



Diphenylmethyl linked high-triplet-energy material as a host for deep-blue phosphorescent organic light-emitting diodes

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ARTICLE INFO

Article history:

Received 23 August 2012

Received in revised form 18 January 2013

Accepted 23 January 2013

Available online 4 February 2013

Keywords:

Diphenylmethyl linkage

High triplet energy

Deep blue phosphorescent organic light emitting diode

Carbazole

Dibenzofuran

ABSTRACT

A diphenylmethyl-linkage-based high-triplet-energy organic material, 3-(dibenzofuran-2-yl)diphenylmethyl-9-phenyl-9H-carbazole (TCDFCz), was synthesized as a host material for deep-blue phosphorescent organic light-emitting diodes (PHOLEDs). High-triplet-energy moieties, 9-phenylcarbazole and dibenzofuran, were connected through diphenylmethyl linkage, and high triplet energy of 2.97 eV was obtained. TCDFCz was effective as the high-triplet-energy host material for deep-blue PHOLEDs, and a high quantum efficiency of 13.1% was achieved.

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1. Introduction

The development of high-efficiency blue phosphorescent organic light-emitting diodes (PHOLEDs) is of great importance for reducing the power consumption of organic light-emitting diode (OLED) displays, because of low quantum efficiency of the current fluorescent blue OLEDs. Theoretically, the quantum efficiency of PHOLEDs can be four-times higher than that of fluorescent PHOLEDs, and external quantum efficiency above 20% has already been reported in blue PHOLEDs [1–9].

Great progress in the quantum efficiency of blue PHOLEDs has mainly been made through the development of high-triplet-energy host materials [10–15]. The development of high-triplet-energy materials was made by interconnecting high-triplet-energy moieties through various linkages that do not increase the conjugation length of host materials. Typical linkages include diphenylsilane [16,17] and diphenylphosphine oxide [18,19]. Benzene and pyridine were also applied as connecting units between high-triplet-energy moieties, but the triplet energy of host materials was degraded because of extended conjugation through those units. Therefore, further development of connecting units that can maintain the high triplet energy of each moiety is strongly required.

In this work, high-triplet-energy host material with diphenylmethyl linkage, 3-(dibenzofuran-2-yl)diphenylmethyl-9-phenyl-9H-carbazole (TCDFCz), was developed as a host material for deep-blue

PHOLEDs, and the device performance of the PHOLEDs was investigated. The TCDFCz was designed to have high triplet energy by separating the conjugation between dibenzofuran and carbazole using diphenylmethyl linkage. A high triplet energy of 2.97 eV was obtained in TCDFCz, and a high quantum efficiency of 13.1% was demonstrated in deep-blue PHOLEDs using TCDFCz host material.

2. Experimental details

2.1. General information

Benzophenone, 9-phenylcarbazole, Eaton's reagent and *n*-butyllithium (Aldrich Chem. Co.) were used without further purification. 2-Bromodibenzofuran was synthesized according to literature method [20]. Tetrahydrofuran (Duksan Sci. Co.) was distilled over sodium and calcium hydride.

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. Transmittance measurements were conducted using KBr pellet method. The mass spectra were recorded using a JEOL, JMS-600W spectrometer in fast atom bombardment mode. Elemental analysis of the materials was carried out using EA1110 (CE instrument). Differential scanning calorimeter (DSC) measurements were performed on a Mettler DSC822e 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

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(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 performed using low temperature PL measurement in liquid nitrogen. Excitation wavelength for PL measurement was 310 nm. The highest occupied molecular orbital of compound was estimated from ionization energy measured with a surface analyzer (Riken Keiki Co. Ltd., AC-2).

2.2. Synthesis

Synthetic scheme of TCDFCz is presented in Scheme 1.

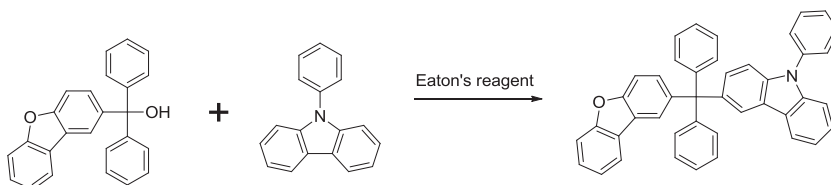
2.2.1. Synthesis of 3-(dibenzofuran-2-yl diphenylmethyl)-9-phenyl-9H-carbazole (TCDFCz)

Dibenzofuran-2-yl diphenylmethanol (2.5 g, 7.13 mmol) and 9-phenylcarbazole (3.48 g, 14.3 mmol) were dissolved in dichloromethane (50 ml) under a nitrogen atmosphere. The reaction mixture was stirred at 0 °C and Eaton's reagent (1.62 ml) was added slowly in the reaction mixture. The reaction mixture was stirred for 4 h at same temperature. The mixture was diluted with dichloromethane and washed with distilled water three times. 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/hexane(5/1). The final white powdery product was obtained in 1.8 g.

