Effective hole-injection layer for non-doped inverted top-emitting organic light-emitting devices

Yanlong Meng, Wenfa Xie, Ning Zhang, Shufen Chen, Jiang Li, Wei Hu, Yi Zhao[*], Jingying Hou, Shiyong Liu[*]

State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012, PR China

Abstract

Non-doped inverted top-emitting organic light-emitting diode with high efficiency is demonstrated through employing an effective hole-injection layer composed of MoO_x. One reason for high efficiency lies on the energy-level matching between MoO_x and hole-transport, and another is due to the Ohmic contact formed between MoO_x and Ag. Both of them lead to an improvement of the hole-injection capability from Ag top anode. Moreover, the symmetrical current of "hole-only" device with MoO_x shows better hole-injection capability, which is independent of the deposition sequence. The optimized device with MoO_x hole-injection layer exhibits maximum current efficiency of 3.7 cd/A at a raised luminance level of 14,900 cd/m^2 and a maximum luminance of 47,000 cd/m^2 under 18 V.
© 2008 Elsevier Ltd. All rights reserved.

PACS: 78.60.Fi; 82.45.Mp; 85.60.Jb

Keywords: ITEOLEDs; MoO_x; Hole-injection

1. Introduction

Since the organic light-emitting diodes (OLEDs) were reported, some structures were developed to enhance the efficiency and satisfy the commercial request in display field [1]. From the standpoint of device integration based on a-Si substrate, it is advantageous to make use of *n*-channel field-effect transistors backplanes in AM-OLEDs, demanding the TEOLED structure featuring the bottom contact as cathode: namely, an inverted TEOLED (ITEOLED) which needs a reflective cathode at the bottom and a transparent anode on the top. However, unlike the conventional OLEDs, changing the formation process of the contact between electrode and organic layer also leads to a degraded carrier-injecting capability [2,3]. Hence, enhancing the charge injection from electrode has become an important issue for ITEOLEDs.

Up to now, there are many methods developed to improve the efficiency of ITEOLEDs [4–6]. To reduce the loss of light outcoupled, it is desirable to use a transparent top anode. Then, ITO top anode, which can meet these requests, was used extensively [7,8]. In spite of the enhancement for carrier injection, from the view of processing, it is still intractable to sputter a transparent ITO anode on the top of the organic layer because of the intense radiate damage [9,10]. To relax the damage brought from the magnetron sputter deposition, various buffer layers are introduced [7,8,11]. The metals with high work function are introduced as top-anode, which can be deposited by thermal evaporation with less radiate damage [12]. But the most of metals with high work function shows strong diffusing capability, which may leads to an unsatisfying result [13].

In earlier reports, transition-metal oxides, such as IrO_x, RuO_x, with a high valence band were used for anode modification in conventional OLEDs, which showed excellent performance for enhancing the efficiency [14–16]. Among these transition-metal oxides, MoO_x is easy to be

---

*Corresponding authors.

E-mail addresses: mengyanlong@yeah.net (Y. Meng),
syliu@jlu.edu.cn (S. Liu).

acquired through thermal evaporation. But it has not been employed in ITEOLEDs yet. Here, we demonstrate the ITEOLEDs employing MoO_x as effective hole-injection layer and protective layer shows excellent performance.

2. Experimental

The structures of devices were shown in Fig. 1. The devices were built on SiO_2-coated Si wafers. 45-nm-thick Ag and 10-nm-thick Mg:Al [17] were deposited on the substrate in sequence. Ag was used to ensure the high reflectivity of the bottom cathode. The sample was then loaded into another evaporation chamber for depositions of LiF and organic layers. Following deposition of an ultrathin LiF layer on the top of Mg:Al cathode, the organic multilayer, which consist of 77-nm-thick Tris-8-hydroxyquinoline aluminum (Alq_3) as the emitting layer and electron-transport layer, 10-nm-thick alpha-napthylphenylbiphenyl diamine (NPB) as the hole-transport layer, 35-nm-thick 4,4′,4″-Tris(3-methylphenylpheny-lamino)triphenylamine (m-MTDATA) as the hole-injecting, were deposited sequentially. A 15-nm-thick MoO_x layer was then deposited in succession. At last, a 15-nm-thick Ag was deposited in another evaporation chamber through shadow mask, defining an active area of $0.04\,cm^2$. The current–voltage (J–V) and luminance–voltage (L–V) characteristics were measured by Keithly 2400 and PR 650, respectively. All measurement proceeded directly in atmosphere.

