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# Efficient multilayer and single layer phosphorescent organic

# light-emitting devices using a host with balanced bipolar transporting properties and appropriate energy level

Chunxiu Zang<sup>a</sup>, Xiaomei Peng<sup>a</sup>, Hui Wang<sup>a</sup>, Ziwei Yu<sup>a</sup>, Letian Zhang<sup>a</sup>, Wenfa Xie<sup>a,\*</sup>, Hongyu Zhao<sup>b,\*</sup>

<sup>a</sup>State key Laboratory on Integrated Optoelectronics, College of Electronics Science and Engineering, Jilin University, Changchun, 130012, People's Republic of China.

<sup>b</sup>Beijing Tuocai Optoelectronics Technology CO. LTD, Beijing, 100086, People's Republic of China.

\*Corresponding author: xiewf@jlu.edu.cn, zhy\_china@126.com

# Abstract:

Efficient multilayer (ML) and single layer (SL) phosphorescent organic light-emitting devices (PHOLEDs) with a bipolar host 9,9'-[4'-(2-ethyl-1*H*-benzimidazol-1-yl) [1,1'-biphenyl]-3,5-diyl] bis-*H*-Carbazole (EBBPC) are fabricated. The photophysical, electrochemical and carrier transporting properties of EBBPC host are investigated. The maximum external quantum efficiency (EQE) can reach 14.9%, 15.6%, 16.0% and 15.4% for red (R), green (G), blue (B), and white ML-PHOLEDs. And the maximum EQE can also reach 10.1%, 14.6% and 9.8% for R, G, and B SL-PHOLEDs. Besides, the SL-PHOLEDs show low efficiency roll-off due to the broader exciton formation zone in SL OLEDs. The excellent performance of ML- and SL-PHOLEDs could be attributed to the balanced bipolar transporting properties and appropriate energy level of the host.

Keywords: bipolar; organic light-emitting device; single-layer; roll-off

### **1. Introduction**

Phosphorescent organic light-emitting diodes (PHOLEDs) have received tremendous scientific and industrial interest in recent years for the high efficiency as they could achieve 100% internal quantum efficiency in principle by utilizing both the singlet and triplet excitons [1-8]. In the meantime, the PHOLEDs reported generally adopt multilayer configuration in which the emission layers are sandwiched within many functional layers to obtain balanced charge fluxes and improve the device efficiency. In these complicated structures OLEDs, however, the presence of heterojunction interfaces in the multilayer structure is detrimental to the device stability due to the accumulation of charges, exciplex formation and the generation of a higher electric field at the interfaces [9, 10]. Therefore, special attention must be paid to match the appropriate energy levels of adjacent layers and to avoid the above phenomena. However, the employing of these complicated device configurations will doubtlessly increase the complexity of the manufacture and the fabrication cost. This presents an intense motivation to develop single-layer OLEDs (SLOLEDs), as much fewer types of material are required and simpler structure is adopted.

Moreover, in PHOLEDs, as triplet excitons have a long lifetime by nature, triplet-triplet annihilation (TTA) and triplet-polaron quenching (TPQ) caused by high concentration of excitons are the dominating loss mechanisms result in efficiency roll-off [11, 12]. Thus, the phosphorescent materials are often doped into a suitable host material that can disperse triplet emitters to reduce aggregation quenching and TTA of guest triplet emitters and plays an indispensable role in energy transfer and carrier transport at the same time [6, 13]. As a high-performance host material, it must fulfill the some certain requirements including a high triplet energy level suitable for dopants to facilitate energy transfer, similar mobility for both electrons and holes (i.e. bipolar materials) to ensure a broad exciton recombination zone, and the energy level [highest occupied molecular orbital (HOMO)/lowest unoccupied molecular orbital (LUMO)] of the host material should be matched with neighboring layers in order to achieve both an effective carrier injection and a good carrier confinement [3, 14-18]. Thus, many hosts do not suit for different guests, e. g., 4,4'-*N*,*N*'-dicarbazole- biphenyl

2

(CBP) [19] and poly-(*N*-vinylcarbazole) (PVK) [20] are unsuitable for blue PHOLEDs, as the blue phosphorescent dye in common use is bis [(4,6-difluorophenyl)-pyridinato- $N,C^{2}$ ] (picolinato) Ir(III) (FIrpic) with the triplet level of 2.62 eV, the triplet levels of CBP (2.56 eV) and PVK (2.5 eV) are lower than that of FIrpic; hence the energy-transfer process from CBP or PVK to FIrpic is endothermic, which limits the device efficiency, operating temperature and long-term operational stability [18, 21].

More importantly, many host materials could not be used as host in SLOLEDs because of the unsuitable HOMO/LUMO level for electrode or unipolar transport property. In SLOLEDs, Poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS) modified Indium tin oxide (ITO) is usually used as anode and excellent hole injection is easier to achieve for most hosts, therefore, the hosts in the SLOLEDs need to have better electron injection ability. In addition, in order to avoid TTA and TPQ caused by the carrier accumulation at the electrode (especially cathode), the host materials need to have balanced bipolar transport properties, excellent electron transporting property in particular. As a result, there are relatively few reports on well-performing SLOLEDs [22-26].

