

Effect of Pressure and MoO₃ Hole Injection Layer on the Current-Voltage Characteristics of Organic Light Emitting Diodes.

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ABSTRACT

We examine the fundamental operation of an Organic Light Emitting Device with emphasis laid on the Hole Transport Layer (HTL) and the optoelectronic properties of the other layers that make up the device. Investigation of the adhesion properties together with surface morphology, electrical and optical characterization of the different layers of the device was carried out. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT: PSS) was used as the conventional HTL material in the first case. This yields the reference device or system under studies. In the second case, PEDOT: PSS was replaced by an inorganic material, molybdenum trioxide (MoO₃). The device performance in case two (2) revealed an improvement in performance. A couple of deposition techniques were examined together with the analysis of their effect on the resultant device properties. With the aid of theoretical models, we quantified the results obtained in terms of average pull-off forces and corresponding adhesion energies. The Derjaguin-Muller-Toporov model was utilized to model the adhesion energies between interfaces of adjacent layers of the device. Results that delineate modeling of charge transport across device interfaces are shown including the effects of pressure on the device optoelectronic properties.

1.0 Introduction

In recent years, much progress has been made in the fabrication of organic light emitting diodes (OLEDs) [1-6]. Low cost methods such as spin casting, screen and inkjet printing are being sort to fabricate organic electronic devices that are relevant for OLED applications such as in display and illumination [6]. Organic light emitting diodes have gained tremendous strides over conventional display and solid state lighting counterparts due to their higher resolution and lower power consumption. In addition, they are thinner, light weight and can be built on flexible substrates. In a typical device, at least one organic material is sandwiched between two electrodes. Charge carriers are injected from the electrode upon the application of a bias across the device. Holes injected from the anode, recombine with electrons from the cathode and produce excitons that can decay radiatively with the emission of photons. Crucial to the device performance are charge injection, charge transport, exciton formation and relaxation [7,8]. Adhesion at the interfaces of the device is also very critical in determining the device power efficiency and lifetime. Compression treatment of the finished device aids in closing up nanovoids at device interface and therefore has the potential of enhancing charge transport, since good adhesion will entail no or negligible charge trapping and exciton quenching that have adverse effects on device efficiency. The most common material used

for the anode is Indium Tin Oxide (ITO) due to its excellent transparency and high electrical conductivity [8]. Poly-(3,4-ethylenedioxythiophene):poly-(p-styrenesulfonate), PEDOT:PSS is the conventional candidate employed as hole transport layer due to its high electrical conductivity. However, this layer has some setbacks on the device, for example, due to the acidic PSS, it etches In and Sn from the ITO over time into the active layer thereby diminishing device efficiency. Its conductivity is anisotropic and its Highest Occupied Molecular Orbital (HOMO) energy is not sufficient enough to bridge the energy offset with ITO. It is as well hygroscopic and precipitates the diffusion of humidity into the device active area. Applying pressure to a complete device can significantly improve its performance [9]. A couple of other materials have been proposed to be used as Hole Transport Layers (HTLs). Transition Metal Oxides (TMOs) such as NiO, WO₃, V₂O₅ and MoO₃ with electrochromic properties show some interesting attributes to be useful as Hole Injection Layer Materials (HILMs) in organic photovoltaics and LEDs [10-13].

In this work, we used molybdenum trioxide (MoO₃) to fabricate some devices and compared the performance with those based on PEDOT:PSS as HTL. The devices were then subjected to compression treatment. Finite element simulation was used to examine how a layer will wrap around a dust particle at an interface. An analytical model was also used to explain the dependence of void length at interface with adhesion energy. The Derjaguin-Muller-Toporov (DMT) model which considers adhesion between stiff elastic spheres with weak adhesion and small tip radii is brought into play [14-16].

