Influence of emissive layer thickness on electrical characteristics of polyfluorene copolymer based polymer light emitting diodes

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Abstract. Polymer light emitting diodes (PLEDs) with a device configuration of ITO/ PEDOT:PSS/ PFONPN01 [Poly[2,7-(9,9'-dioctylfluorene)-co-N-phenyl-1,8-naphthalimide (99:01)]/LiF/Al have been fabricated by varying the emissive layer (EML) thickness (40/65/80/130 nm) and the influence of EML thickness on the electrical characteristics of PLED has been studied. PLED can be modelled as a simple combination of resistors and capacitors. The impedance spectroscopy analysis showed that the devices with different EML thickness had different values of parallel resistance (R_P) and the parallel capacitance (C_P). The impedance of the devices is found to increase with increasing EML thickness resulting in an increase in the driving voltage. The device with an emissive layer thickness of 80nm, spin coated from a solution of concentration 15 mg/mL is found to give the best device performance with a maximum brightness value of 5226 cd/m^2.

1. Introduction

Polymer light-emitting diodes (PLEDs) have been a major area of attraction for the researcher over past few decades due to its potential application in the field of flat-panel displays and solid state lighting sources due to their low cost, ease of fabrication, brightness, speed, wide viewing angle, low power consumption and contrast [1-5]. The performances of such device are critically dependent on the properties of both emissive materials and their processing parameters. Among PLEDs, stable and pure blue light emitting devices is of utmost importance as it plays a vital role in realizing full color displays and high quality white light [6,7]. Polyfluorenes (PFs) are well-known emissive material for blue polymer light emitting diodes (PLEDs) due to their high luminescence efficiency, excellent thermal stability, charge transport property, good processability and color tunability [8,9]. To achieve high efficiency for a given polymer, it is necessary to balance and confine electrons and holes inside the emissive layer [10]. Unfortunately, in case of PFs the hole mobility is much larger than the electron mobility. However, adding electron withdrawing moieties like 1,8 napthalamide (NPN) into the polyfluorene main chain can significantly enhance its electron transporting properties[11,12].

In this paper, we report the fabrication of blue PLEDs using PFONPN01 [Poly[2,7-(9,9'-dioctylfluorene)-co-N-phenyl-1,8-naphthalimide (99:01)] as emissive layer (EML) and the effect of EML thickness on the electrical parametres of PLEDs. The impedance of the devices is found to

increase with increasing EML thickness resulting in an increase in the driving voltage. The device with an EML thickness of 80nm, spin coated from a solution of concentration 15 mg/mL is found to give the best device performance with a maximum brightness value of 5226 cd/m^2.

2. Experimental Section

2.1. *Chemicals and substrate*
Indium tin oxide (ITO) coated glass substrate (sheet resistance 15 Ω/sq.), Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT: PSS),   Lithium fluoride (LiF) and Aluminum wire (99.999% purity) were purchased from Sigma Aldrich and used as received. PFONPN01 used as emissive layer was synthesized using previously reported method [11] and its chemical structure is shown in figure 1(a).

2.2. *Characterization details*
The thin film of PEDOT:PSS and PFONPN01 were deposited on ITO coated glass substrate by spin coating technique using Laurell and Spin 150 spin coater outside and inside the Jacomax glove box respectively. The Veeco Dektak 150 Surface Profilometer was used to measure thicknesses of the thin films. All current density vs voltage (J-V) characteristics of the fabricated devices were measured by Keithley-2400 digital source meter. The luminance values of the PLEDs were measured using LCS 100 integrated sphere. CH 680 Instrument was used to measure the impedance spectroscopies of all the devices. All the device characterizations were carried out inside glove box under inert gas environment.

