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High efficiency green phosphorescent organic light-emitting diodes with a low roll-off at high brightness

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ABSTRACT

Highly efficient green phosphorescent organic light-emitting diodes (PHOLEDs) with low efficiency roll-off at high brightness have been demonstrated with a novel iridium complex. The host material 1,3-bis(carbazol-9-yl)benzene (mCP) with high triplet energy is also used as the hole transporting layer to avoid carrier accumulation near the exciton formation interface and reduce exciton quenching. It provides a new approach for easily fabricating PHOLED with high triplet energy emitter. Moreover, the hole blocking layer is extended into the light emitting layer to form a co-host, realizing better control of the carrier balance and broader recombination zone. As a consequence, a maximum external quantum efficiency of 20.8% and current efficiency of 72.9 cd/A have been achieved, and maintain to 17.4% and 60.7 cd/A even at 10,000 cd/m^2, respectively.

1. Introduction

In the last few years, organic light-emitting diode (OLED) has shown its strong potential in the next generation of display and lighting applications. In particular, owing to the harvest of both singlet and triplet excitons for photon emission, iridium complex based phosphorescent organic light-emitting diodes (PHOLEDs) have boosted the internal quantum efficiency up to 100%, accelerating the mass production of OLEDs. However, the PHOLEDs suffer from a decrease of efficiency when the driving current density increases [1,2]. This roll-off phenomenon is mainly caused by triplet–triplet annihilation (TTA) [3] and triplet–polaron quenching (TPQ) [4], which limits the efficiency of

PHOLEDs at high brightness. To overcome the drawback, some solutions have been employed: (i) Avoiding carrier accumulation at the interface adjacent to light emitting layer (EML). For instance, Wang et al. [5] demonstrated that accumulated space charge at the exciton formation interface causes exciton quenching. In 2011, they further fabricated a simplified device using 4′-bis(carbazol-9-yl)biphenyl (CBP) as both light emitting host and hole transporting layer with low roll-off by eliminating 4,4′-N,N′-dicarbazole-biphenyl (NPB)/CBP heterojunction [6]; (ii) Balancing charge carriers in EML and broadening the recombination zone. Kim et al. [7] mixed 4,4′,4″-tris(N-carbazolyl)triphenylamine (TCTA) and 1,3,5-tris(N-phenyl-benzimidazole-2-yl)benzene (TPBi) as co-host to widen the recombination zone, achieving improved efficiency roll-off, which was attributed to low probability of exciton quenching. Recently, Chen et al. [8] mixed the hole transporting material TCTA and electron transporting material 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPb) as

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co-host to fabricate a highly efficient iridium(III)bis(4,6-difluorophenyl)-pyridinato-N,C^2)picolinate (FIrpic) based blue device. However, with relatively low triplet energy of 2.7 eV, TCTA is not a suitable host for many phosphorescent dyes with higher triplet energy such as deep blue emitters [9].

In this paper, we focus on a newly synthesized iridium based green phosphorescent emitter, iridium(III)bis(2-(4-trifluoromethylphenyl)pyridine)tetraphenylimidodiphosphinate (Ir(tfmppy)$_2$(tpip), inset of Fig. 1a), which was first reported by Zheng group [10]. The incorporation of tetraphenylimidodiphosphinate group into the molecule may enhance the electron mobility of the complex and reduce the TTA in the device due to the two electron transport units of P=O bond. However, the efficiency dropped from the peak value of 67.95 cd/A to 52.74 cd/A at 1000 cd/m^2, which needs to be improved for high brightness application. In addition, an emission at 380 nm from 1,3-bis(carbazol-9-yl)benzene (mCP) host was still observable in the electroluminescence (EL) spectrum, indicating an incomplete energy transfer from mCP to the emitter Ir(tfmppy)$_2$(tpip). To solve these issues, we tried to combine the two approaches mentioned above, using the high triplet energy (2.9 eV) host material mCP mixed with an electron transporting material TPBi as co-host, and utilizing mCP as the hole transporting layer (HTL) as well to remove the barrier between the HTL and EML. An external quantum efficiency (EQE) of 20.8% has been reached with a maximum current efficiency of 72.9 cd/A. The EQE and

current efficiency remain at 17.4% and 60.7 cd/A, respectively, at 10,000 cd/m^2, showing a significantly reduced roll-off.