2. Analytical framework and Finite Element Simulations

The adhesion between two beams suspended by a particle was modeled by Manstrangelo et al. [17]

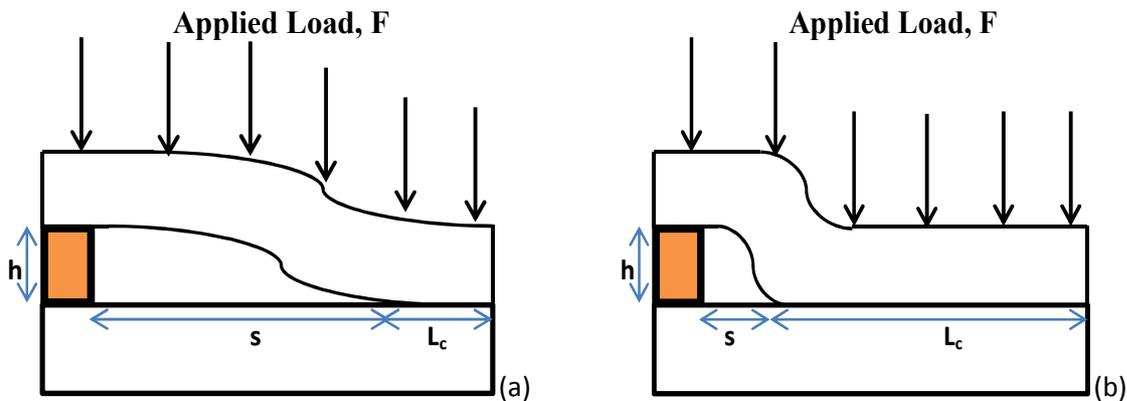


Figure 1 : Beam configurations of suspended beams according to Manstrangelo et al; (a) S-shaped beam; (b) Arc-shaped beam

Figure 1 shows a model akin to this which was adopted in order to examine interaction between the layers in the organic light emitting device. The model assumes interaction in the elastic regime and that attraction occurs only within the contact region of the interfacial area. Within this framework and using an energy balance approach, the void lengths for two different layer orientations, that is an S-shape and an Arc-shape, are given by [17]:

$$s_{s\text{-shape}} = \left(\frac{3Et^3h^2}{2\Gamma} \right)^{1/4} \quad (1)$$

$$s_{arc\text{-shape}} = \left(\frac{3Et^3h^2}{8\Gamma} \right)^{1/4} \quad (2)$$

where E is the beam Young's modulus, h is the particle (dust) height, t is beam thickness and Γ is the adhesion energy which can be determined from Atomic Force microscopy, AFM measurements of pull-off forces are given by:

$$\Gamma_{DMT} = \frac{F_{pull-off}}{2\pi R} \quad (3)$$

where $R = \left(\frac{1}{R_{rip}} + \frac{1}{R_{rms}} \right)^{-1}$ is the effective radius of the AFM tip in contact with the substrate.

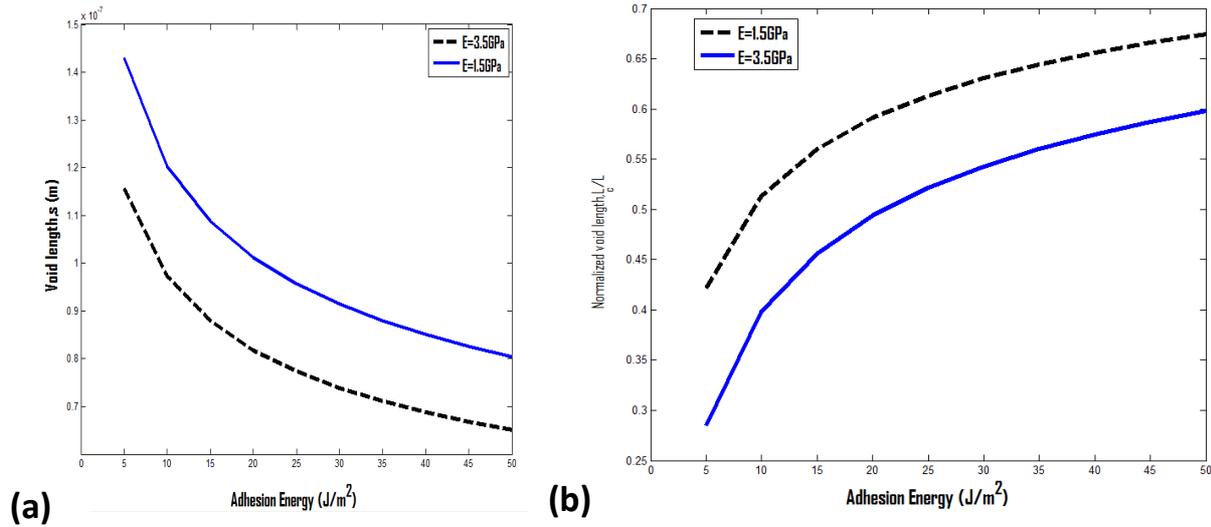


Figure 2: (a) Effect of void length on adhesion energy; (b) Effect of Normalized contact length on adhesion energy.

