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Organic light-emitting diodes with 2-(4-biphenylyl)-5(4-tert-butyl-phenyl)-1,3,4-oxadiazole layer inserted between hole-injecting and hole-transporting layers

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ABSTRACT

Organic light-emitting diodes (OLEDs) were fabricated based on copper phthalocyanine (CuPc) (hole-injecting layer), *N*,*N*′-bis(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) (hole-transporting layer) and tris(8-hydroxyquinoline) aluminum (Alq_3) (emission and electron-transporting layer). A 2-(4-biphenylyl)-5(4-tert-butyl-phenyl)-1,3,4-oxadiazole (PBD) layer was inserted between CuPc and NPB. The effect of different thickness of PBD layer on the performance of the devices was investigated. The device structure was ITO/CuPc/PBD/NPB/Alq_3/LiF/Al. Optimized PBD thickness was about 1 nm and the electroluminescent (EL) efficiency of the device with 1 nm PBD layer was about 48 percent improvement compared to the device without PBD layer. The inserted PBD layer improved charge carriers balance in the active layer, which resulted in an improved EL efficiency. The performance of devices was also affected by varying the thickness of NPB due to microcavity effect and surface-plasmon loss.

1. Introduction

Since Tang and Van Skyle reported the efficient bright emission obtained from a bilayer organic device [1], organic light-emitting diodes (OLEDs) have been intensively investigated in both the scientific community and commercial markets [2]. High-performance OLEDs should have a low operating voltage, high efficiency and relatively good stability. In a typical OLED, the hole experiences a smaller barrier compared to the electron [3], and hole mobility in an organic layer is larger by orders of magnitude than electron mobility [4]. Therefore, there are excessive holes which result in the exciton-quenching in the emission layer. To increase electroluminescent (EL) efficiency, it is necessary to balance charge carriers in the active layer. For undoped OLEDs, various techniques are available to improve the efficiency, such as anode [5,6] or cathode modifications [7–9], electron [10] or hole-blocking [11] and the high electron mobility materials [12]. Cathode modifications have been demonstrated to enhance minority carrier (electron) injection, so as to achieve a balanced injection of carriers. As a result, the total current and efficiency can be improved simultaneously. However, modifications of the anode have been shown to improve the current injection but sacrifice the efficiency, or vice versa [5,13,14]. Insertion of a hole-blocking layer (HBL) between hole-injecting layer (HIL) and hole-transporting layer (HTL) has also been reported to improve the efficiency but not the current [11].

In this study, we report the insertion of a 2-(4-biphenylyl)-5(4-tert-butyl-phenyl)-1,3,4-oxadiazole (PBD) layer between copper phthalocyanine (CuPc) and *N*,*N*′-bis(1-naphthyl)-*N*,*N*′- diphenyl-1,1′-biphenyl-4,4′-diamine (NPB). The efficiency of the device with the optimized thickness of PBD layer was improved by about 48 percent compared to the device without PBD layer. The band diagram [15,16] of our improved design is shown in Fig. 1. The effect of the different thicknesses of PBD and NPB on the performance of the devices was discussed.

2. Experiment

The resistance of ITO-coated glass sheets is about 20 Ω/square. The sheets were used as the substrate for the devices and cleaned by routine procedure included sonication in detergent, de-ionized water, acetone and alcohol, finally irradiated in a UV–ozone chamber. The device with the fundamental structure

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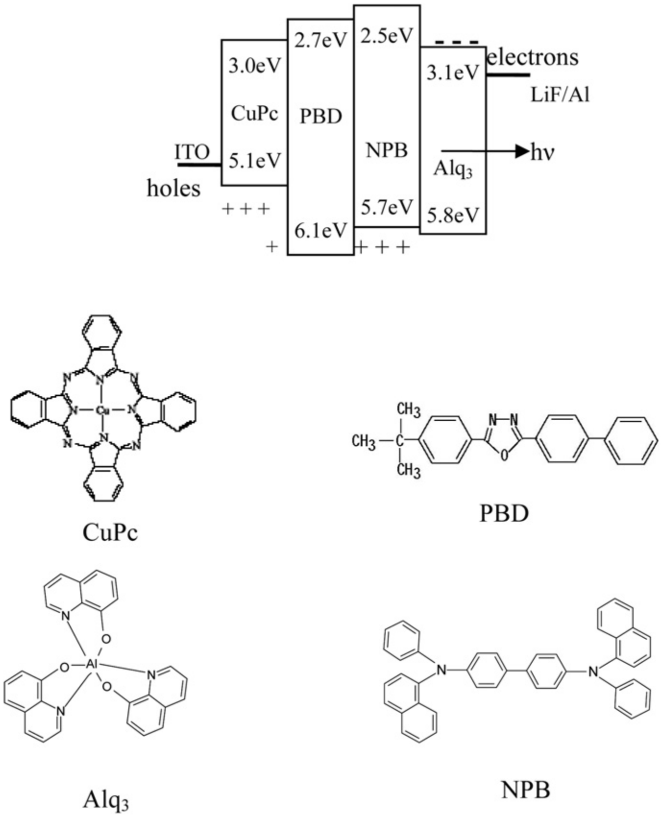

Fig. 1. Band diagram of the devices and chemical structures of materials in the experiment.

