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2015 IOP Conf. Ser.: Mater. Sci. Eng. 73 012046

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## Improved Performance of Organic LEDs with Modified Metal-Organic Interface

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**Abstract.** Incorporation of thin layers of strong acceptor tetrafluoro-tetracyanoquinodi methane (F<sub>4</sub>TCNQ) over conducting substrate Indium-Tin-Oxide (ITO) modified its work function. The enhancement depended on the thickness of F<sub>4</sub>TCNQ film. These modified ITO substrates were used as hole injection contacts in OLEDs which show enhanced device current, lower operating voltages and enhanced current efficiency.

### 1. Introduction

In organic based electronic devices like Organic Light Emitting Diodes(OLEDs) and Organic Photovoltaics(OPVs), the basic processes such as charge injection, charge transport, capture and radiative recombination of opposite charged charge carriers are involved[1,2]. In an organic metal interface, if interface dipoles are absent and vacuum level alignment is assumed then the energy barrier for charge injection is given by the difference between the work function of the electrode and the highest occupied molecular orbital(HOMO) or lowest unoccupied molecular orbital(LUMO) of the organic semiconductor (Schottky-Mott Limit). Often the energy barrier off set is modified by interface dipoles which modify the potential barrier [3-5]. The resultant large interface barrier results in injection limited charge transport. Some of our recent studies on electrode/organic interfaces indicate that it is possible to align the Fermi level electrodes with the negative polaron states of thin layers of strong acceptor molecules [6-7]. This process enhances the work function of the substrates and enables to form ohmic contact with many organic molecular films.

To demonstrate the enhancement in work function, we have fabricated OLED devices using indium tin oxide (ITO) modified with different thickness of tetrafluoro-tetracyanoquinodimethane (F<sub>4</sub>TCNQ) as the hole injecting anodes, 2,2',7,7'-Tetrakis-(diphenylamino)-9,9'-spirobifluorene (Spiro-TAD) as the hole transport material and a thin film of gold as cathode. The J-V characteristics of the device show pure space charge limited conduction showing the formation of ohmic contact between the modified electrode and Spiro-TAD (HOMO=5.4eV). The OLEDs with modified anodes have shown enhanced efficiency and low turn-on voltages.

