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Angle-stable inverted top-emitting white organic light-emitting devices based on gradient-doping electron injection interlayer

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ABSTRACT

By adopting an 4,7-diphenyl-1,10-phenanthroline (Bphen):Ag interlayer with the gradient structure between the cathode and electron injection layer, the electron injection of white inverted top-emitting organic light-emitting devices (WITOLED) was enhanced. The structure of Ag–Ge–Ag anode capping with light outcoupling was used to ensure the spectral stability with the angles. The structure of the emission layer was carefully designed to guarantee the spectral stability with the voltages. The highest current efficiency of the WITOLED can reach 21.7 cd/A. The CIE 1931 coordinates varies only 0.022 in X and 0.015 in Y (0°–60°).

Organic light-emitting device (OLED) is one of the most promising lighting and display applications since its outstanding advantages, such as the superior color quality, high efficiency and compatibility with flexible substrates [1]. White OLED (WOLED) has great potential application in display technology. The method used to manufacture the active matrix OLED (AMOLED) large scale display panel is WOLED cooperated with color filters, which costs less and requires simpler processes. Such method may be a valid way for realizing the widely practical application of OLED in display. One special structure of OLEDs is top-emitting OLED (TEOLED), which is applied widely in high-quality AMOLED displays and relatively cheaper than the conventional bottom-emitting OLED (BOLED) [2]. As low cost display driver circuits are preferentially based on n-channel a-Si TFT technology, then the inverted TEOLED (ITEOLED) is needed since the cathode on the bottom of ITEOLED could work well with the n-channel a-Si TFT [3]. However, there are only a few reports about white ITEOLED. Special structure and stacked structure has been employed to overcome or utilize the characteristics of white ITEOLED. By utilizing the characteristic of microcavity effect, Wang et al. achieved white ITEOLEDs with merely a single blue emitting layer [4]. Schwab et al. [5] and Najafabadi et al. [6] used the stacked structure with four colors (red, blue, green, yellow) or two colors (orange, blue) to fabricate the white ITEOLED.

White ITEOLED faces both the problem of high reflectance metal anode which would lead to strong microcavity effect and the problem of electron injection. Metal anode, which is used widely in OLED with top-emitting structure, has a higher reflectivity and lower transmittance. A strong microcavity effect would be caused when working together with the other opaque metal cathode. Microcavity effect may make the spectra narrowed and weaken the angle stability of devices and it is depended on the reflectivity of the mirrors made of two electrodes, cavity length of the devices, and the losses due to internal absorption [7]. Such feature of ITEOLED applied the metal electrode applied in monochromic OLED would not influence its characteristics greatly and even enhance the quantum efficiency of monochromic TEOLED, but when it comes to white OLED with top-emitting structure, it would lead to the narrow electroluminescent spectra [5].

Several methods have been proposed to modify the optical properties of the metal electrode. A traditional way is to add a capping layer above the semitransparent metal electrode. Thomschke et al. made a white ITEOLED which is angular stability by applying a 55 nm organic capping layer [7]. Another way to ameliorate the optical properties of the metal electrode is to replace the metal electrode with the transparent oxide conductor with a buffer layer, for example, recently Kim et al. using indiumzinc oxide top electrode with 1, 4, 5, 8, 9, 11-hexaazatriphenylene-hexacarbonitrile buffer layer archived angle stable green ITEOLED, of which the CIE 1931 coordinates vary only 0.02 in both X and Y (0°–60°) [8]. The third way is using the multilayer electrodes. Schwab et al. comprised an ultra-thin MoO_3–Au wetting layer system below

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the actual Ag electrode to fabricate a multi-layer thin film metal electrode [5]. Our previous work demonstrates that TEOLEDs with the Ag/Ge/Ag (AGA) cathode achieved excellently angle-stable characteristic [9].

The other question of ITEOLED is the bad behavior of electron injection [10], which leads that the behaviors of similar conventional device are much better than the inverted one. A possible explanation proposed by Scholz et al. for such phenomenon is the process of deposition of metal onto organic semiconductors which create an organic–inorganic mixed interlayer between the organic bulk material and the metal [11]. Several kinds of interlayers, such as Ag:Cs:diphenyl-1,10-phenanthroline (Bphen) [12], TiO_2 [13], ultrathin Alq_3–LiF-Altrilayer [14], MgO [15], and Al/Liq [16] have been applied between the cathode and electron transporting layer (ETL) to improve the electron injection. In our previous work, Bphen:Ag interlayer has been used to improved the electron injection in green ITEOLED [17]. In this work, white ITEOLEDs with Bphen:Ag interlayer (BAI) and AGA anode were demonstrated. The influence of the thickness and Ag concentration of Bphen:Ag interlayers on the performance of the ITEOLED were studied. And the white ITEOLED with a gradient Bphen:Ag interlayer shows a highest efficiency of 21.7 cd/A and angle-stable characteristic, the CIE 1931 coordinates varies only 0.022 in X and 0.015 in Y (0^o–60^o).

