

Enhancing the Electroluminescence Properties of Novel Blue Light Emitting Polymer and MEH-PPV Based White Light Emitting Polymer Devices by Processing Condition Optimization

Rong-Ho Lee, Hsin-Fang Hsu

Department of Chemical Engineering, National Yunlin University of Science & Technology, Yunlin, Taiwan 640, Republic of China

Received 30 October 2006; accepted 14 March 2007

DOI 10.1002/app.26904

Published online 8 August 2007 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: A series of triarylaminooxadiazole-containing tetraphenylsilane light emitting polymer (PTOA) and poly(2-methoxy, 5-(2'-ethyl-hexyloxy)-*p*-phenylene-vinylene) (MEH-PPV) based white light emitting polymer devices (PLEDs) were fabricated to study blue and orange-red emitter composition and light emitting layer processing effects on white emission electroluminescence properties. Color purity, current turn-on voltage, brightness, and current efficiency were strongly determined by MEH-PPV content and the thin film processing condition. The intensity of PTOA blue emission was equal to that of MEH-PPV orange-red emission when the device was fabricated by a polymer composite film containing 10 wt % of MEH-PPV. Color purity [Commission Internationale de L'Eclairage (CIE_{x,y}) coordinates (0.26,0.33)] was nearly white emission under applied 8 V. The brightness and current effi-

ciency of PTOA-MEH-PPV composite film based devices increased as MEH-PPV content increased. Furthermore, white emission blue shifted with increasing spin-rate of thin film coating and applied voltage. Low turn-on voltage, high current density, and high brightness were obtained for the device fabricating with light emitting layer coating with high spin-rate. Moreover, low current efficiency was obtained for the PLED with a thinner light-emitting layer. A white emission CIE (0.28,0.34) was obtained for PTOA-MEH-PPV based white PLED. White PLED brightness and efficiency can be as high as 700 cd/m² and 0.78 cd/A, respectively. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 2863–2869, 2007

Key words: electroluminescence; light emitting polymer blend; white polymer light emitting device

INTRODUCTION

Polymer light emitting displays (PLEDs) fabricated from semiconducting polymers have been extensively studied due to their potential use in flat panel displays.^{1,2} White PLEDs have attracted significant research attention because they can be combined with color filters to create full-color displays.³ Several techniques have been used to prepare white PLEDs, including single layer polymers doped with fluorescent dyes,^{4–10} dye doped in polymer film multilayers,¹¹ polymers mixed with a phosphorescent-Ir complex,^{3,12–15} single polymers with aggregates,¹⁶ exciplex formation in polymer film,^{17–21} single or multi-layer polymer blends,^{22–35} and a single polymer with an emitter moiety chemically doped onto the polymer main chain.^{36,37}

Kido et al. reported that several fluorescent dyes with different colors dispersed in poly(*N*-vinylcarba-

zole) (PVK) could emit white light with high brightness.⁴ Kim et al. showed that bright white PLEDs could be produced using a blend of two fluorescent dyes in a blue emission polyfluorene derivative as a light-emitting layer.⁸ Mikami et al. reported that white PLEDs with high brightness, efficiency, and color purity could be fabricated using a single light emitting layer of fluorescent dye doped polymers.¹⁰ Gong et al. showed that white PLEDs with excellent electroluminescence (EL) properties could be fabricated using a phosphorescent-Ir complex doped polymer as a light-emitting layer.¹² Shen et al. and Yang and coworkers reported that bright and color stable white PLEDs could be produced using MEH-PPV polyalkylfluorene blend as an emissive layer.^{24,25} Lee et al. demonstrated that bright white PLEDs could be produced based on red and blue light emitting fluorene copolymer bands.²⁶ Flexible color PLEDs based on red and blue emission phosphorescent polymer blending have been reported by Tokito et al.³⁴ On the basis of the above, white PLEDs can be easily obtained by blending red and blue emission polymers to form a light-emitting layer. White PLEDs with high brightness, high power efficiency, and good color stability have been developed.^{4,10,12,24–26,34} Most of these white

Correspondence to: R.-H. Lee (lerongho@yuntech.edu.tw).

Contract grant sponsor: National Science Council of Taiwan, ROC; contract grant number: NSC94-2216-E-224-005.