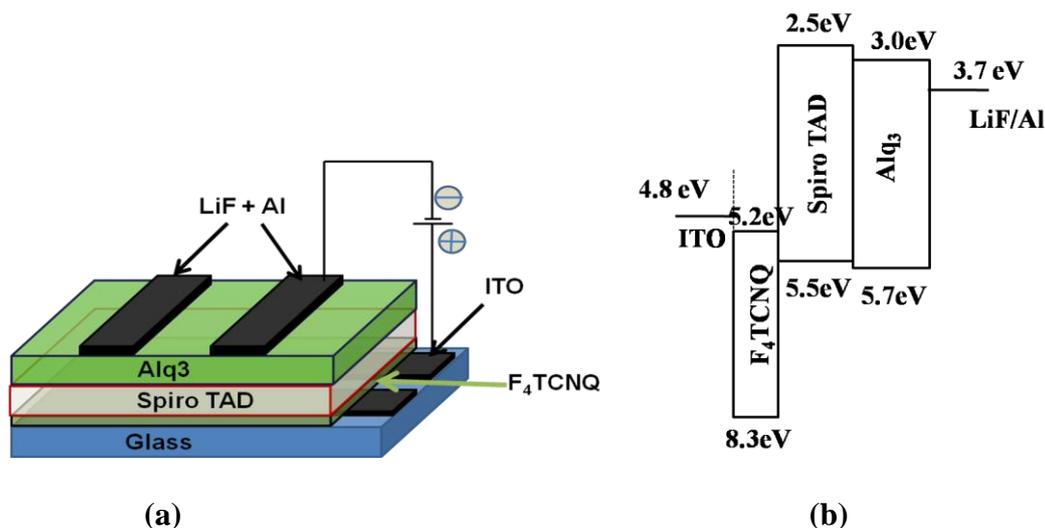


## 2. Experimental Details

The OLEDs were fabricated on ITO coated glass substrates having a sheet resistance of  $20 \Omega/\text{sq}$  and a thickness of 120 nm. Before fabricating the device, ITO coated glasses were patterned and cleaned by a conventional solution cleaning process using deionized water, acetone, trichloroethylene, and isopropyl alcohol for 20 min each were using an ultrasonic bath. Prior to film deposition, to enhance work function the surface of ITO substrate were treated with oxygen plasma for 5 min. Organic layers were deposited onto glass substrates under high vacuum ( $4 \times 10^{-6}$  torr) at a deposition rate of  $0.4 \text{ \AA}/\text{s}$ . Thickness of the deposited layers were measured in situ by a quartz crystal thickness monitor. Three OLEDs were fabricated with different thickness of  $\text{F}_4\text{TCNQ}$  ( $x = 0, 5 \text{ nm}$  and  $7 \text{ nm}$ ). The device structure was ITO (12nm)/  $\text{F}_4\text{TCNQ}$  ( $x$ )/ Spiro-TAD (30nm)/  $\text{Alq}_3$ (50nm)/ LiF(1nm)/ Al(150nm). The size of each pixel was  $5 \text{ mm} \times 5 \text{ mm}$ . The emission spectrum has been recorded with a high resolution spectrometer (ocean optics HR-2000 CG UV-NIR). The current density-voltage-luminescence (J-V-L) characteristics have been measured with a luminance meter (LMT-1009) and Keithley 2400 programmable current-voltage digital source meter. All the measurements were carried out at room temperature under ambient conditions.

## 3. Result and Discussion

The schematic structure of fabricated OLEDs and energy level diagram is shown in Figure 1(a)-(b).



**Figure 1:** (a) Schematic diagram of OLED and (b) energy level diagram of device.

It has been observed from the energy level diagram that there are large energy barrier (0.7 eV) between ITO and Spiro TAD. To reduce the barrier and increase the hole injection into the device it is necessary to modify the ITO/Spiro-TAD interface. We have introduced a thin layer of  $\text{F}_4\text{TCNQ}$  and varied its thickness and observed the effect of the introduction this layer on OLED devices. Three devices (D1( $x=0 \text{ nm}$ ), D2( $x=5 \text{ nm}$ ) & D3( $x=7 \text{ nm}$ )) have been fabricated and their J-V characteristics have been measured.

Figure 2(a) show the J-V characteristics of the devices with and without anode modification. It is clearly seen that the interface modified devices (D2 & D3) gives higher current density as compared to unmodified device (D1) and as we increase the thickness of  $\text{F}_4\text{TCNQ}$  the

current density has also increased and the maximum current density has been observed for device D3.

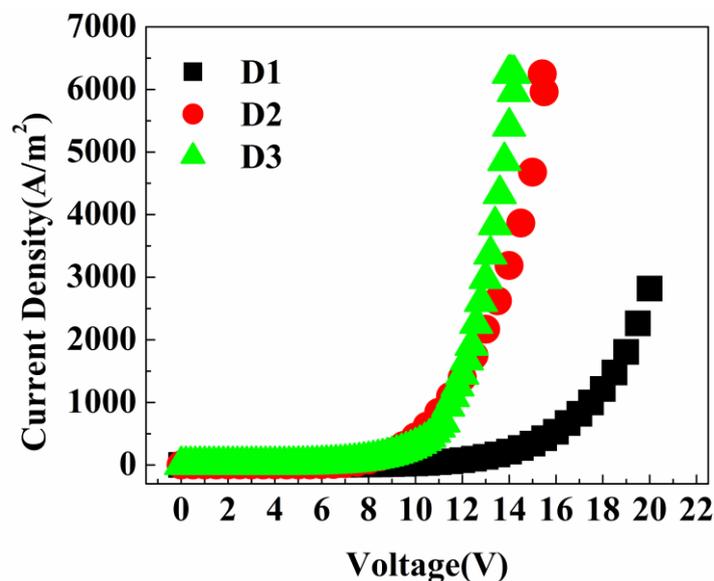
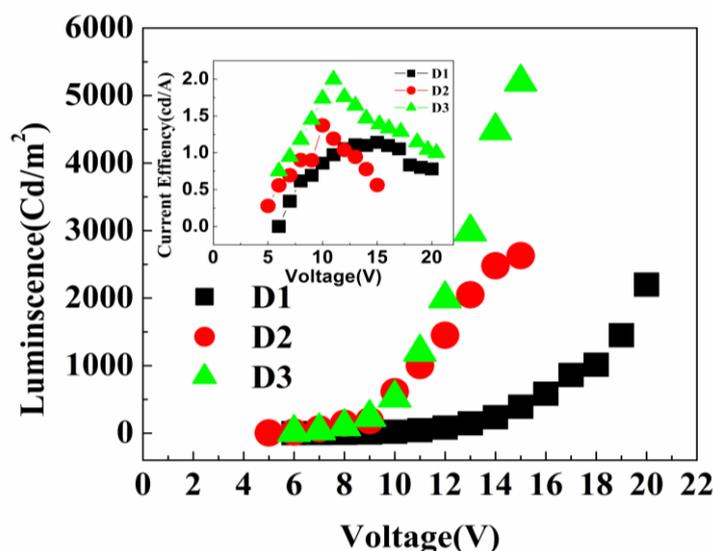