In efficient ML-**SL-PHOLEDs** this paper, and with bipolar 9,9'-[4'-(2-ethyl-1*H*-benzimidazol-1-yl) [1,1'-biphenyl]-3,5-diyl] bis-*H*-Carbazole (EBBPC) host is reported. For comparison, ML-PHOLEDs with CBP, 2,7-bis (diphenylphosphoryl)-9-[4-(*N*,*N*-diphenylamino) phenyl]-9-phenylfluorene (POAPF) and 2,6-bis(3-(carbazol 9,9'-[4'-(2-ethyl-1H-benzimidazol-1-yl) -9-yl) phenyl)pyridine (26DCzPPy) host are also fabricated. Iridium (III) bis-(2-methyldibenzo- [f, h] quinoxaline) (acetylacetonate) [Ir(MDQ)<sub>2</sub>(acac)], tris(2-phenylpyridine) iridium  $[Ir(ppy)_3]$  and Firpic are used as the red, green and blue dopants. EBBPC host shows balanced bipolar transporting properties and appropriate energy level which makes it can be used as efficient host for most common phosphorescent dopants in ML- and SL-PHOLEDs.

# 2. Experimental section

2.1. Materials and instruments

Ultraviolet-visible (UV-vis) absorption and photoluminescence (PL) spectra at room temperature are measured using Cary 500 UV-vis-NIR spectrophotometer and FL-4600 FL spectrophotometer, respectively. The phosphorescence spectrum is studied from the delayed emission of EBBPC at 77 K. Cyclic voltammetry (CV) studies are recorded on a BAS 100 W instrument containing 0.1 M tetrabutylammonium hexa-fluorophosphate in  $CH_2Cl_2$  as a supporting electrolyte with a scan rate of 100 mV/s. All materials and solvents are commercially available and used as received without further purification.

2.2. Synthesis



Scheme1. Synthetic routes of EBBPC.

The synthetic route and structure of EBBPC is shown in Scheme 1. EBBPC can be synthesized according to the method described in Ref. 27 and 28. 11.2 g (70 mmol) carbazole (B), 10 g (31 mmol) 1,3,5-tribromobenzene (A), 4.9 g (35 mmol) K<sub>2</sub>CO<sub>3</sub>, 0.06 g (3 mmol) copper (I) iodide, and 0.06 g (3 mmol) 1,10-phenanthroline are added to 100 mL of dried dimethyl formamide (DMF). The mixture is heated at 160 °C for 12 h under nitrogen atmosphere. After this mixture cooled to room temperature, the reaction is quenched with 200 mL of water and extracted with 100 mL of dichloromethane three times. Combining the organic layer and removing the solvent, the residue is then purified by silica gel column chromatography with petroleum ether/ethyl acetate (10:1) as the eluent to give 7.78 g of white power (C). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, d): 8.13 (d, 2 H), 7.75 (s, 1 H), 7.70 (s, 2 H), 7.54-7.25 (m, 6 H). MS (ESI) [m/z]: calcd for C<sub>18</sub>H<sub>11</sub>Br<sub>2</sub>N, 401.09; found, 401.00. Anal. Calcd for C<sub>18</sub>H<sub>11</sub>Br<sub>2</sub>N: C, 53.90; H, 2.76; N 3.49. Found: C, 53.82; H, 2.84; N 3.53%.

Under an argon atmosphere, 3.0 g (10 mmol) of intermediate C, 4.5 g (10 mmol) of

intermediate D, 0.23 g (0.2 mmol) of Pd(PPh<sub>3</sub>)<sub>4</sub> and 10 ml of an aqueous 2M sodium carbonate solution are added to 50 ml of THF, and the mixture is heated and stirred under reflux. To the resultant, menthol is added to precipitate solids. The solids is collected and dissolved in dichloromethane to obtain an oily matter. The oily matter is purified by means of silica gel column chromatography to obtain 4.3 g of EBBPC (yield 80%). As the eluent of silica gel column chromatography, a solvent of hexane: dichloromethane=10:1, and a solvent of henane: dichloromethane=3:1 are used in this sequence. MS (MALDI-TOF) [m/z]: calcd for C<sub>45</sub>H<sub>32</sub>N<sub>4</sub> 628.76; found, 628.90. <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>  $\delta$  [ppm]): 8.18 (d, J = 7.8 Hz, 4H), 8.00 (d, J = 1.8 Hz, 2H), 7.91 (d, J = 8.4 Hz, 2H), 7.89 (t, J = 1.8 Hz, 1H), 7.82 (d, J = 7.8 Hz, 1H), 7.63 (d, J = 7.8 Hz, 1H), 7 7.8 Hz, 4H), 7.51 (d, J = 8.4 Hz, 2H), 7.48 (t, J = 7.2 Hz, 4H), 7.34 (t, J = 7.2 Hz, 4H), 7.30 (t, J = 7.2 Hz, 1H) 7.22 (t, J = 7.8 Hz, 1H), 7.16–7.18 (m, 1H), 2.86–2.90 (m, 2H), 1.40 (t, J = 7.2 Hz, 3H). <sup>13</sup>C NMR (600 MHz, CDCl<sub>3</sub>,  $\delta$  [ppm]):156.08, 143.25, 142.38, 140.54, 140.12, 139.77, 136.35, 136.14, 129.00, 128.68, 128.19, 127.95, 126.26, 125.26, 124.49, 124.33, 123.73, 122.79, 122.55, 120.57, 120.54, 119.19, 109.85, 109.62, 77.21, 77.00, 76.78, 21.28, 12.06. Anal. Calcd for C<sub>45</sub>H<sub>32</sub>N<sub>4</sub>: C, 85.96; H, 5.13; N 8.91. Found: C, 85.73; H, 5.20; N 8.65%.