Journal of Applied Polymer Science, Vol. 106, 2863–2869 (2007)
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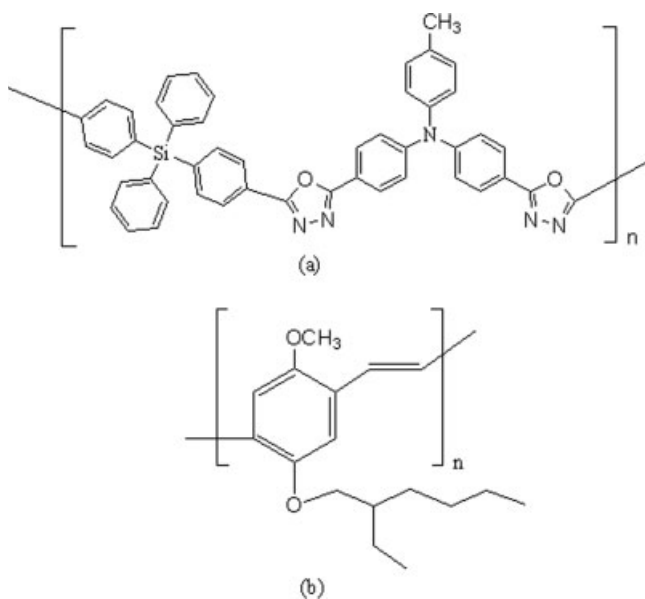


Figure 1 Chemical structures of (a) PTOA and (b) MEH-PPV.

PLEDs generated white emission by mixing two complementary colors.^{10,12,24-26,34} PLEDs exhibit almost the same blue and orange-red emitting band intensity, like two crests, which produce a strong white color. Nevertheless, the influences of light emitting layer composition and thin film processing on white emission color stability and EL properties have not been thoroughly examined.⁴⁻³⁷

This work blends poly(2-methoxy, 5-(2'-ethyl-hexyloxy)-*p*-phenylene-vinylene) (MEH-PPV) with a novel blue light-emitting polymer PTOA to generate a white light emitting solution for spin-coating fabrication. PTOA is synthesized according to the literature.³⁸ The orange-red polymer MEH-PPV is chosen due to its lower turn-on voltage and high brightness.³⁹⁻⁴¹ The orange-red and blue light emitting polymer blend single layer structure is the most attractive due to facile film formation, large-scale production, and cost effective

features. The spin-coated white PLED is further deposited with cathode in the vacuum chamber. The goal of this investigation is to study the relationship among the blue and orange-red emitter composition, light emitting layer processing condition, and white emission EL properties for the PTOA-MEH-PPV composite film based PLEDs.

EXPERIMENTAL

White LEP preparation

Figure 1 shows PTOA and poly(2-methoxy, 5-(2'-ethyl-hexyloxy)-*p*-phenylene-vinylene) (MEH-PPV) chemical structures. The PTOA synthesis procedure is reported in the previous literature.³⁸ PTOA possesses a very high T_g of 218°C. The 5% weight loss point appeared at 373°C. MEH-PPV was purchased from ADS (American Dye Source Corp. Quebec, Canada; $M_w = 150,000$). PTOA and MEH-PPV were dissolved in cyclohexanone, and mixed in different volume ratios to obtain white light emitting polymer (LEP) solutions. The compositions and sample abbreviations for the white LEP solutions are summarized in Table I.

White PLED fabrication

The device configuration of white polymer device (PLED) was ITO glass/PEDOT/white LEP/Cathode. Glass substrates with patterned ITO electrodes were well washed and cleaned by O_2 -plasma treatment. A thin film (600 Å) of hole-transporting material (PEDOT; AI4083, Bayer) was laid on the ITO-coated glass substrate using the spin-casting method. The white LEP thin film was spin-coated from the cyclohexanone solution onto the PEDOT layer to give a light-emitting layer and was dried at 70°C for 1 h in a glove box. A high purity calcium cathode (100 Å) was thermally deposited onto the white LEP thin film, followed by aluminum metal (1000 Å) deposition as the

TABLE I
Composition, Film Thickness, Optical Property, and EL Performances of White Polymer Light Emitting Devices

Device no.	PTOA: MEH-PPV (wt %) ^a	Film thickness (nm) ^b	$\lambda_{\max}^{\text{PL}}$ (B,R) ^c (nm)	$\lambda_{\max}^{\text{EL}}$ (B,R) ^c at 8 V	CIE (<i>x,y</i>) ^d at 8 V	Max. brightness (cd/m ²)	Max. efficiency (cd/A)
I	100 : 0	54.5	(459,-)	(442,-)	(0.18,0.18)	629	0.37
II	100 : 1	54.9	(461,531)	(442,-)	(0.18,0.18)	620	0.32
III	100 : 3	58.8	(459,543)	(444,528)	(0.20,0.21)	656	0.42
IV	100 : 6	58.9	(451,554)	(444,528)	(0.21,0.25)	890	0.47
V	100 : 10	62.4	(453,554)	(443,542)	(0.26,0.33)	1225	0.67
VI	100 : 25	66.2	(445,550)	(445,563)	(0.33,0.42)	1867	1.10

^a Concentration of PTOA-MEH-PPV solution: 10 mg/mL.