2.3. *Device Fabrication Method*
PLEDs were fabricated using PFONPN01 copolymer as emissive layers. The PLED device configuration is composed of a pre-cleaned and pre-patterned ITO as the transparent anode. The ITO surface was ultrasonically agitated and cleaned in 2% detergent, acetone and isopropanol, each for ten minutes. After cleaning, an 40 nm thin layer of poly (3,4-ethylendioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) as a hole injecting layer was spin-coated at 3000 rpm for 60 seconds, then baked for 15 minutes in an argon environment at 130°C. The copolymers were dissolved in chloroform at four different concentration (5/10/15/20 mg/mL) and spin-coated above the PEDOT:PSS layer under an ambient atmosphere and thermally treated at 80°C for 15 minutes for the removal of residual solvent. The film thickness of the emissive layer was~ 80 nm. Finally, LiF and aluminum was thermally evaporated at a rate of 0.1 Å s^{-1} and 10 Å s^{-1} respectively at a base pressure of 10^{-6} mbar to form the cathode electrode. The emissive area of the diodes was 20 mm^2. The schematic device structure is shown in figure 1(b).
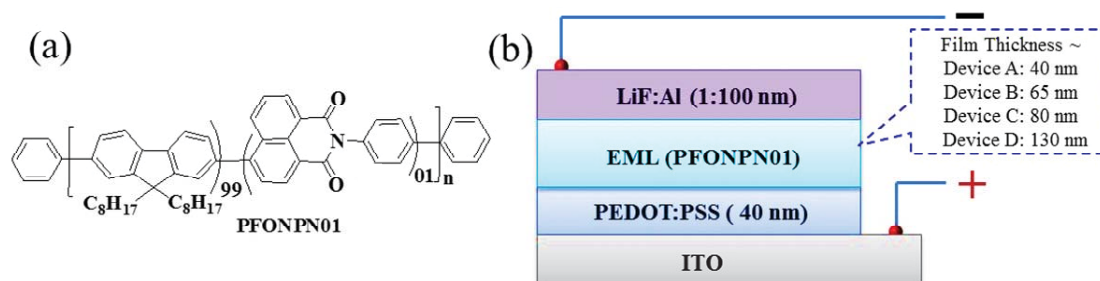

Figure 1. (a) Chemical structure of PFONPN01 (b) Schematic device structure

3. Result and Discussion

The impedance of the fabricated devices with different EML thicknesses were measured using CH680 instrument in the range of 10 Hz to 1 MHz at zero dc bias voltage. The amplitude of the ac test signal was 50 mV. The results of the measurements were analyzed based on the complex impedance equation:

$$Z = \frac{1}{Y} = R + jX = R + \frac{1}{jwC} = Z' + jZ''$$

where Z, Y , R, X and C are the impedance, admittance, resistance, reactance, and capacitance of the device, respectively.
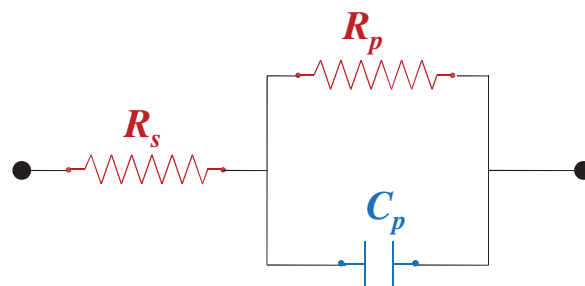

Figure 2. Equivalent circuit model for PLED

Table 1. Comparison of the resistance and the capacitance values for the devices with different emissive layer thickness

Device Name	EML thickness (nm)	R_S (ohm)	R_p (ohm)	C_p (F)
A	40	41.32	1567	1.18E-8
B	65	43.58	2058	9.862E-9
C	80	44.62	2293	6.318E-9
D	130	39.2	4935	4.314E-9

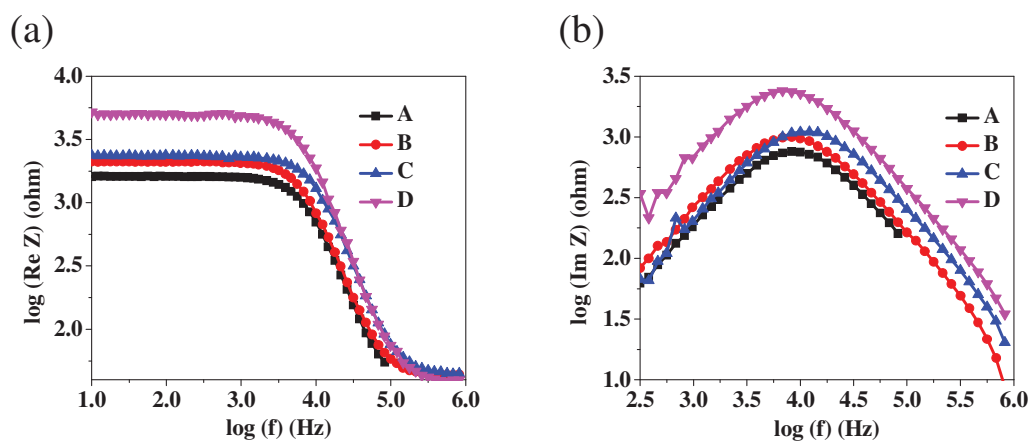(a)   (b)

Figure 3. Frequency-dependent (a) real and (b) imaginary parts of the impedance of the PLEDs at 0 V.

Figures 3(a) and 3(b) show the frequency dependent real and imaginary parts of the impedance of devices with different EML thickness. Both real and imaginary parts of the impedance are changed with the EML thickness of the devices. The real and imaginary part of the impedance of device D increases significantly and can be attributed to the high parallel resistance and low parallel capacitance of the emissive layer due to its large thickness and high roughness which can be due to the less solubility of the polymer at high concentration.

