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Efficient blue-green and green electroluminescent devices obtained by doping iridium complexes into hole-block material as supplementary light-emitting layer



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ABSTRACT

In this work, organic electroluminescent (EL) devices with dominant and supplementary light-emitting layers (EMLs) were designed to further improve the EL performances of two iridium^{III}-based phosphorescent complexes, which have been reported to provide EL devices with slow EL efficiency roll-off. The widely used hole-block material 2,2',2"-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) was selected as host material to construct the supplementary EML. Compared with single-EML devices, double-EMLs devices showed higher EL efficiencies, higher brightness, and lower operation voltage attributed to wider recombination zone and better balance of carriers. In addition, the insertion of supplementary EML is instrumental in facilitating carriers trapping, thus improving the color purity. Finally, high performance blue-green and green EL devices with maximum current efficiencies of 35.22 and 90.68 cd/A, maximum power efficiencies of 26.36 and 98.18 lm/W, and maximum brightness of 56,678 and 112,352 cd/m², respectively, were obtained by optimizing the doping concentrations. Such a device design strategy extends the application of a double EML device structure and provides a chance to simplify device fabrication processes.

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1. Introduction

Organic light-emitting devices (OLEDs) based on iridium^{III} (Ir^{III}) phosphorescent complexes have been intensively studied because of their potential application in large-area flat-panel displays and in solid-state lighting [1–3]. In the past years, many groups have studied the design of device structures and the synthesis of Ir^{III} complexes [4–6]. Significant enhancement on maximum electroluminescent (EL) efficiencies has been realized, but EL efficiencies of most reported devices drop rapidly with increasing current density due to triplet-triplet annihilation (TTA), triplet-polaron annihilation (TPA), and electric field induced dissociation of excitons [7,8]. The roll-off of EL efficiency is quite severe in

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Recently, we reported the synthesis and EL performances of two interesting bis-cyclometalated Ir^{III}-complexes of Ir(dfppy)₂(tpip) [complex 1, dfppy=4,6-difluorophenylpyridine, tpip=tetraphenylimido-diphosphinate] and Ir(tfmppy)₂(tpip) [complex 2, tfmppy=4trifluoromethylphenylpyridine] [9]. By doping complex 1 or 2 into the bipolar host material 1,3-bis(carbazol-9-yl)benzene (mCP), a series of blue-green and green EL devices with single light-emitting layer (EML) was fabricated and investigated. By optimizing the doping concentrations, bright blue-green and green EL devices with maximum current efficiencies of 25.45 and 67.95 cd/A, respectively, were obtained. Interestingly, the roll-off of EL efficiency in these devices was significantly suppressed due to the broadening of the recombination zone. However, hole mobility of mCP is relatively higher than its electron mobility [10], which causes the unbalanced distribution of holes and electrons within the light-emitting layer, thus limiting the improvement of EL efficiency.

In this work, we aim to further improve the EL performances of these two complexes by designing the double EML device

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structure, which has been demonstrated to be efficient in improving EL efficiency, confining recombination zone as well as delaying the roll-off of efficiency [11–14]. A supplementary EML (SEML) was constructed by doping complex 1 or 2 into 2,2',2"-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) layer. This design strategy helps to simplify the device fabrication processes because TPBi functions also as an electron transport/hole-block material in this study. A series of EL devices with or without SEML was fabricated and investigated. Compared with single EML reference devices, double EMLs devices showed higher efficiencies, higher brightness, lower operation voltage, and even improved color purity. By optimizing the doping concentrations, high performance blue-green and green EL devices with maximum current efficiencies of 35.22 and 90.68 cd/A, maximum power efficiencies of 26.36 and 98.18 lm/W, and maximum brightness of 56,678 and 112,352 cd/m², respectively, were obtained.

2. Experimental

Most of the organic materials used in this study were obtained commercially and used as received without further purification, while complexes 1 and 2 were synthesized and purified in our laboratory. Indium-tin-oxide (ITO) coated glass with a sheet resistance of 15 Ω/sq was used as the anode substrate. Prior to film deposition, patterned ITO substrates were cleaned with detergent, rinsed in de-ionized water, dried in an oven, and finally treated with oxygen plasma for 10 min at a pressure of 10 Pa to enhance the surface work function of ITO anode (from 4.7 to 5.1 eV) [15]. All the organic layers were deposited with the rate of 0.1 nm/s under high vacuum ($\leq 3 \times 10^{-5}$ Pa). The EMLs were prepared by co-evaporating dopant and host material from two individual sources, and the doping concentration was modulated by controlling the evaporation rate of the dopant. LiF and Al were deposited in another vacuum chamber ($\leq 8.0 \times 10^{-5}$ Pa) with the rates of 0.01 and 1 nm/s, respectively, without being exposed to the atmosphere. The thicknesses of these deposited layers and the evaporation rate of individual materials were monitored in vacuum with quartz crystal monitors. A shadow mask was used to define the cathode and to make 10 9 mm² devices on each substrate. Current density-voltage-brightness (J-V-B) characteristics were measured by using a programmable Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a silicon photodiode. The EL spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer.

