



Improved power efficiency in blue phosphorescent organic light-emitting diodes using diphenylmethyl linkage based high triplet energy hole transport materials

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ABSTRACT

Improved power efficiency in blue phosphorescent organic light-emitting diodes (PHOLEDs) was demonstrated by using new high triplet energy hole-transport materials based on the diphenylmethyl linkage. Two high triplet energy hole-transport materials with diphenylamine or ditolylamine moieties linked through a diphenylmethyl linkage, 4,4'-(diphenylmethylene)bis(N,N-diphenylaniline) (TCBPA) and 4,4'-(diphenylmethylene)bis(N,N-di-p-tolylaniline), were synthesized and evaluated as hole-transport materials for blue PHOLEDs. The power efficiency of TCBPA was superior to that of standard 1,1-bis[4-[N,N'-di(p-tolyl)amino]phenyl]cyclohexane.

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1. Introduction

High power efficiency is an important aspect of device performance for organic light-emitting diodes (OLEDs) because the power consumption of organic light-emitting diode panels can be reduced by improving their power efficiency. In general, power efficiency can be enhanced by lowering the driving voltage and increasing the quantum efficiency of organic light-emitting diodes. Therefore, high efficiency phosphorescent organic light-emitting diodes (PHOLEDs) with low driving voltage have been developed [1–5]. In particular, many studies have attempted to enhance the power efficiency of blue PHOLEDs due to the low power efficiency of current blue fluorescent OLEDs.

Much effort to improve the power efficiency of blue PHOLEDs was devoted to reducing the driving voltage of blue PHOLEDs by using host, hole and electron transport layers with high triplet energy and good charge transport properties [6–21]. Several electron transport materials based on the pyridine moiety were reported to be effective

in decreasing the driving voltage of blue PHOLEDs because of the high electron mobility of pyridine-based electron transport materials [6–8]. High power efficiency of 56 lm/W at 100 cd/m² was demonstrated using the high mobility electron transport materials. In addition to the electron transport materials, mixed host or bipolar host materials could also improve the driving voltage of blue PHOLEDs [9–14]. Several mixed host structures made up of strong hole and electron transport materials were reported and could lower the driving voltage due to low energy barrier for hole and electron injection and good charge transport properties [9–11]. Similarly, bipolar host materials with strong hole and electron transport groups in the molecular backbone structure were also effective to achieve the low driving voltage [12–14]. In the case of hole-transport materials, 1,1-bis[4-[N,N'-di(p-tolyl)amino]phenyl]cyclohexane (TAPC) is known as a high triplet energy hole-transport material capable of decreasing the driving voltage of blue PHOLEDs because of its high hole mobility [17]. However, few materials have been developed to improve the driving voltage of blue PHOLEDs [18–21]. Therefore, it is strongly required to develop high triplet energy hole-transport materials with good hole-transport

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properties to enhance the power efficiency of blue PHOLEDs.

In this work, two high triplet energy hole-transport materials with good hole-transport properties, 4,4'-(diphenylmethylene)bis(N,N-di-*p*-tolylaniline) (TCBTA) and 4,4'-(diphenylmethylene)bis(N,N-diphenylaniline) (TCBPA), were synthesized and compared with TAPC as hole-transport materials for blue PHOLEDs. The power efficiency of blue PHOLEDs was shown to be improved by more than 30% using TCBPA as the hole-transport material of blue PHOLEDs instead of TAPC.

2. Experimental section

2.1. General information

Triylaniline, sodium-*t*-butoxide, tri-*t*-butyl phosphine, 4,4'-dihydroxytetraphenylmethane and *p,p'*-ditolylamine (TCI Chem. Ind. Co.) were used without further purification. 4-bromotoluene, palladium acetate(II), pyridine, trifluoromethanesulfonic anhydride, 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl, tris(dibenzylideneacetone)dipalladium and diphenylamine (Aldrich Chem. Co.) were used without further purification. All reaction solvents were distilled over drying agent. The ¹H and ¹³C nuclear magnetic resonance (NMR) were recorded on Avance 500 (Bruker). Fourier transform infrared (FT-IR) spectra were recorded on a Nicolet 380 Fourier transform spectrometer. The mass spectra were recorded using a JEOL, JMS-600 W spectrometer in FAB mode. Elemental analysis of the materials was carried out using EA1110 (CE instrument). The DSC measurements were performed on a Mettler DSC822e differential scanning calorimeter under nitrogen at a heating rate of 10 °C/min. Photoluminescence (PL) spectra were recorded on a fluorescence spectrophotometer (HITACHI, F-7000) and the ultraviolet–visible (UV–Vis) spectra were obtained by means of a UV–Vis spectrophotometer (Shimadzu, UV-2501PC). Sample was dissolved in tetrahydrofuran at a concentration of 1.0×10^{-4} M for UV–Vis and PL measurements. Triplet energy analysis was carried out using low temperature PL measurement in liquid nitrogen. The highest occupied molecular orbital of compounds was measured with a cyclic voltammetry. Cyclic voltammetry measurement of organic materials was carried out in acetonitrile solution with tetrabutylammonium perchlorate at 0.1 M concentration. Ag was used as the reference electrode and Pt was the counter electrode.