According to different purposes, devices with different emission layer (EML) and interlayer have been designed. The structures of the devices are shown in Fig. 1. The structures of device A, B, C, D are different in the BAI, while devices D, E, F, G and W are different in EMLs. And the detailed structure of the EML and interlayer is listed in Table 1. Ag(1 nm)/Ge(1 nm)/Ag(12 nm) (AGA), MoO_3, di-[4-(N,N-di-ptolyl-amino)-phenyl] cyclohexane (TAPC), 4,4′,4″-tris(carbazol-9-yl) triphenylamine (TCTA), Bphen, and Ag were used as anode, anode buffer layer, hole-transporting layer (HTL), electron blocking layer, electron transporting layer (ETL) and cathode. 50 nm NPB capping on anode was used as the light outcoupling layer. Devices are fabricated by thermal evaporating on the precleaned glass substrates under the high vacuum (5×10^{-4} Pa). The material deposition rate was monitored with quartz crystal. The typical deposition rate of organic materials and oxide materials is 0.1–0.2 nm/s, while that of metal materials is 0.2–0.3 nm/s. The emission area of the investigated OLEDs is 10 mm^2. Luminance-voltage (LV) and current-voltage (IV) characteristics of unpackaged devices are measured simultaneously using a Keithley 2400 source meter and a Minolta luminance meter LS-110 in air at room temperature. The spectra of the devices are measured with Ocean Optics Maya 2000-PRO spectrometer.

Table 1
The detailed structure of BAI and EML.

Device	ABI	EML
A	BAI1 (2 nm)	EML1(20 wt%, 30 nm)
B	BAI1 (4 nm)	EML1(20 wt%, 30 nm)
C	BAI2 (4 nm)	EML1(20 wt%, 30 nm)
D	BAI2 (2 mn)/BAI1 (2 nm)	EML1(20 wt%, 30 nm)
E	BAI2 (2 mn)/BAI1 (2 nm)	EML1 (15 wt%, 25 nm)/EML2 (10 wt%, 5 nm)
F		EML2 (10 wt%, 5 nm)/EML1 (15 wt%, 25 nm)
G		EML2 (3 wt%, 2 nm)/EML1 (15 wt%, 28 nm)
W		EML3 (10 wt%, 20 nm)/EML1 (15 wt%, 10 nm)

EML1: CBP: Firpic; EML2: CBP: $Ir(MDQ)_2(acac)$; EML3: CBP: PO-01.
BAI1: Ag:Bphen (1:1); BAI2: Ag:Bphen (3:1).
Firpic: bis (3,5-difluoro-2-(2-pyridyl) phenyl-(2-carboxypyridyl)iridium.
$Ir(MDQ)_2$: bis(2-methyldibenzo[f, h]quinoxaline)(acetylacetonate)iridium(III).
PO-O1: iridium(III) bis(4-phenylthieno[3,2-c] pyridinato-N,$C^{2'}$) acetylacetonate.

In our previous work, an efficient inverted green OLED based on Bphen:Ag interlayer was fabricated. Here, a further research about the effect of the BAI on the electron injection had been made. The devices with same EML but different BAI were fabricated. The BAI of device A, B, C, and D is Ag:Bphen (1:1, 2 nm), Ag:Bphen (1:1, 4 nm), Ag:Bphen (3:1, 4 nm) and Ag:Bphen (3:1, 2 nm)/Ag:Bphen (1:1, 2 nm). The EML of these four devices are 4, 4′-bis(carbazol-9-yl)biphenyl (CBP): 20 wt% bis(3,5-difluoro-2-(2-pyridyl) phenyl-(2-carboxypyridyl)iridium(III) (Firpic) (30 nm). Fig. 2 shows the voltage–current density characteristics of devices A–D. It can be seen that the current density of device D is the highest among all device at the same voltage, which means that the gradient structure of Bphen:Ag interlayer has the best electron injection behavior. The lowest current density of device C indicates that high concentration of Ag in Bphen would undermine the effect of Bphen:Ag interlayer. The property of BAI with high concentration of Ag could be treated as a single Ag layer which reduces the effect of BAI. Another phenomenon needed to be noticed is that similar current density–voltage characteristics exist in device A and B, which indicated that the thickness of the Bphen:Ag interlayer have little influence on the electron injection.

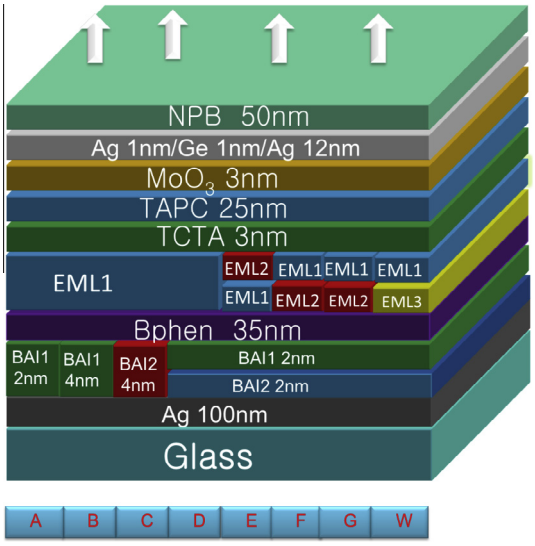

Fig. 1. The structure of the devices.

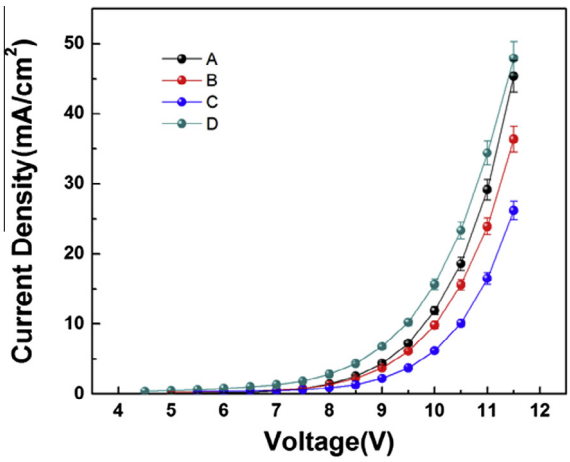

Fig. 2. The voltage–current density characteristics of devices A–D.