Organic Electronics 14 (2013) 2198–2203

Contents lists available at SciVerse ScienceDirect

Organic Electronics

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

Highly efficient green phosphorescent organic light emitting diodes with simple structure

Min Chul Suh *, Hee Young Shin, Seung Ji Cha

Department of Information Display and Advanced Display Research Center, Kyung Hee University, Seoul 130-701, Republic of Korea

ARTICLE INFO

Article history:
Received 6 February 2013
Received in revised form 25 April 2013
Accepted 5 May 2013
Available online 29 May 2013

Keywords:
Highly efficient
Simple structure
Mixed host
Space charge limited current
Mobility change

ABSTRACT

A series of simple structures is investigated for realization of the highly efficient green phosphorescent organic light emitting diodes with relatively low voltage operation. All the devices were fabricated with mixed host system by using 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) and 1,3,5-tri(*p*-pyrid-3-yl-phenyl)benzene (TpPyPB) which were known to be hole and electron type host materials due to their great hole and electron mobilities [μ_h(TAPC): 1×10^{-2} cm^2/V s and μ_e(TpPyPB): 7.9×10^{-3} cm^2/V s] [1]. The optimized device with thin TAPC (5–10 nm) as an anode buffer layer showed relatively high current and power efficiency with low roll-off characteristic up to 10,000 cd/m^2. The performances of the devices; with buffer layer were compared to those of simple devices with single layer and three layers. Very interestingly, the double layer device with TAPC buffer layer showed better current and power efficiency behavior compared to that of three layer device with both hole and electron buffer layers (TAPC, TpPyPB, respectively).

1. Introduction

Phosphorescent organic light-emitting diodes (PHOLEDs) have attracted lots of interests because of their potential to realize extremely high quantum efficiency compared to conventional fluorescent OLEDs because they utilize both singlet and triplet excitons for light emission [1–3]. Indeed, red phosphorescent materials have been applied in the fabrication process of AMOLED for commercial mobile phones or tablet PC since 2007. Recently, green emission is also approaching to the theoretical limitation of efficiency (over 20% of external quantum efficiency, EQE), and the commercialization of which is also currently underway. However, PHOLEDs normally require complicated device structure to maximize the current efficiency compared to fluorescent OLEDs. In principle, hole blocking layers (HBL) and electron blocking layers (EBL) are necessary to confine the holes and electrons inside the emitting layers (EML) [4–7]. Such layers are inevitable because those exciton blocking layers prevent triplet exciton quenching at EML/HBL or EBL/EML interfaces. For suppression of such an exciton quenching effect, we need to use the HBL or EBL materials with higher triplet energies than those of EML materials [8–10]. In addition, the broadening of charge recombination zone could help to improve the device efficiency partially from the reduction of such phenomenon [11,12]. The most common methodology to expand the recombination zone may be a utilization of a mixed host system. Furthermore, the structure including mixed host system with charge blocking layers could enhance the device lifetime as well as efficiency [13–16]. However, such sophisticated device architectures inevitably increase the manufacturing complexity and production cost. Therefore, the simplification of device structure could become very important for the future [17,18]. The devices with simple structure were suggested intermittently. For instance, peak EQE of 18.6% (corresponding to current efficiency of 57.5 cd/A) in three layer PHOLEDs with NPB/TCTA:Ir(ppy)$_3$/Bepp$_2$:Ir(ppy)$_3$ double EML was reported [11]. Besides, it was reported that the high efficiency could also be obtained in the PHOLEDs without charge blocking

* Corresponding author. Tel.: +82 2 961 0694; fax: +82 2 968 6924.
E-mail address: mcsuh@khu.ac.kr (M.C. Suh).

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http://dx.doi.org/10.1016/j.orgel.2013.05.007

layers by controlling the charge transport properties of the host materials [19–23]. For example, the devices can be fabricated without HBL if the host material is electron transport type because the recombination zone could be biased toward the HTL [22,23]. In addition, Lee et al. reported that the single layer red phosphorescent device with EQE up to 8% could be prepared by using mixed host system. But they could not realize the device with EQE over than 8% presumably due to an incomplete charge confinement of their OLED structure [19]. Thus, in our report, assuming that the single layer device cannot confine charge completely, we suggested the simple bi-layer structure OLED devices which control recombination zone properly and have good color stability.

In this study, we report six different types of simple green PHOLEDs having mixed host system without charge blocking layers. The PHOLEDs containing bilayer device with only one TAPC buffer layer showed reasonable current and power efficiency (EQE up to 13.8%).