# 2.3. Device fabrication and characterization

All the devices in this paper are manufactured on glass substrates coated with conducting ITO. The substrates are cleaned with Decon 90 and deionized water, dried in the oven and then handled in plasma for about 5 min. In advance of thermal vapor deposition process, a PEDOT:PSS layer with the thickness to be approximately 40 nm is spin-coated on the top of the ITO anode for SL-PHOLEDs. Finally, organic layers and cathode materials are deposited on the substrates sequentially under vacuum ( $\sim 5.0 \times 10^{-4}$  Pa). A shadow mask is used to define the cathode and to make four identical 10 mm<sup>2</sup> devices on each substrate. Luminance-current-voltage characteristics and spectra of unpackaged devices are measured simultaneously using Goniophotometric Measurement System based on spectrometer (GP-500, Otsuka Electronics Co. Osaka, Japan) in air at room temperature. The impedance studies are carried out using an Impedance Analyzer (TH2829C, Tonghui Co. Changzhou, China).

5

This setup allows frequency sweeps covering a range from 20 to  $10^6$  Hz. The ac oscillator level is set to 100 mVrms for all measurements and the superimposed dc bias is varied between 0 and +10 V.

# 3. Results and Discussion

#### **3.1.** Photophysical properties

Figure 1 (a) shows the UV-vis absorption and emission spectra of EBBPC in CH<sub>2</sub>Cl<sub>2</sub>. Fig.1 (b) and (c) depicts the cyclic voltammogram, thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) curves of EBBPC. The key parameters of EBBPC are summarized in Table 1. The optical energy band gap  $(E_g)$  of EBBPC is calculated to be 3.35 eV from the onset of the absorption spectrum according to the UV-vis absorption curve. The fluorescence emission peak of EBBPC is 400 nm and its singlet state  $(S_1)$  is estimated to be 3.1 eV accordingly. The phosphorescence spectrum is obtained from the delayed emission of EBBPC at 77 K and the triplet energy  $(T_1)$  is estimated to be 2.74 eV (453 nm). The HOMO level of EBBPC is estimated to be -5.71 eV through the cyclic voltammogram measurement and LUMO level is deduced to be -2.36 eV from HOMO and  $E_{\rm g}$ . The oxidation signal of EBBPC exhibit quasi-reversible oxidation, we believed that it is due to that EBBPC is a carbazole-containing complex [29]. At the same time, EBBPC exhibits high glass transition temperature ( $T_g$ ) of 142.6 °C. From the structure of **EBBPC** see that EBBPC contains carbazole we can two and one benzimidazole groups. Such a molecular configuration renders the molecule in a non-planar structure and which is strongly beneficial to the thermal stability. The carbazole group has high triplet energy and hole transporting property, while the benzimidazole is an electron transporting group. The incorporation of ethyl at the 2-position of the benzimidazole ring limits the extent of conjugation which further keeps the triplet energy gap at a very high level. Thus, EBBPC has high triplet energy, excellent charge transport properties and high thermal stability.



Fig.1. (a) Normalized absorption spectrum (Abs), photoluminescence spectrum at 298 K (PLRT), phosphorescence spectrum at 77 K (PLLT) of EBBPC, Abs of FIrpic and PLRT of POAPF in CH<sub>2</sub>Cl<sub>2</sub>. (b) Cyclic voltammogram of EBBPC. (C) Thermogravimetric analysis (TGA) [Inset is differential scanning calorimetry (DSC) curve] of EBBPC.

Table 1 Photophysical characteristics of EBBPC, POAPF, CBP and 26DCzPPy									
Complex	$\lambda_{abs,max}^{a}$	$\lambda_{\mathrm{PL,max}}^{\mathrm{a,b}}$	S <sub>1</sub> <sup>c</sup>	T <sub>1</sub> <sup>d</sup>	E <sub>g</sub> <sup>e</sup>	HOMO <sup>f</sup>	LUMO <sup>g</sup>	Tg	T <sub>d</sub>
	(nm)	( <b>nm</b> )	(eV)	(eV)	(eV)	(eV)	(eV)	(°C)	(°C)
EBBPC	240	400, 483	3.1	2.74	3.35	-5.71	-2.36	142.6	431.2
POAPF	294 <sup>h</sup>	510, N.A.	N.A.	2.75 <sup>h</sup>	2.86 <sup>h</sup>	-5.26 <sup>h</sup>	-2.40 <sup>h</sup>	129 <sup>h</sup>	466 <sup>h</sup>
CBP	341 <sup>i</sup>	355, 485 <sup>i</sup>	3.49 <sup>i</sup>	2.56 <sup>i</sup>	3.10	6.0	2.90	62 <sup>i</sup>	N.A.
26DCzPPy	N.A.	N.A. 457	N.A.	2.71 <sup>j</sup>	3.40	6.05 <sup>j</sup>	2.65 <sup>j</sup>	N.A.	N.A.

