



# A host material consisting of phosphinic amide for efficient sky-blue phosphorescent organic light-emitting diodes



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## ABSTRACT

A new bipolar host material (PBCz-PO) consisting a phosphinic amide is developed to show a resonance effect. It possesses quite high triplet energy (2.76 eV), and this high triplet energy make it suitable to be blue host in phosphorescent organic light-emitting diodes (PHOLEDs). The device with PBCz-PO as a host and Flrpic as a dopant achieved a maximum current efficiency ( $\eta_c$ ) of 31.5 cd/A, a maximum power efficiency ( $\eta_p$ ) of 31.0 lm/W and a maximum external quantum efficiency (EQE) of 13.4% as well as low driving voltage.

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## 1. Introduction

Phosphorescent organic light-emitting diodes (PHOLEDs) are considered as the most promising candidates for the next-generation display and lighting technology due to the ideal characteristics of electrophosphorescence, such as the 100% internal quantum efficiency which result from the highly efficient utilization of triplet excitons [1–8]. Although tremendous efforts have been paid to increase the luminous efficiency of PHOLEDs, the development of a stable and efficient blue PHOLED remains a great challenge. One of the most important problem is concentration quenching and triplet–triplet annihilation, which result in significant efficiency roll-offs in PHOLEDs [9–23].

A basic strategy widely used to restrain these effects is to dope the phosphorescent units into a suitable host materials to reduce their intermolecular interaction. It is desirable that the host materials ought to have a large enough triplet energy gap ( $T_1$ ) for efficient energy transfer to the guest, good carrier transport properties for a balanced recombination of holes and electrons in the emitting layer, and energy-level matching with neighboring layers for effective charge injection [24–32]. These inevitable demands of host materials provide organic chemists with

tremendous opportunities to contribute their expertise to this technology.

The design of bipolar host materials configured with electron-donating groups as donors (D) and electron-withdrawing groups as acceptors (A) seems to be the most appealing strategy because they can provide more balance in electron and hole transport [33–45]. However, these strong D–A interaction will suppress the exciton recombination due to the not high enough  $T_1$  [46–48]. So, Yuan et al. adopted the twisted  $\pi$ -conjugated bridge to partially reduce the electronic coupling of the donor and acceptor [49]. However, it is very difficult for this strategy to achieve hosts both with relative high  $T_1$  values and with strong electrical activities.

Recently, a special insulating linkage based on diphenylphosphine oxide (DPPO) has been utilized and realized excellent device performance, such as ultralow driving voltages (less than 3 V for onset) and high and stable efficiencies [50]. The P=O moieties not only serve as insulating linkages but also modify the electrical properties of the molecules as a kind of electron-withdrawing group. In addition, a special effect of N–P=O on charge redistribution by resonance variation was also observed [51]. Our group recently reported a series of high-performance PO hosts with high  $T_1$  values and relatively balanced carrier injection/transporting capabilities. The device based on Flr6 as dopant can achieve power efficiency of 23.7 lm W<sup>-1</sup>, EQE of 13.6% [17].

Here, in this work, we report another efficient bipolar host for sky-blue electrophosphorescence, based on the structure of the

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3,3'-bicarbazole (BCz) [52–55] as the donor and phosphine oxide as the acceptor named diphenyl(9'-phenyl-[3,3'-bicarbazol]-9-yl) phosphine oxide (PBCz-PO). The greater contribution of enantiotropic  $N^+=P-O^-$  resonances to PBCz-PO results in a much enhanced electron transportation from the polarized carbazolyl. As a result, PBCz-PO endowed its sky-blue PHOLED with impressively low driving voltages: 3.3 V for 100 cd/m<sup>2</sup> and under 4.1 V for 1000 cd/m<sup>2</sup>, as well as favorable electroluminescence (EL) efficiencies, such as the maximum external quantum efficiency (EQE) of up to 13.4%, the current efficiency ( $\eta_c$ ) of 31.5 cd/A, and the power efficiency ( $\eta_p$ , max) of 31.0 lm/W.

## 2. Experimental

### 2.1. Materials and methods

All of the chemicals, i.e., (9-phenyl-9H-carbazol-3-yl) boronic acid, 3-bromo-9H-carbazole, chlorodiphenylphosphine and *n*-butyl lithium were purchased from Bepharma Limited and Alfa Aesar. All of these materials were used without further purification. THF was purified using the PURE SOLV (Innovative Technology) purification system. Chromatographic separations were carried out using silica gel (200–300 nm). All the other reagents were used as received from commercial sources unless otherwise stated. <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded on a Varian Unity Inova 400 spectrometer at room temperature. Mass spectra were recorded on a Thermo ISQ mass spectrometer using a direct exposure probe. UV–vis absorption spectra were recorded on a PerkinElmer Lambda 750 spectrophotometer. PL spectra and phosphorescent spectra were recorded on a Hitachi F-4600 fluorescence spectrophotometer. Differential scanning calorimetry (DSC) was performed on a TA DSC 2010 unit at a heating rate of 10 °C/min under nitrogen. The glass transition temperatures ( $T_g$ ) was determined from the second heating scan. Thermogravimetric analysis (TGA) was performed on a TA SDT 2960 instrument at a heating rate of 10 °C/min under nitrogen. The temperature at 5% weight loss was used as the decomposition temperature ( $T_d$ ). Cyclic voltammetry (CV) was carried out on a CHI600 voltammetric analyzer at room temperature with a conventional three-electrode configuration consisting of a platinum disk working electrode, a platinum wire auxiliary electrode, and an Ag wire pseudo-

