



Diphenylphosphine oxide-based host materials for green phosphorescent organic light-emitting devices



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ABSTRACT

Two novel host materials based on the diphenylphosphine oxide (DPPO) and spirofluorene were designed and facily synthesized by tuning the ratio of donor spirofluorene units and acceptor DPPO units from 1: 1 to 1: 2, i. e., (4-(9,9'-spiro[fluoren]-2-yl)phenyl)diphenylphosphine oxide (SPDPPO) and (9,9'-spiro[fluorene]-2,7-diylbis(4,1-phenylene))bis(diphenylphosphine oxide) (SBPBDPPO). Furthermore, 3,6-disubstituted spirofluorene possesses better thermal stability performance, for instance, higher glass transition temperatures (SPDPPO of 140 °C and SBPBDPPO of 172 °C). The phosphorescent organic light-emitting devices (PhOLEDs) were fabricated by doping tris(2-phenylpyridine)iridium (Ir(ppy)₃) in hosts as light-emitting layer. The PhOLEDs employed SPDPPO as host exhibited a low turn-on voltage of 3.0 V and the maximum current efficiency of 36.5 cd/A, power efficiency of 32.8 lm/W and a maximum external quantum efficiency (EQE) of 10.5%, respectively.

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1. Introduction

The phosphorescent organic light-emitting devices (PhOLEDs) have been applied in full color displays and solid-state lighting due to their superior characteristics such as good flexibility, light weight, wide-viewing angles and self-emission, especially the unique features of theoretically 100% internal quantum efficiency (IQE) by harvesting both singlet and triplet excitons [1–6]. The phosphorescent emitters are usually doped into an appropriate host to avoid concentration quenching and triplet–triplet annihilation (TTA). Thus, it is necessary to design appropriate phosphorescent host material [7,8].

Donor–acceptor (D–A) type molecules have been gradually constructed to be one of the most effective strategy to enhance

charge injection and carrier transport in organic semiconductor materials [9]. Especially, in some D–A material-based OLEDs, triplet excitons have been demonstrated to be almost fully employed, and even more excellent performances were obtained in such OLEDs, so as to tremendously improve the electroluminescence (EL) efficiencies of OLEDs [10].

Recently, the diphenylphosphine oxide (DPPO) functional group has attracted great attention as a strong electron withdrawing group [11]. Its structure allows it to affect only charge transport properties without degrading the triplet energy level (T₁) [12]. Its unique structure is also beneficial to the forming stability amorphous morphology because it is difficult to stack molecules [13]. Therefore, many DPPO-based host materials have demonstrated good performances in PhOLED, such as low driving voltage, high efficiency and low roll-off in efficiency [14].

Spirofluorene will continue to find extensive use in a myriad of material context for PhOLEDs due to their good hole-transporting ability and high T₁ [15]. Molecules based spirofluorene utilize a spiro bridge to connect two conjugated moieties via a central

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tetrahedral bonding atom. This structure minimizes the probability of interchain interactions, prevents the close packing of molecules, and increases the rigidity and glass transition temperatures (T_g) [16]. Lee and co-workers reported two hosts SPPO1 and SPPO11 linked spirobifluorene and phosphine-oxide hybrid molecules as hosts, which acquired a satisfactory electroluminescent performance [17–19]. This can be attributed to the steric hindrance which leads to a twisted geometry in these molecules and makes extension of conjugation between the spirobifluorene and the substituent moiety unlikely [20].

Above all, it is anticipated that the combination of high T_1 and high thermal stability in one molecule is feasible by adopting DPPO and spirofluorene. Herein, we rationally designed and synthesized two phosphorescent host materials, (4-(9,9'-spirobifluorene)-2-yl)phenyl)diphenylphosphine oxide (SPDPPO) and (9,9'-spirobifluorene)-2,7-diylbis(4,1-phenylene))bis(diphenylphosphine oxide) (SBPBDPPO), by using DPPO as acceptor and spirofluorene as the donor with higher T_g by the Suzuki reaction. Their photophysical properties, electrochemical properties and theoretical calculations, were carefully investigated. The phosphorescent devices based on them have been prepared. The synthetic scheme of them is represented in Scheme 1.

2. Experimental section

2.1. General information

All of the chemicals and reagents are used in their original form without further purification. Solvents for chemical synthesis are purified according to the standard procedures. All chemical reactions are performed in a nitrogen atmosphere.