Yield 44%. Tg 113 °C. FT-IR (KBr) 3051.7, 1734.4, 1594.4, 1476.8, 1449.0, 1361.7, 1324.8, 1281.4, 1193.8, 1128.2, 1026.7, 927.9, 843.4, 811.1, 746.0, 699.9 cm^{-1} . $^1\text{H NMR}$ (500 MHz, CDCl_3): δ 7.21–7.31 (m, 10H), 7.34–7.45 (m, 10H), 7.54 (d, 1H, $J=4.3\text{Hz}$), 7.57(d, 4H, $J=2.0\text{Hz}$), 7.79 (d, 1H, $J=4.0\text{Hz}$), 7.92 (s, 1H), 7.95 (d, 1H, $J=3.7\text{Hz}$), 8.07 (s, 1H). $^{13}\text{C NMR}$ (125 MHz, CDCl_3): δ 65.0, 108.9, 109.8, 110.5, 111.6, 119.9, 120.2, 120.7, 122.0, 122.5, 122.5, 122.7, 123.4, 123.6, 124.5, 125.8, 125.9, 126.9, 127.3, 127.5, 129.8, 130.5, 131.3, 131.4, 137.7, 139.0, 141.1, 142.3, 147.6, 154.4, 156.6. DEPT (CDCl_3): 108.9, 109.8, 110.5, 111.6, 119.9, 120.3, 120.7, 122.01, 122.5, 122.7, 125.8, 125.9, 126.9, 127.3, 127.6, 129.8, 130.5, 131.3, 131.4. MS (FAB) m/z 575 [M^+]. Anal. Calcd for $\text{C}_{43}\text{H}_{29}\text{NO}$: C (89.71%), H (5.08%), N (2.43%), Found: C (89.53%), H (5.05%), N (2.43%).

2.3. Device fabrication and measurement

Device structure of blue PHOLED fabricated using TCDFCz was indium tin oxide (ITO, 50 nm)/N,N'-diphenyl-N,N'-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD, 30 nm)/N,N'-di(1-naphthyl)-N,N'-diphenylbenzidine (NPB, 15 nm)/N,N'-dicarbazolyl-3,5-benzene (mCP, 10 nm)/TCDFCz: bis((3,5-difluoro-4-cyanophenyl)pyridine) iridium picolinate (FCNIrpic) (30 nm)/1,3-bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPB, 25 nm)/LiF (1 nm)/Al (200 nm). Doping concentrations of FCNIrpic were 10%, 15% and 20%. All organic materials were thermally deposited on ITO substrate at a vacuum pressure of 1.33×10^{-4} Pa. After thermal evaporation of Al cathode, all devices were encapsulated with a glass lid and CaO getter. Device performances of blue PHOLEDs were measured using Keithley 2400 source measurement unit and CS1000 spectroradiometer.



Scheme 1. Synthetic scheme of TCDFCz.

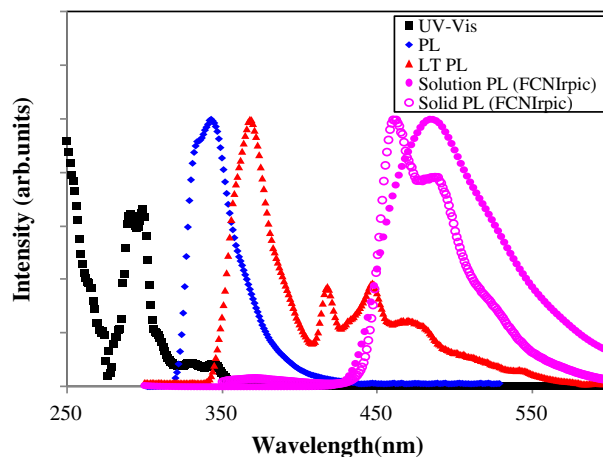


Fig. 1. UV–vis absorption, solution PL and low temperature PL spectra of TCDFCz. Excitation wavelength for PL measurement was 310 nm. Solution PL spectrum of FCNIrpic in tetrahydrofuran and solid PL spectra doped FCNIrpic film were also added.

3. Results and discussion

TCDFCz was designed to exhibit high triplet energy by combining two high-triplet-energy moieties, 9-phenylcarbazole and dibenzofuran, through diphenylmethyl linkage. The diphenylmethyl linkage interconnects two aromatic units using sp^3 carbon, and can maintain the high triplet energy of each aromatic unit. Therefore, the high triplet energy required for deep-blue phosphorescent emitters can be obtained in TCDFCz.

UV–vis and PL measurements were carried out to analyze the photophysical properties of TCDFCz. Fig. 1 shows UV–vis, solution PL, and low-temperature PL spectra of TCDFCz. Strong absorption peaks at 246 nm, 291 nm, and 299 nm assigned to the combined absorption of dibenzofuran and phenylcarbazole were observed, and additional weak UV–vis absorption peaks at 330 nm and 344 nm which correspond to the absorption of 9-phenylcarbazole were detected in the UV–vis absorption spectrum. The bandgap was 3.40 eV according to the absorption edge of the UV–vis spectrum. Solution PL emission of TCDFCz was observed at 371 nm, which was red-shifted to a longer wavelength compared to that of 9-phenylcarbazole (363 nm). Triplet energy was calculated from the first phosphorescent emission peak as 2.97 eV. Considering the triplet energy of 9-phenylcarbazole (3.02 eV) and dibenzofuran (3.04 eV), the triplet energy of TCDFCz was degraded by only 0.05 eV due to the isolation of each unit by the diphenylmethyl linkage. The triplet energy of TCDFCz was high enough for energy transfer to deep-blue emitting FCNIrpic with a triplet energy of 2.74 eV. Solution PL and solid PL spectra of FCNIrpic are also shown in this figure.

The thermal properties of TCDFCz were measured using DSC. The glass transition temperature of TCDFCz was 113 °C, which was higher than that of common mCP host material (55 °C) because of the diphenylmethyl linkage.

Molecular simulation of TCDFCz was performed to study the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO). A suite of Gaussian 03 software and the nonlocal

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