

Effect of Higher Carrier Injection Rate on Charge Transport and Recombination In Mixed-Host Organic Light Emitting Diode

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Abstract— The effect of anode surface modification on performance of uniformly mixed-host (MH) emissive layer (EML) based organic light emitting diode (OLED) has been investigated by the numerical simulation. Indium Tin Oxide (ITO) used as anode in the device. Due to proposed surface modification technique of ITO, the energy barrier at anode/organic layer interface is reduced which consummately enhanced the hole injection rate that leads to balance of carriers transportation and recombination in the EML. Through the numerical simulation, the electrical characteristics and internal device physics of uniform MH-OLED have been analyzed quantitatively. Calculated current balance factor which is related to the external quantum efficiency also confirmed the efficiency enhancement of MH-OLED by the proposed ITO work function modification technique.

Keywords— indium tin oxide (ITO) modification; hole injection; numerical simulation; carrier recombination; current balance factor.

I. INTRODUCTION

As a next generation display technology, organic light emitting diodes (OLEDs) has been drawn enormous attention over other displays, like CRT, LCD etc. due to simple fabrication process, wide viewing angle, very fast switching time ($<1\mu\text{s}$), broad operating temperature range ($-46\text{ }^\circ\text{C}$ to $70\text{ }^\circ\text{C}$) and low turn-on voltage [1]. The first OLED consisting of bi-layer organic materials was fabricated by Tang and Vanslyke in 1987 [2]. Since then, number of research works have been carried out to improve the device performance over the conventional bi-layer OLED via organic materials improvement and development of the layer architecture and composition [3].

In order to improve the conventional heterojunction (HJ) OLED performance and reliability, a mixed-host structure had been introduced where hole transport material (HTM) and electron transport material (ETM) mixed uniformly as the host material in the single emissive layer (EML) [4]. The interface

of hole transport layer (HTL) and electron transport layer (ETL) which formed in HJ structure, can be eliminated by mixing these two transport materials. About 6 times heightening of operation lifetime had been achieved from this MH structure as compared to the conventional HJ-OLED [4].

Current OLEDs research topics being reported were more on charge balance or confinement for better recombination [4-7] and replacement of ITO or its surface modification [8-9] to improve the device performance as well as operation life time. In OLEDs, the physical process involved in charge injection at an electrode/molecule interface has a crucial role. The energy barrier at this interface dictates the functionality of OLEDs. It was recently discovered that the surface work function of metal oxide electrodes is one of the most important parameters in determining the interfacial barrier [3, 9]. Owing to its unique combination of good electrical conductivity and excellent optical transmission properties, tin-doped indium oxide (ITO) has been well developed in industry as electrode which is widely used in opto-electronics devices ranging from solar cells to OLEDs [10-11]. But the potential barrier between ITO and the organic layer severely limits the efficiency of hole-injection. The treatment of the ITO surface to reduce the interfacial energy barrier usually has a strong influence on the performance of the OLED device. A recent breakthrough in boosting the ITO surface work function by O_2 and Cl_2 plasma treatment [9], post annealing treatments, adding buffer layer [8] and UV-ozone treatments [12] have been reported. A simple way, the soaking of ITO in different concentrations of Cesium Fluoride (CsF) solution and subsequent UV-ozone treatment to modify the ITO surface work function has been demonstrated by H.-W. Lu et al [3]. In that work, ITO surface modification technique using CsF solution qualitatively applied to analysis the performance of conventional HJ structure.

As a simple and better performed device over HJ structure, a modified uniform MH structure has been proposed in this work to further improvement of MH-OLED performance and

operation life time. The proposed MH structure is: ITO (modified with various CsF solutions) / NPB : Alq₃ (3:2) (100 nm) / LiF/Al. The feasibility of this proposed MH structure has been analyzed from the simulated current density- luminance - voltage (J-L-V) characteristic and efficiency curves. After that, the effect of ITO surface modification (using different concentrations of CsF solutions) to increase the hole injection efficiency has been explained in terms of distribution of carrier concentration, current density, electric field and carriers recombination.

By the systematic study of the MH-OLED performance, it is possible to qualitatively explain some effects on J-L-V characteristic and efficiency. But, without numerical simulation, it is difficult to explain these effects quantitatively [12]. Numerical simulation is more viable to get any analytical results directly from the OLEDs where complicated working process and several interfaces are involved. Therefore, this work led by numerical simulation which treated as powerful method to retrieve detailed information such as the charge carrier injection, distribution of electric field and recombination those are difficult to measure directly through experiments, investigated the underlying device mechanism and their amplitude which can affect the device performance.