^1H NMR and ^{13}C NMR data were recorded with Switzerland Bruker DR \times 600 NMR spectrometer, and chemical shifts were reported in ppm using tetramethylsilane (TMS) as an internal standard. Elemental analyses of carbon, hydrogen, and nitrogen were performed on a Vario EL elemental analyzer. Thermogravimetric analysis (TGA) was undertaken using a Netzsch TG 209 and the thermal stability of sample was determined by measuring the weight loss while heating at a rate of $10^\circ\text{C}/\text{min}$ with a gas flow rate of $30\text{ mL}/\text{m}^3$ under a nitrogen atmosphere. Differential scanning calorimetry (DSC) was performed on a Netzsch DSC Q2000 unit at a heating rate of $10^\circ\text{C}/\text{min}$ from room temperature to 350°C under a flow of nitrogen and cooling at rate of $20^\circ\text{C}/\text{min}$; T_g were determined from the second heating scan. UV–vis absorption spectra were recorded using a Hitachi U-3900 spectrophotometer and the

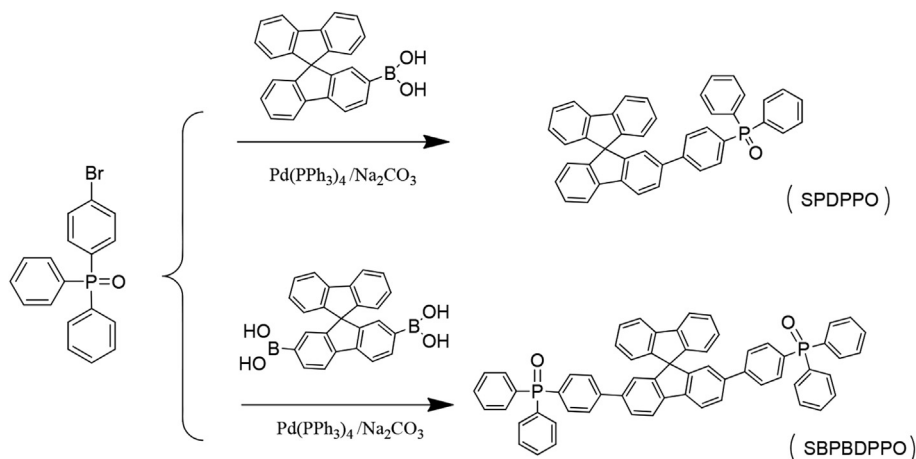
photoluminescence (PL) spectra were examined by HORIBA Fluoromax-4 spectrophotometer in diluted dichloromethane (CH_2Cl_2) solution at room temperature. The low-temperature phosphorescence spectra were measured on an Edinburgh F-980 spectrometer at 77 K in 2-methyltetrahydrofuran (2-MeTHF).

Cyclic voltammetry was performed using a CHI 660E voltammetry analyzer in a one-compartment electrolysis cell consisting of a platinum wire as working electrode, a platinum electrode as counter, and a calomel electrode as reference. Tetrabutylammonium perchlorate was used as a supporting electrolyte (0.1 M), and cyclic voltammetric were monitored at a scan rate of $50\text{ mV}/\text{s}$. The highest occupied molecular orbital (HOMO) levels were calculated according to the equation $E_{\text{HOMO}} = -4.8 - e(E_{\text{ox}}^{\text{1st}})/V$, where E_{ox} was the first oxidation peaks measured from CV curves. On the other hand, the lowest unoccupied molecular orbital (LUMO) levels were calculated based on the equation $\text{LUMO} = \text{HOMO} + E_g$, and E_g is from the absorption and emission spectra. All solutions were purged with a nitrogen stream for 3 min before measurement.

Theoretical calculation of SPDPPO and SBPBDPPO were carried out by using the Gaussian 03 package. The ground state structures were optimized by density functional theory (DFT) in B3LYP/6-31G(d) basis sets [21–23]. Theoretical prediction for molecular orbital distributions were acquired based on the optimized structures.

2.2. Device fabrication and characterization

To investigate the electroluminescent properties of the SPDPPO and SBPBDPPO, PhOLEDs with the configuration of ITO/MoO₃ (3 nm)/mCP (30 nm)/Ir(ppy)₃ (8 wt%): host (18 nm)/TPBi (60 nm)/LiF (1 nm)/Al (200 nm) were fabricated by high-vacuum (3×10^{-4} Pa) thermal evaporation onto a glass substrate pre-coated with ITO. MoO₃ and N,N'-Dicarbazolyl-3,5-benzene (mCP) were used as hole injecting/transporting materials, and tris(2-phenylpyridine)iridium (Ir(ppy)₃) acts as emitting material. 1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi) was employed as electron-transporting and hole-blocking material. LiF was the electron injection layer. ITO (indium tin oxide) and Al (aluminum) were evaporated as the anode and cathode. Luminance-voltage-current density (L-V-J) characteristics of devices were recorded on Keithley 2400 Source Meter and L-2188 spot Brightness Meter. The active area of ITO was $(3 \times 3)\text{ mm}^2$. The electroluminescent spectra were recorded on PR-655 spectrophotometer.



Scheme 1. Synthetic scheme of SPDPPO to SBPBDPPO.

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