2. Experimental

2.1. Materials

1,1-Bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) as a hole transporting host materials, 1,3,5-tri(*p*-pyrid-3-yl-phenyl)benzene (TpPyPB) as an electron transporting host material [24], *fac*-tris(2-phenylpyridinato)iridium(III) [Ir(ppy)$_3$] as a green dopant, lithium fluoride (LiF) as an electron injection layer (EIL) and lithium quinolate (LiQ) as a π-electron donor and/or EIL were purchased from commercial suppliers and used without purification.

2.2. Device fabrication

To fabricate OLED devices, clean glass substrates precoated with a 150-nm-thick indium tin oxide (ITO) layer with a sheet resistance of ∼12 Ω/sq were used. Line patterns of ITO were formed on glass by photolithography process. The ITO glass was cleaned by sonification in an isopropylalcohol, rinsed in deionized water, and finally irradiated in a UV–ozone chamber. All organic materials were deposited by the vacuum evaporation technique under a pressure of $\sim 1 \times 10^{-7}$ Torr. The deposition rate of organic layers was about 1 Å/s. Then, LiF and Al were deposited in another vacuum deposition system without breaking vacuum. Deposition rates of LiF and Al were 0.1 Å/s and 5–10 Å/s, respectively.

2.3. Measurements

The current density–voltage (*J*–*V*) and luminance–voltage (*L*–*V*) data of OLEDs were measured by Keithley SMU 238 and Minolta CS-100A, respectively. The OLED area was 4 mm^2 for all the samples studied in this work. Electroluminescence (EL) spectra and CIE coordinate were obtained using a Minolta CS-1000A spectroradiometer.

3. Results and discussion

3.1. Design of hole only and electron only devices

TAPC has a HOMO value of ∼5.5 eV while TpPyPB has a LUMO value of ∼3.0 eV. Thus, we could easily expect that the recombination zone of mixed host system of those materials could be weighted towards anode side. Separately, we also investigated the bulk property from the characteristics of hole and electron only devices as follows:

HOD 1: ITO/MoO_3 (0.75 nm)/TAPC (100 nm)/MoO_3 (10 nm)/Al (100 nm)
HOD 2: ITO/MoO_3 (0.75 nm)/TAPC: TpPyPB (1:1, 100 nm)/MoO_3 (10 nm)/Al (100 nm)
EOD 1: ITO/LiQ (1.5 nm)/TpPyPB (100 nm)/LiQ (1.5 nm)/Al (100 nm)
EOD 2: ITO/LiQ (1.5 nm)/TAPC: TpPyPB (1:1, 100 nm)/LiQ (1.5 nm)/Al (100 nm)

We designed hole only devices (HOD) of TAPC and TAPC mixed with TpPyPB (1:1, wt./wt.) as well as electron only devices (EOD) of TpPyPB and TpPyPB mixed with TAPC (1:1, wt./wt.) to estimate how much the hole and electron current level change after mixing of materials with totally different nature. Meanwhile, the reference values of hole and electron mobilities of TAPC and TpPyPB by themselves obtained from the TOF (time of flight) experiment were 1.0×10^{-2} and 7.9×10^{-3} cm^2/V s, respectively [1]:

Fig. 1a shows the hole current characteristics of hole only device prepared with TAPC (HOD 1) and the device fabricated with TAPC mixed with TpPyPB (HOD 2). The current density of HOD 2 at 3 V was reduced significantly (>4 orders of magnitude) which means that the hole carriers could become minor carriers if TAPC is mixed with TpPyPB. In contrast, the current density increased after mixing in case of EOD as shown in Fig. 1b. In other words, the electron current density at 3 V even exceeded an order of magnitude after mixing TpPyPB with 50% of TAPC (EOD 2), although the current level of EOD 2 was less than that of EOD 1 below 2.23 V. Thus, the electrons could become major charge carriers and the holes could be minor charge carriers when the TAPC was mixed with TpPyPB at the normal operation region after turn-on voltage. Thus, the recombination zone could be biased toward anode side when we use a mixed host system of TAPC and TpPyPB.

3.2. Design of full device structure

To confirm a position of recombination zone and capability of charge confinement for realization of highly efficient simple devices, we prepared series of green PHOLEDs as follows:

Device A: ITO/TAPC: TpPyPB: Ir(ppy)$_3$ (1:1, 5%, 80 nm)/LiF (0.5 nm)/Al (100 nm)
Device B: ITO/TAPC: TpPyPB: Ir(ppy)$_3$ (1:1, 5%, 75 nm)/TpPyPB (5 nm)/LiF (0.5 nm)/Al (100 nm)