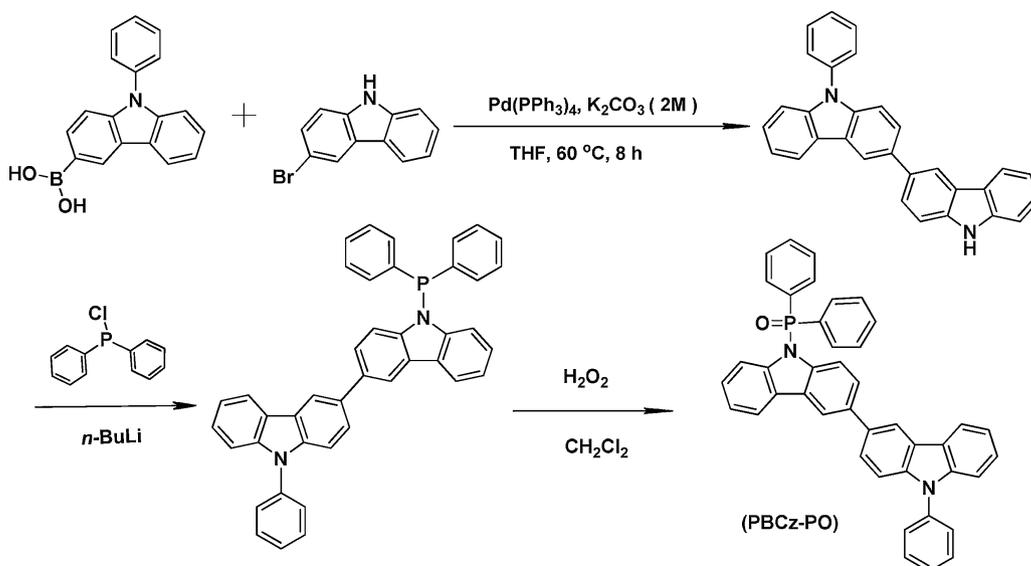
reference electrode with ferrocenium–ferrocene (Fc<sup>+</sup>/Fc) as the internal standard. Nitrogen-purged DCM was used as the solvent for the oxidation scan and DMF was used for the reduction scan, with tetrabutylammonium hexafluorophosphate (TBAPF<sub>6</sub>) (0.1 M) as the supporting electrolyte. The cyclic voltammetry (CV) was obtained at a scan rate of 100 mV/s. The ultraviolet photoemission spectroscopy (UPS) characterization was performed in a Kratos AXIS Ultra<sub>DL</sub> ultrahigh vacuum (UHV) analysis system.

### 2.2. Fabrication of the OLED

The OLED was fabricated through vacuum deposition of the materials at ca.  $2 \times 10^{-6}$  Torr onto ITO-coated glass substrates having a sheet resistance of ca. 30 Ω per square. The ITO surface was cleaned ultrasonically and sequentially with acetone, ethanol, and deionized water, in an ultrasonically bath, dried in an oven, and finally exposed to UV-ozone for about 30 min. Organic layers were deposited at a rate of 2–3 Å/s, and subsequently the HAT-CN and Liq were deposited at 0.2 Å/s and then capped with Al (ca. 4 Å/s) through a shadow mask without breaking the vacuum. For the device, the emitting area was determined by the overlap of two electrodes as 0.09 cm<sup>2</sup>. The EL spectra, CIE coordinates and J–V–L curves of the device were measured with a PHOTO RESEARCH SpectraScan PR 655 photometer and a KEITHLEY 2400 SourceMeter constant current source at room temperature. The EQE values were calculated according to the previously reported methods.

### 2.3. Synthesis of 9-phenyl-9H, 9'H-3, 3'-bicarbazole (PBCz)

3-bromo-9H-carbazole (2.56 g, 10.4 mmol), (9-phenyl-9H-carbazol-3-yl) boronic acid (3.28 g, 11.4 mmol) and tetrakis(triphenylphosphine) palladium (0) (0.6 g, 0.52 mmol) were dissolved in THF/2 M K<sub>2</sub>CO<sub>3</sub> (3:1, v/v). The reaction mixture was heated to 60 °C for 8 h under an argon atmosphere. After cooling to room temperature, the organic layer was separated and evaporated to remove the solvent. The residue was purified by column chromatography with 1:3 (v/v) dichloromethane–petroleum ether as the eluent and recrystallized from dichloromethane–petroleum ether to give a white crystalline powder (3.02 g, 71%). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.40 (s, 1H), 8.25 (d, *J* = 7.7 Hz, 1H), 8.19 (d, *J* = 7.8 Hz, 1H),



**Scheme 1.** Molecular structure and synthesis of PBCz-PO.

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