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High quantum efficiency and color stability in white phosphorescent organic light emitting diodes using a pyridine modified carbazole derivative

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ABSTRACT

A high triplet energy material derived from carbazole and pyridine, 9,9'-(5-(2-methylpyridin-3-yl)-1,3-phenylene)bis(9*H*-carbazole), was synthesized as the host material for blue and white phosphorescent organic light-emitting diodes. The 9,9'-(5-(2-methylpyridin-3-yl)-1,3-phenylene)bis(9*H*-carbazole) host showed a high triplet energy of 2.99 eV for efficient energy transfer to blue triplet emitter. High quantum efficiencies of 19.8% and 17.1% were achieved in blue and white phosphorescent organic light-emitting diodes using the 9,9'-(5-(2-methylpyridin-3-yl)-1,3-phenylene)bis(9*H*-carbazole) host material. In addition, the 9,9'-(5-(2-methylpyridin-3-yl)-1,3-phenylene)bis(9*H*-carbazole) based white phosphorescent organic light-emitting diodes showed good color stability.

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1. Introduction

Phosphorescent white organic light emitting diodes (PHWO-LEDs) are promising as a next generation lighting device due to high quantum efficiency and broad emission spectrum for high color rendering index. In particular, the high quantum efficiency of the PHWOLEDs is an attractive feature for a lighting device because power consumption can be reduced.

In general, the PHWOLEDs are constructed by combining a blue triplet emitter with a yellow triplet emitter, or red, green and blue triplet emitters [1-10]. The energy of the blue triplet emitter is transferred to low energy triplet emitters during the light emission process and the device performance of the PHWOLEDs are mostly dominated by the device performance of the blue phosphorescent organic light-emitting diodes (PHOLEDs). Therefore, the development of high efficiency blue PHOLEDs is important to improve the device performances of PHWOLEDs and one of the most efficient methods is to develop high triplet energy host materials [11-15]. Several high triplet energy host materials were

0143-7208/\$ - see front matter © 2013 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.dyepig.2013.11.020 reported as the host materials for the PHWOLEDs to enhance the quantum efficiency. Although high quantum efficiency was reported using various host materials, it was difficult to obtain both high quantum efficiency and color stability in the PHWOLEDs.

In this work, a high triplet energy host material derived from pyridine and carbazole, 9,9'-(5-(2-methylpyridin-3-yl)-1,3-phenylene)bis(9H-carbazole) (PPBC), was developed to improve the quantum efficiency and color stability of PHWOLEDs. High quantum efficiencies of 17.1% and good color stability with 0.01 change of color coordinate between 100 cd/m² and 10,000 cd/m² were demonstrated in PHWOLEDs using the PPBC host material in addition to high quantum efficiency of 19.8% in blue PHOLEDs.

2. Experimental section

Reagents and solvents were purchased at reagent grade from TCI, Aldrich, and P&H tech and used as received. All reactions were performed under a positive pressure of high purity nitrogen. The ¹H and ¹³C nuclear magnetic resonance (NMR) were obtained on Avance 500 (Bruker). The FT-IR spectra for all the samples were measured using a Nicolet 380 FTIR spectrometer. High resolution mass spectra were measured on JEOL, JMS-600W spectrometer in fast atom bombardment(FAB) mode. Elemental analyses of carbon, hydrogen, and nitrogen were performed on Flash2000 (Thermofisher). Differential





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scanning calorimetry (DSC) was performed on a Mettler DSC822e at a heating rate of 10 °C/min from 20 to 350 °C under argon. The photoluminescence (PL) spectra were recorded on a fluorescence spectrophotometer (HITACHI, F-7000) and the ultraviolet–visible (UV–Vis) spectra were obtained using a UV–Vis spectrophotometer (Shimadzu, UV-2501PC). Low temperature PL measurement for triplet energy analysis recorded on a PerkinElmer LS-55 in liquid nitrogen. Cyclic voltammetry (CV) was carried out in acetonitrile solution (oxidation scan) at room temperature with tetrabutylammonium perchlorate (0.1 M) was used as the supporting electrolyte. The conventional three-electrode configuration consists of a platinum working electrode, a platinum wire auxiliary electrode, and an Ag wire pseudoreference electrode with ferrocenium–ferrocene (Fc⁺/Fc) as the internal standard. The synthetic route of PPBC is shown in Scheme 1.

2.1. Synthesis of 2-(3,5-dichlorophenyl)-3-methylpyridine

3-Bromo-2-methylpyridine (1.9 g, 11.04 mmol) and 3,5dichlorophenyl boronic acid (3.16 g, 16.57 mmol) dissolved in degassed THF (60 mL) were placed into a 250 mL two necked round bottom flask. A solution of potassium carbonate (3.05 g, 22.09 mmol) in water (20 mL) was added slowly to the solution, and bubbled with nitrogen for 30 min. Finally, tetrakis(triphenylphosphine)palladium(0) (1.26 g, 1.17 mmol) was added to the above reaction mixture, then heated under reflux under nitrogen overnight. The solution was cooled to room temperature and the organics were extracted using ethyl acetate and washed with distilled water. The solution was dried over magnesium sulfate and evaporated to remove solvent. The product was further purified by flash chromatography on silica gel using methylene chloride/*n*-hexane as an eluent to produce 2.0 g (76% yield) of 2-(3,5-dichlorophenyl)-3-methylpyridine.

