



## گستره کاملی از رنگ تابشهای مختلف از دیودهای نورتاب پلیمری

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چکیده - در این مقاله، ساخت دیودهای نورتاب آلی با رنگ تابشهای متنوع، به کمک مخلوط کردن و یا آلایش کردن پلیمر میزبان گزارش می-شود. در ابتدا ساخت دیود آلی های پایه با تابشهایی به رنگهای آبی، سبز، نارنجی و سفید مهتاب گونه توصیف می شود. سپس، ساخت نوآورانه دیودهایی با تابش به رنگهای خاص مانند تابش بنفش، قرمز و سفید آفتاب گونه ارائه می شود. برای تنظیم رنگ تابش در دیودهای آلی، تابش آبی از پلی فلورین، تابش سبز از جفت پلیمر پلی فلورین و بنزوئیدازول، تابش نارنجی از پلیمر فنیلین ونیلین به همراه تابش قرمز از لموجن قرمز در ترکیبهای مختلف با یکدیگر به کار گرفته شده اند.

کلید واژه- دیود آلی، دیود نورتاب آلی سفید، پلی فلورین، مخلوط کردن و آلایش کردن، روشنایی حالت جامد.

## Full range of Emission colours from Polymeric Light-Emitting Diodes

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Abstract- We demonstrate the fabrication of organic light-emitting diodes (OLED) with various emission colours by blending and/or doping the polymer host matrix with different dopants. We firstly present the preparation of elementary OLEDs i.e. blue, green, orange, and cool white diodes. Then, we exhibit the fabrication of light-emitting devices with particular emission colours such as magenta, red, and warm white electroluminescence. Blue emission from polyfluorene (PFO), green emission from polyfluorene-alt-benzothiadiazole (F8BT), and orange emission from polyphenylene-vinylene (MEH-PPV), together with the red emission from Lumogen F Red have been mixed in different combination to vary the emission colour in OLEDs.

Keywords: OLED, white organic light-emitting diode, polyfluorene, blending/doping, solid-state lighting.

## 1 Introduction

Intensive researches have been conducted in the field of organic light-emitting diodes (OLEDs) to fabricate low-cost high-quality flat-panel displays and efficient solid-state light sources. Studies concerning OLEDs include synthesizing novel materials, enhancing device structure, and modelling the mechanisms affecting the device characteristics [1-3]. Fabrication of OLEDs with primary-colour emissions *i.e.* pure green, red, and blue is essential to produce portable displays and colour TVs. In addition, OLEDs with cool or warm white illumination are being developed to enable the promise of solid-state lighting. Conjugated polymers have been widely used to fabricate solution-processed OLEDs due to ease in their fabrication procedures such as spin coating, roll-to-roll casting, and ink-jet printing [4]. For instance, poly [2-methoxy-5-(2-ethylhexyloxy)-1, 4-phenylenevinylene] (MEH-PPV) is one of the first materials have been introduced and employing in OLEDs or poly [9, 9-di-n-octylfluorenyl-2, 7-diyl] (PFO) and poly [(9, 9-di-n-octylfluorenyl-2, 7-diyl)-alt-(benzo[2, 1, 3]thiadiazol-4, 8-diyl)] (F8BT) have been studied intensively due to their promising properties. In present paper, we report the fabrication of polymer light-emitting diodes with a varying range of different colour including blue, green, red, orange, magenta, and also warm and cool white emission. By using a polymer host matrix blended with another polymer or doped with a fluorescent dye, we were able to effectively change the emission colour of the device.

## 2 Experiment

We used indium-tin-oxide (ITO) coated polyethylene-terephthalate (PET) slides as transparent anode. In order to clean the electrode, we used sonication procedure and placed the slides in detergent, acetone, methanol, and isopropanol each for six minutes. Next, we coated poly [3, 4-ethylenedioxythiophene] - [styrenesulfonate] (PEDOT:PSS) on the top of ITO at 3000 rpm and baked the sample at 100 °C. The latter layer with thickness of 60nm acts as hole injecting layer in the device structure. Afterward, we employed three different polymers; namely PFO, MEH-PPV, and F8BT, and also a fluorescent dye to prepare various master solutions for the emissive active layer. We used Lumogen® F Red 305 as the red fluorescent dye, which is an organic chromophore based on perylene and presents promising properties such as high stability and high quantum efficiency. The polymers and organic dye were dissolved in toluene with concentration of 10

