



Effects of novel transition metal oxide doped bilayer structure on hole injection and transport characteristics for organic light-emitting diodes

Chi-Ting Tsai^a, Ya-Han Liu^a, Jian-Fu Tang^a, Po-Ching Kao^b, Chung-Hao Chiang^a, Sheng-Yuan Chu^{a,c,*}

^a Department of Electrical Engineering, National Cheng Kung University, Tainan 70101, Taiwan

^b Department of Electrophysics, National Chiayi University, Chiayi 60004, Taiwan

^c Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 70101, Taiwan

ARTICLE INFO

Keywords:

Organic light-emitting diodes
Transition metal oxide
Electrical doping
Graded doping
Hole injection

ABSTRACT

A contemporary hole injection bilayer structure (HIBL) based on molybdenum trioxide (MoO₃)-doped *N,N'*-Di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) has been demonstrated and compared with several efficient transition metal oxide (TMO)-based hole injection layers (HILs). Device performances of OLEDs was significantly improved by the utilization of this HIBL. Results of electroluminescence (EL) spectra, hole-only current density-voltage test and capacitance measurement by impedance spectroscopy (IS) are indicative of enhanced hole injection and transport characteristics. Moreover, ultraviolet photoelectron spectroscopy (UPS) results authenticated a cascading highest occupied molecular orbital (HOMO) energy level contributed to both improved hole injection and transport properties, therefore leads to better carrier balance and device efficiency in OLEDs.

1. Introduction

Innovation of organic light-emitting diode (OLED) has attracted researchers to explore the next generation LEDs. Vast applications like solid state lighting and flat panel display are economically viable by the unique properties of OLEDs [1]. To attain high efficiency in light emission, one primary issue is to overcome the energy losses during electron-photon conversion [2]. Two factors that influence carrier injection and transport characteristics to diminish the energy losses and enhance efficiency are injection barrier at electrode/organic interface and charge carrier transport from this interface to active zone [2,3]. Diverse techniques that assist carrier injection from electrodes to organic layers are being pursued by researchers. To be precise, the effective approaches are interlayer sandwiched between electrode and organic layer [4,5], modification of electrode work function [6–8] and addition of p-type/n-type dopant in carrier transporting layer [9,10]. Particularly, transition metal oxides such as MoO₃, WO₃, V₂O₅, ReO₃ are utilized for hole injection recently due to their high work function [10–13]. These TMOs fulfill their existence as thin hole injection interlayers and also as p-type dopants in hole transport layers (HTLs). These TMO-based thinfilms are practiced effectively to enhance hole injection characteristics in OLEDs and hole-extraction properties in organic photovoltaic devices (OPVs) [14]. On the other hand,

introducing a cascading or graded interface and transport layer are known to improve carrier flow and device performance [15]. Besides, it has also been found that employing a graded doping profile in emissive layer (EML) can substantially extend OLED lifetime by widening the recombination zone [16]. Significant efforts have been made to focus on the graded doping system based on emitting host-guest system [17–24], organic-organic [15,25,26] and metal-polymer materials [27]. Much less attention was paid to other inorganic dopants, especially, TMOs. Moreover, the underlying mechanisms behind its role in augmenting carrier injection and transport are not completely understood.

In this work, we report a novel hole injection bilayer (HIBL) structure based on NPB:MoO₃ for gradual tuning the energy barrier distribution of material with respect to altered doping concentration. Hole transport in this HIBL is analyzed by the current density-voltage characteristics of hole-only devices. Observed by ultraviolet photoelectron spectroscopy (UPS), this HIBL exhibits an energetically stepwise structure which accomplishes effective hole injection and transport properties those are desired to achieve improved OLED characteristics. Additionally, capacitance-voltage measurement was conducted by impedance spectroscopy (IS) to authenticate the charge injection ability by monitoring the carrier dynamics in the emitting layer.

* Corresponding author.

E-mail address: chusy@mail.ncku.edu.tw (S.-Y. Chu).