II. ELECTRICAL MODEL

To analysis the effect of ITO work function modification on the underlying physics and performance of MH-OLED, drift-diffusion electrical model has been implemented using a numerical simulator called ‘SETFOS’ (Semiconducting Thin Film Optics Simulation Software). Poisson equation and continuity equation as presented in “(1),” and “(2),” correspondingly considered in that electrical model to solve the electric field distribution and time evaluation of the system.

$$\frac{dE}{dx} = \frac{q}{\epsilon\epsilon_0}(p-n) \quad (1)$$

$$\frac{dn}{dt} = \frac{dJ_n}{qdx} - R - T \quad (2)$$

where, E is the electric field inside the OLED, q the elementary charge, ϵ the dielectric permittivity of the organic materials, ϵ_0 the dielectric constant, p and n are the hole density and the electron density respectively, J_n the electron current density, R the recombination rate and T the trapping rate for free carriers. The carrier density distribution can be obtained by solving the drift and diffusion equation based on the Langevin recombination (R) process those are calculated using “(3),” and “(4),” respectively. The conventional electric field dependent ‘Poole-Frenkel mobility (PFM)’ model as expressed in “(5),” applied for carrier mobility calculation in the device. The current balance factor (CBF) (b) in “(6),” used to measure the device quantum efficiency [14] which is defined as the recombination current with the total current density.

$$J_n = qp\mu_n E + qD_n \frac{dn}{dx} \quad (3)$$

$$R = \frac{q}{\epsilon\epsilon_0}(\mu_p + \mu_n) \quad (4)$$

$$\mu(E) = \mu_0 \exp(\gamma^* \sqrt{E}) \quad (5)$$

$$b = \frac{\int_0^L qR(x)dx}{J_T} \quad (6)$$

Here, μ_p and μ_n are the hole and electron mobility respectively, D_n the diffusion constant, $\mu(E)$ is the charge carrier mobility under electric field (E), μ_0 is the mobility under zero electric field, γ is the Poole-Frenkel field dependent factor, J_T is the total current density and L is the total device thickness.

III. DEVICE STRUCTURE AND INPUT PRAMETERS FOR SIMULATION

Three uniform MH-OLED structures, namely S_0 , S_1 and S_2 are presented in “Fig. 1,” where NPB (HTM) and Alq₃ (ETM) mixed with a uniform ratio of 3:2 (NPB:Alq₃). The used HOMO and LUMO levels of MH material are 5.4 eV and 3.1 eV [7] and the work function of LiF/Al is 3.0 eV [3] in simulation. The work function of ITO is 4.78 eV, 4.95 eV and 5.11 eV used in simulation for device S_0 (unmodified ITO), device S_1 (ITO modified by 1 wt% CsF solution) and device S_2 (ITO modified by 10 wt% CsF solution) respectively [3]. Considering the unmodified work function based ITO as reference, the effects of proposed ITO work function modification technique using different concentrations of CsF solutions on uniform MH-OLED performance have been analyzed in the next section in terms of hole injection density, carrier recombination rates and current balance factor.

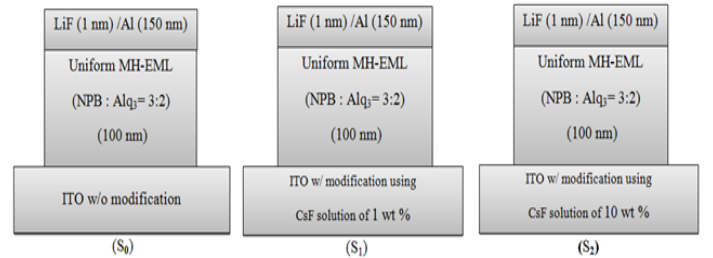


Fig. 1. W/ and W/O Modified ITO based MH-OLEDs

IV. RESULTS AND DISCUSSIONS

A. J-L-V Characteristics and Efficiencies

Figure 2 shows the current density-voltage-luminance (J-V-L) characteristic curve of three MH-OLEDs. 1 wt% and 10 wt% solutions of CsF improved the work function of ITO from normal value of 4.78 eV to 4.95 eV and 5.11 eV respectively. These work function improvement of ITO increased the hole injection rate by which the J-L-V curves influenced significantly. At a constant operating voltage of 8 V (Fig. 2(a)), the current density three MH-OLEDs of S_0 , S_1 and S_2 are 490 mA/cm², 925 mA/cm² and 1480 mA/cm² correspondingly. In contrast, the luminescences of device S_0 , S_1 and S_2 shows the same trend as that of current density with