^b Spin coating rate of light emitting layer: 2000 rpm.

^c The wavelength of maximal PL (or EL) emission band: (B,R) = (Blue emission, Orange-Red emission).

^d Color purity of white emission: CIE (*x,y*).

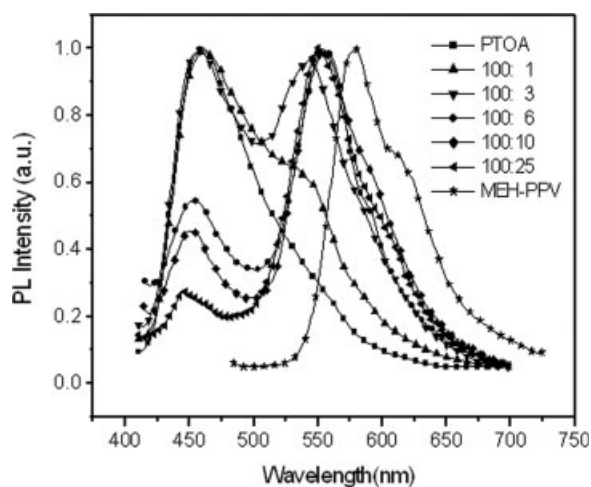


Figure 2 PL spectra of PTOA-MEH-PPV composite films.

top layer in a high vacuum chamber. The device was then encapsulated by a glass cover, and sealed with UV-cured epoxy glue. The cathode deposition rate was determined with a quartz thickness monitor (STM-100/MF, Sycon). Thin film thickness was determined with a surface texture analysis system (3030ST, Dektak). Photoluminescence (PL) and electroluminescence (EL) spectra were recorded on a Hitachi F-4500 fluorescence spectrophotometer. The current-voltage characteristics were measured on a programmable electrometer with current and voltage sources (Keithley). Luminance was measured with a BM-9 luminance meter (Topcon).

RESULTS AND DISCUSSION

Composition dependence of white PLED electroluminescence performances

PL spectra of PTOA-MEH-PPV based composite films with various weight percentage ratios is shown in Figure 2. Composition, thin film thickness, and wavelength of maximum PL emission of PTOA-MEH-PPV films are summarized in Table I. PTOA shows PL with an emission maximum at 460 nm accompanied by a shoulder at around 530 nm. The PL main peak at 580 nm is accompanied by an excimer emission shoulder at ~ 620 nm for the MEH-PPV film. The maximal emission intensity of PTOA decreased significantly with increasing MEH-PPV content for the PTOA-MEH-PPV based composite films. Blue emission band intensity was equal to the orange-red emission band for the composite film containing 3 wt % of MEH-PPV. Excellent compatibility between PTOA and MEH-PPV resulted in significant PTOA emission band change with MEH-PPV incorporation into PTOA film. In addition to reduced PTOA emission intensity, blue emission band FWHM decreased with

increasing MEH-PPV content due to excimer formation suppression by MEH-PPV polymer chain presence. Emission from PTOA excimer was attributed to interaction between oxadiazole and diphenyl(4-tolyl) amine groups.³⁸ Additionally, orange-red emission band was also sensitive to varying MEH-PPV content. Orange-red emission blue shifting from MEH-PPV was enhanced with increasing PTOA content due to PTOA polymer chain dilution.

