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Green phosphorescent organic light emitting diodes with simple structure to realize an extremely low operating voltage

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ABSTRACT

Relatively simple phosphorescent OLED with extremely low operating voltage (~3.1 V) was devised. To eliminate some components of OLED device, the biased charge injection design was required. To confine the recombination zone toward the anode side, relatively large hole injection barrier (~0.7 eV) and small electron injection barrier (<0.2 eV) were applied. To realize extremely low operating voltage, we utilized materials with relatively high mobility. Two different emitting layers containing hole transporting material (TAPC) mixed with two different electron transporting materials (TpPyPB and TmPyPB) showed slightly different device behavior. Two layer device with the highest efficiency was obtainable from the biased injection design (TAPC:TpPyPB system), while the biased injection was inefficient in case of TAPC:TmPyPB system.

1. Introduction

Phosphorescent organic light-emitting diodes (PHOLEDs) have widely utilized to maximize internal quantum efficiency up to 100% [1–3]. However, PHOLEDs normally require complicated device structure to maximize the current efficiency compared to fluorescent OLEDs. Generally, hole blocking layers (HBL) and electron blocking layers (EBL) are essential to confine the holes and electrons inside the emitting layers (EML) because exciton can be migrated to the neighboring layers if recombination zone is formed widely [4–7]. Thus, such layers are inevitable because those exciton blocking layers help to prevent triplet exciton quenching at EML/HBL or EBL/EML interfaces [8–11]. However, such sophisticated device architectures inevitably increase the manufacturing complexity and production cost. Also, even though current efficiency could be enhanced by confining exciton efficiently, driving voltage increases unavoidably. Therefore, the simplification of device structure could become very important for the future and OLED with simple stack were reported intermittently. For instance, PHOLEDs with NPB/TCTA:$Ir(ppy)_3$/$Bepp_2$:$Ir(ppy)_3$ double EML were reported and this device showed extremely low operating voltage of ~3.3 V [12] and red PHOLED without electron injection layer was also reported with external quantum efficiency (EQE) about 12.5% [13]. Furthermore, there are several reports of PHOLEDs showing high efficiency without charge blocking layers by controlling the charge transport properties of the host materials [9–16]. For example, the devices can be realized without HBL by using electron transporting material as host material and the recombination zone could be biased toward anode side [14,15]. In addition, Joo et al. reported that the single layer red PHOLEDs with external quantum efficiency up to 8% could be prepared by using mixed host system [16]. And this group reported about triplet mixed host EML which contains hole transport type host with good hole injection properties and electron transport type host with good electron injection properties [17,18]. In this study, we report various simple green PHOLEDs having mixed host system 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 low operating voltage (~3.1 V) at given constant luminescence of 1000 cd/m^2.

2. Experimental

2.1. Materials

1,1-Bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) as material for hole transporting layer (HTL) as well as host of emission layer (EML), 1,3,5-tri(*p*-pyrid-3-yl-phenyl)benzene (TpPyPB) and 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) as materials for electron transporting host in EML as well as electron transport layer (ETL), *fac*-tris(2-phenylpyridinato)iridium(III) [$Ir(ppy)_3$] as green dopant, lithium fluoride (LiF) as electron injection layer, lithium quinolate (LiQ) as a π-electron donor and/or EIL and

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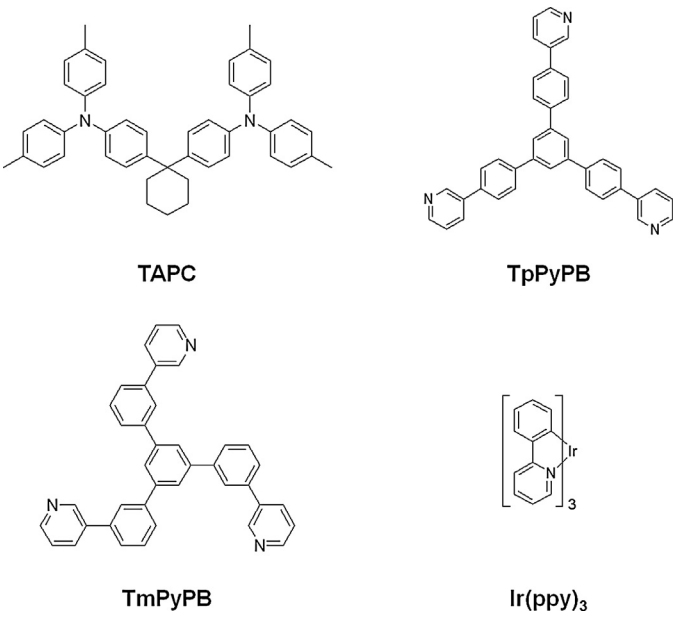

Fig. 1. Chemical structures of host and dopant materials utilized in this study.

aluminum (Al) as cathode were purchased and used without purification. Chemical structures of host and dopant materials used in this study are summarized in Fig. 1.