the same operating voltage, which indicates that high luminescence from a device can be obtained with high current density. 10 wt% CsF solution based MH-OLED shows the highest current density and luminance due to higher hole injection rate which also result of higher efficiency as evidence as presented in Fig. 2(b). The improved values of electroluminescence (EL) efficiency, γ of modified ITO based MH-OLEDs by 1 wt% and 10 wt% CsF solutions are 1.91 cd/A and 2.43 cd/A over without ITO modification based OLED where the value of EL efficiency is 1.40 cd/A. These significant performance improvements of uniform MH-OLED using ITO surface modification by CsF solutions can be visualized and explained from the carrier distribution and recombination profiles.

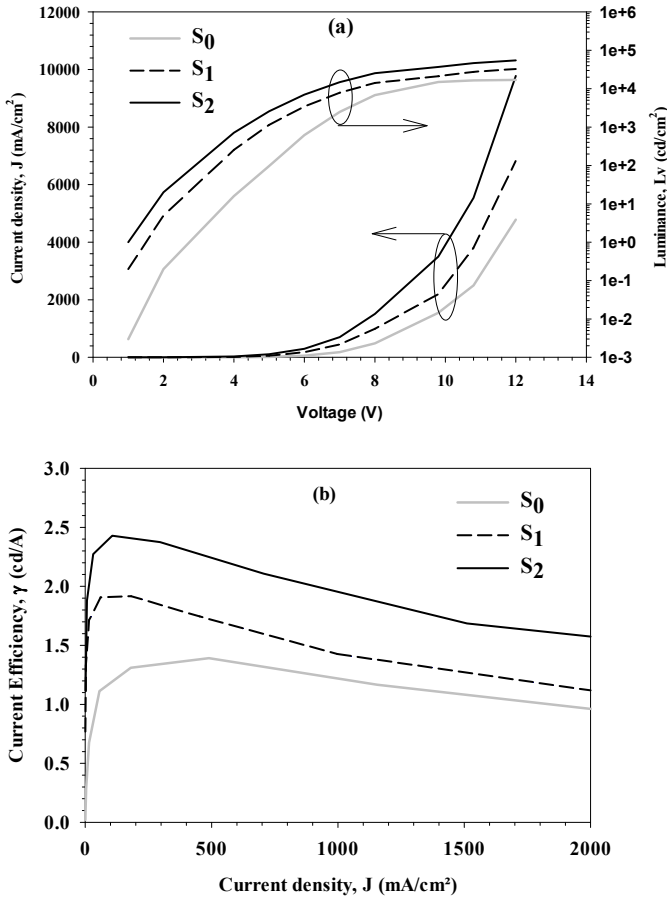


Fig. 2. (a) Current density-Luminance-Voltage and (b) Electroluminescence Efficiency vs Current density curves for unmodified and modified ITO based MH-OLEDs.

B. Effects of ITO surface modification on carrier injection, transport and recombination

From the carrier concentration profile as shown in Fig. 3 (a), it is observed that at operation voltage of 8 V, the concentration of hole at anode/EML interface are increased around 1 and 1.7 orders due to ITO surface modification by 1 wt% and 10 wt% CsF solutions respectively, where the interfacial energy barrier between ITO (4.95 eV and 5.11 eV) and EML (HUMO=5.4 eV) reduced from 0.62 eV (in unmodified condition (4.78 eV)) to 0.45 eV, and 0.29 eV

correspondingly. Therefore, these significant reduction of interfacial energy barrier improved the hole current density at the ITO/EML interface (Fig. 3(b)) around 1.5 and 1.8 times which linked to increase the total current density (Fig. 2(a)) of the modified ITO based MH-OLEDs over unmodified ITO based MH-OLED. Furthermore, the electric field distribution along the EML as shown in Fig. 3(c) increased steadily from anode/EML interface due to this proposed CsF solution based ITO surface modification technique which harmoniously influenced the recombination rate at the EML.

