



A novel, bipolar host based on triazine for efficient solution-processed single-layer green phosphorescent organic light-emitting diodes



Bin Huang^{a,b}, Wei Jiang^{a,*}, Jinan Tang^a, Xinxin Ban^a, Ruigang Zhu^b, Huang Xu^a, Wen Yang^a, Yueming Sun^{a,*}

^aSchool of Chemistry and Chemical Engineering, Southeast University, Nanjing 211189, Jiangsu, PR China

^bDepartment of Chemical and Pharmaceutical Engineering of Southeast University Chengxian College, Nanjing 210088, Jiangsu, PR China

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ABSTRACT

A novel N-phenyl carbazole substituted 2, 4,6-trisphenyl-triazine host material (TPCPZ) for solution processed green phosphorescent organic light-emitting devices (PhOLEDs) was synthesized by a Suzuki-cross coupling reaction. The optical, electrochemical and thermal properties of TPCPZ have been characterized. TPCPZ exhibits a high glass transition temperature of 165 °C and a triplet energy of 2.63 eV. The appropriate HOMO energy level (−5.39 eV) and LUMO energy level (−2.16 eV) matching with the HOMO energy level of PEDOT:PSS (−5.35 eV) and the LUMO energy level of Cs₂CO₃/Al bilayer cathode (−2.2 eV), facilitate the transfer of holes and electrons. The solution-processed single-layer device using TPCPZ as the host for fac-tris(2-(4-phenylpyridine)iridium (Ir(ppy)₃) exhibited a low turn-on voltage of 3.5 V, a maximum current efficiency of 20.8 cd A^{−1} and a maximum luminance of 18,000 cd m^{−2}. These results demonstrated that TPCPZ as a host material is advantageous for fabrication of highly efficient single-layer green PhOLEDs.

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1. Introduction

Since efficient phosphorescent emitters for organic light-emitting diodes (PhOLEDs) was discovered by Forrest's group, tremendous efforts have been made in the development of highly efficient devices [1–5]. In PhOLEDs, the emitting layer consists of a phosphorescent dye which is doped into a host material to avoid concentration quenching and to optimize the charge balance. High efficiencies are obtained only when the energy is efficiently transferred from the host to the phosphorescent dye. To achieve efficient electrophosphorescence, the choice of host is of vital importance [6]. Generally, the triplet energy of the host material is higher than that of the guest. For efficient OLEDs, well balanced charge carrier transport and a broad recombined zone are desirable. In general, the electron mobility of many host materials is much lower than the hole mobility because of the fact that they consist electron donors such as aromatic amines, carbazoles. This task is mostly solved by a design of a bipolar material [7,8]. Introduction of strong electron donors such as aromatic amines or carbazoles [9] to electron accepting N-heterocycles like pyridine [10–

12], oxadiazole [13–16], phenanthroline [17–19], benzimidazole [20–24], is well described. Triazines, are known to be good electron conductors and their derivatives have been used as electron transport layers in OLEDs [4,25–28]. Recently, some bipolar host materials based on triazines for PhOLEDs are reported [29–34]. A series of donor substituted 1,3,5-triazine derivatives as host for blue or green PhOLEDs show good performance.

In recent years, many groups are devoted to improve the performance of solution-processed PhOLEDs which are highly desirable to simplify the fabrication process and reduce the cost of larger area displays [35–44]. Even though polymer light-emitting diodes can be easily fabricated by solution casting, they are generally difficult to synthesize and purify for electronic devices. Therefore, it is a good strategy to develop OLEDs based on solution-processed small molecules. However, the largest challenge for small molecules lies in the poor solubility accompanied by the tendency to crystallize and failure to form high quality films. Consequently, it is of significant importance to design and synthesize amorphous small molecules with high purity and solubility. To date, few works reported on bipolar host based on 1, 3,5-triazine for solution-processed PhOLEDs.

In this work, a bipolar host composed of electron-transporting 2, 4,6-trisphenyl-triazine and hole-transporting N-phenyl carbazole,

* Corresponding authors. Tel./fax: +86 25 52090621.

