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Effect of bulk and planar heterojunctions based charge generation layers on the performance of tandem organic light-emitting diodes

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ABSTRACT

Tandem organic light-emitting diodes (OLEDs) were fabricated using organic planar and bulk heterojunctions based charge generation layers (CGLs), which were composed of cobalt phthalocyanine (CoPc) and fullerene (C_{60}). The electroluminescent (EL) characteristics of these two kinds of devices were systematically studied. The results showed that, compared to the corresponding devices with planar heterojunction (PHJ) based CGL, the tandem OLEDs with bulk heterojunction (BHJ) based CGL exhibited a dramatic improvement of performance. By investigating the electrical characteristics of CGLs, it was found that more hetero-interfaces introduced in the BHJ blend were beneficial for generating more interfacial dipoles and charge carriers, and the optimized charge transport pathways were favorable to promote both electron and hole mobilities. As a result, the improved charge carrier balance led to the efficiency enhancement of device performance. The results demonstrated the advantageous effect of BHJ blend film for the rational design of CGLs on the realization of high OLEDs performance.

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1. Introduction

Tandem organic light-emitting diodes (OLEDs) have been highlighted as a viable alternative to conventional OLEDs due to their high efficiency, high luminance, tunable color, and long lifetime for application in display and solid-state lighting [1–3]. In a tandem OLED, the adjacent electroluminescent (EL) units are electrically connected by using a charge generation layer (CGL) [4]. The CGL acts as effective anode and cathode to generate charge carriers and to facilitate the injection of charge carriers into adjacent EL units, thus it plays a crucial role in the device performance of tandem OLEDs [5,6]. The property of CGLs should meet the following prerequisite, such as matched energy levels, effective injection channels for charge carriers, high transparency, low series resistance, deposition compatibility and operational stability [7,8]. Thus, the development of high performance CGLs and the exploration of intrinsic properties of CGLs become a critical issue of tandem OLEDs [9–12].

Recently, the organic heterojunction composed of n-type organic semiconductor and p-type organic semiconductor is proposed as an effective CGL in tandem OLEDs to overcome the problem of unmatched deposition compatibility and low operational stability, instead of the previous CGLs incorporated with metals, inorganic and organic materials [13–16]. The donor (D)/acceptor (A) based planar heterojunction (PHJ) and D:A based bulk heterojunction (BHJ) are two main promising charge generation resources for tandem OLEDs [17–19]. Great effort has been made to design effective CGLs so as to realize high efficiency tandem OLEDs. With PHJ based CGL, such as metal-free-phthalocyanine (H_2Pc)/fullerene (C_{60}), zinc phthalocyanine (ZnPc)/C_{60}, 5, 5‴-bis(naphth-2-yl)-2, 2′:5′, 2″:5″, 2‴-quaterthiophene (NaT_4)/C_{60}, pentacene/C_{60} and 4, 4′, 4″-tris(N-(3-methylphenyl)-N-phenylylamino) triphenylamine (m-MTDATA)/1, 4, 5, 8, 9, 11-hexaazatriphenylene hexacarbonitrile (HAT-CN), high efficiency tandem OLEDs are obtained [20,21]. This phenomenon is attributed to optimized energy level alignment, charge transfer and charge carrier transport. For BHJ based CGL, such as ZnPc:C_{60}, H_2Pc:C_{60}, boron subphthalocyanine chloride (SubPc):C_{60}, and 1, 1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC):HAT-CN, the tandem OLEDs with outstanding performance have been achieved, due to bipolar charge generation characteristics and charge carrier transport channels [18,20,22]. However, little work has been focused on comparison of the BHJ and PHJ based CGLs, which are consisted of the same n-type and p-type organic semiconductor, especially the detailed investigation of

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CGL mechanism.

In this work, we simultaneously utilized both BHJ and PHJ based CGLs to construct high efficiency tandem OLEDs, and systematically studied the effect of CGLs architecture on device performance. A p-type organic semiconductor, cobalt phthalocyanine (CoPc), was used to fabricate CGL, and the device performance was dependent on mixing ratio and CGL architecture. The external quantum efficiency, current-limited devices and single carrier devices were used to investigate the effect of different CGLs on device performance. It was found that the improved hetero-interfaces, optimized charge transport pathways and rational charge carrier mobility were responsible for charge carrier balance in tandem device. The results further demonstrated that BHJ base CGL was an effective way for the fabrication of high performance tandem OLEDs.

2. Experimental

Indium-tin-oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω/sq and film thickness of 150 nm were ultrasonically cleaned with detergent, acetone, ultra-purified water and ethanol. Afterwards, the substrates were dried with nitrogen gas flow and were treated using O_2 plasma in a vacuum chamber under a pressure of 25 Pa for 15 min. Then, the organic, inorganic and metallic layers were deposited in separate vacuum chambers under a pressure below 3×10^{-4} Pa and 3×10^{-3} Pa, respectively. The deposition rates and film thickness were monitored using a quartz crystal oscillator. A typical area of the OLEDs is 28 mm^2.

