Organic Electronics 14 (2013) 1177–1182

Contents lists available at SciVerse ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Enhancement of electroluminescence efficiency and stability in phosphorescent organic light-emitting diodes with double exciton-blocking layers

Chun-Hong Gao, Dong-Ying Zhou, Wei Gu, Xiao-Bo Shi, Zhao-Kui Wang *, Liang-Sheng Liao *

Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Institute of Functional Nano & Soft Materials (FUNSOM), Soochow University, Suzhou, Jiangsu 215123, China

ARTICLE INFO

Article history:
Received 21 December 2012
Received in revised form 7 February 2013
Accepted 13 February 2013
Available online 27 February 2013

Keywords:
Double exciton-blocking layer
Organic light-emitting diode
Phosphorescence
Operational stability
Charge carrier
Exciton formation zone

ABSTRACT

A high efficiency phosphorescent organic light-emitting diode (OLED) has been fabricated by introducing a double exciton-blocking layer (d-EBL) between the hole-transporting layer and the light-emitting layer in the device. The device exhibits a yellow emission with a maximum current efficiency of 58.5 cd/A at 117 cd/m^2, corresponding to the power efficiency of 50.9 lm/W, which is two times improved compared with that of devices having only one traditional single exciton-blocking layer (s-EBL). The efficiency improvement has been investigated through the electroluminescence (EL) spectral analyses in the phosphorescent guest-doped and the non-doped OLEDs. The results demonstrate that the electrons are blocked and the excitons are confined more effectively in the d-EBL-based devices than that in the s-EBL-based devices. In addition, over two times improvement in the lifetime is also achieved in the devices with the d-EBL compared with the devices having a traditional s-EBL.

1. Introduction

Since the first double-layer organic light-emitting diode (OLED) was reported by Tang and VanSlyke in 1980s [1], many kinds of device structures, such as structures with carrier confining layers [2–4], triplet quantum well [5], emissive layer with double dopants [6,7], emissive layer with mixed hosts [8], emissive layer with different doping profile [9,10] and tandem structure with multiple emission units [11–13] have been developed to achieve high efficiency and long-term stability in OLEDs. However, continual improvement in device structure to further enhance efficiency and stability in OLEDs is still necessary for widely commercial applications of the OLEDs in displays and solid-state lighting.

In general, two types of excitons (singlet and triplet) can be created upon charge carrier recombination during the light-emitting process in OLEDs. For a phosphorescent OLED, both the singlet and triplet excitons can be harvested by introducing heavy metal atom (such as iridium) in the phosphorescent dopant, leading to almost 100% internal quantum efficiency [14–16]. Three energy processes dominate the light-emitting mechanism in phosphorescent OLEDs. The first is a Förster energy transfer process that can make the energy of host singlet transfer to dopant singlet. The second is an intersystem crossing process in the dopant corresponding to the decaying from the singlet to the triplet. The last one is Dexter energy transfer process in which the triplet of the host can transfer to the triplet of the dopant. The singlet excitons formed in phosphorescent OLEDs will decay transiently because of its shorter lifetime than the triplet. If the triplet energy of the adjacent layer of the emitting layer (EML) is smaller than that of the host in EML, triplet excitons are inclined to diffuse into the adjacent layer owing to their longer diffusion

* Corresponding authors. Tel.: +86 512 65880945 (L.-S. Liao).
E-mail addresses: zkwang@suda.edu.cn (Z.-K. Wang), lsliao@suda.edu.cn (L.-S. Liao).

1566-1199/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.orgel.2013.02.013

length [17–20]. Therefore, confining the excitons in EML is very important for high efficiency OLEDs. Two conditions need to be satisfied to confine the triplet excitons in the EML. Firstly, the host material should have higher triplet energy than that of the dopant. Secondly, the triplet energy of the material near the EML should be higher than that of the host material. Otherwise, it is possible for the triplet excitons in the EML to diffuse into the hole-transporting layer (HTL) or the electron-transporting layer (ETL) due to their long diffusion length caused by long lifetime. A single exciton-blocking layer (s-EBL), with triplet energy higher enough than that of the host material, is usually inserted at the HTL/EML interface [21–25], or the EML/ETL interface [26–28] to block the excitons out of the EML. However, we found that a s-EBL cannot block the exciton diffusion completely.

In this paper, we demonstrated a phosphorescent OLED with improved device efficiency and stability by introducing a double exciton-blocking layer (d-EBL) comprised of different materials between the hole-transporting layer and the light-emitting layer. Besides the role of exciton confinement, the other functions of the d-EBL relating to the improvement of the device efficiency and stability were also discussed.