2. Experimental details

All materials used in this work were purchased from commercial vendors (purities > 99.9%) and were used as received. Patterned indium tin oxide (ITO)-coated glass substrates (Global Tech International) with sheet resistance of approximately 11 Ω /square were used. Diverse device structures for various purposes were described in the text. ITO was used as anode, molybdenum trioxide (MoO_3) was used as anode buffer layer or HIL dopant, N,N' -Di(1-naphthyl)- N,N' -diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) was used as hole transport layer and the host material in HIL, tris(8-hydroxyquinoline)aluminum (Alq_3) was used as light-emitting layer and electron transport layer, and lithium fluoride (LiF)/Al was used as cathode. In phosphorescent OLEDs, 4,4'-cyclohexylidenebis[N,N -bis(4-methylphenyl)benzenamine] (TAPC) was used as hole transport layer, bis[3,5-di(9H-carbazol-9-yl)phenyl]diphenylsilane (SimCP₂) and bis[2-(4,6-difluorophenyl)pyridinato- C^2,N] (picolinato)iridium(III) (FIrpic) were used as host and guest materials of phosphorescent blue emitting layer, and 2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1- H -benzimidazole) (TPBi) was used as electron transport layer. In advance of organic film deposition, the substrates were immersed in an ultrasonic bath of detergent solution, isopropanol, ethanol and de-ionized (DI) water for 15 min each sequentially, then followed by a rinse of DI water. Eventually, the substrates should undergo UV-ozone (Jelight UVO-42) treatment for 20 min before use. All organic layers, inorganic oxides, metal electrodes and doped layers were deposited at room temperature by thermal evaporation under high vacuum of 5×10^{-7} Torr. Electrical doping of MoO_3 and NPB was implemented by thermal co-evaporation from two individual source boats. The active area of devices was $2.5 \times 2.5 \text{ mm}^2$ defined by the overlap of ITO and Al cathode. Deposition rates, thicknesses and doping concentrations were monitored and controlled via separated quartz crystal oscillators (INFICON, SQM-160) and further calibrated by surface profiling (KLA Tencor, Alpha-Step IQ). The deposition rate of organic materials was 1.0 $\text{\AA}/\text{s}$, while deposition rates of LiF and Al were 0.1 $\text{\AA}/\text{s}$ and 3–5 $\text{\AA}/\text{s}$, respectively. For NPB: MoO_3 layers, deposition rate of dopant MoO_3 was controlled as 0.1 $\text{\AA}/\text{s}$ and the deposition rate of host material was adjusted according to different volume ratio. Devices with different configurations for the same measurement were fabricated in the same batch by switching shadow masks. Current density-voltage-luminescence (J - V - L) characteristics were observed using a program-controlled system incorporated by a source meter (Keithley 2400) and a photometer (Photo Research PR-655) in the ambient atmosphere without protective encapsulation. The UV-vis absorption spectrum was procured by a spectrophotometer (HITACHI U-3310) with scan rate of 600 nm/s. UPS analysis was measured using an electron spectroscopy analysis system (ULVAC-PHI 5000 Versaprobe II) equipped with ultra violet sources providing He-I photons (21.22 eV). The samples with deposition in chronological order for UPS measurement were fabricated in the same batch. To avoid possible ambient oxygen-mediated oxidation and/or ambient-induced chemical changes, samples for UPS were stored in a vacuum sealed box after organic layer deposition, then transferred into the UPS chamber. In order to observe the high binding energy cutoff, a -5 V bias was applied during UPS measurements. Since the depth resolution of UPS is at the range of 0.5–5 nm, the last deposited layer prepared for the UPS in each sample was controlled to be 5 nm to avoid the interruption of background signals from underlying layers. The impedance spectroscopy analysis was investigated by Agilent E4990A precision impedance analyzer at frequency range of 20 Hz to 1.2 MHz with AC oscillation level of 100 mV. And the applied DC bias was varied between -2.5 to +5 V. To ensure reliability and reproducibility of our data, all the experiments were repeated several times.

