Current Applied Physics 14 (2014) 1465–1469

Contents lists available at ScienceDirect

Current Applied Physics

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

Field- and thickness-dependent performance of tandem organic light-emitting diodes with buffer-modified fullerene/copper phthalocyanine heterojunction as interconnector

Zhaoyue Lü [a, *], Xiong Li [b], Yue Wang [c], Jing Xiao [d], Penglai Xu [a]

[a] Department of Physics, School of Science, East China University of Science & Technology, Shanghai 200237, China
[b] School of Science, Beijing Technology and Business University, Beijing 100048, China
[c] Key Laboratory of Luminescence and Optical Information, Ministry of Education, Institute of Optoelectronic Technology, Beijing Jiaotong University, Beijing 100044, China
[d] Physics and Electronic Engineering College, Taishan University, Shandong Province 271021, China

ARTICLE INFO

Article history:
Received 13 June 2014
Received in revised form
15 August 2014
Accepted 21 August 2014
Available online 30 August 2014

Keywords:
Organic heterojunction
Electric-generated-charge
Photo-generated-charge
Fullerene
Copper phthalocyanine

ABSTRACT

Tandem organic light-emitting diodes (OLEDs) with the buffer-modified fullerene/copper phthalocyanine (LiF/C60/CuPc/MoO_3) as interconnector have been investigated. The properties of tandem OLEDs are dependent on electric field, which is induced by electric-field-dependence of charge generation (by electric filed and photons) and separation in such an interconnector structure. At low electric field, because of less generated-charges and light-absorption of C60/CuPc film, current efficiency and brightness of tandem OLEDs with two emissive units exhibit less than twice higher than those of conventional single-unit device. But more than twofold enhancement of current efficiency and brightness is observed at higher electric field due to a large number of charges generated in the interconnector. Meanwhile, the performance of tandem devices is affected by the thickness of C60 and CuPc. The optimal thickness is related to their exciton diffusion length.

1. Introduction

During the last few years, tandem organic lighting emitting diodes (OLEDs) have been attracting much attention for the application of displays and solid state lighting sources due to their advantages of high current efficiency and long lifetime [1–6]. In general, a tandem OLED consists of multiple electroluminescent (EL) units, which are vertically connected electrically in series via interconnectors between adjacent EL units. Both luminance and current efficiency at a fixed current density scale linearly with the number of EL units for tandem OLEDs. Upon the application of electric field, electrons/holes, generated in interconnectors, are injected into neighboring electron-/hole-transport layer (ETL and HTL) of the individual EL units and then recombine with holes/electrons injected from anode/cathode side or injected from side for light emission, respectively. Therefore, the interconnectors function as charge generation layer and are very important to realize high-performance tandem OLEDs.

Interconnectors are typically formed by metal–metal (or metal oxide) bilayer [7–11], organic-metal (or metal oxide) bilayer [8,12], or *n*–*p* organic heterojunction (HJ) [13–15]. However, there are some disadvantages for interconnectors with metals. The deposition of metal is not usually compatible with organic layers since many kinds of metals cannot be deposited by thermal evaporation at a temperature of less than 300 °C, which results in fabrication complexity. Additionally, metals can cause pixel crosstalk in dot-matrix displays. Hence, *n*–*p* junction interconnectors have been attracting much more attention. Nevertheless, problems still remain. In the *n*–*p* junction connector, alkali metal/alkali metal complex (such as Li [14], Li_2CO_3 [16], CsN_3 [17])-doped ETL is usually exploited as *n*-type organic layer. Alkali metals are highly chemical reactive with organics, resulting in exciton quenching of emissive layer. Furthermore, it has been found that dopants in *p*-type organic layer, such as $FeCl_3$ [14] or 2, 3, 5, 6-tetrafluoro-7, 7, 8, 8-tetracyanoquinodimethane (F_4-TCNQ) [2,18], are not stable.

More recently, a novel concept interconnector based on organic photovoltaic HJ has been reported to circumvent the above problems [3,4,16,19]. These organic HJs have been widely used to generate charges under solar illumination in organic photovoltaic (OPV) cells. In the reports by Chen et al. [3,4,16,20], such organic HJ

* Corresponding author. Tel.: +86 21 6425 2839.
E-mail address: lvzhaoyue@ecust.edu.cn (Z. Lü).

http://dx.doi.org/10.1016/j.cap.2014.08.016
1567-1739/

can effectively generate charges at an external electric field as well. For example, a tandem OLED based the interface-modified C60/pentacence (TPBi:Li_2CO_3/C60/pentacence/TCTA:MoO_3) as interconnector has a better performance than the device with conventional *n*–*p* junction (TPBi:Li_2CO_3/TCTA:MoO_3) interconnector. TPBi and TCTA are 1, 3, 5-tris(2-N-phenylbenzimidazolyl) benzene and 4, 4′, 4′-Tri(N-carbazolyl) triphenylamine, respectively [16]. The reason is that amounts of charges are generated in interface-modified C60/pentacence interconnector.

Chen et al. also found that an approximate twofold enhancement of power efficiency in tandem OLED based LiF/C60/pentacence/MoO_3 interconnector can be achieved [4,20]. Similar effects were observed by using ZnPc or copper phthalocyanine (CuPc) instead of pentacence [4,20]. Liu et al. [19] further reported that the interconnector of C60/CuPc can absorb a portion of photons radiated from emissive zone and form excitons which dissociate into free charges. The overall performance of tandem device is enhanced due to the utilization of internal radiation which should be wasted through various channels in normal devices.