<sup>a</sup>Measured in CH<sub>2</sub>Cl<sub>2</sub> (10<sup>-5</sup> M) at 298 K and <sup>b</sup>77 K. <sup>c</sup>Estimated from the emission spectrum at 298 K. <sup>d</sup>Estimated from the emission spectrum at 77 K. <sup>e</sup> Estimated from the UV-vis absorption spectrum. <sup>f</sup>Calculated from onset oxidation potential. <sup>g</sup>Deduced from HOMO and  $E_g$ . <sup>h</sup>Obtained from [30]. <sup>i</sup> Obtained from[31]. <sup>j</sup> Obtained from [32].

### 3.2. Carrier transportation properties

Admittance spectroscopy is a powerful method to infer charge transport properties of the material by analyzing the frequency-dependent capacitance of a device [33-35]. The average transit time  $\tau_{dc}$  of the carriers can be extracted from a plot of the negative differential susceptance against the frequency [33] or the frequency of imaginary part of impedance [ImZ(*f*)] [35]. Here, the frequency dependence [ImZ(*f*)] is used to

determine the mobility of EBBPC and the carrier mobility could be obtained from the equation of  $\mu = f_p d^2 / (\mathbf{V} \cdot \kappa)$ , where  $f_p$ ,  $\kappa$ ,  $\mu$ , d and  $\mathbf{V}$  represents the peak frequency, constant numerical factor, carrier mobility, the thickness of organic material and the applied voltage, respectively. Device A with structure of ITO/TAPC (3 nm)/EBBPC (50 nm)/TAPC (3 nm)/MoO<sub>3</sub> (3 nm)/Ag (100 nm) and device B with structure of ITO/LiF (1 nm)/TmPyPB (3 nm)/EBBPC (70 nm)/Bphen (3 nm)/LiF (1 nm)/Ag (100 nm) is fabricated to measure the frequency dependence of imaginary part of impedance. Where TAPC. TmPyPB, and **B**phen are 1,1-bis-(4-bis(4-tolyl)-aminophenyl) cyclohexene, 1,3,5-tri [(3-pyridyl)-phen-3-yl] benzene, and 4,7-diphenyl-1,10-phenanthroline, respectively. TAPC (TmPyPB and Bphen) and MoO<sub>3</sub> (LiF) are inserted between electrodes and EBBPC to facilitate the injection of hole (electron) and block the transport of electron (hole) for device A (B), and the time that carrier transport through the insert layer could be neglected since the insert layers are thin enough. Hole- and electron-only devices also are fabricated to give a qualitative explanation. The configuration of the hole-only device is  $ITO/MoO_3(3)$ nm)/TAPC (40)nm)/EBBPC(20 nm)/TAPC(40  $nm)/MoO_3(3)$ nm)/Ag(120 nm), while the electron-only device has the structure of ITO/LiF(0.5 nm)/Bphen(65 nm)/EBBPC(20 nm)/Bphen(40 nm)/Ag(120 nm). As the work function of MoO<sub>3</sub> being high enough to block electron injection [36], and Bphen is used as electron-transporting/hole-blocking layer [37], so it can be assumed that only single carriers can be injected and transported through the devices.



Fig.2 (a) and (b) are the frequency dependence of imaginary part of impedance [ImZ(f)] for different voltage of device A and B, respectively. (c)Current density-voltage characteristics of the hole-only and electron-only devices based on EBBPC, hole and electron mobility for EBBPC calculated from ImZ(f).

Fig.2 (a) and (b) show the ImZ(*f*) of device A and B at different DC voltage with frequency. Thus, the hole and electron mobility of EBBPC could be calculated and is shown in Fig. 2 (c). The electron mobility of the EBBPC is  $\sim 2 \times 10^{-5}$  cm<sup>2</sup> v<sup>-1</sup>·s<sup>-1</sup> and this value is about twice of its hole mobility. Fig.2 (c) depicts the current density-voltage curves of the single-carrier devices. We also could see that the current density of the electron-only device is higher than that of the hole-only device. Above results indicated that EBBPC is a bipolar transport material with better electron transporting property.



# 3.3. Monochromatic ML-PHOLEDs with EBBPC host

Fig.3 (a) The device structures of PHOLEDs with different hosts and guests. (b) Chemical structures of part of organic materials.