3. Results and discussion

Owing to closely related energy values of highest occupied

molecular orbital (HOMO) level of NPB (5.5–5.7 eV) [28,29] and work function of MoO_3 (5.6–5.7 eV) [11,30], charge transfer (CT) complexes can be easily procured by co-deposition of NPB and MoO_3 . As shown in Supplementary Fig. S1, an additional absorption peak can be evidently observed in the UV-vis absorption spectra at 495 nm [13], authenticating the presence of CT complexes of $\text{MoO}_3^+:\text{NPB}^-$ [31]. While the doping concentration of MoO_3 increased, intensity of this absorption peak was also increased, which is attributed to more CT complexes produced, suggesting that extra free holes were generated in NPB: MoO_3 . As seen in the inset of Supplementary Fig. S1, intensity of this additional peak promptly increased as the dopant concentration increased, then seemed to gradually saturate by further rising MoO_3 concentration over 33%. In accordance with the increase in free holes, the Fermi level shift of NPB toward the HOMO level was projected [32], hence lowered the energy difference between Fermi level and HOMO energy state [33]. Consequently, by gradually employing different MoO_3 doping concentrations from high to low from the anode to HTL, a descending arrangement of HOMO energy level should be expected. To confirm our concept, appropriate concentration selection of each sub-layer in HIBL is crucial for the best hole injection. We therefore fabricated a batch of OLEDs with a wide variety of concentration combinations. The thickness of HIBL was nominated as 6 nm, which is at a typical thickness range of injecting layers. Supplementary Fig. S2 presents the J - V characteristics of OLEDs with various HIBL concentration combinations. Device with combination 35%/20% demonstrated the optimum J - V properties compared to other devices under identical processing conditions, indicating superior hole injecting capability. Based on these results, we selected 35%/20% as the concentration combination of our HIBL.

Fig. 1 (a) and (b) shows the J - V - L characteristics of the fluorescent green OLEDs with the structure of ITO/HIL/NPB(40 nm)/ Alq_3 (40 nm)/LiF(1 nm)/Al(140 nm). For comparison, devices with typical TMOs-based HILs consist of pure MoO_3 , uniformly doped NPB: MoO_3 (25%) and also device without HIL (normal device) were fabricated. According to literature, optimum thickness of pure MoO_3 HIL is usually around 1–2 nm, therefore, 1-nm is chosen as the thickness of pure MoO_3 HIL. Diverse HIL schematic diagrams of devices A–D are shown in Fig. 2. It can be clearly seen that both light emission and charge injection properties in device D showed inferior behavior. This is attributed to a rather high hole injection barrier between ITO anode and hole transport layer NPB. In contrast, with MoO_3 -based HILs, devices A–C demonstrated enhanced charge injection characteristics and also much higher luminance at a given current density. Particularly, device with the stepwise structure (device A) showed extraordinary hole injection characteristics and light emission compared to other devices. Regarding J - V curves, driving voltage at current density of 100 mA/cm^2 for device A was 5.88 V, while for device B was 6.20 V, for device C was 6.42 V and for device D was 8.89 V. In case of luminance, the maximum luminance achieved for device A was 31,150 cd/m^2 (at 8.5 V), which was superior to device B 22,010 cd/m^2 (at 10 V), device C 20,311 cd/m^2 (at 10 V) and device D 11,930 cd/m^2 (at 10.5 V). Moreover, luminance at 300 mA/cm^2 for devices A–D were 10943, 9757, 9401 and 9266, respectively. These results denote that device with our stepwise HIBL should exhibit improved luminance efficiency. Fig. 1(c) presents the current efficiency and power efficiency as a function of current-density (η - J) for devices A–D. It is evident that device D showed a lower efficiency (2.59 cd/A at 100 mA/cm^2) due to higher hole injection barrier at anode/organic interface. On the other hand, devices A–C showed 3.37, 3.10 and 3.11 at 100 mA/cm^2 , respectively. The power efficiency also showed similar tendency with efficiencies 2.02, 1.57, 1.45 and 0.98 lm/W for devices A–D respectively at 100 mA/cm^2 . The enhanced current efficiency is due to more holes injected from anode as hole injection is significantly improved by stepwise HIBL. However, the maximum current efficiency increased was very little, i.e., from 3.13 (Device D at 380 mA/cm^2) to 3.65 (Device A at 315 mA/cm^2) cd/A , which we inferred it as a result influenced by unbalanced charge effects

Download English Version:

<https://daneshyari.com/en/article/7873435>

Download Persian Version:

<https://daneshyari.com/article/7873435>

[Daneshyari.com](https://daneshyari.com)