 99.6:0.1:0.3 wt% showed less intense red emission owing to the carrier trapping in the low-gap dopant polymer. As the concentration of M-red increased, the typical features of M-red emission were enhanced. This behavior was also reflected in the CIE coordinates, as shown in Fig. 2(b). The color coordinates dramatically move with only small changes in the amount of M-red added to the M-blue matrix. At a concentration of 0.5 wt% M-red, the CIE coordinates were (0.33, 0.36), corresponding to whitish emission.

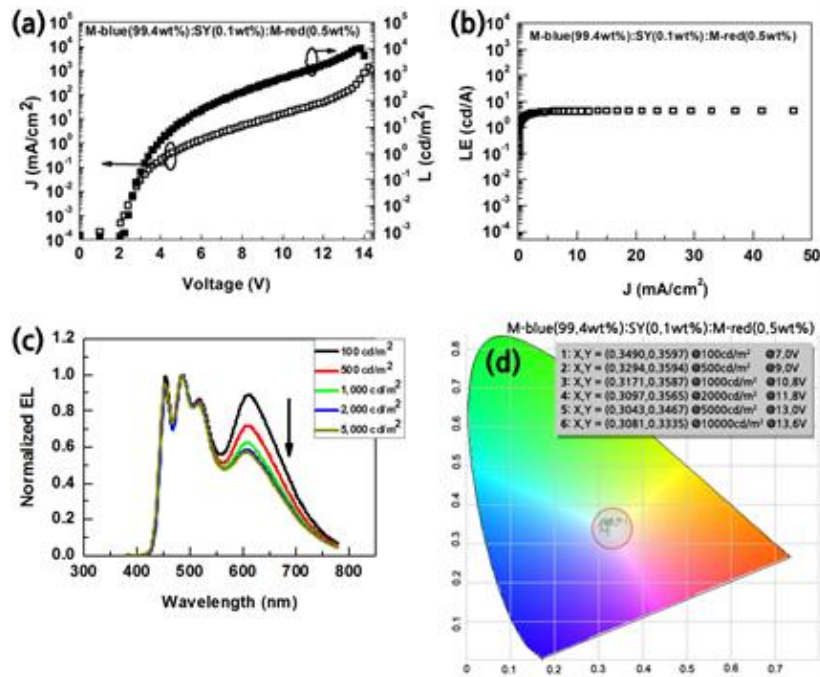


Fig. 3. (a) Current density (J) and luminance (L) versus applied voltage (V), (b) luminous efficiency (LE) versus current density (J), (c) color stability, and (d) variation in CIE color coordinates with applied voltage for white emission device with the structure ITO/PEDOT:PSS/M-blue (99.4 wt%):SY (0.1 wt%):M-red (0.5 wt%)/LiF/Al.

The current density (J)–voltage (V)–luminance (L) characteristics of the WPLEDs with 0.5 wt% M-red, as shown in Figs. 3(a) and 3(b), indicated a maximum luminance of 10,000 cd/m^2 (at 13.6V), maximum luminous efficiency of 4.7 cd/A (at 10.8V), and turn-on voltage of 2.4 V. Furthermore, as shown in Fig. 3(c–d), the white EL spectra and CIE coordinates slightly changed under different applied voltages from ($x = 0.349$, $y = 0.3597$) at 7.0 V to ($x = 0.3081$, $y = 0.3335$) at 13.6 V. This is because the low concentration of M-red in the blend results in partial saturation of emission from M-red at high current densities [32,33]. However, no significant color variation was observed with the applied voltage owing to the homogeneous mixing of the RGB polymers, as shown in Figs. 3(c) and 3(d). The detailed device characteristics including luminance, luminous efficiency, turn-on voltage, and CIE coordinates are summarized in Table 1.

Table 1. Detailed device characteristics of PLEDs with the structure ITO/PEDOT:PSS/M-blue(99.4wt%):SY(0.1wt%):M-red(0.5wt%)/LiF/Al at various applied voltages.