EL spectra of PTOA-MEH-PPV composite film based PLEDs is shown in Figure 3. Maximum EL emission wavelength, and color purity of PTOA-MEH-PPV film based devices are summarized in Table I. The PTOA-MEH-PPV films were coated with 10 mg/mL cyclohexanone solution onto the PEDOT layer. EL spectra composition dependence is similar to PL spectra, two complimentary emission bands corresponding to the PTOA and MEH-PPV are observed at around 440 and 550 nm, respectively. Orange-red emission band maximum significantly varied with increasing MEH-PPV content due to PTOA polymer chain dilution. Blue emission band intensity was equal to the orange-red emission band for the composite solid film containing 10 wt % of MEH-PPV. The color purity CIE (0.26,0.33) of device V is nearly white emission under applied voltage of 8 V. White PLED color purity was found dependent on the composition of PTOA and MEH-PPV based composite film. Current density, brightness, and current efficiency versus PTOA-MEH-PPV applied voltages are shown in Figure 4. Brightness and current efficiency maximum of white PLEDs are also summarized in Table I. Brightness and current efficiency of PTOA-MEH-PPV composite film based devices increased with increasing MEH-PPV content due to MEH-PPV high brightness.³⁹⁻⁴¹

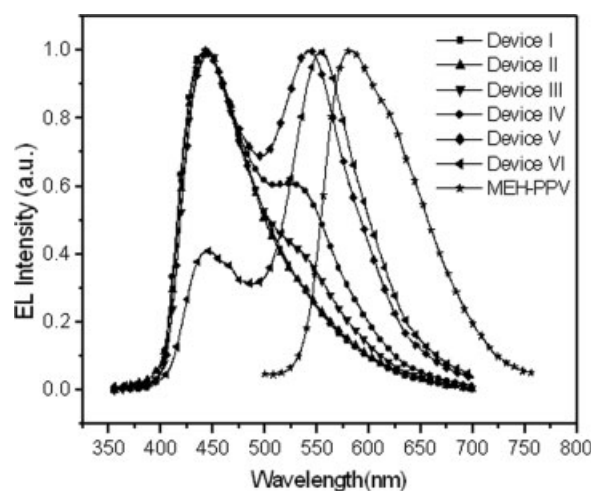


Figure 3 EL spectra of PTOA-MEH-PPV composite films based white light emitting devices under applied voltage of 8 V.

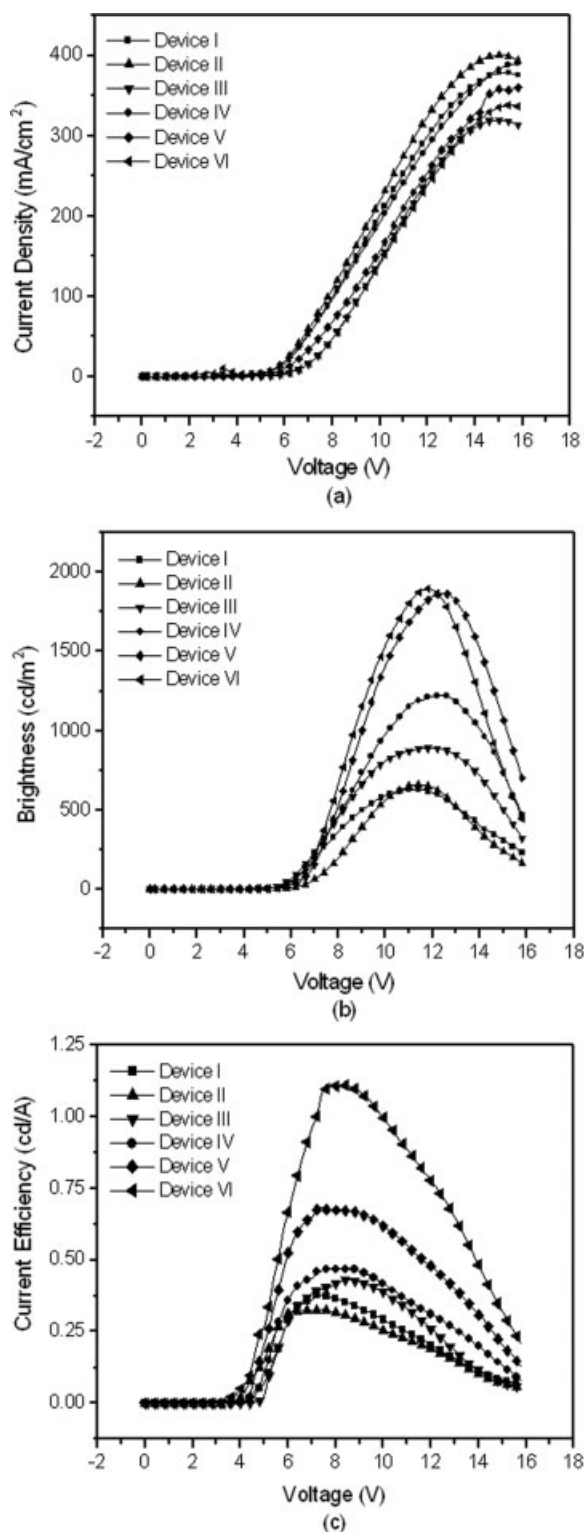


Figure 4 EL performances of PTOA-MEH-PPV composite films based white light emitting devices I-VI.