mg/mL to form the master solutions (~2mg/mL for MEH-PPV). Next, various solutions with different ratio concentrations were prepared by diluting and mixing the master solutions. Final solutions were spin-casted at 3000 rpm on the top of the hole injecting layer. All the prime materials were purchased from Sigma Aldrich Co. except for the Lumogen F Red which was obtained from BASF Company. The fabrication of the devices was completed by thermal evaporation of Al cathode electrode in the vacuum chamber with base pressure of  $\sim 10^{-5}$  mbar. Finally, we recorded the current density against voltage ( $J$ - $V$ ) curves by means of a Keithley 238 source-measurement unit whilst we captured emission intensity by using a calibrated photodiode stage.

## 3 Results and Discussion

Employing PFO for the active layer in a device architecture as ITO/PEDOT:PSS/PFO/Al leads into blue electroluminescence (EL) from the conjugated polymer. The diode has a turn-on voltage of 10V and also its current density increases to be around 1500 A/m<sup>2</sup> when a bias voltage of 17V is applied. It is noteworthy that we can dramatically enhance the device characteristic by using a low-work function metals such as Ca or by inserting a thin layer of LiF in the device structure. We mentioned we can alter the colour emission of the device by blending the active layer with other light-emitting polymers. F8BT is a green emitting copolymer based on PFO. Using F8BT or F8BT blended with PFO in concentrations higher than 5 wt. % as the emissive layer leads into green emission from the organic device. This diode begins to light in voltage of 7V and when a bias voltage of 11V is applied, a current density of around 1500 A/m<sup>2</sup> steadily flows through the device. As another choice, we can use fluorescent dyes to modify the emission in OLEDs. We prepared another emitting diode by doping the polyfluorene host polymer with the Lumogen F Red with rather high concentration of ~50 wt. % to effectively active the fluorescence from the organic dye. Lumogen F Red has a strong emission peak at around 600 nm. On the other hand, the emission peak of PFO is around 417nm whereas the absorption peak of Lumogen F Red is at 576nm. This leads to weak overlap between the two spectra and so partial energy transfer from the host into the guest dopant. Therefore, the blue emission from PFO does not be suppressed completely by the dopant and in combination with the red emission from organic dye yields in a bright magenta emission from the diode. The electrical

characteristic of magenta diode is similar to that of blue device; for instance, the turn-on voltage of magenta diode is around 11V and a current density of 900 A/m<sup>2</sup> flows through the device at bias voltage of 18V. Chromospheres doped into or blended in the host matrix trap mobile carriers and form charged centres which attract the carriers with opposite charge; therefore, recombination mainly takes place in low band-gap guest emitter leading to wavelength conversion. In addition, conjugated polymers as dopants can facilitate charge trapping due to their semiconducting nature and so can enhance electron-hole recombination in host-guest systems. Incidentally, the fluorescent dye dopant in magenta device was not able to suppress the short wavelength peak emission by the host matrix and the overall electroluminescence from the device was a combination of the emissions from both the host and the guest moieties. Nevertheless, we can dominate the emission from the fluorescent guest dopant; if we employ another host polymer with photoluminescence (PL) close to the absorption spectrum of the dye. We replaced PFO with MEH-PPV which has an emission peak of around 554nm. We know employing the MEH-PPV as the emissive layer leads into orange emission from the organic device; however, incorporating the fluorescent dye into MEH-PPV as the host matrix changes the colour tune into red emission. Electrical characteristic of the red and orange diodes are quite similar. For instance, current density of around 10000 A/m<sup>2</sup> flows through the two diodes for a bias voltage of around 10V. Low barrier potential between the electrodes and the active layer cause the current density to be higher in MEH-PPV-based diodes in comparison with diodes employing PFO. We mentioned white light-emitting diodes are of paramount importance to develop displays and solid-state light sources. As the first attempt to fabricate white light-emitting organic diode, we combined the emission spectra of PFO, F8BT, and Lumogen F Red in a single active layer (with weight ratio of 94.5:5:0.5 for PFO:F8BT:Lumogen). In principle, we can finely tune the concentration of the dopants to achieve pure white emission from the device. However, in an alternative structure we can mix the blue emission from polyfluorene with the complementary orange emission from polyphenylene-vinylene to prepare a promising white polymer-LED. We added MEH-PPV into PFO solution with weight ratio of 1:99 in the next OLED. The electroluminescence spectrum of the latter device simultaneously possesses two peaks, namely at 417nm due to PFO host matrix and at 554nm due to MEH-PPV guest polymer and

therefore, a strong white illumination is observed from the polymer-blend. This white OLED begins to illuminate at voltage of 7V and its current density increases to be 2000 A/m<sup>2</sup> at bias voltage of 11V. In chromaticity diagram presented in Fig. 1, we observe the device has a CIE coordinate of (0.31, 0.33) and a colour correlated temperature of 6492 K which proves this kind of organic device has a considerable potential to be employed in the

