

Role of MoO₃-modified organic photovoltaic-type charge generation layer in tandem organic light-emitting diodes



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ABSTRACT

The role of each part in MoO₃-modified organic photovoltaic (OPV)-type charge generation layer (CGL) was investigated by incorporation into hole-only devices. The OPV heterojunction (HJ) consists of fullerene (C60) and copper phthalocyanine (CuPc). According to the electrical characteristics of hole-only devices, it was found that charge generation mainly took place at the interface between MoO₃ and hole transport layer rather than the interface of C60–CuPc HJ under an electric field. The C60–CuPc HJ exhibited good charge generation in the presence of photons due to photovoltaic effect, despite its poorer charge generation upon an application of electric field. In addition, charges can be also generated at CuPc/MoO₃ interface upon a relatively high electric field since the absorption band of charge transfer complex formed by CuPc and MoO₃ is within visible region. These results, which had been rarely reported in the previous studies on OPV-type CGL, can well explain the experimental results of tandem organic light-emitting diodes (OLEDs) with MoO₃ modified OPV-type CGLs.

1. Introduction

Tandem organic light-emitting diodes (OLEDs), consisting of two or more electroluminescent (EL) units vertically connected electrically in series via charge generation layer (CGL) between neighboring EL units, have attracted much attention due to their advantages of high current efficiency and luminance at low current density, as well as the prolonged lifetime as compared to conventional single-unit OLEDs [1–6]. In tandem OLEDs, electrons and holes are generated in CGLs under the application of external electric field and then injected into adjacent EL units for light emission. Therefore, CGLs are critical for high-performance tandem OLEDs.

As a basic connector in tandem OLEDs, CGLs should also have a high optical transmission and a low electrical resistance to minimize power loss. Traditional CGLs are typically comprised of ultrathin conductive metal layer [7,8] and *p-n* junctions [9–11], where *p*- and *n*-type doped organic materials are widely employed as *p*- and *n*-type layer, respectively. CGLs with a metal layer have some disadvantages, such as low optical transparency and high deposited temperature. By contrast, organic *p-n* junctions have been attracting more and more attention due to their advantages of excellent optical and electrical properties, as well as the ease of fabrication by thermal evaporation. In order to

reduce the wastage induced by the absorption of CGLs in tandem OLEDs, usually used candidates are wide band gap organic materials, such as 4, 4', 4''-tris(*N*, *N*-phenyl-3-methylphenylamino) triphenylamine (m-MTDATA) [12,13] and 4, 7-diphenyl-1, 10-phenanthroline (Bphen) [9,14].

However, a series of novel *p-n* junction CGLs with low band gap organic materials, e.g. fullerene (C60)/pentacene [15–17], C60/copper phthalocyanine (CuPc) [18–20], and C60:phthalocyanine derivatives [21,22], have been recently explored. As a matter of fact, this type of CGLs are composed of so-called organic photovoltaic (OPV)-type heterojunctions (HJs), which have been widely used in organic solar cells due to charge generation under solar illumination, *i.e.* photovoltaic effect. These OPV-type HJs have poor optical transparency but can still be served as an efficient CGL in tandem OLEDs. One of the explanations is that OPV-type HJs can generate charges at an external electric field, referred to as electric-generated charges [16,18,23]. On the other hand, OPV-type CGLs can absorb a portion of photons from emissive zone to form excitons and dissociate into free charges at the interface between the donor (D) and the acceptor (A) in OPV-HJs [19,20]. These charges are called photo-generated charges [19,23]. Based on electric- and photo-generated charges, OPV-HJs can be utilized as efficient CGLs in tandem OLEDs. Although these results are impressive, the understand-

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ing of OPV-type CGLs is still limited.

In this work, in order to deeply understand the functionality of MoO₃-modified OPV-type CGLs (C60/CuPc/MoO₃), each part of CGLs was incorporated into hole-only devices. With the help of current density–voltage (J – V) characteristics, it has been demonstrated that the main charge generation process took place at the interface between MoO₃ and hole transport layer (HTL) at an external electric field. Then, tandem OLEDs with MoO₃-based CGLs were fabricated by vacuum sublimation. After device characterization, the best efficiency of tandem OLED with MoO₃-only as a CGL was about twice of that of single EL unit device. Additionally, tandem OLEDs with MoO₃-modified C60–CuPc CGL exhibit lower (higher) efficiency than MoO₃-only based tandem device at low (high) luminance. These results clarified excellent charge-generation of C60–CuPc HJ under light condition.

2. Experiments

The role of each part of MoO₃-modified C60–CuPc CGL was first investigated by incorporation into hole-only devices. The detailed structures are as follows:

- A1: ITO/NPB(40 nm)/Alq₃(60 nm)/CuPc(20 nm)/NPB(40 nm)/Al;
 A2: ITO/NPB(40 nm)/Alq₃(60 nm)/C60(15 nm)/CuPc(20 nm)/NPB(40 nm)/Al;
 B1: ITO/NPB(40 nm)/Alq₃(60 nm)/MoO₃(3 nm)/NPB(40 nm)/Al;
 B2: ITO/NPB(40 nm)/Alq₃(60 nm)/C60(15 nm)/CuPc(20 nm)/MoO₃(3 nm)/NPB(40 nm)/Al;
 B3: ITO/NPB(40 nm)/Alq₃(60 nm)/LiF(0.5 nm)/C60(15 nm)/CuPc(20 nm)/MoO₃(3 nm)/NPB(40 nm)/Al.