To confirm the effectiveness of EBBPC as host for PHOLEDs, red (R), green (G) and blue (B) ML-PHOLEDs whose structures are shown in Fig.3 have been fabricated successfully. They share the similar structure: ITO/MoO<sub>3</sub> (3 nm)/TAPC (35 nm)/TCTA (5 nm)/emitting layer (EML, 20 nm)/Bphen (50 nm)/LiF (0.5 nm)/Mg:Ag (15:1, mass ratio, 120 nm), the EML is the host materials doped with different phosphorescent dyes. Where TCTA is 4,4',4"-tris (carbazol-9-yl) triphenylamine. TAPC, TCTA are hole transporting layers (HTL), Bphen is electron transporting layer (ETL). For example, the blue PHOLEDs are demonstrated in detail. And the corresponding devices are marked as device  $B_1$  (EBBPC host), device  $B_2$  (POAPF host), device  $B_3$  (CBP host) and device  $B_4$  (26DCzPPy host). Previous literatures showed that POAPF can be used as bipolar host in blue ML-PHOLED and SL-PHOLED [30, 38]. Thus, the devices with POAPF host are used as references. Fig. 4 shows the normalized electroluminescence (EL) spectra of the devices  $B_1$  and  $B_2$  at

6 V. As can be seen, both the devices exhibit the typical FIrpic EL characteristics with a maximum intensity at *ca*. 471 nm. No extra emission is observed, indicating that efficient energy transfer from host to guest has been realized.



Fig.4. Normalized EL spectra of device B<sub>1</sub> and B<sub>2</sub> at 6 V [Inset: External quantum efficiency (EQE)-current density curves].

Fig. 5 (a) depicts the current density-voltage-luminance curves of the blue devices. The current density of device B<sub>2</sub> is higher than that of device B<sub>1</sub>, and the turn-on voltage (driving voltage at a brightness of 1 cd/m<sup>2</sup>) of device  $B_2$  (ca. 2.5 V) is also lower than that of device  $B_1$  (ca. 2.9 V). The low driving voltage and high current density of device B<sub>2</sub> could be attributed to the low HOMO level of POAPF. The HOMO/LUMO of EBBPC and POAPF is 5.71/2.26 eV and 2.4/5.26 eV, respectively. While the HOMO of TcTa (5.7 eV) is higher than that of POAPF and the electron injection barrier between Bphen and POPAF (0.4 eV) is smaller than that between Bphen and EBBPC (0.54 eV) as the LUMO of Bphen is 2.8 eV, which results in an easier injection of carrier in devices used POAPF host and a lower turn-on voltage. Although the current density of device  $B_1$  is lower than that of device B2, the maximum brightness and current efficiency (CE) of device B<sub>1</sub> is higher than those of device B1. The maximum brightness of device  $B_1$  (22050 cd/m<sup>2</sup>) is about twice of device  $B_2$  (11320 cd/m<sup>2</sup>). As could be seen in Fig.1 (a), there is a larger spectra overlap between absorption band of FIrpic and fluorescence band of EBBPC than POAPF, which means there should be a better energy transfer between FIrpic and EBBPC compared to POAPF and we can obtain a higher efficiency blue OLED using FIrpic as dopant and EBBPC host than using POAPF host. Fig.5 (b) shows the

efficiency-luminance characteristics of devices  $B_1$  and  $B_2$ . At brightness of 100, 1000 and 10000 cd/m<sup>2</sup>, the CE of device  $B_1$  are 28.7, 22.5 and 13.8 cd/A, while those of device  $B_2$  are 21.1, 19.7 and 5.8 cd/A, and power efficiencies (PE) of  $B_1$  are 26.2, 15.7 and 6.3 lm/W, while those of  $B_2$  are 22.1, 15.5 and 2.5 lm/W, respectively. The similar results are also observed for the devices using Ir(ppy)<sub>3</sub> or Ir(MDQ)<sub>2</sub>(acac) as dopant. The characteristics of the devices are summarized in Table 2. As can be seen, the devices based on EBBPC host show higher efficiencies than those using POAPF as host at the same brightness.



Fig.5 (a) Current density-voltage-luminance curves of blue devices  $B_1$  and  $B_2$ . (b) Current efficiency-power efficiency-luminance characteristics of blue devices  $B_1$  and  $B_2$ .