Yellow powder, m.p. 87 °C, FT-IR (KBr) 3051, 1557, 1423, 1407, 1383, 1293, 1254, 1190, 1097, 1034, 884, 853, 793, 765, 736, 695 cm⁻¹. ¹H NMR (500 MHz, CDCl₃): δ 2.50 (s, 3H), 7.19 (t, 1H, J = 2.5 Hz), 7.21 (s, 2H), 7.39 (s, 1H), 7.47 (d, 1H, J = 4.5 Hz), 8.54 (d, 1H, J = 3.2 Hz). ¹³C NMR (125 MHz, CDCl₃): δ 23.2, 121.0, 127.4, 127.6, 134.3, 135.0, 136.8, 142.8, 148.8, 155.5. HRMS calcd for C₁₂H₉Cl₃N (M⁺ + H) 238.0185, found 238.0190.

2.2. Synthesis of 9,9'-(5-(3-methylpyridin-2-yl)-1,3-phenylene) bis(9H-carbazole)

2-(3,5-Dichlorophenyl)-3-methylpyridine (2.0 g, 8.40 mmol), carbazole (3.51 g, 21.00 mmol), sodium *tert*-butoxide (4.04 g, 42.00 mmol), 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl (0.86 g, 2.10 mmol), tris(dibenzylideneacetone)dipalladium (0.48 g, 0.52 mmol) and xylene (140 mL) were combined and heated under reflux under nitrogen for 36 h. The solution was cooled to room temperature and the organics were extracted into ethyl acetate, washed with distilled water, dried over magnesium sulfate, filtered. The solvent was removed in vacuo, and the product was isolated by column chromatography on silica gel (methylene chloride/*n*-

hexane) to give 9,9'-(5-(3-methylpyridin-2-yl)-1,3-phenylene) bis(9*H*-carbazole) (1.95 g, 46% yield). Additional purification by sublimation (at 200 °C and 10^{-5} mm Hg) resulted in 1.5 g of pure compound (36% overall yield).

Light yellow powder, m.p. 132 °C, T_g 94 °C, FT-IR (KBr) 3047, 1590, 1450, 1334, 1311, 1229, 1155, 1120, 1026, 1002, 925, 806, 748, 710, 647 cm⁻¹. ¹H NMR (500 MHz, CDCl₃): δ 2.75 (s, 3H), 7.21 (t, 1H, J = 4.2 Hz), 7.30 (t, 4H, J = 5.2 Hz), 7.44 (t, 4H, J = 5.3 Hz), 7.58 (d, 4H, J = 4.0 Hz), 7.65 (s, 2H), 7.69 (d, 1H, J = 4.8 Hz), 7.85 (s, 1H), 8.13 (d, 4H, J = 4.0 Hz), 8.55 (d, 1H, J = 3.3 Hz). ¹³C NMR (125 MHz, CDCl₃): δ 23.8, 109.5, 120.5, 121.3, 123.7, 124.1, 126.1, 126.2, 135.1, 137.1, 139.5, 140.5, 143.4, 148.9, 155.6. HRMS calcd for C₃₆H₂₅N₃ (M⁺ + H) 500.2121, found 500.2127. Element analysis Calcd. for C₃₆H₂₅N₃ C(86.55%) H(5.04%) N(8.41%); found C(86.04%) H(5.08%) N(8.52%).

2.3. Device fabrication and measurements

Device structure of blue PHOLEDs was indium tin oxide (50 nm)/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, 60 nm)/4,4'-cyclohexylidenebis[*N*,*N*-bis(4-methyl phenyl)aniline] (TAPC, 20 nm)/1,3-bis(N-carbazolyl)benzene (mCP, 10 nm)/PPBC:iridium(III) bis[(4,6-difluorophenyl)-pyridinato-*N*,*C*²] picolinate (FIrpic) (25 nm)/diphenylphosphine oxide-4-(triphenyl silyl)phenyl (TSPO1, 35 nm)/LiF(1 nm)/Al(200 nm). Doping concentrations of FIrpic were 3, 5, and 10%. PHWOLEDs had the device structure of ITO (50 nm)/PEDOT:PSS (60 nm)/TAPC (20 nm)/mCP (10 nm)/PPBC:FIrpic (15 nm)/1,3,5-tris(N-phenylbenzimidazole-2yl)benzene (TPBI):iridium(III) bis(2-phenylquinoline) acetylacetonate(Ir(pq)₂acac) (15 nm, 5%)/TSPO1 (35 nm)/LiF (1 nm)/Al (200 nm). The doping concentrations of FIrpic were 3, 5 and 10%. The doping concentration of FIrpic and Ir(pq)₂acac was controlled by managing the evaporation rate of host and dopant materials. Hole only device with a device structure of ITO (50 nm)/PEDOT:PSS (60 nm)/TAPC (20 nm)/mCP (10 nm)/PPBC (25 nm)/TAPC (10 nm)/ Al and electron only device with a device structure of ITO (50 nm)/ Ca (5 nm)/PPBC (25 nm)/TSPO1 (35 nm)/LiF (1 nm)/Al (200 nm) were also prepared as single carrier devices. All devices were fabricated by vacuum thermal evaporation and were encapsulated with a glass lid after cathode deposition. The substrate and the glass lid were sealed using epoxy adhesive and CaO desiccant was inserted inside the device to capture moisture and oxygen. Current density-voltage characteristics of all devices were measured by Keithley 2400 source measurement unit and luminance performances were characterized by CS1000 spectroradiometer.

3. Results and discussion

The design of PPBC was based on carbazole and pyridine unit to obtain bipolar charge transport properties from the PPBC host material. The carbazole unit can play a role of a hole transport unit, while the pyridine unit can act as an electron transport unit, which can induce bipolar charge transport character in the PPBC host. In addition, a methyl substituent was attached to the pyridine unit to



Scheme 1. Synthetic route of PPBC.

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