Figure 2(a): J-V characteristics of OLEDs with and without anode modification.

This increase in current density can be attributed to a decreased hole injection barrier at the anode-organic interface and the formation of an ohmic contact for holes at this interface and consequently an enhanced injection of holes from ITO to Spiro-TAD. Since the measured electrical current density depends mainly on the organic electrode injection barrier which depends on the difference in the work function of metal electrode and the HOMO/LUMO of the organic materials, this increase in current density indicates a modification of the ITO's work function. Recently it has been shown by Brown and Li et al [8-10] that in the case organic films grown at low vacuum where the metal surface is passivated by oxide layer formation or hydrocarbon coverage, the metal wave function is not coupled with the organic wave function and interface states are not formed preventing partial charge transfer. Charge transfer take place only through quantum mechanical tunneling by integer charge transfer process. In this process if the metal Fermi level is in between the organic HOMO and LUMO, charge transfer does not occur and the SM limit is followed. However if the metal work function is lower than the negative polaron state of the organic, integer electron transfer occur from the metal in to the organic and positive interface dipoles are formed which pins the metal Fermi level with the negative polaron state. They have also shown that in the case of deposition of F<sub>4</sub>TCNQ on to low work function metals, the effective work function is enhanced up to 5.5eV. Similarly if the Fermi level is higher than the positive polaron state of the organic, integer electron transfer occur from the organic into the metal resulting in a negative interface dipole formation and subsequent pinning of the metal work function on the positive polaron state. The increased current density in the present case may be due to this effect

Figure 2(b) shows the Luminescence-Voltage (L-V) characteristics of the devices D1, D2 and D3. It has been observed that the interface modified devices show higher luminescence and as we increase the thickness of the F<sub>4</sub>TCNQ luminescence increases and saturates at about 7nm.



**Figure 2(b):** *L-V characteristics of OLEDs with and without anode modification. Inset shows the current efficiency-voltage of these OLEDs.*

The highest luminescence ( $5000 \text{ cd/m}^2$ ) was achieved by device D3. Also, the operating voltage decreases in case of interface modified devices (D2 and D3). Similar enhancement has been observed for current efficiency (as shown in the inset of Fig 2(b)).

#### 4. Conclusions

In conclusion, we have explicitly shown that modification of conducting substrates using strong acceptor when used as anodes in OLEDs gives higher current density, lower operating voltages and higher luminescence.

#### Acknowledgements

The authors would like to thank and greatly acknowledge to Director, National Physical Laboratory India to encourage us for novel research, Council of Scientific and Industrial Research (CSIR) for the financial support from the project NWP-55 (TAPSUN). One of the author (OR) acknowledges University Grant Commission (UGC), New Delhi, for providing the Senior Research Fellowship.

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