Finite element simulations were carried out using finite element code ABAQUS CAE v 6.9 to predict how dust particles at interfaces will affect the nanovoid created. Plain strain conditions with different heights of dust particles was used. The void length increases with dust particle height.

3. Experimental Methods

Two categories of devices were fabricated: a bilayer device made of Glass/ITO/PEDOT:PSS/MEH:PPV/Al, and another with constituents Glass/ITO/MoO₃/MEH:PPV/Al. Glass slides of dimensions 2.5cm by 2.5cm by 1mm were rinsed in de-ionized (DI) water, later cleaned in decon-90 and re-rinsed in DI water. They were then placed in a beaker containing isopropyl alcohol (IPA) before cleaning in ultrasonic bath at room temperature for 10min. The cleaned glass slides were dipped in ethanol before blow drying with Nitrogen. 200nm thick ITO anode was deposited on clean glass slides using Edwards Auto 306 magnetron sputtering system that was operated at an initial vacuum pressure of 4.0×10^{-5} Torr, rf power of 50 W and average deposition rate of $0.8 \text{ \AA}/s$. The sheet resistivity obtained using the Signatone Four Point Probe was $50 \Omega/sq$. The ITO was annealed at $250^\circ C$ for 1h. PEDOT: PSS solution obtained from Sigma Aldrich Company was filtered using a $0.25 \mu m$ mesh-size filter paper into a 10ml beaker. Using a Laurell Spin coater, the polymer solution was spin casted on the ITO-coated glass at 2000 rpm for 20 seconds with one end of the ITO covered with a Scotch tape to create the anode contact. It was then placed in Carbolite oven to dry it at $100^\circ C$ for 10mins. Other samples of ITO-coated glass are used to deposit MoO₃ thin film using an Edwards Auto 306 thermal vacuum evaporator. The MoO₃ powder (99.99% pure) obtained from Sigma Aldrich Company was placed in a molybdenum boat in the chamber and evaporated unto the ITO at a high vacuum of 4.5×10^{-5} Torr and a substrate temperature of $200^\circ C$ before spin coating poly(2-methoxy-5-(2'-ethylhexyloxy)-1,4-

phenylenevinylene) (MEH-PPV) on it. Prior to this, 100mg of the MEH: PPV powder was dissolved in 20ml of the solvent chloroform. The solution was then stirred for 6 hours at a speed of 700rpm. The resultant orange-red solution was filtered using a 0.25 μm mesh-size filter paper and spin-coated onto the PEDOT:PSS/ITO/Glass substrate at a speed of 800rpm for 60s. It was also spin-coated on MoO₃/ITO/Glass. An appropriate shadow mask was used to deposit the Al cathode by thermal vacuum evaporation under a high vacuum of 4.0×10^{-5} at room temperature. The active area of the device was 3mm². Some of the final devices were then taken for compression treatment by applying pressure on them using dead weights. Dead weights of 100N and 150N were applied on different devices for 5min each. Polydimethyl siloxane, PDMS, smeared with a self-assembled monolayer (SAM), which was placed on the device prior to compression, is then gently removed from without affecting its active area due to the low adhesion between the SAM and the Al cathode. Both the compressed and uncompressed devices were then taken for electrical characterization using a Keithley 2400 Source Measure Unit.

4. Results and Discussion

The results from the analytical model shows that as the adhesion energy increases, the void length decreases implying that stronger adhesion is essential to close up interfacial voids at the interfaces of OLED layers. This will therefore limit charge trapping at the interface during device operation. From the finite element simulations (Figure 3), it was noticed that as the dust particle height increases, the void length increases under the same load.