2. Experimental

Prior to evaporation, patterned indium tin oxide (ITO) substrates were cleaned in order by detergent, acetone and isopropyl alcohol, then dried by nitrogen. All devices in this work were fabricated by vacuum thermal evaporation onto ITO substrates under a base pressure of 5×10^{-6} Torr and the deposition rates were measured with quartz crystals. The MoO$_3$ was evaporated from a tungsten boat with a deposition rate of 0.1 Å/s. The organic materials in quartz crucibles, except for the dopants, were evaporated at a rate of 0.5–1 Å/s. At the end, 100-nm-thick aluminum was deposited as the cathode. The current–voltage–luminance characteristics of the devices were measured using a computer controlled Keithley 2400 and Topcon BM-7A measurement system. All the measurements were carried out at room temperature under ambient atmosphere [11].

3. Results and discussion

As a common host for blue phosphorescent dyes, mCP has a high triplet energy and a very deep highest occupied molecular orbital (HOMO) level as well. It is very difficult to inject holes from ITO or normal hole injection layer to mCP. Therefore, mCP is usually not used as HTL. To study the feasibility of mCP as HTL, three devices were fabricated with a basic structure of ITO/MoO$_3$ (1 nm)/HTL/mCP:Ir(tfmppy)$_2$(tpip) (15 nm, 6 wt%)/4,7-diphenyl-1,10-phenanthroline (BPhen) (50 nm)/8-Hydroxyquinolinolatolithium (Liq) (1 nm)/Al (100 nm) , where MoO$_3$ is the hole injection layer, which can modify the work function of ITO [12]. The HTL is as follows: H1, NPB (35 nm); H2, mCP (35 nm); H3, NPB (25 nm)/mCP (10 nm). NPB, a commonly used HTL material, is used for comparison with mCP.

Fig. 1 shows the current density and current efficiency curves of device H1–H3. With the help of MoO$_3$, holes can be easily injected from the ITO into either NPB or mCP. The HOMO levels of NPB and mCP are 5.4 eV and 5.9 eV, respectively, resulting in difficult hole injection from NPB to mCP. With this energy barrier, device H1 and H3 demonstrate lower current densities than H2. However, device H1 and H3 show completely different *I–V* characteristics. The main difference is that in device H1, NPB HTL has direct contact with Ir(tfmppy)$_2$(tpip) doped mCP EML, whereas in device H3, an undoped mCP is inserted between NPB and EML. The current of device H1 is significantly higher than that of device H3, indicating that there may exist other channels for hole transporting. The Ir(tfmppy)$_2$(tpip), whose HOMO of 5.4 eV is similar to that of NPB, may facilitate the hole transporting, which was observed on another phosphorescent emitter tris(2-phenylpyridine)iridium (Ir(ppy)$_3$) [13]. On the other hand, device H2 achieves the highest current, confirming that the mCP, with a high hole mobility of 10^{-4} cm^2/V s [14], can also be a HTL by inserting a thin layer of MoO$_3$ at the ITO
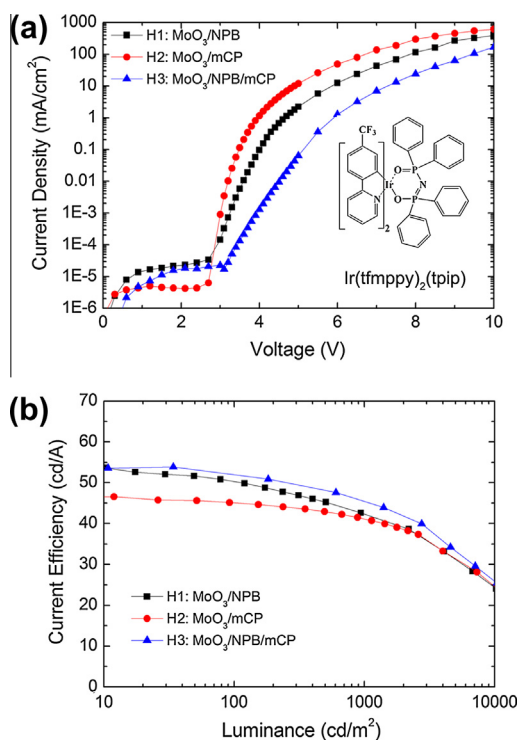

Fig. 1. (a) Current density–voltage characteristics of H1–H3 and chemical structures (inset) of Ir(tfmppy)$_2$(tpip) used in this work. (b) Current efficiency–luminance characteristics of H1–H3.