Despite these meaningful results, there still remains room to further investigate tandem OLEDs with such organic HJ. In this paper, tandem OLEDs with buffer-modified C60/CuPc interconnector (LiF/C60/CuPc/MoO_3) were demonstrated. The introduction of LiF and MoO_3 buffer layer can modify the interface energy alignment to facilitate the injection of generated electrons/holes into the adjacent emissive units. Some studies have shown that the thickness of interconnector has a strong effect on device efficiency. In this work, the influence of C60 and CuPc thickness on the performance of tandem OLED is also presented. After device characterization, the optimal thickness of C60 and CuPc is determined to be 15 and 20 nm, respectively, which is related to the excition diffusion length of C60 and CuPc.

2. Experimental

The organic materials, used to fabricate OLEDs in this work, are all common: N, N′-di(naphthalene-1-yl)-N, N′-diphenyl-benzidine (NPB), tris(8-hydroxyquinoline) aluminum (Alq_3), C60 and CuPc. The chemical structures of these materials can be found in elsewhere [2,21]. All organic materials were commercially available from Nichem Fine Technology Co. Ltd. and used without further purification.

To investigate the effect of LiF/C60/CuPc/MoO_3 as interconnector, we constructed two series devices (series A and B). In series A, two kinds of devices, a conventional single-unit device and tandem devices having an interconnector of LiF/C60/CuPc/MoO_3 with various thicknesses of C60, were fabricated. The schematic configurations of OLEDs are shown in Fig. 1. Based on the optimum C60 thickness, the effect of CuPc thickness in the interconnector on device performance, named as series B, was further investigated. There are two EL units in all tandem OLEDs, where the first and second EL units, basically consisting of NPB/Alq_3, are identical.

All devices were fabricated on the indium-tin oxide (ITO)-coated glass substrates. The substrates were routinely cleaned by ultrasonic bath in de-ionized water, isopropanol and 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, Alq_3, LiF, C60, CuPc and MoO_3 were evaporated on the substrates in turn under a pressure of $2–5 \times 10^{-4}$ Pa. Next, the substrates were transferred to another chamber without breaking the vacuum to thermally evaporate Al cathode at a pressure of around 3×10^{-4} Pa. The thickness and deposition rate of the materials are monitored *in situ* by a 6 MHz quartz-crystal oscillator during evaporation. The device emissive area was 2 mm × 2 mm defined by the overlap of the ITO anode and Al cathode.

The optical and electrical characteristics, including current–voltage–brightness and EL spectra were simultaneously recorded using a software-controlled Keithley 2400 source meter combined with a CS2000 spectrometer. The absorption of C60/CuPc films was measured with Shimadzu UV-3101PC spectrometer. The photoluminescent (PL) spectrum of Alq_3 film was recorded by Spex Fluorolog-3 spectrometer. All measurements were immediately carried out after preparation at room temperature under ambient conditions without encapsulation.

3. Results and discussion

First, the characteristics of series A are discussed. Series A includes a conventional single-unit device: ITO/NPB(40 nm)/Alq_3(60 nm)/LiF(0.5 nm)/Al, and three tandem OLEDs with various thicknesses of C60 in LiF/C60/CuPc/MoO_3 interconnector: ITO/NPB(40 nm)/Alq_3(60 nm)/LiF (0.5 nm)/C60(10, 15 and 23 nm)/CuPc(5 nm)/MoO_3(3 nm)/NPB(40 nm)/Alq_3(60 nm)/LiF(0.5 nm)/Al, labeled as A1–A3, respectively. The current density–voltage–brightness and current efficiency–current density characteristics of series A devices are shown in Fig. 2(a) and (b), respectively. It can be seen from Fig. 2(a) that the driving voltage of tandem OLEDs is much higher than that of single-unit device due to the large vertical stack structure. For instance, the driving voltages at a current density of 20 mA/

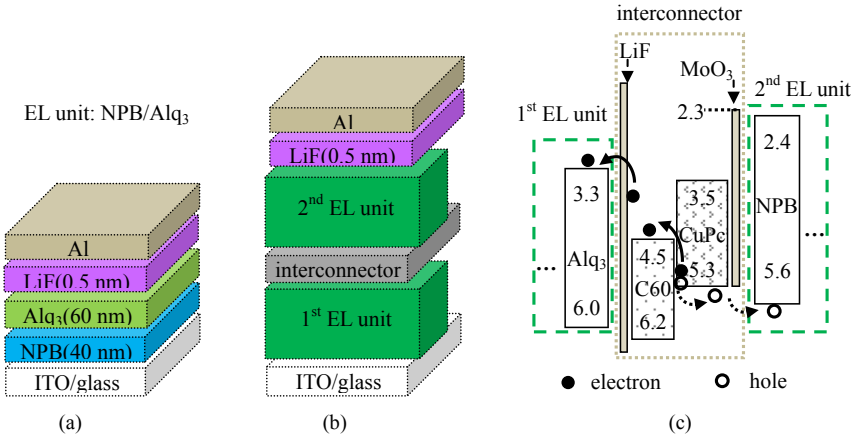

Fig. 1. Schematic configurations of (a) single-unit OLED and (b) tandem OLED with two EL units; (c) principle of charge generation and injection in tandem OLEDs.