3. Results and discussion

To demonstrate the capability of the MoO_x for hole-injection, two "hole-only" devices were fabricated firstly: (1) Ag/MoO_x (15 nm)/NPB (120 nm)/MoO_x (15 nm)/Ag;
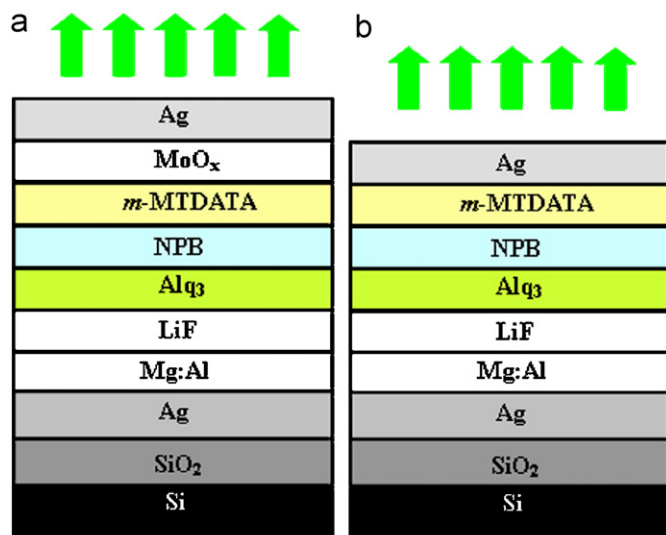
(2) Ag/Ag_2O/NPB (120 nm)/MoO_x(15 nm)/Ag. Seen from Fig. 2(a), the hole-injection current from the Ag/MoO_x bottom electrode with forward-bias voltage is symmetrical with that from MoO_x/Ag top electrode with reverse-bias voltage, indicating the independence of the depositing sequence. But, the hole-injection current from MoO_x/Ag top electrode is stronger than that from the Ag/Ag_2O bottom electrode as shown in the current density–voltage curve of Device 2, confirming the better hole-injection characteristic of the MoO_x layer than Ag_2O which is known as excellent hole-injection layer [18]. MoO_x is a wide-gap semiconductor with a band gap of 3–3.1 eV, and an electron affinity of around 2.2 eV, which implied a valence band at around 5.3 eV [19]. The level matching
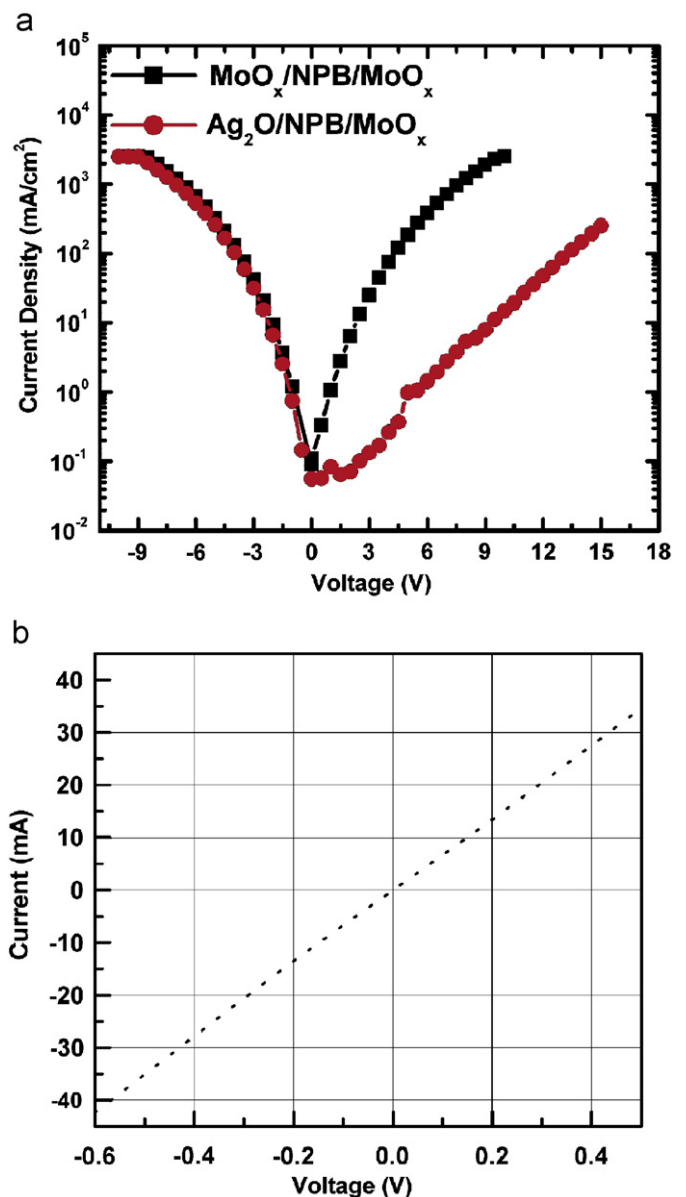

