



Electroluminescence enhancement of glass/ITO/PEDOT:PSS/MEH-PPV/PEDOT:PSS/Al OLED by thermal annealing

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ABSTRACT

Manufacturing of glass/ITO/PEDOT:PSS/MEH-PPV/PEDOT:PSS/Al organic light emitting diode (OLED) by depositing PEDOT:PSS/MEH-PPV/PEDOT:PSS using spin coating has been reported. The roles of PEDOT:PSS in the structure have been reported. It allows transportation of holes from ITO to the highest occupied molecular orbit (HOMO) of MEH-PPV. In additions, it allows transportation of electrons from Al to lowest unoccupied molecular orbit (LUMO) of MEH-PPV. Further, it confines electrons in the LUMO of MEH-PPV due to the higher barrier of PEDOT:PSS of LUMO. The effect of thermal annealing on the current-voltage curve as well as on the electroluminescence intensity has been reported. The results show that the current increased from 25 mA to 52 mA at 7 V, when the sample was thermally annealed at 150 °C. Such enhancement in electrical injection leads to enhancement of the electroluminescence to a factor of 4.7 at the peak luminescence wavelength (~592 nm). Reasons for electroluminescence improvement caused by thermal annealing have been proposed.

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1. Introduction

Organic semiconductors have been studied extensively because of their various potential optoelectronic applications like organic light emitting diodes (OLEDs) [1–3], field effect transistors [4,5], solar cells [6–9], sensors [10], biosensors [11], and photodetectors [12,13]. Further, recent realization of optically pumped polymer-based lasers [14–19] has motivated extensive studies of conjugated polymers as potential candidates for electrically pumped lasers [20–23]. The attractive attributes of OLEDs such as ease of fabrication, low manufacturing cost, flexibility, and light weight have grown interest both in academic and industrial fields. These advantages provide applications in various optoelectronic devices such as car lighting, mobile phone, computer monitors, digital camera, and TV monitors. The layer structure of OLEDs is composed of a stack of several layers. The main challenges in OLEDs are two issues, namely the device lifetime and efficiency. The organic semiconductor materials can be degraded by interacting with oxygen in the environment. Hence it is necessary to use a cap layer to prevent such degradation. Another important issue is the electroluminescence efficiency that can be extracted from the device. The

efficiency improvement needs layer structure design that provides efficient current injection, in other words low resistive layer structure should be used. Several effects have been investigated in order to improve the extracted electroluminescence efficiency from the device. These effects include thermal treatment, interlayer, and blending effects. The effect of the thermal treatment conditions during fabrication of the EL device on quantum efficiency have been reported by Lee and Park [24]. MEH-PPV has been used as an emissive amorphous polymer. Devices have been annealed twice, before and after Al deposition. They have found that the EL output decreases as the pre-deposition annealing temperature increases and the EL device with only post-deposition annealing is the most efficient one. They have concluded that the post-deposition annealing was the most efficient method for improving the EL characteristics as it has altered the polymer/Al interface. Pre-deposition annealing can increase the maximum optical output due to increased thermal endurance. However, the quantum yield is lowered. For blending effect, Cea et al. have improved the EL characteristics of MEH-PPV-based PLEDs by incorporation of 2,5-bis [5-(4-*tert*-butylphenyl)-2-pyridyl]- 1,3,4-oxadiazole (PPyD) as an electron transport material into MEH-PPV. They have concluded that the single mixed layer devices showed higher external quantum efficiencies than dual layer devices, with values up to 0.26% has been achieved [25]. For interlayer effect, Guo et al. [26] have shown the effect of different interfacial layer on the luminescence

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efficiency of PLED based on MEH-PPV with structure ITO/PEDOT:PSS/MEH-PPV/poly(ethylene oxide) PEO/LiF/Al. They have reported that the EL efficiency for the device with PEO/Al cathode was 1.50 cd/A, which is two orders of magnitude higher than that of the Al cathode device, 0.017 cd/A, and was increased from 1.34 cd/A (Ca/Al cathode) to 2.48 cd/A (PEO/Ca/Al cathode). The luminescence efficiency was 2.01 cd/A for the device applying LiF/Al cathode. The efficiency has been achieved as high as 4.96 cd/A with the PEO/LiF/Al cathode. They have concluded that formation of specific interaction and reaction at the PEO layer during the cathode deposition, lowering the injection barrier height or creating a tunneling path for minority carriers, which results in the enhanced injection of electrons.

The conjugated polymers that have been used here are Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and Poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene-vinylene] (MEH-PPV). The function of the PEDOT:PSS is improving carrier transportation and MEH-PPV is the emission unit in the device. PEDOT:PSS is a mixture of two polymers. The first one is poly(3,4-ethylenedioxythiophene) or PEDOT, which is a conjugated polymer and carries a positive charge. The other polymer is sodium polystyrene sulfonate or PSS which is a sulfonated polystyrene. Part of the sulfonate groups are deprotonated and carry a negative charge. PEDOT:PSS is one of the most promising materials due to its excellent conductivity, solubility, stability, and optical characteristics [27]. MEH-PPV is one of Poly(*p*-phenylene vinylene) (PPV) derivative that and represents an important family of *p*-conjugated materials used for device production. It offers the advantages of being soluble in organic solvents, good environmental stability, easy conductivity control and the possibility of low-cost and large-area fabrication.

