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Correlation between electroluminescence, charge transport and photophysical properties of polymer blends

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ABSTRACT

A comprehensive study of the electrical and optical performance of white polymer light-emitting diodes (PLEDs) based on blends of polyfluorene (PFO) and poly (2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) is reported in this paper. MEH-PPV concentration was varied from 0 to 15 wt.%. Maximum luminance and current efficiency was obtained at 2.0 wt.% of MEH-PPV. To understand the origin of this enhancement, electrical and emission properties of the blends were investigated over the entire range of MEH-PPV concentration. Charge carrier mobility was estimated using an electroluminescent transient technique (ELT) which was found to be increased at the lower concentrations of MEH-PPV, however, at higher concentration it was lower than that of pristine PFO.The highest carrier mobility of $2.6 \times 10^{-5}\,cm^2\,v^{-1}\,s^{-1}$ was observed at 1.2 wt.% MEH-PPV in PFO. Fowler–Nordheim (F-N) model was used to estimate the potential barrier height at anode interface. It was found that the blending with MEH-PPV reduced the hole injection barrier in the PLEDs up to 5.0 wt.% of MEH-PPV. At concentrations greater than 5.0 wt.%, barrier values started increasing again. Correlating these properties with the current density-voltage curves of PLED, it was observed that current density in the device increased up to 5.0 wt.% of MEH-PPV, this was in good agreement with the behavior of barrier height with MEH-PPV concentration. Therefore, it was inferred that reduction in the barrier height is the dominant effect in improving the luminance of blend based PLEDs. However, it was also observed that maximum current efficiency (1.92 cd/A) occurred at 2.0 wt.% of MEH-PPV blend in PFO with Commission Internationale de l'Enclairage (CIE) coordinates close to white light (0.33, 0.33). This meant that emissive characteristics of the blends were also changing with wt.% of MEH-PPV. To investigate this, photophysical properties, photoluminescence (PL) and photoluminescence quantum efficiency (PLQE) of the binary blends were also studied. It was observed that blending lead to lower PLQE and hence would lead to decrease in the current efficiency with MEH-PPV wt.%, if energy transfer from host to guest was the only mechanism for exciton formation in blend based PLEDs. However, comparing the electroluminescence (EL) and PL behavior of blends, enhancement in current efficiency was observed due to direct excitation of MEH-PPV in PLEDs. These two opposite effects, reduction in PLQE of the blend and direct excitation of MEH-PPV, resulted in best current efficiency of PLED at 2.0 wt.%.

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1. Introduction

It has been shown that performance of polymer electronic devices, such as light emitting diodes [1–6], thin film transistors [7–11] and solar cells [12–15], improves by blending two conjugated polymers. In PLEDs, blending of two polymers enables tuning of the CIE coordinates of light emission and enhances efficiency. Polyfluorene (PFO) and poly (2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) are two well known conjugated polymers extensively used in fabricating single layer white polymer light emitting diodes. Polyfluorene is known to be highly fluorescent bluish- green emitting polymer having hole mobility ($\sim 10^{-4}$–$10^{-5}\,cm^2\,v^{-1}\,s^{-1}$) [16]. It also shows high photoluminescence quantum efficiency (~0.45–0.55) with good thermal and chemical stability [17,18], whereas MEH-PPV is an orange–red emitting polymer having good solubility and luminescent properties. In blend PLEDs, EL from both the polymers is obtained due to partial energy transfer from the host (PFO having larger band gap)

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to guest polymer (MEH-PPV having smaller band gap) resulting in white emission.

Although there have been several reports of white PLED using PFO and MEH-PPV, there is a lack of understanding regarding the correlation between electrical and optical properties of these blends over a large range of MEH-PPV concentrations. Shen et al. studied the characteristics PLEDs by blending 0.6 wt.% of MEH-PPV with PFO, and achieved current efficiency of 2.3 cd/A with CIE coordinate of (0.33, 0.31) at 7 V [19]. Similarly, Alam et al. studied different properties of the blends of these two polymers for 5.0–60 wt.% of MEH-PPV concentrations [20]. Ternary blends of these two polymers with inert PMMA have also been investigated by Ananthakrishnan et al. for a fixed concentration [21]. Owing to high hole injection barrier (0.6–0.7 eV) from the anode side, low device efficiency of the blended white PLED is attributed to an imbalance in charge injection. To overcome this problem, many researchers have tried a hole transport layer (HTL)/electron blocking layer (EBL) at anode side or electron transport layer (ETL)/hole blocking layer (HBL) at cathode side or both by making multilayer diode structure [22–26]. Most of them have used MEH-PPV concentration below 2.0 wt.% [22–25], some have looked at a concentration up to 3.0 wt.% [19,26] and there was only one study reporting results in the range 5.0–60 wt.% [20]. The best efficiency was reported in the range 0.5–1.0 wt.% of MEH-PPV [19,22–26] except when the investigated range was 5.0–60 wt.% (In that study, the best efficiency was obtained at 5.0 wt.% as expected) [20]. The efficiency improvement in the polymer blends has been attributed to several effects – improvement in carrier mobility, trapping of charges, improvement in emission efficiency. However, it is not obvious what is the role of each effect.