Thin film processing condition influence on white PLED electroluminescence performance

EL spectra of PTOA-MEH-PPV composite film based white PLEDs are shown in Figure 5. The light emitting layer PTOA-MEH-PPV was coated with 15 mg/mL

cyclohexanone solution onto the PEDOT layer with different spin rates. The light emitting layer spin-coating conditions and film thickness, and EL emission band wavelengths for devices V1-V3 are summarized in Table II. The intensity and wavelength of maximal

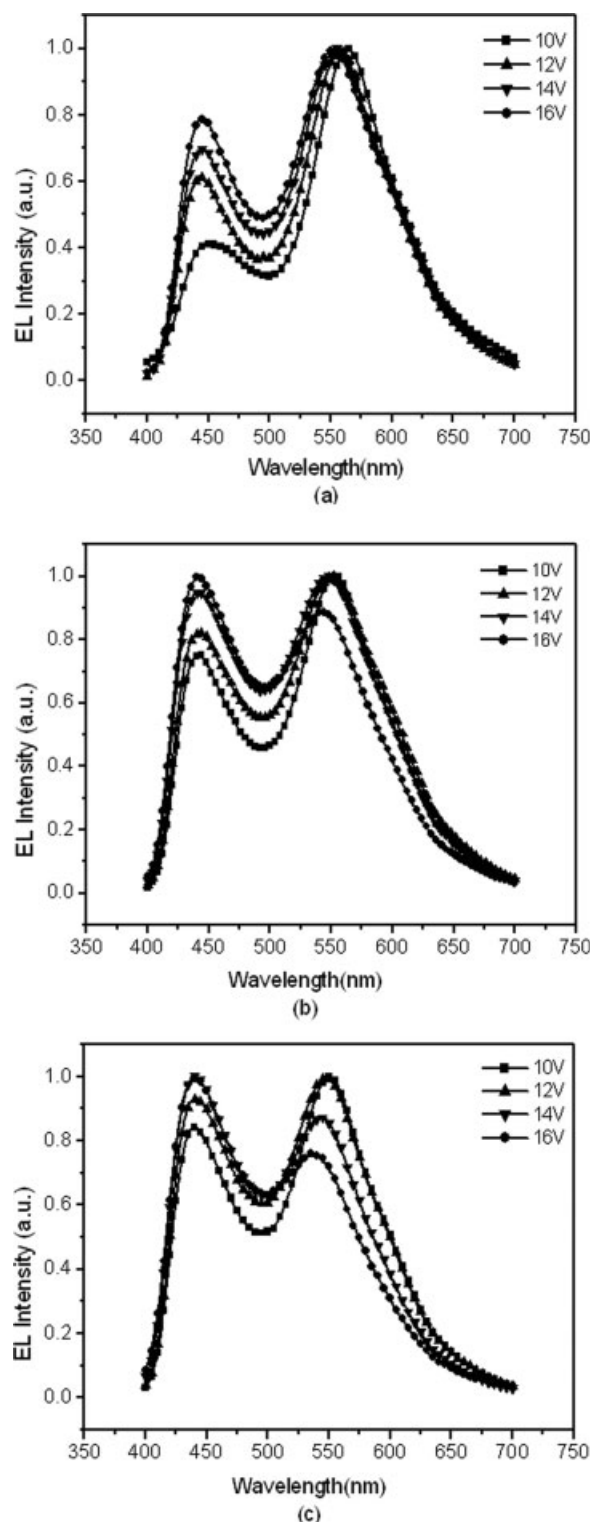


Figure 5 EL spectra versus applied voltages for the PTOA-MEH-PPV based white polymer light emitting devices [(a) device V1, (b) device V2, and (c) device V3].

