



# Efficient organic blue fluorescent light-emitting devices with improved carriers' balance on emitter molecules by constructing supplementary light-emitting layer



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## ABSTRACT

In this work, we demonstrated an efficient device design strategy in improving the electroluminescent performances of blue fluorescent emitter 4,4'-bis(2,2-diphenylvinyl)-1,1'-biphenyl (BCzVBi) by constructing predominant and supplementary light-emitting layers. 4,4',4''-tris(carbazole-9-yl)triphenylamine (TcTa) and 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy) were selected as host materials. Experimental results revealed BCzVBi doped TcTa film functions as predominant light-emitting layer, while BCzVBi doped 26DCzPPy film functions as supplementary light-emitting layer, which helps to broaden the recombination zone, improve the trapping of carriers and balance the distribution of holes and electrons on emitter molecules. Finally, the 15 wt% doped device with the Commission Internationale de l'Eclairage coordinates of (0.143, 0.189) obtained the maximum current efficiency, power efficiency, external quantum efficiency and brightness up to 11.99 cd/A, 10.91 lm/W, 8.0% and 36,060 cd/m<sup>2</sup>, respectively. Even at the high brightness of 1000 cd/m<sup>2</sup>, current efficiency as high as 9.76 cd/A can be retained by the same device.

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## 1. Introduction

Organic light-emitting devices (OLEDs) have attracted great interest throughout the world owing to their potential applications in solid-state lighting and full-color flat panel displays [1–3]. Although most reported phosphorescent devices displayed relatively higher electroluminescent (EL) performances compared with fluorescent devices, fluorescence emitting compounds are deemed to be worth studying because of the difficulty of realizing long lifetime blue phosphorescent devices [4]. For commercial application, three primary colors of blue, green and red are basically required [5]. In the past years, green and red EL devices have been improved significantly; consequently, their efficiencies and brightness have basically satisfied the requirements of application [6,7]. However, blue EL devices have remained the most challenging of the primary emissive colors in terms of efficiency and brightness.

In the past decades, much important effort has demonstrated the efficacy of doping emitters into proper host materials in balancing carriers' distribution, broadening recombination zone, and even lowering operation voltage [8–10]. To ensure the well trapping of both holes and electrons on emitter molecules within light-emitting layer (EML), energy gaps of the utilized host materials should be wide enough [11]. Therefore, it is more difficult to select proper host materials for blue emitters due to their wider energy gaps than those of red and green emitters [12]. In addition, the wide energy gaps of most blue emitters especially the deep blue emitters are generally accompanied with the low-lying highest occupied molecular orbital (HOMO) level or the high-lying lowest unoccupied molecular orbital (LUMO) level, which results in high hole injection or electron injection barrier. As a result, most reported blue devices possessed unbalanced distribution of holes and electrons on emitter molecules, thus displayed high drive voltage, low EL efficiencies, rapid roll-off of EL efficiency and low brightness [13].

In the past years, many groups have paid their attentions to the design of efficient blue fluorescent emitters and the optimization of device structures [14,15]. For example, Kim et al. reported the fabrication and investigation of blue EL devices by doping the well-

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known blue fluorescent emitter 4,4'-bis(2,2-diphenylvinyl)-1,1'-biphenyl (BCzVBi) into hole/electron transport layers (HTL/ETL). The high-lying LUMO level of BCzVBi makes it difficult for BCzVBi molecules to trap enough electrons, thus causing the unbalanced carriers' distribution and the narrow recombination zone. Consequently, the obtained devices displayed high operation voltage and rapid roll-off of EL efficiency, and the optimized device realized the maximum brightness and current efficiency of only 13,190 cd/m<sup>2</sup> and 7.64 cd/A, respectively [14]. Based on a highly efficient novel blue fluorescent emitter, Lee et al. realized blue EL device with the Commission Internationale de l'Eclairage (CIE) coordinates of (0.144, 0.297). Although the maximum current efficiency up to 8.17 cd/A has been realized, the maximum brightness of the same device was only 9989 cd/m<sup>2</sup> due to the rapid roll-off of EL efficiency [15].

In this work, we aim to improve the EL performances of the well-known blue fluorescent emitter BCzVBi by constructing predominant and supplementary EMLs with matched energy levels, which was expected to be helpful in balancing the distribution of holes and electrons, and broadening the recombination zone [16–19]. Considering the high-lying LUMO level of BCzVBi, the predominant EML was designed by doping BCzVBi into p-type host material 4,4',4''-tris(carbazole-9-yl)triphenylamine (TcTa), while the supplementary EML was inserted between predominant EML and hole block layer (HBL) by doping BCzVBi into bipolar host material 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy). The doping concentrations of BCzVBi in predominant and supplementary EMLs were optimized in sequence. Finally, bright pure blue EL device with the maximum current efficiency, power efficiency, external quantum efficiency (EQE) and brightness as high as 11.99 cd/A, 10.91 lm/W, 8.0% and 36,060 cd/m<sup>2</sup>, respectively, was obtained.

