



Carbazole-diphenylimidazole based bipolar material and its application in blue, green and red single layer OLEDs by solution processing



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ABSTRACT

Single layer structure is an ideal way to realize the low cost solution processed organic light-emitting diodes (OLEDs). Efficient electroluminescent material with hole and electron transport characters as well as suitable triplet energy levels is highly desired to realize single layer OLEDs with good performance. In this work, a bipolar compound with carbazole as electron donor and 4,5-diphenylimidazole as electron acceptor has been successfully synthesized and applied as emitting layer for both non-doped and doped single layer OLEDs fabricated by solution processing. A pure blue emission achieved in the non-doped single layer OLED with a current efficiency of 0.38 cd A^{-1} , luminance up to 672 cd m^{-2} . The single layer doped OLEDs with the bipolar compound as host achieved blue, green and red phosphorescence OLEDs with current efficiency of 0.083, 13.42, 2.58 cd A^{-1} and luminance up to 108, 17103, 1347 cd m^{-2} , respectively. The performance of single layer OLEDs has been greatly improved by bipolar emitting materials which show a great potential to simplify the configuration of OLEDs.

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1. Introduction

The development of organic light-emitting diodes (OLEDs) have been divided into two ways according fabrication process since very beginning of last century [1–3]. So far, the vapor deposition method still dominates for allowing the complicated multiple layers fabrication which always give an excellent device performance [4–6]. However, the finite space of vapor deposition chamber limited the development of large-sized OLEDs, not to mention the low utility of materials. The solution process is another way for fabricating OLEDs mainly used in polymer materials, with which the fabrication of large-sized OLEDs is realizable [7–9]. While, the multilayer structure is still a huge challenge.

The single layer structured OLEDs is ideal for low cost and simple fabrication processes, especially for solution processing such as inkjet printing or roll-to-roll, which can be used in large scale

fabrication [10–16]. In the earlier reports, the performances of single layer OLEDs were too poor [17–19]. The extra quantum efficiency (EQE) was lower than 1.5% even with the phosphorescent emitters.

Recently, the bipolar emitting materials with electron donor-acceptor (D-A) or D- π -A structures have been reported [20–24]. These kinds of materials characterized with triphenylamine or carbazole moieties as donor and the phenanthroimidazole or triazine as acceptor [25–30]. The donor part (D) services as the hole-transport unit and acceptor part (A) as the electron-transport unit [31]. The photochemical properties and energy levels of bipolar emission materials can be adjusted by changing the conjugation degree of D and A, as well as the structure of D and A moieties [32,33].

Bipolar emitting materials greatly improved the performance of single layer OLEDs. As reported by Tai-Hsiang Huang et al., the non-doped green single layer OLED achieved a current efficiency (CE) of 7.5 cd A^{-1} [34]. The bipolar emitting material reported in that work has S,S-dioxide moieties as the electron acceptor and diarylamine as the electron donor.

The performance of single layer OLED was further improved by

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using phosphorescent emitter tris[2-phenylpyridinato- $C^{2,N}$]iridium(III) ($Ir(ppy)_3$) as the guest [35]. The maximum luminance (L_{max}) of ~ 16400 $cd\ m^{-2}$ and power efficiency of $4.4\ lm\ W^{-1}$ achieved with bipolar host material Me-TBBI (tris(2-methyl-3'-(1-phenyl-1*H*-benzimidazol-2-yl)biphenyl-4-yl)amine). Observably, the bipolar emitting material is a promising way to improve the performance of the single layer OLEDs. While, up to now the blue and red single layer OLEDs are rarely reported.

It is known that the intermolecular charge transfer of bipolar materials always decrease the energy gap (E_g) [36]. The reported bipolar host for phosphorescence OLEDs (PhOLEDs) mainly use the carbazole substituent as the donor moiety which derivatives has sufficient high triplet energy level (E_T) to ensure the energy transfer to the dopant [37–42]. The phenanthroimidazole (PPI) derivatives showed good thermal stability, relatively high E_T and high electron mobility which is always applied as acceptor in bipolar emitting materials [25,43–45]. Previously, our lab reported a bipolar blue emitter with acceptor moiety of 4,5-diphenylimidazole (DPI) and donor moiety of carbazole, which showed reducing of π -configuration and increase of E_g Ref. [25]. In this work, with considering of the solution processing fabrication, a tertiary butyl moiety was introduced to improve the solubility and film forming ability [46]. The molecular structure of the bipolar emitting material 9-(4'-(1-(4-(*tert*-butyl)phenyl)-4,5-diphenyl-1*H*-imidazol-2-yl)-[1,1'-biphenyl]-4-yl)-9*H*-carbazole (Cz-BP-DPI) is shown in Scheme 1. The blue, green and red PhOLEDs fabricated with Cz-BP-DPI as host and with Iridium(III) bis[(4,6-difluorophenyl)-pyridinato- $C^{2,N}$]picolinate (Flpic), bis(2-phenylpyridine)iridium(III)-acetylacetonate ($Ir(ppy)_2(acac)$) and platinum octaethylporphyrin (PtOEP) as phosphorescent guests, respectively. This phosphorescence OLEDs (PhOLEDs) achieved L_{max} of 108, 17103, 1347 $cd\ m^{-2}$ and the maximum CE (CE_{max}) of 0.083, 13.42 and 2.58 $cd\ A^{-1}$, respectively. The L_{max} and CE_{max} of the green single layer PhOLEDs is 1.04 and 1.79 times larger than reported one [35]. In addition, the single layer non-doped OLED with Cz-BP-DPI emitting layer showed a pure blue light with CE_{max} of 0.38 $cd\ A^{-1}$ and L_{max} of 673 $cd\ m^{-2}$.