TABLE II
Spin-Coating Condition, Thin Film Thickness and EL Performances of PTOA-MEH-PPV based White PLEDs

Device no.	Spin-rate ^a (rpm)	Film thickness ^b (nm)	$\lambda_{\max}^{\text{EL}}$ (B,R) ^c at 14 V (nm)	CIE (x,y) at 10 V	CIE (x,y) ^d at 12 V	CIE (x,y) ^d at 14 V	CIE (x,y) ^d at 16 V
V1	2000	67.6	(445,555)	(0.26,0.32)	(0.33,0.38)	(0.32,0.37)	(0.31,0.36)
V2	4000	62.5	(443,548)	(0.31,0.37)	(0.30,0.36)	(0.28,0.34)	(0.26,0.32)
V3	6000	55.6	(440,543)	(0.29,0.35)	(0.28,0.34)	(0.26,0.31)	(0.25,0.30)

^a Spin-rate of light emitting layer coating process.

^b Concentration of PTOA-MEH-PPV polymer solution: 15 mg/mL.

^c The wavelength of maximal EL emission band: (B,R) = (Blue emission, Orange-Red emission).

^d Color purity of white emission: CIE (x,y).

emission peaks (λ_{\max}) varied with increasing applied voltage. Moreover, emission band change with increasing applied voltage was attributed to light emitting layer morphological modification for devices V1–V3. Applied voltage dependence of wavelength and intensity for the blue and orange–red emission bands are shown in Figure 6. In Figure 6(a), the change of λ_{\max} blue emission band with increasing applied voltage was not pronounced as compared to the orange–red emission band. For the blue emission band, the lower λ_{\max} value was observed for devices V2–V3 in comparison with device V1. Nevertheless, thin film processing condition influence on λ_{\max} blue emission band value was not significant. Applied voltage effect on λ_{\max} value can be neglected except for device V1 under applied voltage of 10 V. On the other hand, the orange–red emission band λ_{\max} blue-shifted significantly by increasing the spin-coating rate for devices V1–V3. The λ_{\max} values also decreased significantly with increasing applied voltage. The MEH-PPV polymer chain extended thoroughly during the thin film coating process high spin rate.^{38,42} Aggregation and excimer formation between MEH-PPV polymer chains subsequently reduced due to high centrifugal force on the thin film. The orange–red emission band λ_{\max} resulting blue-shifted with increasing spin-coating rate, corresponding to reduced MEH-PPV inter-chain exciton emission intensity, especially for the device under applied high electric field. Blue emission band emission intensity in Figure 6(b) increased with increasing applied voltage, while red-range emission band intensity decreased with increasing applied voltage. This is due to the fact that the electron hole pair dominantly recombined in the MEH-PPV polymer chains at the lower applied voltage. Carrier recombination would focus on the PTOA chains under higher applied voltage. In addition, electron hole pair recombined in different polymer chains by changing the applied voltage because of the different bandgaps of the constituents.^{5,27,43} Emission intensity of both blue and orange–red emission bands resultingly varied with increasing applied voltage. Two complimentary emission intensity changes were not significant for device V2 with thin film spin coating with a 4000 rpm

rate. White emission good color purity CIE (0.28,0.34) was observed for device V2 under applied 14 V as shown in Table II. The emission intensity of blue emission enhanced with increasing spin rate coating process,