Fig. 1. Schematic of the devices structures with (a) and without (b) MoO_x layer.

Fig. 2. (a) Compares J–V characteristic of "hole-only" devices: Ag/MoO_x (15 nm)/NPB (120 nm)/MoO_x (15 nm)/Ag (closed square); Ag/Ag_2O/NPB (120 nm)/MoO_x (15 nm)/Ag (closed circle); (b) current–voltage (I–V) characteristic of Ag/MoO_x (600 nm)/Ag device.

between MoO_x and NPB leads to an excellent hole-injection. Since the Ag was deposited by thermal evaporation on top of MoO_x, the diffusion of Ag likely to form a doping layer at the interface. The width of the depletion region between metal and MoO_x junction decreases as the doping concentration increases. As a result, the probability of tunneling the barrier increases [20], and an Ohmic contact is likely to be achieved at the metal and MoO_x interface just as shown in the $I-V$ characteristic of Ag/MoO_x (600 nm)/Ag device (see Fig. 2(b)).

For comparison, a controlled device without MoO_x but the same cathode and organic structure was fabricated. The thickness of each organic layer in devices is optimized to meet the resonant conditions according to microcavity effect. Fig. 3 shows the $J-V$ characteristic of devices with and without MoO_x. It is evident that the presence of MoO_x leads to a substantial improvement for hole-injection which is enhanced two times approximately. The improvement is due to the preferable energy-level matching between MoO_x and m-MTDATA [20,21]; seen from $L-V$ curve in Fig. 3, the threshold-voltage of device is debased from 13 to 5.5 V as a result of improvement of hole-injection. At a driving voltage of 9 V, the device with MoO_x has a current density of 6.6 mA/cm^2 leading to a brightness of 100 cd/m^2. By contrast, there are almost no emissions in the controlled device. Moreover, the controlled device shows a very quick degradation. We speculate the major reason for degradation is the energy-level mismatching between Ag and m-MTDATA. In addition, the diffusion of Ag when it was deposited on the top of organic layer also leads instability of device. At last, it is obvious the insertion of MoO_x between Ag and m-MTDATA not only improve the