E-mail address: 101011462@seu.edu.cn (W. Jiang).

namely 2, 4,6-tris (3-((9-phenyl) carbazol-3-yl)-phenyl) -triazine(TPCPZ) is designed and synthesized. The dipolar nature of TPCPZ can promote the strong π - π intermolecular stacking of molecules in the solid state, which can facilitate high charge transport. Moreover, the electron-donating N-phenyl carbazole moieties are conjugated to the electron accepting 2, 4,6-trisphenyl-triazine core; the donor-acceptor intramolecular interaction tends to decrease the energy gap. TPCPZ has a triplet energy of 2.62 eV, which make it suitable as host material for green phosphorescent emitters. The material exhibits high thermal stability and good sublimation properties. The glass transition temperature of 165 °C ensures morphological stability of the host-guest emission layer during the operation of the device. Additionally, TPCPZ has good solubility in common solvents such as CH_2Cl_2 , CHCl_3 , 1,2-dichloroethane et al., which make it suitable for the fabrication of solution-processed PhOLEDs. The single-layer device with TPCPZ as host has low turn-on voltage of 3.5 V, a high maximum luminance efficiency of 20.8 cd A^{-1} and maximum luminance of 18,000 cd m^{-2} in a solution-processed green phosphorescent OLED using an emitter of fac-tris(2-(4-phenylpyridine)iridium ($\text{Ir}(\text{ppy})_3$)).

2. Experimental

2.1. General

All reactants and solvents were purchased from commercial sources and used without further purification. ^1H NMR and ^{13}C NMR spectra were recorded on a Bruker ARX300 NMR spectrometer with $\text{Si}(\text{CH}_3)_4$ as the internal standard. Elemental analysis was performed on an Elementar Vario EL CHN elemental analyzer. Mass spectra were obtained using a Thermo Electron Corporation Finnigan LTQ mass spectrometer. UV-vis absorption spectra were recorded with a spectrophotometer (Agilent 8453) and PL spectra were recorded with a fluorospectrophotometer (Jobin Yvon, FluoroMax-3). Thermogravimetric analysis (TGA) was performed using a Netzsch simultaneous thermal analyzer (STA) system (STA 409PC) under dry nitrogen atmosphere at a heating rate of 10 °C min^{-1} . Glass transition temperature was recorded by differential scanning calorimetry (DSC) at a heating rate of 10 °C min^{-1} with a thermal analysis instrument (DSC 2910 modulated calorimeter). Cyclic voltammetry measurements were performed on a Princeton Applied Research potentiostat/galvanostat model 283 voltammetric analyzer in CH_2Cl_2 solutions (10^{-3} M) at a scan rate of 100 mV s^{-1} with a platinum plate as the working electrode, a silver wire as the pseudo-reference electrode, and a platinum wire as the counter electrode. The supporting electrolyte was tetrabutylammonium hexafluorophosphate (0.1 M) and ferrocene was

selected as the internal standard. The solutions were bubbled with a constant argon flow for 10 min before measurements.

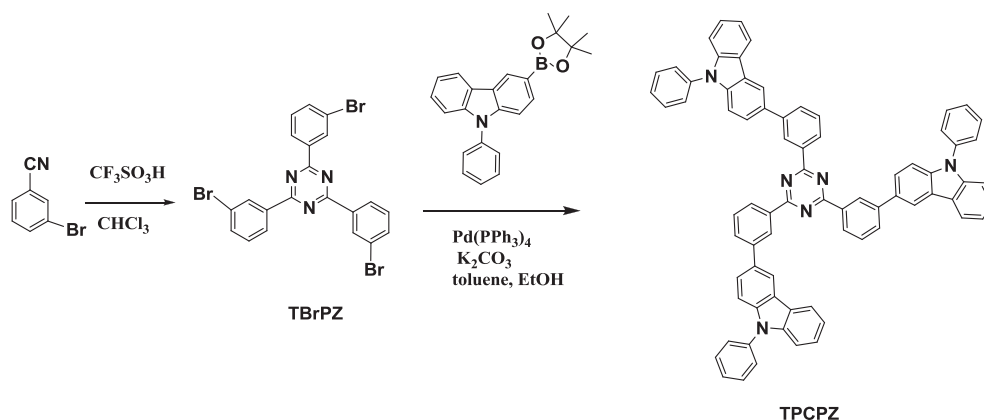
DFT calculations of TPCPZ were performed using the Gaussian 03 program package. The calculation was optimized at the B3LYP/6-31G(d) level of theory. The molecular orbitals were visualized using Gaussview [45].