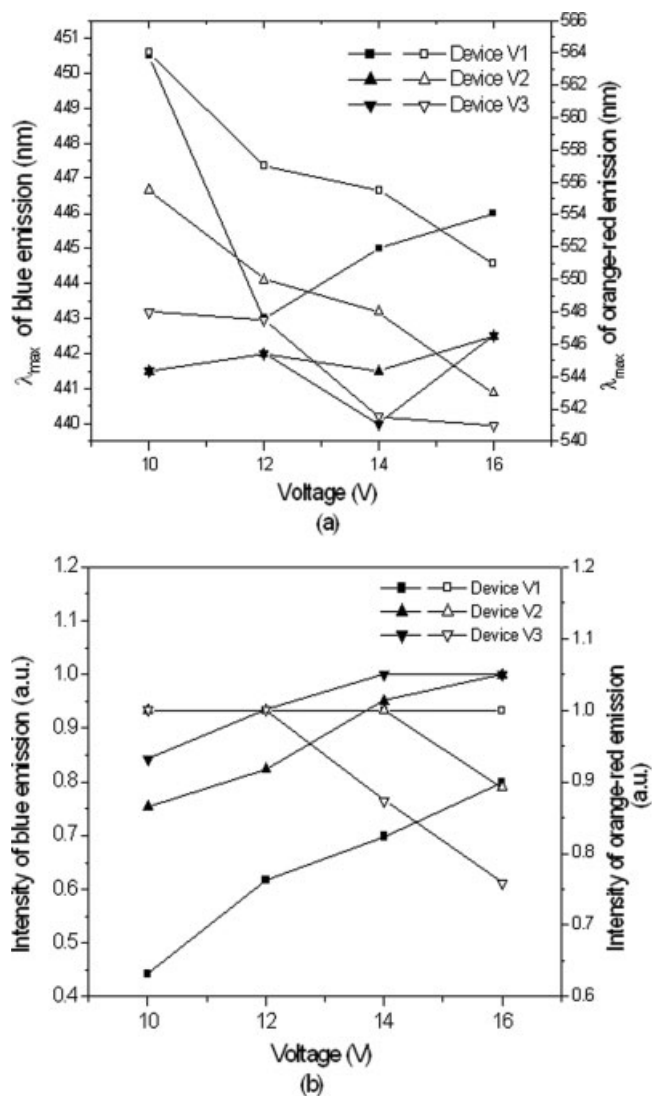


Figure 6 Applied voltage dependence of wavelength and intensity for the blue and orange–red emission bands of polymer white light emitting devices V1–V3 (filled symbol: blue emission; open symbol: orange–red emission).

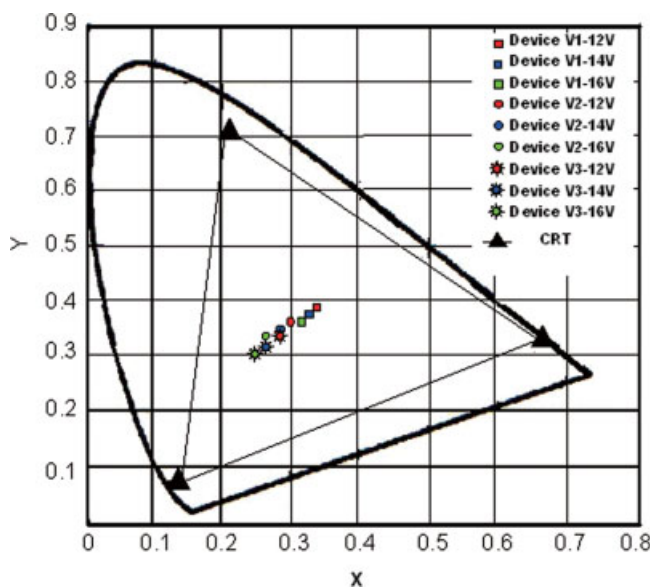


Figure 7 CIE 1931 chromaticity diagram of white PLEDs V1–V3 under various applied voltages. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

while the orange–red emission band decreased with increasing spin-coating rate. This is due to the thoroughly extended polymer chain during the high spin rate coating process, favorable for carrier transporting and recombination within the PTOA polymer chains. Therefore, PTOA blue emission intensity enhanced with increasing spin-coating rate. Blue emission intensity enhancement corresponding to enhanced current density also partially contributed from reduced light emitting layer thickness. Conversely, the orange–red emission intensity decreased with increasing spin-coating rate, especially for device V3 under high-applied voltages. Blue emission intensity increased for devices V1–V3 with increasing spin-coating rate and applied voltage, attributed to carrier recombination enhancement in PTOA. Subsequently, the electron-hole pair recombination in MEH-PPV relatively reduced with increasing spin-coating rate and applied voltage. Therefore, orange–red emission intensity reduced with increasing applied voltage, especially for thinner light emitting layer devices. The blue and orange–red emission intensities mentioned above were changed with varied thin film processing conditions, corresponding to light emitting layer morphology modification. The CIE 1931 chromaticity diagram of devices V1–V3 under applied various voltages are shown in Figure 7. The white emission blue shifts with increasing applied voltage and thin film coating process spin-rate.

Current density, brightness, and current efficiency versus devices V1–V3 applied voltage are shown in Figure 8. Current density and brightness increased with decreasing light emitting layer thickness, corresponding to increasing spin-coating rate. Moreover, turn-on volt-

age decreased with spin-coating rate of light emitting layer for devices V1–V3, due to better contact of hole transporting and light emitting layers. This is favorable for hole injection into the light emitting layer. Subsequently, lower turn-on voltage of device V3 was observed in comparison with devices V1–V2. However,