3. Synthesis

Synthetic scheme of TCBTA and TCBPA is presented in Scheme 1.

3.1. Synthesis of 4,4'-(diphenylmethylene)bis(4,1-phenylene) bis(trifluoromethanesulfonate) (TC-OTf)

4,4'-Dihydroxytetraphenylmethane (10.0 g, 28.4 mmol) and pyridine(9.0 g, 113.5 mmol) were added to 100 mL dichloromethane and the solution was cooled in an ice salt

bath. Trifluoromethanesulfonic anhydride (24.0 g, 85.1 mmol) in 50 mL of dichloromethane was added dropwisely to the solution under nitrogen. The reaction was allowed to proceed for 2 h and quenched by adding methanol and water followed by dilution with dichloromethane. The crude solution was washed with dichloromethane and water. The organic layer was dried over anhydrous magnesium sulfate and evaporated in vacuo to give the crude product, which was purified by column chromatography using dichloromethane/hexanes eluent. The final white powdery product was obtained in 16.0 g.

TC-OTf yield 91%. ¹H NMR (500 MHz, CDCl₃): δ 7.13 (d, 4H, *J* = 4.0 Hz), 7.18 (d, 4H, *J* = 4.0 Hz), 7.23–7.31 (m, 10H). MS (FAB) *m/z* 616 [M⁺].

3.2. 4,4'-(diphenylmethylene)bis(N,N-di-*p*-tolylaniline) (TCBTA)

The mixture of TC-OTf (3.00 g, 4.87 mmol), *p,p'*-ditolylamine (2.40 g, 12.2 mmol), sodium *t*-butoxide (2.34 g, 24.4 mmol), 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl (S-Phos, 0.40 g, 0.975 mmol), and tris(dibenzylideneacetone)dipalladium (Pd₂(dba)₃, 0.23 g, 0.244 mmol) in 100 mL of dry xylene was refluxed under N₂ for 36 h. Then reaction mixture was cooled down to room temperature, diluted with 100 mL water and extracted with ethyl acetate. The organic extract was dried over anhydride magnesium sulfate, filtered, and evaporated to yield a brown solid. The crude material was purified by column chromatography on silica gel using dichloromethane/*n*-hexane as eluent. Additional purification by sublimation (290 °C. at 10⁻⁵ mm Hg) resulted in 1.9 g of pure white compound.

TCBTA yield 55%. mp 306 °C. Tg 100 °C. FT-IR (KBr) 3030.1, 1739.2, 1601.1, 1499.1, 1444.9, 1369.0, 1320.4, 1272.8, 1213.2, 1110.6, 1036.1, 815.6, 759.4, 723.0, 702.1 cm⁻¹. ¹H NMR (500 MHz, CDCl₃): δ 2.29 (s, 12H), 6.86 (d, 4H, *J* = 4.3 Hz), 6.98–7.00 (m, 12H), 7.04 (d, 8H, *J* = 4.0 Hz), 7.15–7.25 (m, 10H). ¹³C NMR (125 MHz, CDCl₃): δ 20.8, 64.1, 120.9, 124.7, 125.8, 127.3, 129.8, 131.2, 131.7, 132.4, 139.9, 145.3, 145.8, 147.2. MS (FAB) *m/z* 710 [M⁺]. Anal. Calcd. for C₅₃H₄₆N₂: C(89.54%); H(6.52%); N(3.94%). Found: C(89.47%); H(6.52%); N(3.94%).

3.3. 4,4'-(Diphenylmethylene)bis(N,N-diphenylaniline) (TCBPA)

The mixture of TC-OTf (3.00 g, 4.87 mmol), diphenylamine (2.06 g, 12.2 mmol), sodium *t*-butoxide (2.34 g, 24.4 mmol), 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl (S-Phos, 0.40 g, 0.975 mmol), tris(dibenzylideneacetone)dipalladium (Pd₂(dba)₃, 0.23 g, 0.244 mmol) in 100 mL of dry xylenes was refluxed under N₂ for 36 h. Then reaction mixture was cooled down to room temperature, diluted with 100 mL water and extracted with ethyl acetate. The organic extracts were dried over anhydride magnesium sulfate, filtered, and evaporated to yield a brown solid. The crude material was purified by column chromatography on silica gel using dichloromethane/*n*-hexane for eluent. Additional purification by sublimation

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