## 2. Experimental

All the organic materials used in this study were obtained commercially from Luminescence Technology Corp. and used as received without further purification. Indium-tin-oxide (ITO) coated glass with a sheet resistance of 10 Ω/sq was used as the anode substrate. Prior to film deposition, patterned ITO substrates were cleaned with detergent, rinsed in de-ionized water, and finally dried in an oven. All the organic layers were deposited with the rate of 0.1 nm/s under high vacuum ( $\leq 2.0 \times 10^{-5}$  Pa). The EMLs were prepared by co-evaporating BCzVBi and host material from two individual sources, and the doping concentration was modulated by controlling the evaporation rate of BCzVBi. MoO<sub>3</sub>, LiF and Al were deposited in another vacuum chamber ( $\leq 8.0 \times 10^{-5}$  Pa) with the rate of 0.01, 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 ten emitting dots with the active area of 9 mm<sup>2</sup> on each substrate. Current density-brightness-voltage (*J-B-V*) characteristics were measured by using a programmable Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a silicon photodiode. The photoluminescence (PL) and EL spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer. The external quantum efficiency (EQE) of EL device was calculated based on the photo energy measured by the photodiode, the EL spectrum, and the current pass through the device.

## 3. Results and discussion

Device structure and HOMO/LUMO levels diagram of the

designed OLEDs are depicted in Fig. 1. In this case, BCzVBi was selected as the blue emitter because of its pure blue emission, high luminescent efficiency and excellent thermal stability. Due to the high hole mobility ( $1 \times 10^{-2}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) and high-lying LUMO level (−1.8 eV), di-[4-(*N,N*-ditolyl-amino)-phenyl]cyclohexane (TAPC) was utilized as hole transport/electron block layer (HTL/EBL) [20]. Meanwhile, 1,3,5-Tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) was used as hole block/electron transport layer (HBL/ETL) due to its low-lying HOMO level (−6.7 eV) and high electron mobility ( $1 \times 10^{-3}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) [21]. To facilitate the balance of holes and electrons on emitter molecules, p-type material TcTa and bipolar material 26DCzPPy were chosen as the host materials of EML1 and EML2, respectively [22]. The molecular structures of BCzVBi, TcTa and 26DCzPPy were also shown in Fig. 1.

As shown in Fig. 2, holes and electrons inject from anode and cathode into EMLs via HTL and ETL, respectively. According to the previous reports, the stepwise HOMO levels of TAPC (−5.5 eV), TcTa (−5.7 eV), and 26DCzPPy (−6.1 eV) are beneficial for the injection and transport of holes, while the stepwise LUMO levels of TmPyPB (−2.7 eV), 26DCzPPy (−2.6 eV), and TcTa (−2.4 eV) are beneficial for the injection and transport of electrons [7,21]. Therefore, balanced distribution of carriers (holes and electrons) and wide recombination zone could be expected. In addition, the LUMO level of TAPC is 0.6 eV higher than that of TcTa, while the HOMO level of TmPyPB is 0.6 eV lower than that of 26DCzPPy; as a result, holes and electrons are well confined within EMLs [23]. In this case, most holes within EMLs should be well trapped by BCzVBi molecules due to its high-lying HOMO level (−5.4 eV), which is 0.3 and 0.7 eV higher than those of TcTa and 26DCzPPy, respectively. The high-lying LUMO level of BCzVBi (−2.4 eV), which equals to that of TcTa and is even 0.2 eV higher than that of 26DCzPPy (−2.6 eV), makes it difficult for electrons to transfer onto BCzVBi molecules within EMLs. As a result, unbalanced distribution of holes and electrons on BCzVBi molecules was expected.

Firstly, a series of single-EML devices with the structure of ITO/MoO<sub>3</sub> (3 nm)/TAPC (50 nm)/BCzVBi (x wt%):26DCzPPy (10 nm)/TmPyPB (50 nm)/LiF (1 nm)/Al (100 nm) were fabricated and examined to optimize the doping concentration of BCzVBi within 26DCzPPy layer. Fig. 3(a) depicted the doping concentration dependence of EL efficiency. Current density-brightness-voltage characteristics of these devices are depicted in the insert of Fig. 3(a). The 15 wt% doped device (defined as device A) gave the highest brightness of 20,958 cd/m<sup>2</sup>, the highest current efficiency of 12.30 cd/A (EQE = 7.6%), and the highest power efficiency of 12.53 lm/W, respectively. At the certain brightness of 1000 cd/m<sup>2</sup>, current efficiency of device A was only 8.62 cd/A due to the rapid roll-off of efficiency, which eventually causes the low brightness. EL spectra of these devices operating at the current density of 10 mA/cm<sup>2</sup> were measured and shown in Fig. 3(b). Besides the main emission peaked at about 455 nm, which agrees well with the PL spectrum of BCzVBi, another shoulder emission peaked at about 480 nm was also observed. According to previous reports [24], the 480 nm shoulder peak was attributed to the aggregation of BCzVBi molecules. Pure BCzVBi emission was observed in all these devices with BCzVBi at different doping concentrations, indicates the completed Förster energy transfer from host to emitter molecules.

To further improve the electroluminescent performances of BCzVBi, a series of double-EMLs devices with the structure of ITO/MoO<sub>3</sub> (3 nm)/TAPC (50 nm)/BCzVBi (x wt%):TcTa (10 nm)/BCzVBi (x wt%):26DCzPPy (10 nm)/TmPyPB (50 nm)/LiF (1 nm)/Al (100 nm) were also fabricated and examined. Compared with single-EML devices, as shown in Fig. 4(a), double-EMLs devices displayed slower roll-off of efficiency and thus higher brightness. EL spectra of these double-EMLs devices operating at the current density of 10 mA/cm<sup>2</sup> were measured and shown in Fig. 4(b). Pure BCzVBi

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