hole-injection but also protect the organic layers from the further diffusion of Ag. Finally, the maximum luminance of 47,000 cd/m^2 is acquired at 18 V, which is comparable with the conventional ITO device. Fig. 4 shows current efficiency and power efficiency (inset of Fig. 4) versus voltage characteristics, respectively. The efficiency of device with MoO_x ascends linearly as the voltage increase,
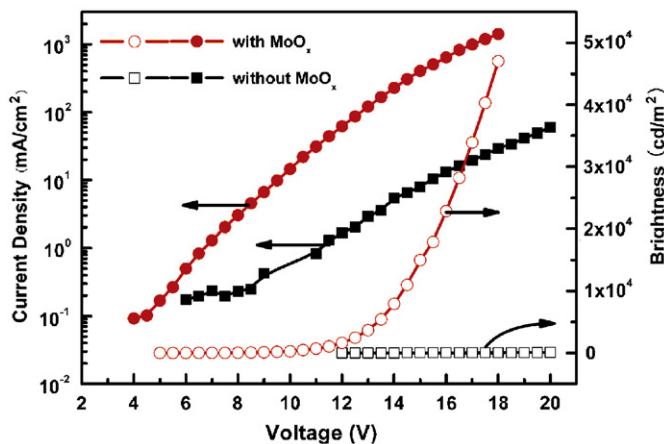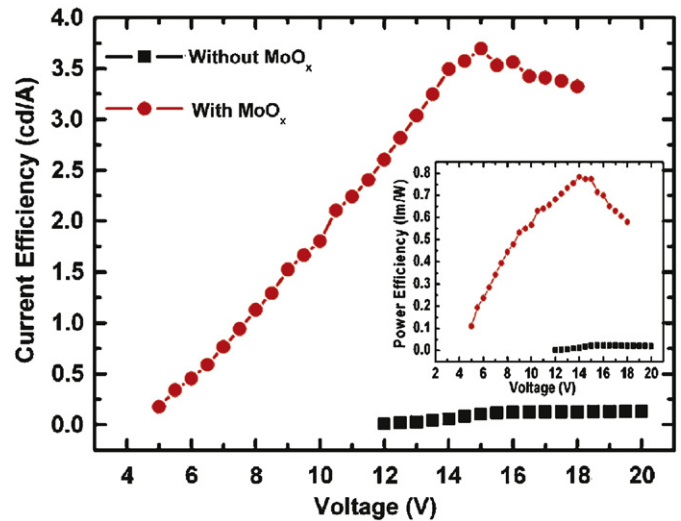
Fig. 4. Compare current efficiency versus voltage and power efficiency versus voltage (inset) characteristic of devices with and without MoO_x.
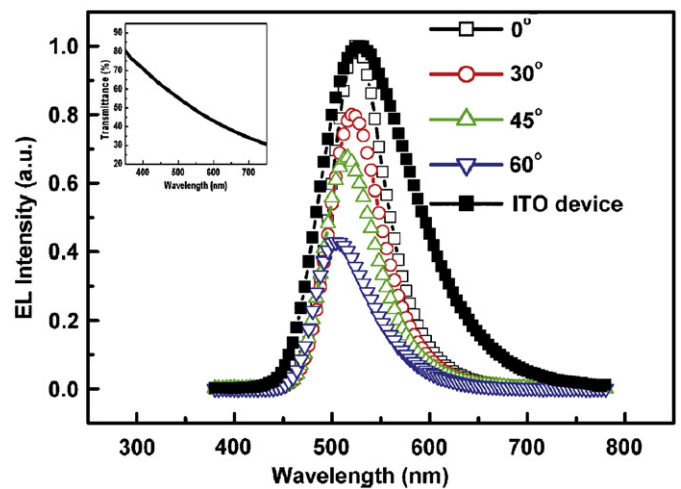
Fig. 5. EL spectra of the inverted top-emitting organic light-emitting device with different viewing angle (0°, 30°, 45°, and 60°) off the surface normal in comparison with conventional ITO bottom-emitting device. Inset: Transmittance of 15-nm-thick Ag anode.

while the controlled device keeps a bad efficiency, which is less than 0.2 cd/A. The inverted top-emitting device with MoO_x achieves a maximum current efficiency of 3.7 cd/A at 15 V and power efficiency of 0.8 lm/W at 14 V. Since the employing of MoO_x enhances the hole-injection capability of top Ag anode in the inverted top-emitting device, it seems, unlike the conventional top-emitting devices, which have more efficiency electron injection, the unbalanced electron injection is the dominant reason for enhancing the efficiency further.

Fig. 5 illustrates the normalized electroluminescence (EL) spectra at viewing angles of 0°, 30°, 45°, and 60° off the surface normal. The microcavity effect attributed to bottom electrode with high reflectance and 15-nm-thick top electrode exhibiting 50% transmittance (see inset of Fig. 5) at 520 nm changes the spectra outputted. As a result, the spectra are narrowed compared with conventional device with ITO anode based on

Fig. 3. Current density versus voltage ($J-V$) and luminance versus voltage ($L-V$) characteristics of devices with and without MoO_x layer.

glass substrate, and the peak of the EL spectra shifts 20 nm as the viewing angle increase from $0°$ to $60°$.