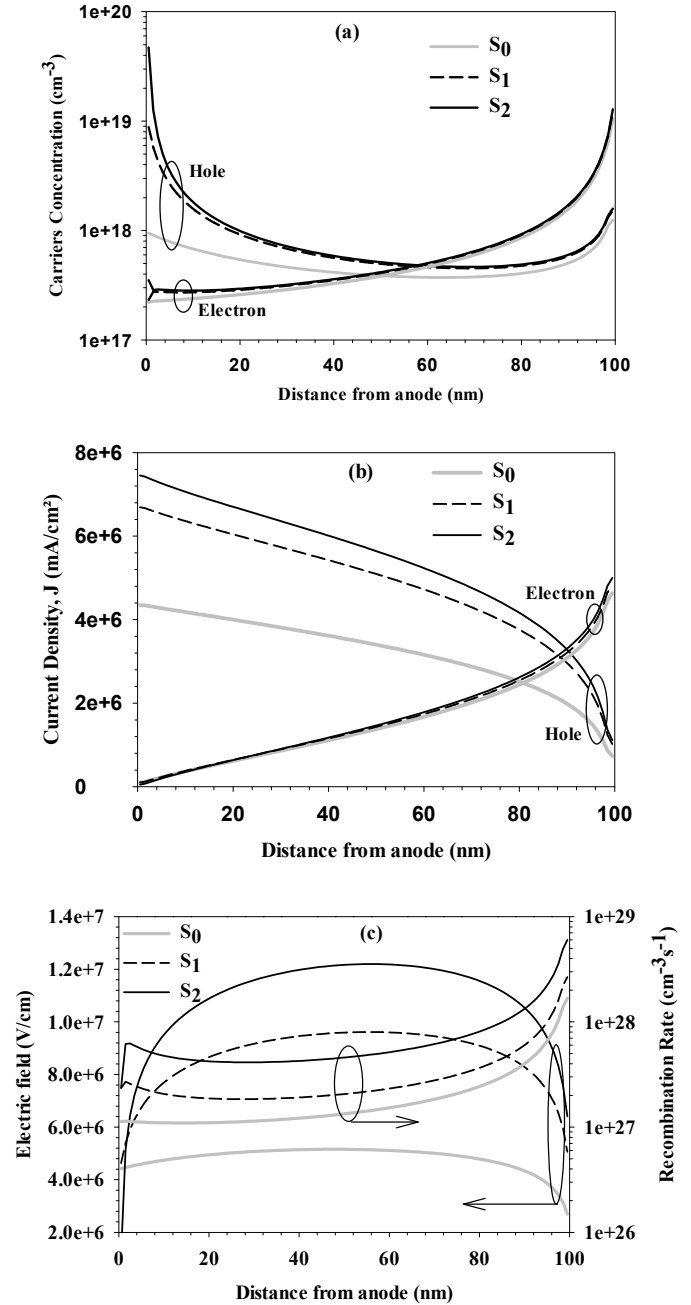


Fig. 3. Effect of hole injection rate (using treated ITO) on (a) Carriers density profile, (b) Current density (for hole and electron) and (c) Electric field and Carrier recombination zone in the EML at 8V with respect to untreated ITO based MH-OLEDs.

Fig. 4 presents the highest calculated values of current balance factor (using equation “(6),”) of modified ITO based MH-OLEDs with 1 wt% ($b=0.77$) and 10 wt% ($b=0.83$) CsF solutions. The CBF of MH-OLEDs based on ITO surface modification reached steady state position before unmodified ITO based MH-OLED which CBF is 0.71.

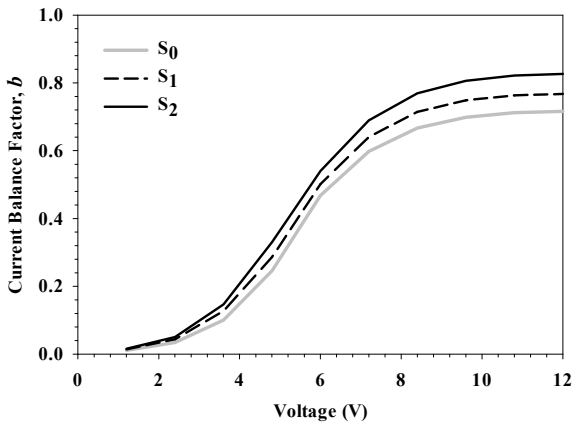


Fig. 4. Ameliorant of Current Balance Factor due to higher hole injection rate (treated ITO) over lower hole injection rate (untreated ITO) based MH-OLEDs at different operating voltage.

C. Discussions

Through this numerical simulation, the influence of CsF solution on ITO work function modification and device performance have been explained from the hole current density, recombination rate and current balance factor. Due to the ITO surface modification by ITO soaking in 1 wt% and 10 wt% CsF solutions, the energy barrier at anode/emissive layer interface reduced by 0.17 eV and 0.33 eV respectively from the original value (0.62 eV) which ultimately boost up the hole injection efficiency through that interfacial region. Therefore, electroluminescence efficiencies of treated ITO based devices are improved by 1.36 and 1.73 times. Around 1.2 times improvement (with 10 wt% CsF solution) in current balance factor over normal condition also confirmed the improvement of device external quantum efficiency which is related to device life time.