In this report, the effect of post annealing on improving the extracted electroluminescence intensity from glass/ITO/PEDOT:PSS/MEH-PPV/PEDOT:PSS/Al organic light emitting diode has been investigated. The role of PEDOT:PSS as an interlayer has been reported. The mechanism of post annealing on improving the efficiency has been discussed.

2. Experimental section

The materials used in this work are Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV), purchased from Sigma–Aldrich, UK with an average molecular weight M_n from 70,000 to 100,000 and used as received without further purification, High-conductivity Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) as an aqueous solution and indium tin oxide coated glass substrate (ITO) ($\text{In}_2\text{O}_3:\text{SnO}_2$) (surface resistivity 8–12 Ω/sq) purchased from Sigma–Aldrich, and toluene (analytical grade, ACS), purchased from Scharlab, SL (Barcelona, Spain).

The substrate surfaces were thoroughly cleaned through the following steps: Cleaning with warm acetone by ultrasonic cleaner for 10 min at 50 °C, followed by warm isopropyl alcohol for 10 min in ultrasonic cleaner at 50 °C, then the substrate was rinsed in distilled water at least 3 times. Then, the solvent residues were removed by immersing the substrates in boiling distilled water for 10 min. Finally, the substrates were dried in a hot plate for 10 min to be ready for the deposition process.

A transparent film of PEDOT:PSS with approximately 80 nm in thickness was obtained on the substrate by spin-coating aqueous solution at 4000 rpm for 30 s. To increase the conductivity of the PEDOT:PSS film, the sample was baked at 120 °C for 15 min by using Multi-stage Programmable Furnace (Neytech, VULCAN model 3–550) in air. MEH-PPV in toluene at a concentration of 4 mg mL^{−1} was sonicated in ultrasonic bath at 50 °C for at least 2 h until the

polymer is fully dissolved. The MEH-PPV solution was spin coated onto ITO/PEDOT:PSS film at 2000 rpm for 30 s. Then, the film was baked at 100 °C for 5 min to remove the solvent completely by using Multi-stage Programmable Furnace (Neytech, VULCAN model 3–550) in air, with film thickness approximately 46.5 ± 1.4 nm as measured by optical surface profilometer (ZYGO Maxim-GP200 profilometer). Another layer of PEDOT:PSS has been deposited using the same condition mentioned above. The last layer of Aluminum (Al) electrodes were deposited by conventional thermal evaporation technique at a base pressure of 10^{-7} Torr onto the organic layers through a shadow mask using a high vacuum coating unit (Edwards Co. model E306A, England). The electrode active area was 7.1 mm² (with circular patterns). After evaporation, the chamber was allowed to cool down for ~30 min and the samples were removed and used for testing.

The absorption spectra of films were measured using a single beam UV-VIS Spectrophotometer (Camspec model M501 UV-VIS). The photoluminescence was measured using JASCO FP-6300 spectrofluorometer, with monochromatized Xe lamp used as the source for PL excitation. The excitation wavelength was 500 nm. The current-voltage (*I*–*V*) characteristics of OLED devices (without annealing and with annealing) were measured using a Keithley 2401. The electroluminescence (EL) measurements were carried out by using ScopeTek Digital Fiber Spectrometer model STDFSM3666 (USB 2.0) and Keithley 2401 as electrical excitation source.

Post-deposition thermal annealing for the manufactured devices at 150 °C for 1 h was carried out.

3. Results and discussion

Fig. 1 demonstrates a sketch of the MEH-PPV-based OLED. ITO and Al act as positive and negative electrodes, respectively, as the work function of ITO and Al are 4.7 and 4.3 eV, respectively. PEDOT:PSS layers were spin coated below and above MEH-PPV layer. The function of PEDOT:PSS layer is twofold: Not only does it provide transportation of carriers (electrons and holes) to the emissive layer (and thus improves electrical properties of the device) but also it protects the emissive layer from degradation caused by oxidation (and thus it improves the lifetime of the device [28,29]. In different studies PEDOT:PSS/Al has been used in optoelectronic applications [30–32]. A work reported by H.Y. Jeong et al. was published to consider the interaction at PEDOT:PSS/Al interface in more details [32]. According to [32], aluminum atoms can attach to the PEDOT:PSS film, and react with organic molecules containing carbon, oxygen and sulfur, leading to Al^{3+} . The oxidation state (Al^{3+}) strongly attracts the insulating PSS chains in the active organic film. In additions, Al can penetrate into PEDOT:PSS layer. The absorbance and fluorescence spectra of MEH-PPV with

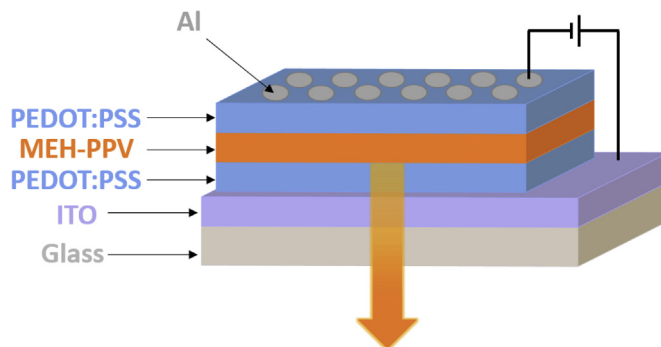


Fig. 1. Sketch demonstrating the layer structure of the device.

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