4. Conclusions

In summary, the using of MoO_x thin layer as an effective hole-injection layer for inverted top-emitting organic light-emitting devices with Ag anode is reported. The better hole-injection capability, which is independence of deposition sequence with the Ag anode is demonstrated. In such device, the presence of MoO_x leads to a maximum luminance and efficiency of $47,000 \, cd/m^2$ (18 V) and $3.7 \, cd/A$ (15 V), respectively. The improvement is attributing to the satisfying energy-level matching and Ohmic contact formed between Ag and MoO_x. Furthermore, MoO_x also provided a protection for organic layer from the diffusion of Ag when it was deposited by thermal evaporation. The study here afford us more candidates for choosing better anodes which are easier to be deposited with less irradiation and diffusion for inverted top-emitting devices.

Acknowledgments

References

[1] V. Bulović, G. Gu, P.E. Burrows, S.R. Forrest, M.E. Thompson, Nature (London) 380 (1996) 29.
[2] H. Heil, J. Steiger, S. Karg, M. Gastel, H. Ortner, H. von Seggern, J. Appl. Phys. 89 (2001) 420.
[3] L.S. Hung, S.T. Lee, Mater. Sci. Eng. B 85 (2001) 104.
[4] Ta-Ya Chu, Szu-Yi Chen, Jenn-Fang Chen, Chin H. Chen, Jpn. J. Appl. Phys. 45 (2006) 4948.
[5] S. Kho, S.Y. Sohn, D.G. Jung, Jpn. J. Appl. Phys. 42 (2003) L552.
[6] Lintao Hou, Fei Huang, Wenjin Zeng, Junbiao Peng, Yong Cao, Appl. Phys. Lett. 87 (2005) 153,509.
[7] T. Dobbertin, M. Kroeger, D. Heithecker, D. Schneider, D. Metzdorf, H. Neuner, E. Becker, H.-H. Johannes, W. Kowalsky, Appl. Phys. Lett. 82 (2003) 284.
[8] V. Bulović, P. Tian, P.E. Burrows, M.R. Gokhale, S.R. Forrest, Appl. Phys. Lett. 70 (1997) 2954.
[9] Ho Won Choi, Soo Young Kim, Woong-Kwon Kim, Jong-Lam Lee, Appl. Phys. Lett. 87 (2005) 082,102.
[10] S.-W. Park, J.-M. Choi, E. Kim, S. Im, Appl. Surf. Sci. 244 (2005) 439.
[11] T. Dobbertin, O. Werner, J. Meyer, A. Kammoun, D. Schneider, T. Riedl, E. Becker, H.-H. Johannes, W. Kowalsky, Appl. Phys. Lett. 83 (2003) 5071.
[12] X. Zhou, M. Pfeiffer, J.S. Huang, J. Blochwitz-Nimoth, d.S. Qin, A. Werner, J. Drechsel, B. Maennig, K. Leo, Appl. Phys. Lett. 81 (2002) 922.
[13] X.Z. Wang, Z.T. Xie, X.J. Wang, Y.C. Zhou, W.H. Zhang, X.M. Ding, X.Y. Hou, Appl. Surf. Sci. 253 (2007) 3930.
[14] Shizuo Tokito, Koji Noda, Yasunori Taga, J. Phys. D: Appl. Phys. 29 (1996) 2750.
[15] Chenfeng Qiu, Zhilang Xie, Haiying Chen, Man Wong, Hoi Sing Kwok, J. Appl. Phys. 93 (2003) 3253.
[16] Soo Young Kim, Jeong Min Baik, Hak Ki Yu, Jong-Lam Lee, J. Appl. Phys. 98 (2005) 093707.
[17] S.Y. Kim, J.-L. Lee, Appl. Phys. Lett. 88 (2006) 112,106.
[18] Chieh-Wei Chen, Ping-Yuan Hsieh, Huo-Hsien Chiang, Chun-Liang Lin, Wu Han-Ming Wu, Chung-Chih Wu, Appl. Phys. Lett. 83 (2003) 5127.
[19] K.J. Reynolds, J.A. Barker, N.C. Greenham, R.H. Friend, G.L. Frey, J. Appl. Phys. 92 (2002) 7556.
[20] Chih-Wei Chu, Sheng-Han Li, Chieh-Wei Chen, Vishal Shrotriya, Yang Yang, Appl. Phys. Lett. 87 (2005) 193508.
[21] Shih-Fang Chen, Ching-Wu Wang, Appl. Phys. Lett. 85 (2004) 765.