Device			L <sub>max</sub>	ηc.max, ηc.1000,	η <sub>p.max</sub> , η <sub>p.1000</sub> ,	EQE <sub>max</sub>	CIE(x,y)
		Von (V)	(cd/m²)	η <sub>c.5000</sub> (cd/A)	η <sub>p.5000</sub> (Im/W)	(%)	@6V
	B <sub>1</sub>	2.9	22050	31.9, 22.5, 17.3	33.4, 15.7, 9.3	16.0	(0.16,0.32)
	B <sub>2</sub>	2.5	11320	21.2, 19.7, 12.1	22.2, 15.5, 6.9	10.4	(0.16,0.33)
	B <sub>3</sub>	2.8	16080	15.5, 10.8, 7.3	16.2, 7.2, 3.6	7.9	(0.16,0.32)
PHOLEDs	B <sub>4</sub>	3.4	30640	22.9, 21.7, 18.9	20.5, 13.8, 9.9	11.2	(0.16,0.33)
with	G <sub>1</sub>	2.8	81160	54.1, 53.9, 50.2	54.5, 43.1, 32.4	15.6	(0.32,0.61)
POAPF, CBP and DCzPPy host	G <sub>2</sub>	2.4	49120	50.0, 48.5, 43.6	52.4, 37.5, 27.2	14.5	(0.32,0.61)
	G <sub>3</sub>	2.9	73930	44.5, 44.0, 41.9	46.5, 32.3, 24.8	13.0	(0.31,0.62)
	G4	3.4	63020	42.1, 42.0, 39.4	31.3, 25.0, 19.6	12.6	(0.30,0.62)
	R <sub>1</sub>	3.2	39100	23.5, 18.7, 15.3	21.1, 11.0, 7.1	14.9	(0.61,0.39)
	R <sub>2</sub>	2.9	16970	14.7, 11.6, 8.3	14.1, 7.3, 4.1	10.5	(0.61,0.38)
	R₃	3.0	32450	23.7, 21.7, 17.4	21.3,14.8, 9.3	15.0	(0.61,0.39)
	R4	3.8	25440	12.6, 12.1, 9.6	7.9, 6.3, 4.0	7.8	(0.60,0.39)
PHOLEDs	G-a	3.6	60480	50.2, 49.7, 44.6	38.5, 28.5, 21.2	14.6	(0.31,0.61)
with	G-b	3.4	54630	43.6, 43.5, 38.5	33.4, 27.6, 20.2	12.7	(0.31,0.61)
different	G-c	3.4	63080	48.8, 47.0, 40.8	38.3, 31.2, 22.8	14.2	(0.31,0.61)
ETL	G-d	3.4	48930	43.0, 39.5, 42.3	27.0, 26.6, 24.1	12.4	(0.31,0.61)
WOLED	W	3.4	37800	40.2, 35.0, 29.6	36.1, 20.0, 13.7	15.4	(0.34,0.42)

Table 2. EL characteristics of the devices.

To investigate the above phenomena, the energy level of the materials is shown in Fig.6 (a). The HOMO and LUMO energy level of POAPF are -2.40 and -5.26 eV [30], respectively. Thus, the  $E_g$  value of POAPF is 2.86 eV. It could be seen that EBBPC has better energy levels matching with the adjacent organic layer (TCTA and BPhen), especially the HOMO, which results in that there is less energy loss in the charge transporting of the devices using EBBPC host than those with POAPF host, as a result, device R<sub>1</sub>, G<sub>1</sub>, and B<sub>1</sub> show a higher luminance than device R<sub>2</sub>, G<sub>2</sub>, and B<sub>2</sub>, respectively.



Fig.6 (a) Scheme of energy level of the materials. (b) EQE performance of devices based on different hosts.(for example, REQE<sub>5000</sub> means the EQE of red OLED at 5000 cd/m<sup>2</sup>)

The high efficiency of the devices possibly results from the precise confinement of triplet excitons within the emissive layers. Triplet excitons generally have long diffusion lengths [39]. Good confinement of triplet excitons within the emitting layer (EML) can be achieved when all corresponding materials, i.e., the host materials and the adjacent HTL and ETL have higher  $E_T$  than that of the triplet emitter [40]. The  $E_T$  of POAPF and EBBPC are 2.75 [30] and 2.74 eV, respectively, which are higher compared to FIrpic (2.62 eV),  $Ir(ppy)_3$  (2.42 eV) [41], and  $Ir(MDQ)_2acac$  (2.05 eV) [42]. As we mentioned above, the electron-transporting character of EBBPC is superior to its hole-transporting character, so the triplet excitons mainly formed at the interface of EML/HTL. The  $E_T$  of TCTA (2.85 eV) [43] and Bphen (2.6 eV) [44] are high enough to confine the formed triplet excitons at the interface. As is shown in Fig.6 (a), the LUMO levels of the dopants are lower than the hosts (EBBPC and POAPF), so the electrons transport mainly on the dopants. The HOMO of POAPF is

deeper than that of dopants while the HOMOs of  $Ir(ppy)_3$  and  $Ir(MDQ)_2acac$  are deeper than that of EBBPC and there is only a small energy gap (0.09 eV) between FIrpic and EBBPC, which results in that the holes transport mainly on POAPF in B<sub>2</sub>, G<sub>2</sub> and R<sub>2</sub> while in G<sub>1</sub> and R<sub>1</sub> it mainly transport on dopants, so it's more difficult for the formation of the excitons in B<sub>2</sub>, G<sub>2</sub> and R<sub>2</sub> than B<sub>1</sub>, G<sub>1</sub> and R<sub>1</sub> because that the hole and electron transport on different molecules. As a result, the efficiencies of devices with EBBPC host are higher than those of devices with POAPF host.

The EQE characteristics of the devices based on different hosts and dopants are summarized in Fig.6 (b). As can be seen, CBP is unsuitable for blue PHOLED, while POAPF and 26DCzPPy are unsuitable for red PHOLED. Thus, as a host material for R, G, and B ML-PHOLEDs, EBBPC shows a relatively excellent performance compared to POAPF, CBP and 26DCzPPy. It should be attributed to that EBBPC possesses balanced bipolar transport characteristic, well-matched HOMO/LUMO levels with adjacent layers and high  $E_T$  at same time.

