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Reduced turn-on voltage and improved efficiency with free interfacial energy barrier in organic light-emitting diodes

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ABSTRACT

Interfacial doping is introduced in order to fabricate both fluorescent and phosphorescent organic light-emitting diodes (FOLEDs and PhOLEDs) with low turn-on voltage, high efficiency and low efficiency roll-off, which employed the doping profile of transporting layer with the host of the light emitting layer (EML). Both hole and electron transport layer doped EML are investigated. Through this method, the injection and transport of carriers can be fine-tuned, and the interfacial energy barriers from the transporting layer to EML are effectively eliminated. Furthermore, a better balance of holes and electrons in the combination zone can be obtained by manipulating the molar ratio of the interfacial doping layer. By combing all these factors, the FOLEDs achieved a very low turn-on voltage of 2.3 V, a high current efficiency of 9.90 cd/A and an external quantum efficiency of 3.12% with low roll-off. In addition, a low turn-on voltage of 2.8 V and a high current efficiency of 54.06 cd/A, which increased by 45% compared to reference device in PhOLEDs have also been achieved.

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1. Introduction

Organic light-emitting diodes (OLEDs) have many advantages of high electroluminescence (EL) efficiency, flexibility and low manufacturing costs, holding great potential for flat-panel displays and solid-state lighting applications [1–3]. For commercial use, it is important to improve the efficiency and lifetime of both fluorescent and phosphorescent OLEDs [4].

One of the most fundamental characteristics of OLEDs is the electrical one represented by the current density–voltage (J-V) relationship, which is controlled by the charge injection at the electrode/organic and organic/organic interfaces [5]. And the charge injection is determined by the energy level of the materials. Therefore, lowering the energy barrier at the interfaces becomes a very important factor to facilitate efficient charge injection through the interfaces [6].

It can be achieved through proper modification of a metal electrode with matched work function of the metal, and the energy level of the organic material. The synthesis of organic materials

which possess superior hole or electron mobility for efficient charge transport has been investigated. Yang et al. employed the ultrathin light emitting layer (EML) to increase the efficiency, but it allowed charge and exciton leakage outside the emissive region, resulting in a decrease in efficiency at high current densities [7]. Many other methods and techniques, including insertion of a thin metal oxide layer [8–10], emissive layers with mixed hosts [11,12], gradient p- and n-doping or nonlinear crossfading doping [13], bipolar host structure and interfacial doping in hole- and electroninjection layers (EIL) [14,15] have also been proposed to improve J-V characteristics with enhanced performance of the devices up to now. Using a bipolar host in the device has many advantages. A great advantage is the improvement of carrier mobilities and balance of electron and hole fluxes in the emitting layer. Bipolar host material with the incorporation of hole-transporting and electron-transporting moieties combines the advantages of both functional units. Many research groups engaged in the design and synthesis of new bipolar materials with good thermal, photophysical and electronchemical properties, and obtained great results [16-21]. However, it has its own limitation: (1) a bipolar molecule has the hole and electron transporting properties simultaneously. But the transporting rate cannot be manipulated to get a better carrier balance; (2) employing bipolar host material may easy to form a heterojunction interface, which may leads to







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carrier accumulation and excitons quench; (3) it is difficult to find a proper bipolar host and the synthesis process is complex.

It is hard work to find a totally matched bipolar host material, so the other kinds of methods from the new design on device structures should be considered. Here in this paper, interfacial doping, a method of hole/electron transporting layer (HTL/ETL) doped EML in OLEDs, is described to lower the interfacial energy barrier and improve device efficiency and operational lifetime. Moreover, by manipulating the molar ratio of the interfacial doping layer, we can obtain a better doping profile and balanced charge combination zone. There are two advantages with this approach. Firstly, the energy barrier is mostly eliminated by interfacial doping, holes and electrons can be directly injected to the EML without encountering any injection barrier. Secondly, the transporting rates of holes and electrons can be adjusted to get balanced charge carriers with a proper doping ratio [22–25]. With the strategy of HTL doped EML, FOLEDs achieve a high current efficiency of 9.90 cd/A and a high external quantum efficiency (EQE) of 3.1% and a low efficiency roll-off with over 2.2% at 1000 cd/ m². Furthermore, employing the same method, PhOLEDs exhibit superb performance, with a 0.2 V lower turn-on voltage and a 45% higher current efficiency compared to the reference device.

2. Experimental details

The OLEDs were fabricated by conventional vacuum deposition of the organic layers and cathode onto an indium–tin–oxide (ITO, 10Ω /sheet, 150 nm) coated glass substrate under a base pressure lower than 2.0×10^{-4} mbar. The ITO glass substrates were cleaned with detergent, de-ionized water, acetone, and isopropanol in turn.

Immediately prior to film formation in vacuum thermal evaporation chamber, the substrates were exposed to a UV–ozone environment for 15 min. The deposition rates and doping concentration of the films were monitored in situ by the calibrated thickness monitors. The typical deposition rates of organic materials and aluminum (AI) were 0.6, and 5.0 Å/s, respectively.

The active area of the devices defined by the overlap between the electrodes was 2 mm × 2 mm in all cases. The current density– voltage–luminance (*J*–*V*–*L*), current efficiency–luminance–power efficiency (Ce–L–Pe) characteristics, and EL spectra were measured and recorded by a testing setup comprising of a Keithley 2400 source-meter and a Minolta PR-650 spectrometer. The luminance was measured using PR-650. All measurements were carried out at room temperature and the devices were kept in N₂ atmosphere to prevent the negative effects of oxygen and humidity.

The structures and energy levels of the devices are shown in Fig. 1. The FOLEDs were fabricated using N,N'-di(naphthalene-1-yl)-N,N'-diphenylbenzidine (NPB) as the HTL, tris-(8-hydroxyquinoline) aluminum (Alq₃) as the ETL, 2,9-dimethyl-4,7diphenyl-1,10-phenanthroline (BCP) as the hole blocking layer (HBL), 8-hydroxyquinolatolithium (Liq) as the EIL and 5,6,11,12tetraphenylnaphthacene (Rubrene) as the dye dopant. In addition, Alq₃ and NPB were served as the host material in Devices A-series and B-series, respectively.

Devices A-series adopted the method of HTL doped EML, employing the structures of ITO/NPB(30 nm)/NPB:Alq₃(x:y,20 nm)/ Alq₃:3 wt%Rubrene(20 nm)/Alq₃(30 nm)/Liq(1 nm)/Al(120 nm). The molar ratios of NPB to Alq₃ were 1:0, 1:1, 2:1 and 1:2 as Devices A0 to A3, respectively. Moreover, ETL doped EML was investigated



Fig. 1. The structures and energy levels of the Devices A(a), B(b) and C(c). The colorful parts show the interfacial doping layer of the corresponding devices.

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