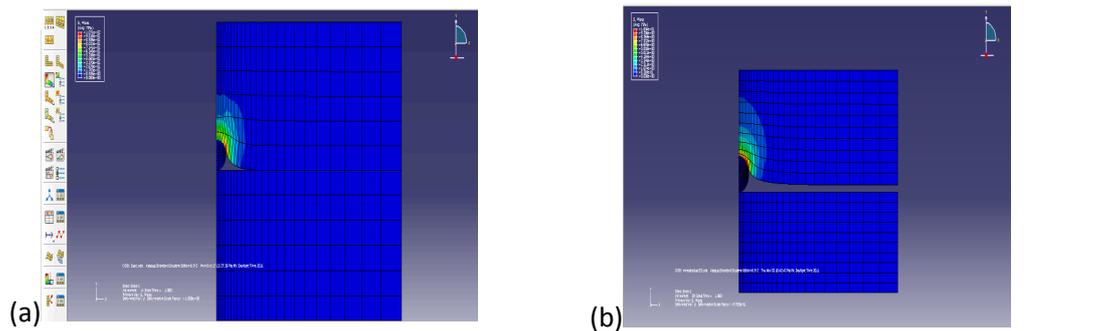


Figure 3: FES results of PEDOT:PSS (100nm) layer on MEH-PPV layer (100nm) sandwiched by dust particle of height: (a) 20nm and (b) 25nm, under the same loading conditions.

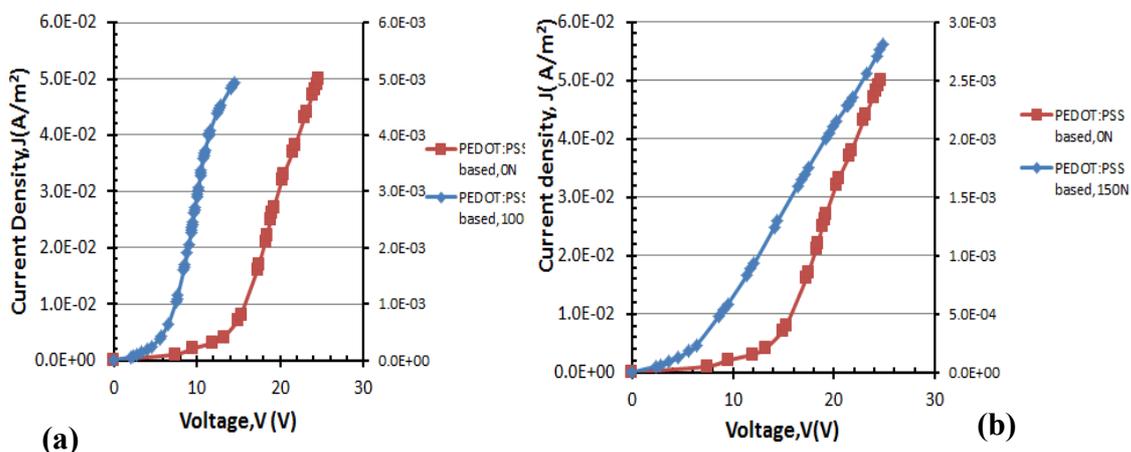


Figure 4: J-V plots for PEDOT:PSS-based device treated with: (a) 0N, 100N and (b) 0N, 150N loads

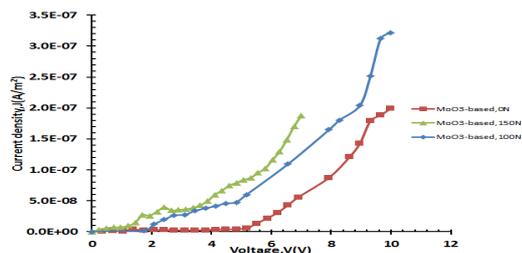


Figure 5: J-V plots for MoO_3 -based device treated with : 0N,100N and ,150N loads.

Figure 4 shows the J-V characteristics obtained for the PEDOT: PSS based devices that were uncompressed had turn-on voltages of 12V and 10V for the compressed ones. The MoO_3 based device which was not compressed gave turn-on voltage of 6V. The device compressed with a load of 100N gave a turn-on voltage of 4V whilst that compressed with 150N gave a turn-on voltage of 3V. This is shown in Figure 5. Since compressing the device reduces the device turn-on voltage, it implies that the nanovoids at the device interfaces are definitely closed resulting in better charge transport during device operation. It was also noticed that the uncompressed MoO_3 based devices gave better performances than the uncompressed PEDOT:PSS based ones.

5. Summary and Conclusion

Bilayer organic light emitting devices were fabricated based on PEDOT:PSS as hole transport layer (HTL) and also others based on MoO_3 as HTL. Devices were subjected to physical compression treatment using dead weights and the resulting J-V characteristics examined to compare their performances. Devices that were based on MoO_3 recorded better performances observed by their lower turn on voltages. The devices that were compressed depicted improved performances relative to the uncompressed. This shows that compressing the device creates more intimate contact at the interfaces by closing up interfacial voids and thereby facilitating charge transport.

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