2.2. Device fabrication

To fabricate OLED devices, 150-nm-thick indium tin oxide (ITO) layer with a sheet resistance of ~12 Ω/sq were used. Line patterns of ITO were formed by photolithography process. The ITO glass was cleaned by sonification in an isopropyl alcohol, 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 0.5 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 3 Å/s, respectively.

2.3. Measurements

The current density–voltage–luminance (*J–V–L*) data of OLEDs were measured by Keithley 2635A and Minolta CS-100A, respectively. Electroluminescence (EL) spectra and CIE coordinate were obtained using a Minolta CS-2000 spectroradiometer.

3. Results and discussion

3.1. Design of hole only and electron only devices

We investigated the bulk property of intrinsic materials and mixed host system of hole transporting materials and electron transporting materials and compared by evaluating current characteristics of hole only devices (HODs) and electron only devices (EODs). The structure of HODs and EODs were as in the following:

HOD: ITO/MoO_3 (0.75 nm)/Host (100 nm)/MoO_3 (10 nm)/Al (100 nm)

where TAPC for HOD 1, TAPC:TpPyPB (1:1, wt./wt.) for HOD 2, and TAPC:TmPyPB (1:1, wt./wt.) for HOD 3 were used as host.

EOD: ITO/LiQ (1.5 nm)/Host (100 nm)/LiQ (1.5 nm)/Al (100 nm)

where host were TpPyPB, TmPyPB, TAPC:TpPyPB (1:1, wt./wt.), and TAPC:TmPyPB (1:1, wt./wt.) for EOD 1, EOD 2, EOD 3, and EOD 4, respectively.

TAPC was mixed with TpPyPB or TmPyPB in the weight percent ratio of 1:1 to estimate how much the hole and electron current level changes after mixing of materials with totally different charge transporting nature. The reference values of hole mobility of TAPC is 1.0×10^{-2} cm²/V s, and electron mobilities of TpPyPB and TmPyPB are 7.9×10^{-3} and 1.0×10^{-3} cm²/V s as reported before [1].

Fig. 2(a) shows the hole current characteristics of fabricated HODs. The current density of HOD 2 and HOD 3 (mixed host system) at 3 V reduced almost 4 orders of magnitude compared with HOD 1 fabricated with intrinsic hole transporting materials, TAPC. This means that hole carriers become minor if TAPC is mixed with electron transporting materials, TpPyPB or TmPyPB. On the other hand, in case of EOD, the electron current density increased sharply over 2.2 V regions when two types of charge transporting material are mixed as shown in Fig. 2(b). Curiously, the electron current density over 3 V even exceeded (in case of TpPyPB) and become similar (in case of TmPyPB) after mixing TpPyPB or TmPyPB with of TAPC (EOD 3 and EOD 4). Contrary to hole carriers, the electrons seem to be major carriers when the TAPC was mixed

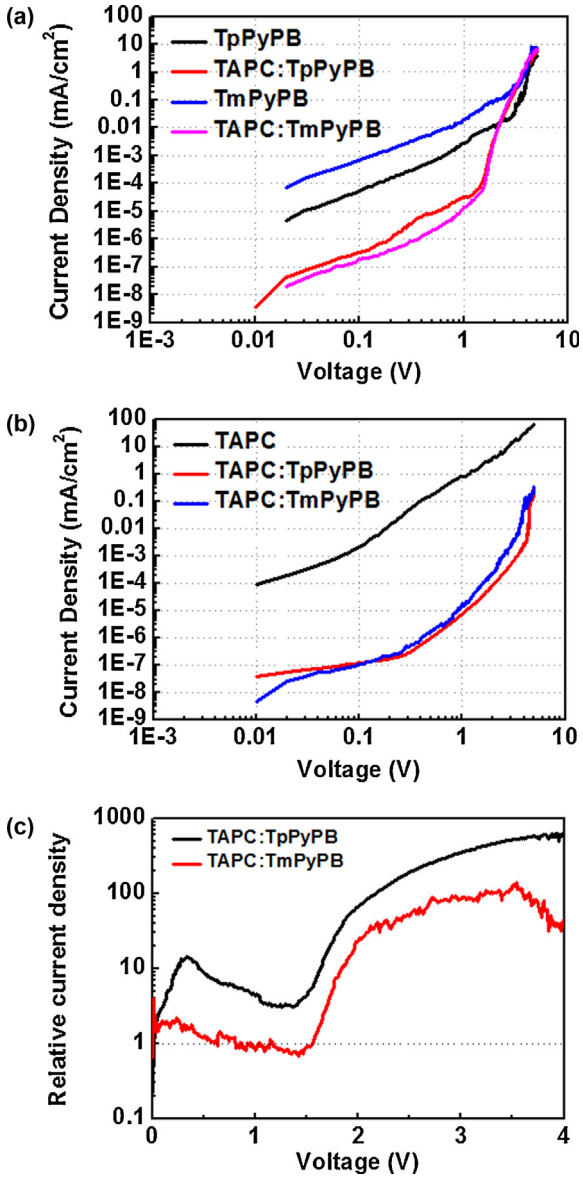

Fig. 2. *J–V* characteristics of (a) HOD and (b) EOD fabricated in this study and (c) relative current density behavior of mixed host system.