# 3.4. ML-PHOLEDs with EBBPC as ETL



Fig.7 (a) Current density-voltage-luminance curves of device G-a to G-d. (b) Efficiency-luminance characteristics of device G-a to G-d.

Inspired by the better electron-transporting properties of EBBPC, it could be an excellent electron transport material for OLEDs. Thus, the green OLEDs employing EBBPC (G-a) as ETL and the control devices based on Bphen (G-b), TmPyPB (G-c) or 1,3-Bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPhB) (G-d) are fabricated to convince the feasibility of EBBPC as ETL in OLEDs. The current density-voltage-luminance curves, current efficiency-power efficiency-luminance characteristics and EQE-current density curves of the devices are depicted in Fig. 7

and Fig. 8, respectively. EBBPC has a higher LUMO level (-2.36 eV) compared to the other ETLs (-2.8 eV for Bphen, -2.7 eV for TmPyPB, -2.6 eV for BmPyPhB). As a result, device G-a shows lower current density than devices G-b to G-d under the same voltage. However, the maximum luminance of device G-a can still reach 60480 cd/m<sup>2</sup>, which is 54630 cd/m<sup>2</sup>, 63080 cd/m<sup>2</sup> and 48930 cd/m<sup>2</sup> for device G-b to device G-d, respectively. Besides, the maximum current efficiency is 50.2, 43.6, 48.8, and 43.0 cd/A for device G-a to G-d, and the maximum power efficiency is 38.5, 33.4, 38.3, and 27.0 for device G-a to G-d, respectively. It means that device G-a shows comparable or even better performance to the other three devices. All above results confirmed that it is feasible to employ EBBPC as ETL in OLEDs.



Fig. 8. EQE-current density curves of device G-a to G-d (The inset is the EL spectra at 6V).

### 3.5. White ML-PHOLEDs with EBBPC as host and ETL

The excellent performances of monochrome devices employing EBBPC as host or ETL provide the possibility of its applications in high efficiency white OLEDs (WOLEDs). WOLED (marked as device W) with EBBPC as host and electron transport layer is fabricated. Efficient WOLED with complementary colors of blue and yellow is achieved by carefully adjusting the thickness of each emitting layers. Fig.9 (a) shows the normalized EL spectra of WOLED under different biases. The spectra almost has no change under the biases from 5 to 10 V, which means that device W possess excellent color stability. The CIE color coordinates just change from (0.341, 0.424) at 5 V to (0.335, 0.424) at 10 V, which should be attribute to a stable recombination zone in the emitting layer. As is known, the probability of 14

excitons formation is proportional to the density of electrons and holes. Thus, there is usually potential barrier between EML and HTL or ETL, which results that carriers accumulate at the interface of EML and HTL or ETL, and the excitons often formed at these interfaces, this contribute to a narrow recombination zone and it may move its position with the rise of voltage and lead to the change of spectra for multi-layer WOLEDs. Since we employing the same material-EBBPC as ETL and host, there should be no barrier at the interface of EML and ETL and the same situation occurs between EML and HTL as is depicted in Fig.5, which brings about little carriers accumulation and that most carriers are injected to EML, this lead to a wide recombination zone even cover the whole EML and generate a stable spectra, finally.



Fig. 9 (a) Normalized EL spectra of the white OLEDs (Inset: External quantum efficiency-current density curves). (b) Efficiency-luminance characteristics of WOLED (Inset is current density-voltage-luminance curves of the device).

Fig. 9 (b) depicts the efficiency-luminance and current density- voltage-luminance characteristics of device W. It can be seen that device W also possesses a low turn-on voltage of 3.4 V. The driving voltages are 4.3 and 5.5 V at brightness of 100 and 1000 cd/m<sup>2</sup>, and the maximum brightness of the device can reach 37800 cd/m<sup>2</sup> (at 11 V). The maximum efficiencies of the device can reach 40.2 cd/A and 36.1 lm/W. As is shown in Fig. 9 (b), the device also shows a relatively low efficiency roll-off. At a luminance of 1000 cd/m<sup>2</sup>, the current efficiency is 12.9% off its peak values, and the power efficiency also can reach 20 lm/W.

# 3.6. Single-layer PHOLED with EBBPC host

Considering the balanced hole and electron transport properties and suitable LUMO energy level, EBBPC is adopted to realize efficient SL-PHOLEDs as host materials,

and blue (marked as device SL-B), green (marked as device SL-G) and red (marked as device SL-R) SL-PHOLEDs is fabricated successfully, and device SL-G with a configuration of ITO/ITO (40 nm)/EBBPC:10wt%Ir(ppy)<sub>3</sub> (90 nm)/LiF(0.5 nm)/Mg:Ag(15:1, mass ratio, 120 nm) is demonstrated in detail.



Fig.10 (a) Current density-voltage-luminance curves. (b)Current efficiency-luminance-power efficiency characteristics of SL OLED (Inset: External quantum efficiency-current density curves).