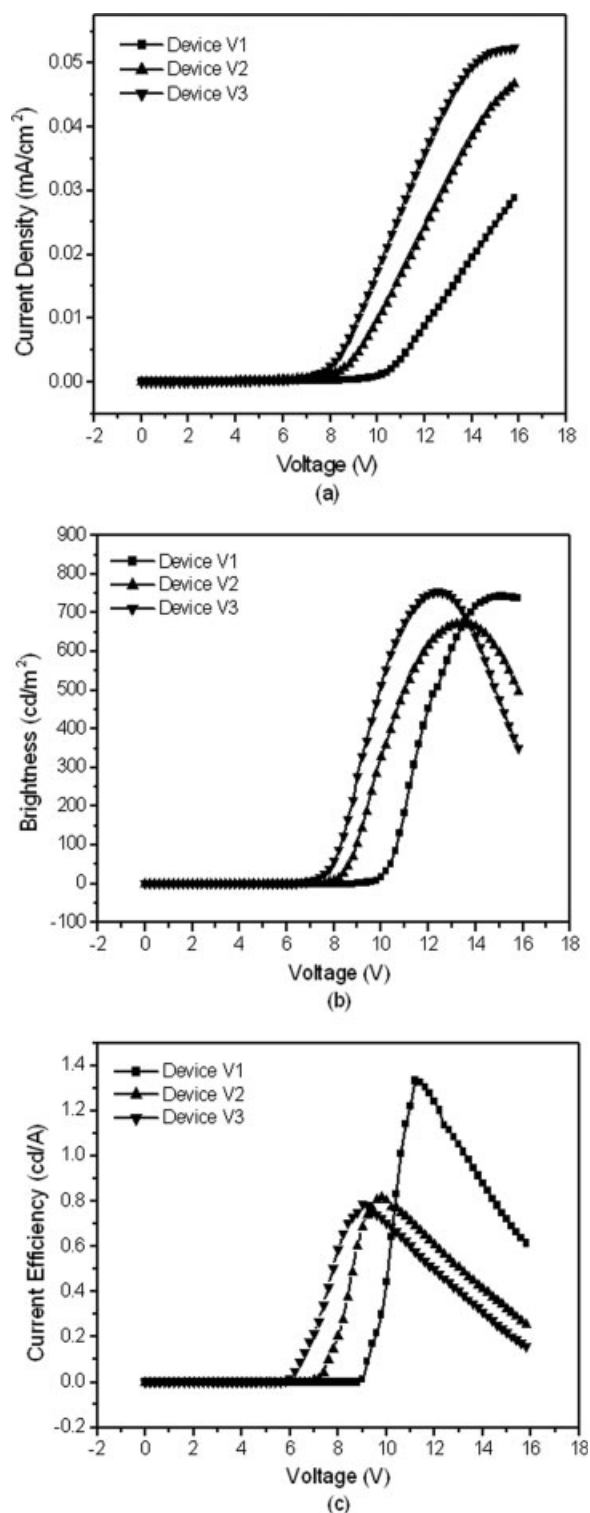


Figure 8 EL performances of white polymer light emitting devices V1–V3.

current efficiency reduced with decreasing light emitting layer thickness. Particle presence on the ITO glass surface, and ITO surface roughness could not be smoothed down by a thin polymer film. Electric leakage is likely to occur as a result during PLED operation processes because of defect presence.⁴⁴ Consequently, low current efficiency was obtained for PLED with a thinner light-emitting layer.

CONCLUSIONS

A series of white light emitting PLEDs based on a novel blue light emitting polymer PTOA and an orange-red polymer MEH-PPV were fabricated for studying the influences of blue and orange-red emitter composition, and thin film processing condition on white emission EL properties. In addition to PTOA-MEH-PPV composition, color purity, current turn-on voltage, brightness, and current efficiency of white PLEDs show the strong thin film processing condition dependence. White emission high color purity was observed for the device based on light emitting polymer composite film containing 10 wt % of MEH-PPV. Brightness and current efficiency of PTOA-MEH-PPV composite film based devices increased with increasing MEH-PPV content. Thin film processing condition influence on λ_{\max} blue emission band value was not significant, while orange-red emission band λ_{\max} significantly blue-shifted with increasing spin-coating rate. The intensity of blue emission enhances with increasing spin-coating rate, while that of orange-red emission band decreased with increasing spin-coating rate. Blue and orange-red emitting wavelength changes and intensities were attributed to varied orientation of polymer chains. High brightness, low turn-on voltage, and low current efficiency were observed for the device based on light emitting layer with a high spin-coating rate. It is concluded that EL properties were determined by both blue and orange-red emitter composition and the spin coating rate of the white light emitting layer.

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