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Highly efficient all phosphorescent white organic light-emitting diodes for solid state lighting applications

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ABSTRACT

We report highly efficient all phosphorescent white organic light-emitting diodes (OLEDs) with an exciton-confinement structure. By stacking two emissive layers (EMLs) with different charge transporting properties, effective charges as well as exciton confinements were achieved. Accordingly, efficient blue OLEDs with a peak external quantum efficiency (EQE) over 22% and power efficacy (PE) over 50 lm/W were developed by using iridium(III) bis(4,6-(difluorophenyl) pyridinato-N,C2′)picolinate (FIrpic) as an electro-phosphorescent dopant. When the optimized orange and red EMLs were sandwiched between the stacked two blue EMLs, white OLEDs with an EQE and PE of 24.3% and 45.9 lm/W at a luminance of 1000 cd/m^2 were obtained without the use of any out-coupling techniques. In addition, these white OLEDs exhibit a color rendering index (CRI) value of 84 with high efficacy.

1. Introduction

White organic light-emitting diodes (OLEDs) are attracting widespread attention as next-generation low-cost and high-efficiency thin-film electroluminescent devices for both flat panel displays and lighting applications due to recent rapid technical evolution [1–3]. White OLEDs for solid state lighting applications require high efficiency, high color rendering index, high color stability, and appropriate color temperature [4–6].

Impressive scientific and technological approaches for developing WOLEDs have been reported, such as blending lumiphores emitting at different colors into a single layer, down conversion of blue OLEDs with yellow or red phosphores, using multiple monochromic OLED stacks connected by charge-generating layer (tandem OLEDs), and using a single device stack with multi-emissive layer (EML) structure doped with different color emitting dopants [4–9]. Among them, the multi-EML structure has advantages over other architectures in terms of efficiency and color controllability because the recombination current, singlet and triplet energy transfer and light-emitting performance of each layer can be easily controlled by thickness, and doping concentration of each EML, respectively. [4,7]

Recently, OLEDs have shown rapid improvements in terms of light-emitting performance through the use of electrophosphorescent materials and *p-i-n* doped structures [10–13]. Especially, to use phosphorescent materials is one effective way to get high efficiency in green and red OLED due to their ability to efficiently utilize both singlet and triplet excitons. For instance, Meerheim et al. have reported highly efficient bottom emitting green OLEDs with 25.5% of EQE by introduction of enhanced microcavity structure. We have also demonstrated 24.2% EQE in bidirectional red OLEDs by precise cavity design [13]. However, as one of the three primary colors, the corresponding light-emitting efficiency of blue OLEDs is much lower than those reported green or red OLEDs, and thus restricting the performance of white OLEDs [14,15].

In this work, we developed high performance blue phosphorescent OLEDs by using a stacked emitting structure to get highly efficient white OLEDs. Consequently, high efficiency phosphorescent white OLEDs with a maximum EQE of 25.6% and a PE of 47.0 lm/W without the use of any out-coupling techniques, were demonstrated through the insertion of orange and red emitting layers sandwiched between two blue emitting layers.

2. Experimental

A series of OLEDs in this study were fabricated using the configuration: indium tin oxide (ITO) (70 nm)/1,1-bis[(di-4-

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tolylamino)phenyl]cyclohexane (TAPC) (55 nm)/EML/1,3-bis(3,5-dipyrid-3-yl-phenyl)benzene (BmPyPB) (15 nm)/BmPyPB:Cs (40 nm)/LiF (1 nm)/Al (120 nm). Two EMLs, containing 7 wt% FIrpic co-deposited with the hole-transport type host material 4,4′,4″-tri(N-carbazolyl)triphenylamine (TcTa, 10 nm) and 10 wt% FIrpic co-deposited with the electron-transport type host material 2,6-bis(3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy, 10 nm) [16], were deposited successively. To fabricate a white OLED, an x nm thick red EML of 7 wt% red dopant (RD)-doped TcTa and a y nm thick orange EML of 7 wt% bis(2-phenylbenxothiozolato-N, $C^{2\prime}$)iridium (III)(acetylacetonate) (Bt_2Ir(acac))-doped TcTa were inserted between two blue EMLs [7] (White A: $(x, y) = (0.5, 0.5)$, White B: $(x, y) = (0.5, 1.0)$), respectively. The schematic diagrams of the structures of the OLEDs and energy level diagram of materials used in this study were shown in Fig. 1. TAPC and BmPyPB/4 mol% cesium doped BmPyPB were used as a hole-transporting layer (HTL) and as an electron transporting layer (ETL), respectively. The overall active area of a single OLED device is 4.0 mm^2.

ITO was cleaned by the standard oxygen plasma treatment. The OLED grade materials were purchased and used without further purification. All organic layers were deposited in a high vacuum chamber below 5×10^{-7} Torr and thin films of LiF and Al were deposited as a cathode electrode. The OLEDs were transferred directly from vacuum into an inert environment glove-box, where they were encapsulated using an UV-curable epoxy, and a glass cap with a moisture getter. The electroluminescence spectrum was measured using a Minolta CS-1000. The current density–voltage (J–V) and luminescence–voltage (L–V) characteristics were measured with a current/voltage source/measure unit (Keithley 238) and a Minolta CS-100.

