



# Tunable microcavities in organic light-emitting diodes by way of low-refractive-index polymer doping

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## ABSTRACT

A method for enhancing the light out-coupling efficiency of organic light-emitting devices (OLEDs) has been demonstrated by blending a low-refractive-index polymer, poly(2,2,3,3,3-pentafluoropropyl methacrylate) (PPFPMA), into the emission layer. The resonant wavelength of the weak microcavity devices blueshifted accompanied with a decrease in refractive indices of the light-emitting layers after the addition of PPFPMA. Stronger directed emission toward the surface normal was obtained when the resonant wavelength became closer to the peak wavelength of intrinsic emission spectrum of the organic emitters. The luminous efficiency of the devices was enhanced by more than 20%. The results suggest that the microcavity properties of the OLEDs can be tunable through blending low-refractive-index materials.

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## 1. Introduction

Organic light-emitting diodes (OLEDs) are playing an increasingly conspicuous role in the next generation display and solid-state lighting technologies because of their high efficiencies, low cost, and their potential to act as large-area and flexible devices [1–3]. Meanwhile, nearly 100% internal quantum efficiencies have been realized by doping phosphorescent molecules to harvest triplet excitons [4,5]. Nevertheless, because of the mismatch of the refractive indices, a large proportion of the generated photons is trapped in the devices and the light out-coupling efficiency ( $\eta_{out}$ ) of conventional OLEDs is limited at *ca.* 20% [6]. Therefore, substantial enhancement in the light extraction efficiencies will be necessary if we are to further improve the device efficiencies.

Many approaches, including using high-refractive-index substrates [3,7], microlense arrays [8,9], photonic crystals [10], and plasmonic nanostructures [11], have been studied to enhance  $\eta_{out}$ . Meanwhile, the conventional structure of OLEDs naturally behaves as a weak microcavity because the dimension of their functional layers is similar to the wavelength of the emitting photons. While both electrodes exhibit high reflectivities, strong microcavity effects can be observed. Therefore, the microcavity effects redistribute the optical modes in the devices, and the  $\eta_{out}$  can be enhanced along the normal emission direction under certain conditions [12–18]. For instance, better light extraction can be achieved by setting the resonant wavelength of the cavity near the peak wavelength ( $\lambda_{em}$ ) of the intrinsic emission spectrum [ $S(\lambda)$ ] of the organic emitters [13,14]. To the best of our knowledge, however, most studies adjust the optical cavity length ( $L$ ) of the microcavities through altering the thickness ( $d$ ) of the organic layers [17,18]. No report tunes the values of  $L$ , which are equal to the products of the refractive indices [ $n(\lambda)$ ] and  $d$ , by altering the refractive indices of the

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emission layers. Adjusting the values of  $L$  through varying the thicknesses of the active layers, however, inevitably affects other electrical properties, such as the internal electrical field strength and the width and location of the recombination zone, resulting in a rather complicated situation for structure designs [19]. An alternative way is to change the refractive indices of the emission layers. In this work, we demonstrated a simple, effective method for enhancing  $\eta_{out}$  of polymer light-emitting diodes (PLEDs) by blending a low-refractive-index polymer, poly(2,2,3,3,3-pentafluoropropyl methacrylate) (PPFPMA), into the emission layer. The microcavity effects of the devices can be optimized by tuning the  $n(\lambda)$  of the emission layer with the amount of PFPMA. As a result, the luminous efficiency incorporating PFPMA was enhanced by approximately 20%.