of ITO/CuPc(15 nm)/NPB(40 nm)/Alq_3(60 nm)/LiF(0.5 nm)/Al was fabricated by using successive vacuum-depositions of hole-injecting layer(HIL, CuPc), hole-transporting layer (HTL, NPB), bi-functional (emissive and electron-transporting layer, EML&ETL) layer (tris(8-hydroxyquinoline) aluminum, Alq_3), and bilayer cathode (LiF/Al) on the top of the ITO glass substrate. As shown in Fig. 1, PBD layer with the thicknesses of 0, 1, 3 and 5 nm was inserted at the CuPc/NPB interface. The devices with structures of ITO/CuPc(15 nm)/PBD(0, 1, 3 and 5 nm)/NPB (40 nm)/Alq_3 (60 nm)/LiF (0.5 nm)/Al and ITO/CuPc (15 nm)/PBD (1 nm)/NPB (10, 20, 30 and 40 nm)/Alq_3 (60 nm)/LiF (0.5 nm)/Al were prepared. The evaporation rates were about 0.1 nm/s for CuPc, PBD, NPB and Alq_3, 0.04 nm/s for LiF and 0.2–0.3 nm/s for Al, respectively, under a base pressure of about 1×10^{-4} Pa without breaking the vacuum. Except for PBD and NPB, all other materials were deposited simultaneously for each four devices. This avoided the uncertainties in comparing devices fabricated with different evaporation processes. A quartz–crystal oscillator monitored the thickness and deposition rate of the materials.

The current–voltage and luminance characteristics of these devices (active area of each device is 3×3 mm^2) were measured by a Keithley 2410 source meter and a PR-650 spectrometer. All the measurements were carried out in the air at room temperature without encapsulation.

3. Results and discussion

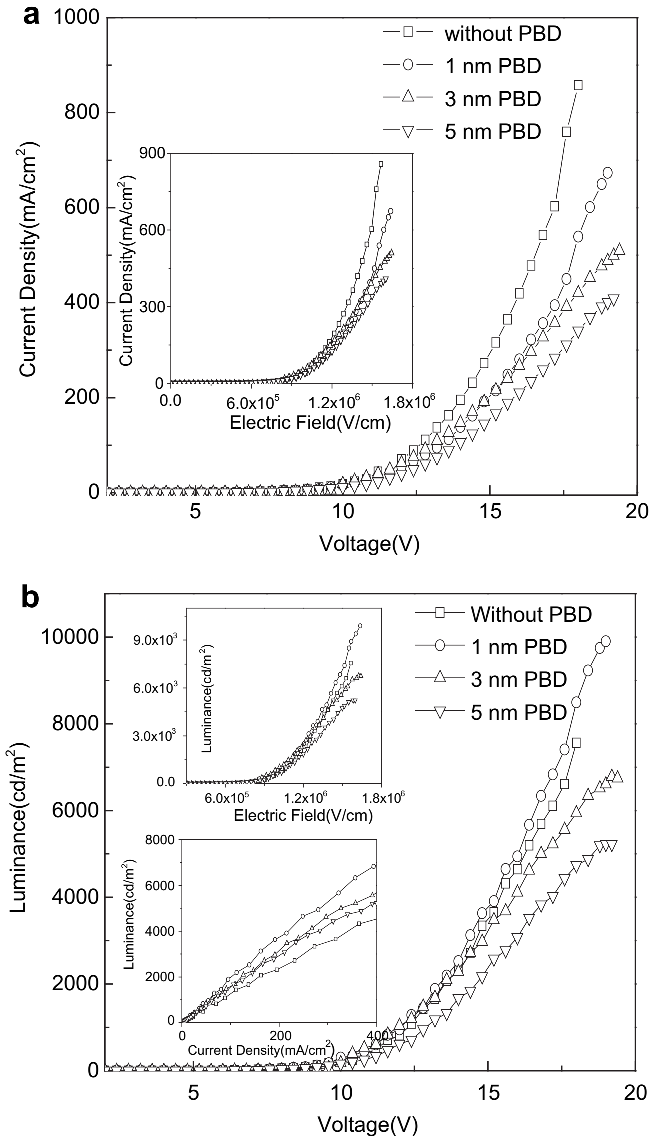

Fig. 2. (a) Current Density vs. Voltage (*J–V*) and Current Density vs. Electric Field (*J–E*) (inset) characteristics, and (b) Luminance vs. Voltage (*L–V*), Luminance vs. Electric Field (*L–E*) (above inset) and Luminance vs. Current Density (*L–J*) (below inset) characteristics for devices with various thicknesses of PBD layer, respectively.

Fig. 2(a) and (b) show the current density–voltage (*J–V*) and luminance–voltage (*L–V*) curves of the devices with different PBD thicknesses, respectively. From Fig. 2, it can be seen that both current density and luminance shift toward higher voltages as the thickness of PBD layer increases. As shown in the insets of Fig. 2, the behavior of current density–electric field (*J–E*) is identical to *J–V*. If the PBD layer does not block holes, the current densities should be identical for all devices at the same electric field. Therefore, the reason why the current density shifts toward higher voltage is not that the PBD layer consumes partial voltage, since the thickness of PBD layer is very thin (less than 5 nm), far less than that of the whole device (115 nm). The highest occupied molecular orbital (HOMO) of PBD is higher than that of NPB (shown in Fig. 1), so the PBD layer blocked holes injecting into NPB layer. The probability of hole injection from CuPc to NPB would be decreased with the increase of PBD thickness. The number of blocked holes is increased with the thickness of PBD. Consequently, in order to achieve the same current density in the devices, the applied voltage was increased as the thickness of PBD increased. The turn-on voltage (5.23, 5.29, 5.40 and 5.67 V for 0, 1, 3 and 5 nm PBD layers at 1 cd/m^2, respectively) did not shift significantly. The current densities to obtain a luminance of 4000 cd/m^2 for devices with 0, 1, 3 and 5 nm