3. Results and discussion

It is crucial to confine effectively both charge carriers and triplet excitons for achieving efficient phosphorescent OLEDs [15,16]. The lowest un-occupied molecular orbital (LUMO) level of HTL material should be controlled for blocking of electron leakage from EML, while the highest occupied molecular orbital (HOMO) level of ETL should be tuned to block hole leakage from EML. Additionally, the triplet energy of both HTL and ETL materials should be high to suppress triplet energy transfer from EML material to the charge transport materials. This energy level design of HTL and ETL materials gives a higher feasibility to confine both charges and excitons in the EML, leading to high EQE in phosphorescent OLEDs. [14–16]

Therefore, in this study, TAPC and BmPyPB with wider triplet energy level (T_1 = 2.9 and 2.69 eV, respectively, which are higher than that of FIrpic (T_1 = 2.62 eV)) and low-lying the HOMO and high-lying the LUMO energy levels, respectively, were used as an HTL and as an electron transporting layer (ETL), respectively. Thus, effective confinement of charges and triplet excitons inside the EML was achieved. [14,15]

Fig. 2 shows EQE (left axis) and PE (right axis) versus luminance characteristics of blue OLEDs in this study. As an approach to improve light-emitting efficiency of white OLEDs, efficient blue OLEDs with device architecture of a stacked emitting structure have been developed and optimized in advance. The blue OLED with a stacked EML structure is comprised of two EMLs with a hole transport-type host of TcTa and an electron transport-type band-gap host of 26DCzPPy both doped with a blue electro-phosphorescent dopant of FIrpic. The optimized efficiency was obtained at a FIrpic concentration of 7 wt% in TcTa layer and 10 wt% in 26DCzPPy layer, respectively. The detailed investigation of this blue OLED will be reported elsewhere. By stacking two blue EMLs with different charge transporting properties, the expansion of hole/electron recombination zone and the reduction of triplet quenching process in the EML were obtained. Consequently, the blue OLED with a peak EQE of 22.7% and PE of 50.5 lm/W without any out-coupling enhancement was demonstrated as shown in Fig 2. The electroluminescence (EL) spectra of the blue OLED is depicted in inset of Fig 2, exhibiting a luminescence wavelength near 468 nm, which originates from the triplet emission of FIrpic.

To establish white OLEDs based on this high efficiency blue OLED, we introduced thin red and orange EMLs between two blue EMLs. Fig. 3 shows current density–voltage–luminance (J–V–L) characteristics of blue and white OLEDs. As shown in Fig 3, the turn-on and driving voltages at 1000 cd/m^2 of the blue OLED were around 3.0 and 4.0 V, respectively. Even though additional two red and orange EMLs were inserted, the current density–voltage (J–V) characteristics of two white OLEDs were almost identical to that of blue OLED because of very thin thickness of red and orange EMLs.

Fig. 4 shows the normalized EL spectra for the white OLEDs together with blue OLED. The EL spectra exhibited the peak wavelengths at 472, 558, and 620 nm, which correspond to the emission from FIrpic, Bt_2Ir(acac), and RD dopants, respectively. The Commission Internationale de L'Eclairage (CIE) coordinates of these white OLEDs are (0.424, 0.421), and (0.434, 0.436) for White A, and B, respectively. These slight spectral changes indicate that the insertion of thin red/orange EML at the TcTa/26DCzPPy interface could control overall emission spectra, color rendering index (CRI),

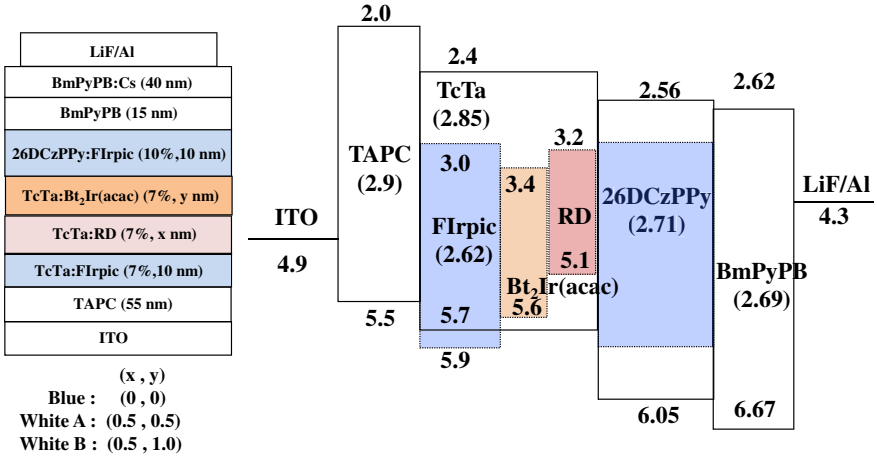

Fig. 1. Schematic diagrams of the structures of the white PHOLEDs and energy level diagrams for the materials tested in this study.