Voltage (V)	Luminance (cd/m^2)	Current density(mA/cm^2)	Luminous efficiency (cd/A)	CIE coordinates (X,Y)
7.0	100	2.9	3.78	(0.3490,0.3597)
9.0	500	9.5	4.52	(0.3294,0.3594)
10.8	1000	23.6	4.67	(0.3171,0.3587)
11.8	2000	46.8	4.62	(0.3097,0.3565)
13.0	5000	125.5	4.24	(0.3043,0.3467)
13.6	10000	284.8	3.39	(0.3081,0.3335)

The surface morphology of the blend with 0.5 wt% M-red was observed by atomic force microscopy (AFM), as shown in Fig. 4. The root-mean-square (RMS) roughness was low (0.228 nm), and no significant phase separation was observed, confirming the good

compatibility of the RGB fluorescent polymers. Moreover, we confirmed that there was no degradation in device performance parameters such as an increase in the turn-on and operation voltage or an abrupt decrease in the device efficiency.

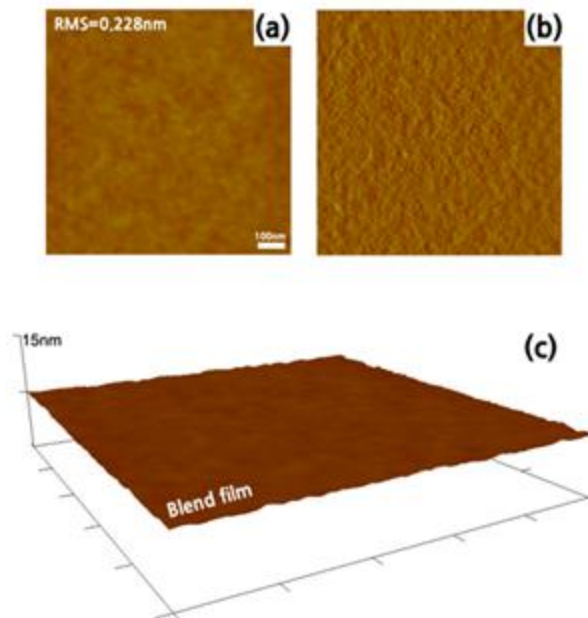


Fig. 4. Topographical image ($1 \mu\text{m} \times 1 \mu\text{m}$) in (a) height mode, (b) amplitude mode, and (c) three-dimensional height mode for RGB blend film with composition of M-blue:SY:M-red = 99.4:0.1:0.5 wt%.

The schematic device configurations for the proposed highly CP white EL device comprising the WPLED and a right-handed CLC (RH-CLC) reflector with a wide bandgap are shown in Fig. 5(a). To confirm polarization, a right or left circular polarizer, which is a sheet with a combination of a linear polarizer and a quarter wave retarder, was inserted between the device and the detector. Generally, PLEDs produce unpolarized EL, but when laminated with an RH-CLC reflector, selective reflection within the photonic bandgap (PBG) region occurs: left CP white light is transmitted through the RH-CLC reflector and right CP white light is reflected back by the RH-CLC reflector toward the Al electrode. This right CP white light reflects off the Al electrode and changes to the left CP white light that can pass through the RH-CLC reflector. As a result, only the left CP white light is obtained from the device shown in Fig. 5(a). As expected, we confirmed experimentally that the left CP light was dominant, whereas the intensity of the right CP light was extremely low within the PBG region, as shown in Fig. 5(b). The wide range of the PBG covering the visible range (from 350 nm to 750 nm) was also confirmed from the transmittance spectra, as shown in Fig. 5(b).