The current density-voltage-luminance characteristics of device SL-G is shown in Fig.10 (a), it could be seen that the turn-on voltage of SL-G is ~3.4 V, which indicates an efficient injection of electrons and holes in the devices. At a low current density of 5 mA/cm<sup>2</sup>, the SL-G depicts a high brightness of 2580 cd/m<sup>2</sup>, and the peak brightness of the device can reach 28990 cd/m<sup>2</sup> (at 10.5 V). Such results demonstrate that the charges could transport well in the devices and the excitons could recombine efficiently. We attribute above results to the balanced bipolar transporting properties and well-matched HOMO/LUMO levels with electrodes, and also the high E<sub>T</sub> of EBBPC, which paves the way to achieve excellent energy transfer between EBBPC and guest material.

As shown in Fig.10 (b), the maximum EQE, current and power efficiencies of device SL-G can reach 14.6%, 52.3 cd/A and 35.4 lm/W, respectively. The current efficiency and EQE of device SL-G are compared with those of device G<sub>1</sub>. But, the power efficiency of Device SL-G is lower due to relatively high drive voltage. However, device SL-G shows a low efficiency roll-off. At a luminance of 1000 and 5000 cd/m<sup>2</sup>, the current efficiency is 0.5% and 4.9% off its peak values, respectively, and the power efficiency also can reach 23.3 lm/W at 5000 cd/m<sup>2</sup>. The performances

of the single-layer devices based on EBBPC host are shown in Table 3. Due to the low current density, the maximum luminances of SL-PHOLEDs are lower than those of ML-PHOLEDs. Although the maximum EQEs of blue and red SL-PHOLEDs are much lower than those of corresponding multilayer devices, the maximum EQEs of SL-B and SL-R also can reach 9.8% and 10.1%, respectively. We think that the lower EQEs of single-layer devices are due to the fact that the guests destroy the balanced carrier transporting property of EBBPC. However, the efficiency roll-off of SL-PHOLED is lower than that of ML-PHOLEDs. Table 4 lists the efficiency roll-off of ML- and SL-PHOLEDs. As can be seen, the efficiency roll-off of ML-PHOLEDs is more serious than that of corresponding SL-PHOLED. For example, the EQE roll-off of blue ML-PHOLED is 45.6% @5000 cd/m<sup>2</sup>, while that is only 19.4% for blue SL-PHOLED. We attributed it to the broader exciton formation zone in SL OLEDs.

Table 3. EL characteristics of the single-layer devices.								
Device	V <sub>on</sub> (V)	L <sub>max</sub> (cd/m <sup>2</sup> )	ηc.max, ηc.1000, ηc.5000 (cd/A)	η <sub>p.max</sub> , η <sub>p.1000</sub> , η <sub>p.5000</sub> (Im/W)	EQE <sub>max</sub> (%)	CIE(x,y) @8V		
SL-B	5.3	7130	21.6, 20.8, 17.6	10.4, 8.2, 5.4	9.8	(0.15,0.39)		
SL-G	3.4	28990	52.3, 52.0, 49.7	35.4, 30.8, 23.3	14.6	(0.30,0.64)		
SL-R	7.7	4920	16.6, 13.6, N.A.	5.5, 3.2, N.A.	10.1	(0.62,0.38)		

Table 4. Efficiency roll-off of ML- and SL-PHOLEDs.
CERO, PERO and EQERO are CE, PE and EQE roll-off

Device		FOF	CERO(%)	PERO(%)	EQERO(%)	
		EQEmax	@1000, 5000 cd/m <sup>2</sup>	@1000, 5000 cd/m <sup>2</sup>	@1000, 5000 cd/m <sup>2</sup>	
Plue	ML	16.0%	29.5, 45.8	53.0, 72.2	29.4, 45.6	
SL	SL	9.8%	3.7, 18.5	21.2, 48.0	4.1, 19.4	
Green ML SL	MĹ	15.6%	0.4, 7.2	20.9, 40.6	0.6, 7.7	
	SL	14.6%	0.6, 5.0	13.0, 34.2	0.7, 4.8	
Pad	ML	14.9%	20.4, 34.9	48.8, 66.4	24.2, 38.3	
Reu	SL	10.1%	18.1, N.A.	41.8, N.A.	18.8, N.A.	

# 4. Conclusions

In summary, efficient multilayer and single-layer phosphorescent OLEDs have

been demonstrated based on bipolar EBBPC host. For example, the maximum external quantum efficiency of the green ML- and SL-PHOLEDs with EBBPC host can reach 15.6% and 14.6%, respectively. Besides, the device with ETL of EBBPC shows comparable efficiency with the devices with ETL of Bphen, TmPyPB, or BmPyPhB. Furthermore, efficient WOLED applying EBBPC as host and ETL is also realized. The WOLED shows maximum external quantum efficiency of 15.4% and good color stability. The excellent performance of the devices should be attribute to that EBBPC possesses balanced bipolar transport characteristic, well-matched HOMO/LUMO levels with adjacent layers and high  $E_T$  at same time.

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► ML- and SL-PHOLEDs with a bipolar host EBBPC are fabricated. The maximum EQE are 14.9%, 15.6%, 16.0% and 15.4% for R, G, B, and white ML-PHOLEDs. The maximum EQE are 10.1%, 14.6% and 9.8% for R, G, and B SL-PHOLEDs. It could be attributed to the balanced bipolar transporting properties and appropriate energy level of EBBPC.