2. Experiment

All the phosphorescent OLEDs are fabricated on precleaned and UV-ozone treated indium-tin-oxide (ITO) electrode on a glass substrate. The organic and metallic layers are subsequently deposited under a base pressure of 2×10^{-6} Torr. N,N′-bis(naphthalen-1-yl)-N,N′-bis(phenyl)-benzidine (NPB) is used as the HTL, 4,4′,4″-tri(N-carbazolyl)triphenylamine (TCTA, $E_T = 2.85$ eV) or 4,4′-bis(carbazol-9-yl)biphenyl (CBP, $E_T = 2.65$ eV) is used as an EBL, 2′,2″-(1,3,5-benzinetriyl)-Tris(1-phenyl-1-H-benzimidazole) (TPBi, $E_T = 2.74$ eV) is used as both the ETL and the host of the EML, Iridium(III)bis(4-phenylthieno[3,2-c]pyridinato-N,$C^{2\prime}$)acetylacetonate [29–31] (PO-01, $E_T = 2.23$ eV, shown in Fig. S1 in the Supporting Information) is used as a phosphorescent dopant. Liq and Al are deposited as a bilayer cathode. The chemical structures of the materials used in this work are shown in Fig. S2 in the Supporting Information. Three groups of devices (listed in Table 1) are fabricated to investigate the role and the mechanism of d-EBL in OLED. To exhibit the remarkable enhancement in the EL performance of d-EBL, four phosphorescent OLEDs (Group A) with or without different EBLs are fabricated as: ITO/MoO_3 (10 nm)/NPB (75 nm)/X/TPBi: PO-01 3 wt.% (20 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm), where "X" stands for 10 nm NPB (Device A-1, without any EBL), 10 nm TCTA (Device A-2, with s-EBL), 10 nm CBP (Device A-3, with s-EBL), and 10 nm TcTa/4 nm CBP (Device A-4, with d-EBL), respectively. To further verify the thickness dependence of EBLs on the electron blocking behavior, two kinds of devices in Group B with or without 4 nm CBP using TcTa as HTL and DCJTB as a charge sensing layer are fabricated with the following structures: ITO/MoO_3 (10 nm)/TcTa ($85 - x_1$ nm)/DCJTB (0.2 nm)/TcTa (x_1 nm)/CBP (y_1 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm), where $(x_1, y_1) = (4, 0), (7, 0), (10, 0), (20, 0), (4, 4), (7, 4), (10, 4)$ and (20, 4) for Devices B-1, B-2, B-3, B-4, B-5, B-6, B-7 and B-8, respectively. To evaluate the triplet exciton diffusion length in devices with or without 4 nm CBP, PO-01 is introduced as an exciton diffusion sensing layer in ten non-doped devices (Group C): ITO/MoO_3 (10 nm)/TcTa ($85 - x_2$ nm)/PO-01 (0.2 nm)/TcTa (x_2 nm)/CBP (y_2 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm), where $(x_2, y_2) = (1, 0), (4, 0), (7, 0), (10, 0), (20, 0), (1, 4), (4, 4), (7, 4), (10, 4)$ and (20, 4) for Devices C-1, C-2, C-3, C-4, C-5, C-6, C-7, C-8, C-9 and C-10, respectively.

A constant current source (Keithley 2400 SourceMeter) combined with a photometer (Photo Research SpectraScan PR 655) is used to measure the electroluminescence (EL) characteristics. An optical microscope is used to calculate the active area of each device (0.1 cm^2) to reduce the possible device-to-device error. The half lifetime (T_{50}), defined as the duration time from the start of testing to the time at which luminance is reduced to 50% of the initial value under a given testing condition.

3. Results and discussion

3.1. Improved device performance by double exciton-blocking layers (d-EBLs)

The EL performances of group A devices (Devices A-1, A-2, A-3 and A-4) are shown in Fig. 1. The current density–voltage (J–V) characteristics are shown in Fig. 1a. All the devices exhibit similar trend in J–V curves and shows small voltage difference at every current density. The voltage difference in the J–V characteristics is attributed to the different carrier mobility and injection barrier height. The reference Device A-1 without EBL exhibits the lowest voltage at each specific current density owing to the high hole mobility (1×10^{-3} cm^2/V s) [32] and a suitable HOMO le-

Table 1
Device layer structures.

Group	Device structure
A	ITO (110 nm)/MoO_3 (10 nm)/NPB (75 nm)/X/TPBi: PO-01 3 wt.% (20 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm) X: 10 nm NPB (A-1); 10 nm TCTA (A-2); 10 nm CBP (A-3); 10 nm TCTA/4 nm CBP (A-4)
B	ITO (110 nm)/MoO_3 (10 nm)/TcTa ($85 - x_1$ nm)/DCJTB (0.2 nm)/TcTa (x_1 nm)/CBP (y_1 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm) $(x_1, y_1) = (4, 0), (7, 0), (10, 0), (20, 0), (4, 4), (7, 4), (10, 4)$ and (20, 4) for Devices B-1, B-2, B-3, B-4, B-5, B-6, B-7 and B-8, respectively
C	ITO (110 nm)/MoO_3 (10 nm)/TcTa ($85 - x_2$ nm)/PO-01 (0.2 nm)/TcTa (x_2 nm)/CBP (y_2 nm)/TPBi (50 nm)/Liq (2 nm)/Al (100 nm) $(x_2, y_2) = (1, 0), (4, 0), (7, 0), (10, 0), (20, 0), (1, 4), (4, 4), (7, 4), (10, 4)$ and (20, 4) for Devices C-1, C-2, C-3, C-4, C-5, C-6, C-7, C-8, C-9 and C-10, respectively