A control device A_0, which is single emitting unit OLED, was fabricated with a structure of ITO (150 nm)/N,N'-diphenyl-N,N'-bis(1-napthyl)-1, 1′-biphenyl- 4, 4′-diamine (NPB, 30 nm)/4, 4′-bis(carbazol-9-yl) biphenyl (CBP):8 wt% bis[2-(4-tert-butylphenyl)-benzothiazolato-N,$C^{2'}$]iridium (acetylace tonate) (denoted as $(t\text{-bt})_2Ir(acac)$, 15 nm)/4, 7-diphenyl-1, 10-phenanthroline (Bphen, 35 nm)/Mg:Ag (100 nm). The tandem devices based on BHJ based CGL were fabricated with an architecture of ITO/NPB (30 nm)/CBP:8 wt% $(t\text{-bt})_2Ir(acac)$ (15 nm)/BPhen (35 nm)/LiF (1 nm)/CoPc:C_{60} (x:y) (5 nm)/MoO_3 (5 nm)/NPB (30 nm)/CBP:8 wt% $(t\text{-bt})_2Ir(acac)$ (15 nm)/BPhen (35 nm)/Mg:Ag (100 nm). Where, the mixing ratios of x to y are 8:1, 4:1, 2:1 and 1:1, corresponding to devices A, B, C and D, respectively.

Meanwhile, a tandem device E based on planar heterojunction CGL was fabricated with a structure of ITO/NPB (30 nm)/CBP:8 wt% $(t\text{-bt})_2Ir(acac)$ (15 nm)/BPhen (35 nm)/LiF (1 nm)/CoPc (2.5 nm)/C_{60} (2.5 nm)/MoO_3 (5 nm)/NPB (30 nm)/CBP:8 wt% $(t\text{-bt})_2Ir(acac)$ (15 nm)/BPhen (35 nm)/Mg:Ag (100 nm). The device architectures are shown in Fig. 1(a). Therein, NPB, CBP and Bphen were used as hole transport, host material and electron transport layer, respectively, while CoPc and C_{60} were used as the p-type and n-type materials in CGL. The interface modification layer of LiF (1 nm) and MoO_3 (5 nm) were located next to the CGL, which were used to enhance the injection of generated charges into the corresponding EL units. The energy level diagrams of emitting units (top) and tandem OLEDs (bottom) are shown in Fig. 1(b).

The current density-voltage-luminance (*J-V-L*) curves were measured with a Keithley 4200 semiconductor characterization system and a ST-86LA luminance meter under ambient atmosphere without encapsulation. The EL spectra were recorded with an OPT-2000 spectrophotometer. The transmittance and absorption spectra of the organic films were measured by using a Shimadzu UV-1700 ultraviolet–visible spectrophotometer. The external quantum efficiencies (*EQEs*) of the OLEDs were calculated on the basis of theory reported in the literature [23,24].

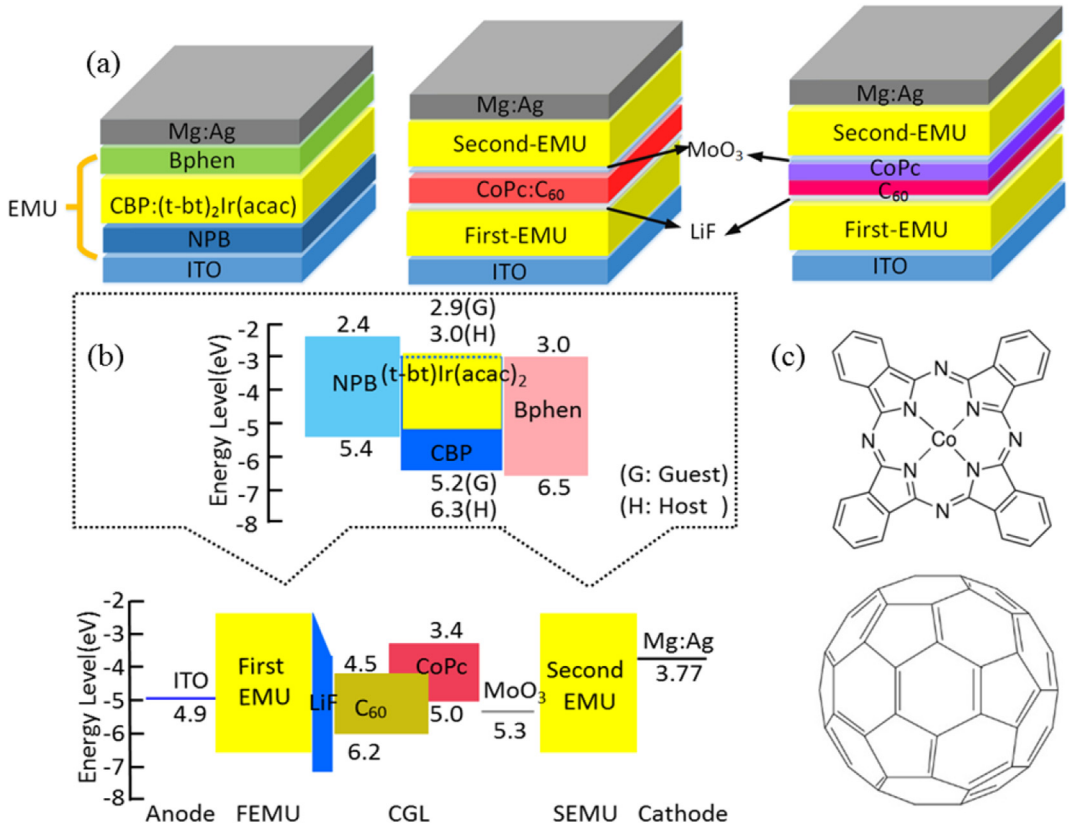

Fig. 1. (a) Three dimensional architecture of single-emitting unit OLEDs and tandem OLEDs. (b) Energy level diagram of tandem OLEDs (bottom) and emitting unit (top). (c) Chemical structure of CoPc and C_{60}.

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