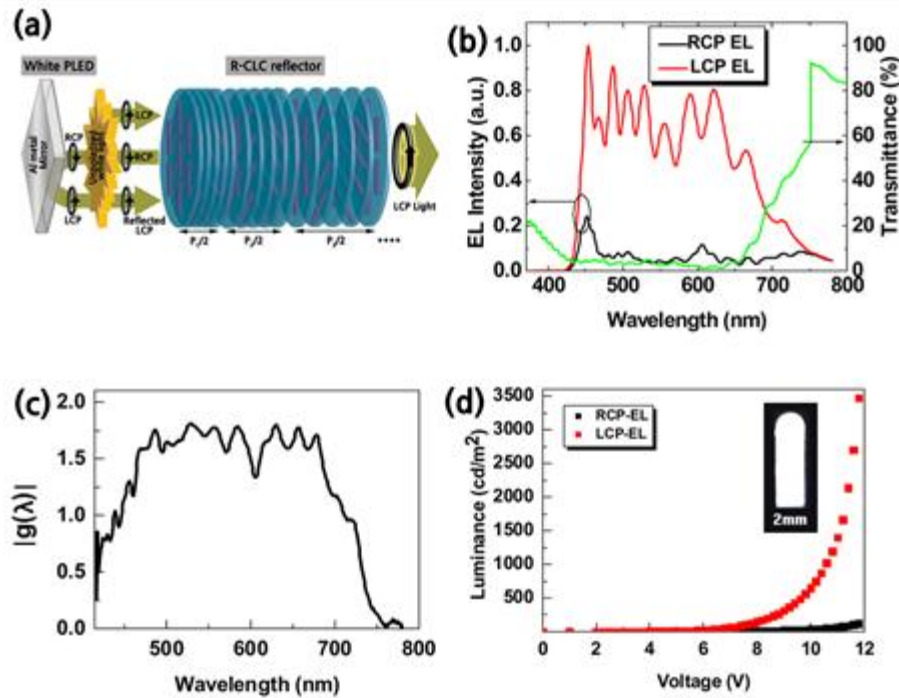


Fig. 5. (a) Schematic device configuration for highly CP white EL from WPLED and corresponding structure of R-CLC reflector with wide bandgap covering visible range. (b) Right (R)- and left (L)-CP-EL spectra. (c) Calculated g -factor over the entire wavelength range of the emission band of WPLED. (d) Luminance versus the applied voltage (L - V) for different polarized emissions from the WPLED combined with RH-CLC reflector. R and L polarizers were inserted to measure the intensity of R-CP-EL and L-CP-EL, respectively.

To quantify the degree of circular polarization, the wavelength-dependent g -factor is defined as

$$g(\lambda) = 2 \frac{r(\lambda) - 1}{r(\lambda) + 1} \quad (2)$$

where $r(\lambda) = I_L(\lambda) / I_R(\lambda)$ is the intensity ratio between the left and the right CP light. By definition, $|g(\lambda)| \leq 2$; $|g(\lambda)| = 0$ corresponds to the non-CP light and $|g(\lambda)| = 2$ corresponds to the perfectly single-handed CP light.

The g -factor over the entire wavelength range of the emission band of the WPLEDs was obtained using the result of Fig. 5(b), as shown in Fig. 5(c). The maximum value of $|g(\lambda)|$ was 1.8 at the center of the PBG, and the shape of the $|g(\lambda)|$ plot resembled the shape of the PBG for the RH-CLC used in this experiment.

The L - V characteristics in Fig. 5(d) confirm the polarization of EL emission. The luminance value for the left CP emission from the combination of WPLED and RH-CLC reflector was greater than 3500 cd/m^2 at 12 V whereas that of RCP emission was only approximately 120 cd/m^2 at 12 V. This confirms that the left CP white light was successfully obtained from the WPLEDs by attaching the RH-CLC reflector with a wide PBG.

4. Conclusion

We demonstrated a straightforward approach to generate highly left CP white light from WPLEDs attaching a wide PBG RH-CLC reflector. The wide PBG (from 350 nm to 750 nm) was realized by introducing a pitch gradient by controlling the twisting power within the CLC medium. Highly efficient white light emission from PLEDs was realized from a homogeneous blend of ternary polymers. The CIE coordinates were (0.33, 0.36), the maximum luminance was 10,000 cd/m² (at 13.6 V), the maximum luminous efficiency was 4.7 cd/A (at 10.8 V), the turn-on voltage was 2.4 V, and color stability was quite good. Furthermore, the degree of circular polarization was very high, as the maximum value of $|g(\lambda)|$ was 1.8 at the center of the PBG and the shape of the $|g(\lambda)|$ plot value matched that of the PBG. Thus, the novel architecture could be useful for various display and photonics applications where highly CP light is required.

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