## 2. Experimental

Fig. 1 shows the device structure and the chemical structures of the materials used in this work. For device fabrication, indium tin oxide (ITO)-coated glass substrates were cleaned with detergent, followed by sequential sonication in deionized water, acetone, and isopropanol. They were then dried in an oven overnight. Prior to use, the substrates were further treated with UV-ozone. Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) was spin coated onto the ITO substrates and then baked at 120 °C for 1 h. The emission layer comprised poly(vinylcarbazole) (PVK), 2-(4-biphenyl)-5-(4-tert-butyl-phenyl)-1,3,4-oxadiazole (PBD), and tris[2-(4-tolyl)phenylpyridine]iridium [Ir(mppy)<sub>3</sub>] at a weight ratio of PVK:PBD:Ir(mppy)<sub>3</sub> = 70:29:1 [20–22]. The emission layer was deposited on top of the PEDOT:PSS layer from a solution of 1,2-dichlorobenzene inside a N<sub>2</sub>-filled glovebox. The polymer film was then annealed at 80 °C for 30 min. To complete the devices, a cathode comprising 30 nm Ca and 100 nm Al was thermally deposited under a vacuum of ca.  $5 \times 10^{-6}$  Torr. The control devices were defined as those prepared without PFPMA. For the devices containing PFPMA, the low-refractive-index polymer was blended into the emission layer at various weight of 0.1–0.5% (w/w). For example, 0.1% PFPMA indicated that the weight ratio of the components in the emissive layer was PVK:PBD:Ir(mppy)<sub>3</sub>:PFPMA = 70:29:1:0.1. The refractive index of PFPMA was 1.140, which was measured by using an ellipsometer (Sopra GES-5). The thicknesses of the

emission layer prepared with and without the addition of PFPMA were precisely controlled and all of them were almost identical (ca. 70 nm); the thicknesses were obtained by using an atomic force microscope (AFM). The electrical characteristics of the PLEDs were recorded by a Keithley 2400 source measure unit. The brightness and the electroluminescent (EL) spectra were measured by using a PR650 SpectraScan colorimeter. The photoluminescent (PL) spectra were obtained using an Ocean Optics HR4000CG-UV-NIR spectrometer. A N<sub>2</sub>-laser was used as the light source. The reflectivity spectra were obtained using a Perkin Elmer UV-Vis Lambda 650 spectrometer. The viewing angle dependence of luminance was determined using a ConoScope™ (Autronic-Melchers, GmbH), where a measuring cone of 80° over an azimuthal angle range of 360° could be obtained. The thin film morphology was analyzed using a DI 3100 series AFM.

## 3. Results and discussion

Fig. 2(a) shows the current density–brightness–voltage ( $J$ – $B$ – $V$ ) curves of the PLEDs containing various amount of PFPMA. The turn-on voltage ( $V_T$ ), defined for a luminance of 0.1 cd/m<sup>2</sup>, was 4.6 V for the control device. The devices prepared with 0.1% and 0.2% PFPMA exhibited almost identical electrical characteristics, suggesting that the addition of PFPMA did not affect the electrical properties of the devices significantly. However, as the concentration of PFPMA was increased beyond 0.2%, we could see that the current density started to decrease obviously. The values of  $V_T$  became 4.8 V for the devices containing 0.3% PFPMA, respectively. The increase in  $V_T$  could be attributed to the insulating nature of PFPMA. The electrical properties of the devices fabricated under different conditions are summarized in Table 1. Fig. 2(b) and (c) display the luminance efficiency–current density ( $\eta$ – $J$ ) and power efficiency–current density ( $L_p$ – $J$ ) curves of the PLEDs prepared with and without PFPMA. The maximum  $\eta$  and  $L_p$  of the control device were 20.5 cd/A and 9.3 lm/W, respectively. After adding PFPMA into the emission layer, substantial enhancement in the luminance efficiency and power efficiency could be obtained. Compared with the control device, the peak  $\eta$  of the device containing 0.2% PFPMA increased approximately by 22% to the 25.1 cd/A. Meanwhile, the  $L_p$  was also enhanced to 11.4 lm/W while the driving voltage was not significantly affected by adding

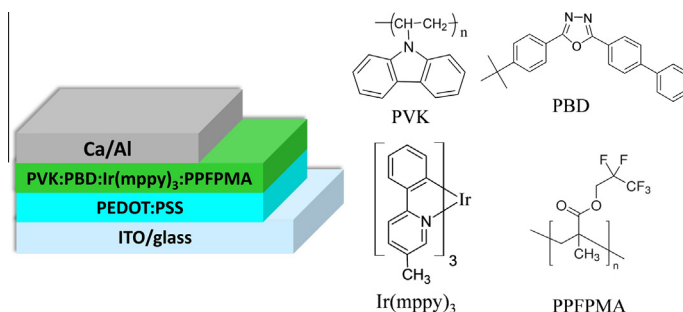


Fig. 1. Device structure and chemical structures of the organic materials used in this work.

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