3. Results and discussion

The device structure and the highest occupied molecular orbital (HOMO)/the lowest unoccupied molecular orbital (LUMO) levels diagram of the designed OLEDs are depicted in Fig. 1. Molecule structures of complexes 1 and 2 are also shown in Fig. 1. Di-[4-(N,N-ditolyl-amino)-phenyl]cyclohexane (TAPC) was used as hole transport and electron block layers (HTL/EBL) due to its high hole mobility $(1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$ and high-lying LUMO level (-1.8 eV) [16]. TPBi was used as hole block and electron transport layers (HBL/ETL) due to its low-lying HOMO level (-6.3 eV) [17]. As has been reported previously, complex 1 or 2 was doped into the bipolar host material mCP as the first EML [9]. By doping complex 1 or 2 into TPBi, a second EML was constructed and inserted between the first EML and hole-block layer (HBL). Injection of holes from the first EML into the second EML is really difficult because of the n-type characteristic of TPBi, whereas the injection of electrons from the second EML into the first EML is relatively easier because of the bipolar characteristic of mCP. Therefore, the first EML is in fact the dominant EML (DEML) whereas the second EML acts as the SEML. In addition, the LUMO level of TAPC is 0.6 eV higher than that of mCP, and the HOMO level of TPBi is 0.4 eV lower than that of mCP [10,17]; hence holes and electrons can be well confined within EMLs, and the width of the recombination zone can be minutely adjusted by controlling the thicknesses of DEML and SEML. The HOMO and LUMO levels of complexes 1 and 2 are all within those of mCP and TPBi. Therefore, carrier trapping was expected to be the dominant EL mechanism of these devices [18].

Based on complexes 1 and 2, two single-EML devices with the structures of ITO/TAPC (40 nm)/complex 1 (10%): mCP (20 nm)/ TPBi (40 nm)/LiF (1 nm)/Al (100 nm) (device A) and ITO/TAPC (40 nm)/complex 2 (6%): mCP (20 nm)/TPBi (40 nm)/LiF (1 nm)/ Al (100 nm) (device D), respectively, were firstly fabricated as reference devices by selecting mCP as the host material and designing both HTL and ETL to be 40 nm. In a previous paper [9], the doping concentrations of complexes 1 and 2 in mCP were optimized to be 10% and 6%, respectively, while the thickness of DEML was fixed at 20 nm. As listed in Table 1, devices A and D reached the maximum forward viewing current efficiencies (η_{cmax}) of 25.45 and 67.95 cd/A, the maximum forward viewing power efficiencies (η_{pmax}) of 23.52 and 69.90 lm/W, and the maximum brightness (B_{max}) of 38963 and 64351 cd/m², respectively. At 5.6 and 5.1 V, devices A and D reached a certain brightness of 1000 cd/m² with current efficiencies of 23.70 and 52.75 cd/A. respectively.

Having kept the thicknesses of HTL and DEML constant, devices B (ITO/TAPC (40 nm)/complex 1 (10%): mCP (20 nm)/complex 1 (10%):TPBi (10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm)) and E (ITO/TAPC (40 nm)/complex 2 (6%): mCP (20 nm)/complex 2 (6%): TPBi (10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm)) were fabricated and characterized by doping complex 1 (10%) and complex 2 (6%), respectively, into the 10 nm TPBi layer as the SEML, Meanwhile, the thickness of HBL/ETL was reduced to 30 nm in order to maintain the constant total thickness of these devices. As shown in Figs. 2 and 3, the insertion of SEML caused a slight shift of current density but also a significant enhancement of current efficiency and brightness. As listed in Table 1, the $\eta_{c max}$, $\eta_{p max}$, and B_{max} of device B are 29.77 cd/A, 22.40 lm/W, and 53,750 cd/m², respectively, whereas the $\eta_{c max}$, $\eta_{p max}$, and B_{max} of device E are 83.72 cd/ A, 89.62 lm/W, and 108,046 cd/m², respectively. At 5.3 and 4.7 V, devices B and E reached a certain brightness of 1000 cd/m^2 with current efficiencies of 28.79 and 60.26 cd/A, respectively. Interestingly, device E displayed lower turn-on voltage (2.9 V) than that (3.1 V) of device D, whereas devices A and B displayed an equivalent turn-on voltage of 3.2 V. At relatively low current density, interestingly, device B showed even lower EL efficiency than that of device A. These phenomena can be attributed to the higher doping concentration of complex 1 than that of complex 2, which causes a relatively easier electron transport in the devices based on complex 1. Therefore, the insertion of SEML has more significant influence on the devices based on complex 2.

To further improve device performance, we have also optimized the doping concentrations of complexes 1 and 2 in the SEML. As listed in Table 1, Figs. 2 and 3, further improved device performances were obtained when the doping concentrations of complexes 1 and 2 in SEML were increased to 12% (device C: ITO/TAPC (40 nm)/complex 1 (10%): mCP (20 nm)/complex 1 (12%): TPBi (10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm)) and 7% (device F: ITO/TAPC (40 nm)/complex 2 (6%): mCP (20 nm)/complex 2 (7%):TPBi (10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm)), respectively. The η_c max, η_p max, and B_{max} of device C are 35.22 cd/A, 26.36 lm/W, and 56,678 cd/m², respectively, which are 38.39%, 12.07%, and 45.47% higher than those of device A. On the other hand, the η_c max, η_p max, and B_{max} of device F are 90.68 cd/A,

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