V. CONCLUSIONS

The effects of proposed CsF solution based ITO surface modification technique on uniform MH-OLED performance have been analyzed from the ‘SETFOS’ based numerical simulation results. Due to ITO surface modification with 10 wt% CsF solution, the energy barrier for hole injection reduced by about 2.37 times (from 0.69 eV to 0.29 eV) which significantly increased hole injection rate at anode/EML interface. Therefore, EL. efficiency is increased around 1.73 times. The value of current balance factor is also improved from 0.71 to 0.83 which is proportional to the experimental quantum efficiency. Sooth to say, the quantitative results of modified ITO based uniform MH-OLEDs performances those are obtained from the numerical simulation results, have proved the experimental feasibility of this proposed uniform

MH structure to improve the device performance and operation life time.

REFERENCES

- [1] N. C. Erickson and R. J. Holmes, “Highly efficient, single-layer organic light-emitting devices based on a graded-composition emissive layer,” *Appl. Phys. Lett.*, vol. 97, no. 8, p. 083308, 2010.
- [2] C. W. Tang and S. a. VanSlyke, “Organic electroluminescent diodes,” *Appl. Phys. Lett.*, vol. 51, no. 12, p. 913, 1987.
- [3] H.-W. Lu, C.-W. Huang, P.-C. Kao, S.-Y. Chu, and Y.-D. Juang, “Effects of ITO Electrode Modification Using CsF Solution on Performance of Organic Light-Emitting Diodes,” *ECS J. Solid State Sci. Technol.*, vol. 4, no. 3, p. R54–R59, Jan. 2015.
- [4] D. Ma, C. S. Lee, S. T. Lee, and L. S. Hung, “Improved efficiency by a graded emissive region in organic light-emitting diodes,” *Appl. Phys. Lett.*, vol. 80, no. 19, p. 3641, 2002.
- [5] C.-H. Hsiao, Y.-H. Chen, T.-C. Lin, C.-C. Hsiao, and J.-H. Lee, “Recombination zone in mixed-host organic light-emitting devices,” *Appl. Phys. Lett.*, vol. 89, no. 16, p. 163511, 2006.
- [6] R. Chowdhury, T.S. Ong, Y.Y. Kee, S.S. Yap, T.Y. Tou, “Numerical and experimental studies of mixed-host organic light emitting diodes,” *Curr. Appl. Phys.* vol.15, no.11, p. 1472–77, 2015.
- [7] C. Riku, Y. Y. Kee, T. S. Ong, S. S. Yap, and T. Y. Tou, “Simulation of mixed-host emitting layer based organic light emitting diodes,” in *AIP Conference Proceeding*, vol. 1657, no. 1, p. 070002, 2015.
- [8] H. Wang, K. P. Klubek, and C. W. Tang, “Current efficiency in organic light-emitting diodes with a hole-injection layer,” *Appl. Phys. Lett.*, vol. 93, no. 9, p. 093306, 2008.
- [9] X. A. Cao and Y. Q. Zhang, “Performance enhancement of organic light-emitting diodes by chlorine plasma treatment of indium tin oxide,” *Appl. Phys. Lett.*, vol. 100, no. 18, p. 183304, May 2012.
- [10] Baldo, M. A., Thompson, M. E. & Forrest, S. R., “High-efficiency fluorescent organic light-emitting devices using a phosphorescent sensitizer,” *Nature*, vol. 403, p. 750–753, 2000.
- [11] Helander, M. G., Wang, Z. B., Qiu, J., Greiner, M. T., Puzzo, D. P., Liu, Z. W. & Lu, Z. H., “Chlorinated indium tin oxide electrodes with high work function for organic device compatibility,” *Science*, vol. 332, p. 944–947, 2011.
- [12] P.-R. Huang, Y. He, C. Cao, and Z.-H. Lu, “The origin of the high work function of chlorinated indium tin oxide,” *NPG Asia Mater.*, vol. 5, no. 8, p. e57, Aug. 2013.
- [13] E. Knapp and B. Ruhstaller, “Numerical analysis of steady-state and transient charge transport in organic semiconductor devices,” *Opt. Quantum Electron.*, vol. 42, no. 11–13, p. 667–677, Feb. 2011
- [14] J. C. Scott, S. Karg, and S. A. Carter, “Bipolar charge and current distributions in organic light-emitting diodes,” *J. Appl. Phys.*, vol. 82, no. 3, p. 1454, Aug. 1997.