The values of the contact resistance (R_S), the parallel resistance (R_P), and the parallel capacitances (C_P) extracted from the frequency-dependent real and imaginary parts of the impedance of the devices with different EML thickness are summarized in Table 1.
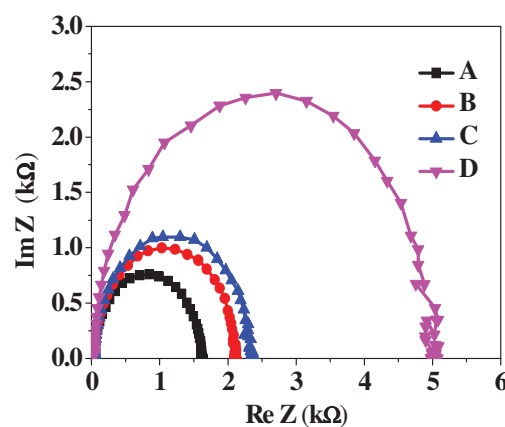

Figure 4. Impedance Cole-Cole plots for the devices with different emissive layer thickness

Figure 4 shows the impedance Cole-Cole plot for the devices with different EML thickness. The horizontal and the vertical axes represent the real (Z') and the imaginary (Z") parts of the impedance of the devices, respectively. All the devices have a very low series resistance suggesting efficient carrier injection from electrode as well as good interface charge transfer. However, as the thickness increases, the shunt resistance of the devices increases. As a result the impedance increases thereby increasing the driving voltage of the devices.
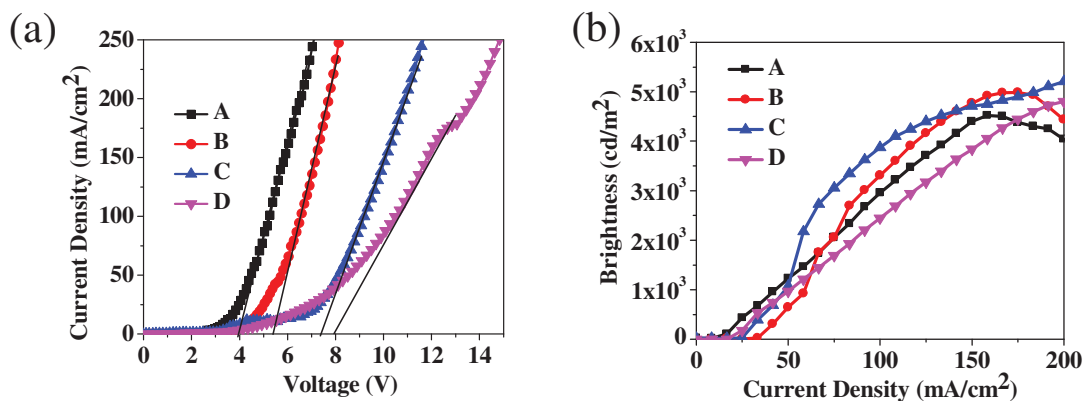

Figure 5. (a) Current density vs voltage (J-V) characteristics and (b) brightness vs current density (B-J) characteristics plot for the devices with different EML thicknesses

Figure 5(a) and 5(b) shows the current density vs voltage (J-V) characteristics and brightness vs current density (B-J) characteristics plot for the devices with different EML thicknesses. The driving voltage of the devices is found to increase with the increase in the EML thickness. However, in case of brightness vs current density curve, it has been noticed that the brightness is maximum for the device C having an EML thickness of approximately 80 nm. The key device properties of all the devices are listed in table 2. The turn on voltages of the PLEDs were calculated by extrapolating the straight portion of the J-V curves as shown in figure 5 (a) and is the voltage at which the device starts emitting light. It is observed that the turn-on-voltage varies from ~4 to 7.9 V when the EML thickness increases from 40 to 130 nm.

Table 2. Key device properties of the devices

Device Name	EML Thickness (nm)	Turn on Voltage (V)	Maximum Brightness (cd/m^2)
A	40	3.95	4516
B	65	5.40	5019
C	80	7.35	5225
D	130	7.93	4791

4. Conclusion

In conclusion, we have demonstrated the influence of EML thickness on the electrical characteristics of polymer light emitting diode (PLED). The impedance spectroscopy analysis showed that the devices with different EML thickness had different values of parallel resistance (R_P) and the parallel capacitance (C_P). The impedance of the devices is found to increase with increasing EML thickness resulting in an increase in the driving voltage. The device with an emissive layer thickness of 80nm, spin coated from a solution of concentration 15 mg/mL is found to give the best device performance with a maximum brightness value of 5226 cd/m^2.

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