Type A devices (A1 and A2) do not introduce MoO₃ layer, which is incorporated into type B devices (B1–B3). The thickness of C60 and CuPc was optimized in the Ref. [23]. A control device (CD: ITO/NPB(40 nm)/Alq₃(60 nm)/NPB(40 nm)/Al) without any part of CGL was also prepared for comparison. The cross sections of device structures are depicted in Fig. 1.

Then, tandem OLEDs with various CGLs (MoO₃, CuPc/MoO₃ and C60/CuPc/MoO₃) were fabricated. The structures of tandem OLEDs were ITO/EL/CGL/EL/Al. There were two EL units in all devices, called the anode- and cathode-side EL units, respectively. The two EL units were the same, basically consisting of NPB(40 nm)/Alq₃(60 nm)/LiF

(0.5 nm). NPB and Alq₃ are short for *N*, *N'*-di (naphthalene-1-yl)-*N*, *N'*-diphenyl-benzidine and tris (8-hydroxyquinoline) aluminum, respectively. For comparison, conventional single EL unit device (ITO/EL/Al) was also prepared.

All organic materials were commercially available from Nichem Fine Technology Co. Ltd. and not purified prior to device fabrication. Commercial indium-tin oxide (ITO)-coated glass substrates, with a sheet resistance of 10 Ω/square, were employed to fabricate devices. The substrates were routinely cleaned by ultrasonic bath in de-ionized water, alcohol and isopropyl alcohol, finally treated by oxygen plasma for 7 min at a power of 75 W under a pressure of 50–100 Pa. The cleaned substrates were then loaded into an evaporation chamber. NPB, Alq₃, LiF, C60, CuPc and MoO₃ were evaporated on the substrates in turn at a pressure of 10^{−6} Torr. Next, the substrates were transferred to another chamber without breaking the vacuum to thermally evaporate Al cathode from a tungsten boat through a shadow mask, defining an active area of 2 mm × 2 mm.

The optical and electrical characteristics, including current density–luminance–voltage (J – L – V) and EL spectra, were simultaneously recorded by a software-controlled Keithley 2400 source meter combined with a CS2000 spectrometer. The absorption spectra were measured by Varian Cary 500 ultraviolet–visible spectrophotometer. All measurements were carried out at room temperature under ambient conditions.

3. Results and discussion

3.1. Role of each part of CGL

Firstly, each part of MoO₃-modified C60/CuPc CGL was introduced into hole-only devices, which has been proven to be an effective approach for investigating the effect of CGLs in a tandem OLED [21,24]. The device structures are displayed in Fig. 1. Fig. 2 shows the current density as a function of applied voltage for devices of types A and B. In all hole-only devices, few electrons can be injected into organic layer from Al cathode due to high energy barrier (~2 eV) at NPB/Al interface, seen in Fig. 3. Thus, electron injection current density (J_e)

$$J_e \rightarrow 0. \quad (1)$$

For CD, any section of CGL was not introduced. No excitons can be formed in the emissive layer and no light emission can be detectable since electrons were seldom injected from the cathode. For A1 device, light emission was also not observed. Assuming that there was charge generation caused by CuPc film upon an external applied voltage (that is, $J_{h1} > 0$, $J_{e1} > 0$, where J_{h1} and J_{e1} are current density for holes and electrons generated in OPV-HJ injection into anode- and cathode-side EL units shown in Fig. 3, respectively), hole extraction current density to the cathode (J_h) would increase. J_e can be negligible because

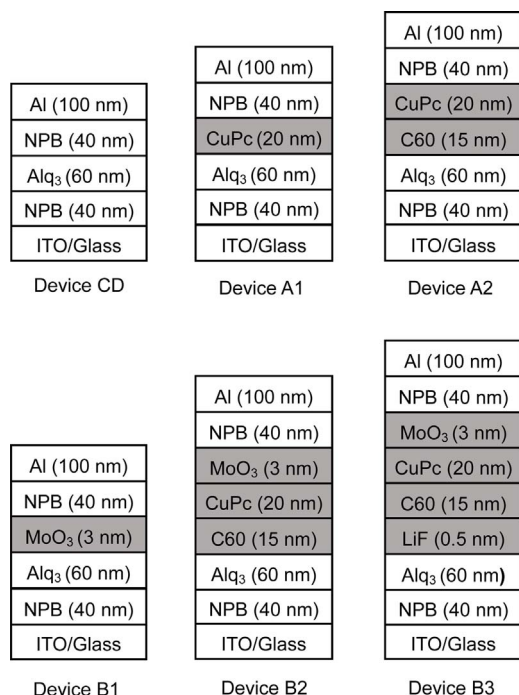


Fig. 1. Cross sections of device structures of types A & B.

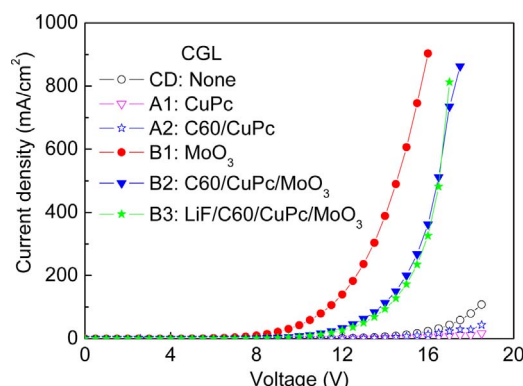


Fig. 2. (Color online) J – V plots of type A & B devices.

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