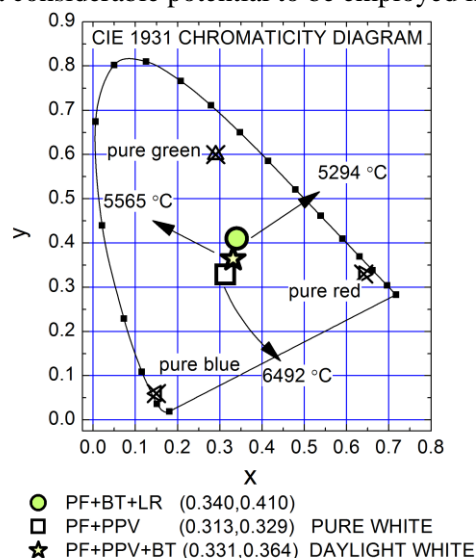


Figure 1. The chromaticity coordinates ( $x$ ,  $y$ ) for the white OLEDs and also the corresponding correlated color temperatures (CCT).

fabrication of solid-state light sources. We should mention the obtained CIE coordinate are estimated by firstly approximates an RGB coordinate from a preliminary image processing protocol. Then, we applied a mathematical transformation to calculate the CIE coordinate values. It is noteworthy that we might enhance the orange peak of the spectrum by slightly increase the concentration of the MEH-PPV in the polymer blend, to prepare a device with warm-white emission colour; however, as a novel approach, we apply a well-controlled enhancement in the long wave-length region by introducing another peak into the spectrum. Practically, we fabricate an efficient warm white OLED by incorporating both F8BT and MEH-PPV into the PFO host polymer with weight ratios of ~5 wt.% and ~1 wt.% respectively to acquire the emissive active layer. Fig. 2, reveals the strong warm white emission from the diode in operation. The device has a CCT value of 5565 K and CIE coordinate of (0.33, 0.36) which are slightly off-centre towards to the red region.

## Conclusion

In summary, we have reported the fabrication of OLEDs with various emission colours. In

particular, we showed based on a wide-band-gap host polymer; namely polyfluorene (F8=PFO), and with a careful doping/blending protocol of the host layer with different emissive materials, we can effectively fabricate a full range of colourful OLEDs and since this fabrication procedure is reasonably accessible, it seems solution-based



Figure 2. Illumination from the white OLED, based on the combination of prime blue emission and the complementary orange emission.

OLEDs are ideal candidates for the future solid-state lighting application. Novel results of present paper discloses that employing Lumogen F Red as the dopant in PFO polymer matrix yields in bright magenta electroluminescence from the OLED whereas doping the MEH-PPV with Lumogen causes strong red emission from the diode. Also, we fabricated a white OLED by blending the PFO with F8BT and MEH-PPV with weight ratio of (94:5:1). This yields a warm white-emitting diode with CCT of 5565 K and CIE coordinate equals to (0.33, 0.36).

## References

- [1] A. Köhler, and H. Bässler, "Triplet states in organic semiconductors," *Materials Science and Engineering: R: Reports*, vol. 66, no. 4, pp. 71-109, 2009.
- [2] M.-R. Fathollahi, F. Akbari Boroumand, F. Raissi, and M.-J. Sharifi, "Quantitative characterization of carrier injection across metal-organic interfaces using Bardeen theory," *Organic Electronics*, vol. 13, no. 5, pp. 905-913, 2012.
- [3] S. Xiao, M. Nguyen, X. Gong, Y. Cao, H. Wu, D. Moses, and A. Heeger, "Stabilization of semiconducting polymers with silsesquioxane," *Advanced Functional Materials*, vol. 13, no. 1, pp. 25-29, 2003.
- [4] S.-R. Tseng, H.-F. Meng, K.-C. Lee, and S.-F. Horng, "Multilayer polymer light-emitting diodes by blade coating method," *Applied Physics Letters*, vol. 93, no. 15, pp. 153308, 2008.