2. Experimental

2.1. Materials

All materials were used as received from commercial supplier

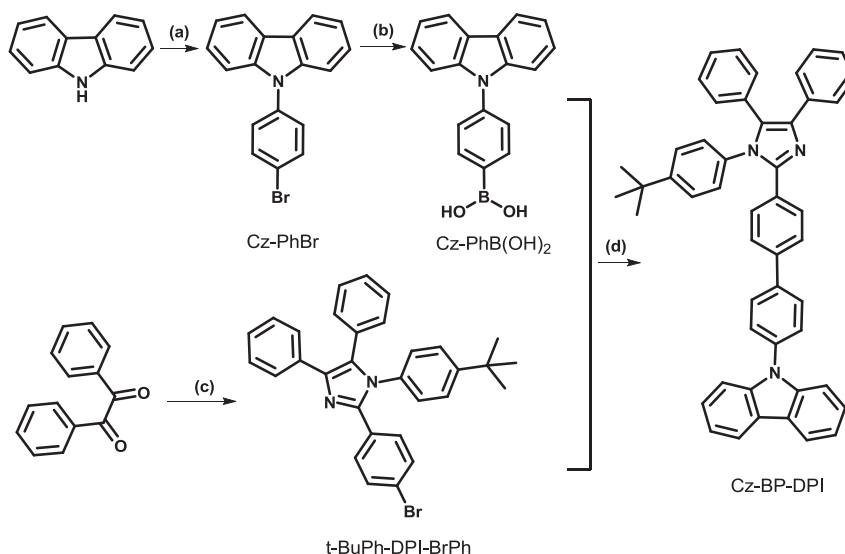
(Tianjin Heowns Biochemical Technology Co., Ltd.) without further purification. All solvents were purchased from Tianjin Guangfu Fine Chemical Research Institute. THF was freshly distilled from sodium/benzophenone under argon (Ar) atmosphere before use.

2.2. General procedures

Ultraviolet–visible absorption (UV–Vis) and photoluminescence (PL) spectra were obtained on a Thermo Evolution 300 UV–Visible spectrometer and a HitachiF-4500 fluorescence spectrometer, respectively. The phosphorescence spectrum was measured in dilute THF solution at 77 K using a Horiba Fluorolog3 fluorescence spectrometer. Differential scanning calorimetry (DSC) was recorded on a TA Q20 instrument operated with heating rate of $10\ ^\circ C\ min^{-1}$ from $25\ ^\circ C$ to $350\ ^\circ C$ in nitrogen atmosphere (N_2). The T_g was determined from the second heating scan loop. Thermal gravity analysis (TGA) was recorded on a METTLER TOLEDO TGA/DSC1 Thermogravimetric Analyzer heating rate of $10\ ^\circ C\ min^{-1}$ from $30\ ^\circ C$ to $600\ ^\circ C$ in a N_2 . X-ray diffraction curves were recorded on a RigakuMiniflex 600 with 2θ range of 3° – 20° , scanning at a rate of $1^\circ\ min^{-1}$. 1H NMR and ^{13}C NMR were recorded with a Bruker ACF400 (400 MHz) spectrometer in chloroform- d ($CDCl_3$) or dimethyl sulfoxide- d_6 ($DMSO-d_6$) with tetramethylsilane as reference. High resolution mass was performed with a Bruker MicroTOF-QII. The element analysis was performed with a VarioMICRO CHNOS elemental analyzer.

2.3. Device fabrication and measurement

Indium tin oxide (ITO) glass substrates with sheet resistance of $10\ \Omega$ per square were cleaned with acetone, methanol and diluted water, then dried in an oven. After O_2 plasma treatment for 5 min, an anode modified layer (~ 40 nm) was achieved by spin-coating the PEDOT:PSS colloidal solution on the ITO side at 3000 rpm, followed with baking at $120\ ^\circ C$ for 30 min. The $10\ mg\ mL^{-1}$ concentrated Cz-BP-DPI THF solutions was spin-coated on the PEDOT:PSS film at 1500 rpm. A 40 nm thick emitter layer was achieved after baking at $80\ ^\circ C$ for 10 min. Then above substrate was immediately transferred into the deposition chamber with a vacuum degree of 2×10^{-6} torr. Then 1.5 nm cesium fluoride (CsF) and 100 nm aluminium (Al) were deposited successively at a rate of $\sim 0.1\ \text{Å}\ s^{-1}$ and $\sim 4\ \text{Å}\ s^{-1}$, respectively. The thicknesses of the spin-coated films



Scheme 1. Synthesis of Cz-BP-DPI. Conditions (a) 1-bromo-4-iodobenzene, Copper powder, K_2CO_3 , 18-Crown-16, DMF, reflux 16 h, (b) triisopropyl borate, $n-BuLi$, THF, $-78\ ^\circ C$, Ar, (c) 4-(*tert*-butyl)aniline, 4-bromobenzaldehyde, CH_3COONH_4 , CH_3COOH , reflux 12 h, (d) $Pd(PPh_3)_4$, K_2CO_3 , Toluene, Reflux.

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