In this paper, we have systematically investigated electroluminescent and photophysical properties of the blends of these two conjugated polymers by varying MEH-PPV concentrations from 0 wt.% to 15 wt.%. The white PLED having 2.0 wt.% of MEH-PPV showed the maximum efficiency of 1.92 cd A^{-1} with CIE coordinate value of (0.31, 0.32) at the injection current density of 69 mA/cm^2. To understand whether improvement in electrical or optical properties of materials was leading to this behavior, we have measured the effective mobility of blends, barrier height at the anode, energy transfer characteristics in PL and EL, and PLQE of the blends as a function of wt.% MEH-PPV. Correlation of these properties with the PLEDs device performance gave insight into why 2.0 wt.% MEH-PPV in PFO resulted in maximum efficiency. Another contribution of this paper is to correlate several electrical and optical properties of blends over the entire concentration range, this has allowed us to draw insights that will not be possible otherwise. This work advances the understanding the basic phenomenon underlying PFO: MEH-PPV based white PLEDs and will be useful in improving them further.

2. Experimental

PFO and MEH-PPV were purchased from American Dye Source Inc. and Sigma-Aldrich respectively. The PLEDs were made on indium tin oxide (ITO) coated glass substrate. ITO thickness was 150 nm and sheet resistance 20 Ω □$^{-1}$. Patterned ITO substrates were cleaned ultrasonically in deionized (DI) water, acetone, isopropyl alcohol (IPA) sequentially and dried in a vacuum oven. Then a layer of positive photo resist was coated and pixels were defined using photolithography technique [27]. The pixelated substrates were placed to ozone treatment for 20 min to enhance the work function of anode. A filtered poly-3,4-ethylenedioxy-thiophene-poly-styrene sulfonate (PEDOT: PSS) solution was spin coated on the pixellized ITO substrate and dried in nitrogen furnace at the temperature of 120 °C for 1 h. The blend of PFO and MEH-PPV solution were prepared in chloroform and toluene (50:50) mixture and spin coated on top of the PEDOT: PSS hole injection layer and subsequently annealed in a vacuum at the pressure of 5×10^{-6} mbar and temperature 125 °C for 1 h. MEH-PPV concentration was varied from (0.8–15 wt.%). Finally, calcium (Ca) and aluminum (Al) thin films were deposited as a cathode on the emitting layer in the vacuum of 2×10^{-6} mbar. The resulting device structure was Glass/ITO (150 nm)/PEDOT: PSS (40 nm)/PFO: MEH-PPV (0.8–15 wt.%)/Ca (20 nm)/Al (120 nm). The square shaped devices were encapsulated inside the nitrogen glove box. The emissive area of the device was 0.16 cm^2. Fig. 1 shows the device structure and energy band diagram of PLEDs having configuration ITO (150 nm)/PEDOT: PSS (40 nm)/PFO: MEH-PPV (0–15 wt.%) (65 nm)/Ca (15 nm)/Al (120 nm).

For electrical and optical characterization of fabricated devices, current-voltage-luminance (I-V-L) characteristics, electroluminescence (EL) spectra and CIE color coordinates were measured using CS-1000 spectro-radiometer (Konica Minolta) and Keithley 2400 source meter. All the measurements were carried out on the encapsulated devices at room temperature under ambient conditions. The details of ELT technique for determining mobility have been reported elsewhere [28]. Horiba Jobin Yvon FluoroLog 3 spectrofluorometer and Lamda −750 UV/Vis/NIR spectrophotometer were used to obtain the photoluminescence (PL) and optical absorption spectra of the blend thin films respectively. Alpha Step profilometer (KLA Tencor) was used to measure the thickness of the films.

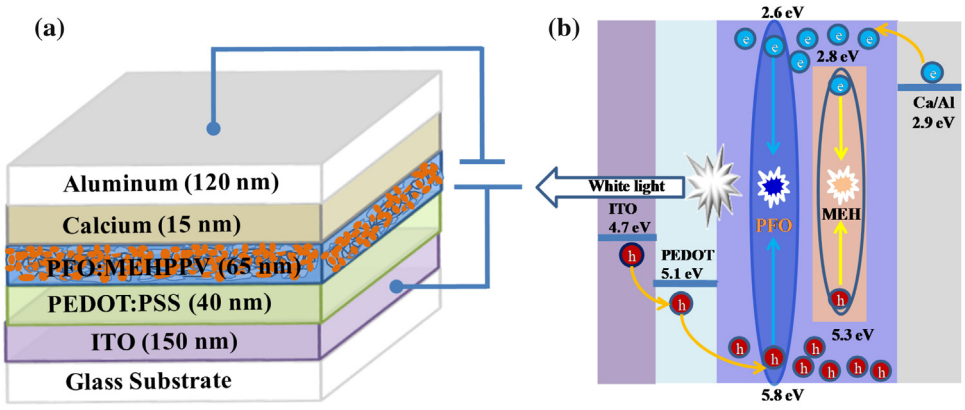

Fig. 1. Schematic of (a) device structure (b) energy level diagram of white PLEDs. MEH-PPV wt.% was varied from 0 to 15 wt.% in the active layer.

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