2.2. Synthesis of 2,4,6-tris(3-((9-phenyl)carbazol-3-yl)-phenyl)-triazine(TPCPZ)

To a solution of 2,4,6-tris(3-bromophenyl)-triazine (TBrPZ) (0.546 g, 1.0 mmol) and N-phenyl-3-(4,4,5,5-tetramethyl-1,3,2-dioxaborolane-2-yl)-6H-carbazole (1.107 g, 3.0 mmol) in 20 mL of toluene and 4 mL of ethanol was added 2 mL of 2.0 M aqueous K_2CO_3 solution. The reaction mixture was then purged with nitrogen for 10 min before adding tetrakis(triphenylphosphine) palladium(0) (0.055 g, 0.048 mmol). After refluxing for 24 h under nitrogen, the resulting mixture was cooled to room temperature and then poured into water and extracted with 60 mL (3×20 mL) CH_2Cl_2 . The combined organic phase was then washed with 20 mL (2×10 mL) saturated aqueous NaCl solution and dried with anhydrous Na_2SO_4 . After removal of the solvent by rotary evaporation, the residue was purified by silica gel column chromatography to afford TPCPZ as a white solid. Yield: 0.60 g (58.08%). ^1H NMR (300 MHz, CDCl_3): δ (ppm) 9.17 (s, 3H), 8.81–8.78 (d, 3H), 8.50 (s, 3H), 8.16–8.19 (d, 3H), 7.96–7.98(d, 3H), 7.80–7.83(m, 3H), 7.67–7.73 (t, 3H), 7.65–7.56 (m, 12H), 7.52–7.43 (m, 6H), 7.40–7.35 (m, 6H), 7.16–7.20 (m, 3H). ^{13}C NMR (75 MHz, CDCl_3) δ (ppm): 109.50, 109.78, 118.67, 119.71, 120.03, 123.06, 123.62, 125.18, 125.80, 126.71, 126.97, 127.14, 127.39, 128.78, 129.54, 131.16, 132.68, 136.51, 137.29, 140.18, 141.02, 142.18, 171.57. MS (MALDI-TOF) [m/z]: calcd for $\text{C}_{75}\text{H}_{48}\text{N}_6$, 1033.22; found, 1033.48. Anal. Calcd. for $\text{C}_{75}\text{H}_{48}\text{N}_6$ (%): C, 87.18; H, 4.68; N 8.13. Found: C, 87.34; H, 4.72; N 8.38.

2.3. Device fabrication and performance measurements

The solution-processed single-layer device using TPCPZ as host with a configuration ITO/PEDOT:PSS (40 nm)/TPCPZ: $\text{Ir}(\text{ppy})_3$ (90 wt%:10 wt%,100 nm)/ Cs_2CO_3 (2 nm)/Al (120 nm) has been fabricated by spin-coating. In a general procedure, indium-tin oxide (ITO)-coated glass substrates were pre-cleaned carefully and treated by UV ozone for 4 min. A 40 nm Poly(3,4-ethylenedioxythiophene) doped with Poly(styrene-4-sulfonate)(PEDOT:PSS) aqueous solution was spin coated onto the ITO substrate and baked at 210 °C for 10 min. The substrates were then taken into a nitrogen glove box, where $\text{Ir}(\text{ppy})_3$ -doped TPCPZ layer was spin coated onto the



Scheme 1. Synthetic route of TPCPZ.

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