

Effect of nitrogen position of carboline on the device performances of blue phosphorescent organic light-emitting diodes



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ARTICLE INFO

Article history:

Received 24 April 2015

Received in revised form 6 June 2015

Accepted 11 June 2015

Available online 2 July 2015

Keywords:

Carboline

Blue device

Power efficiency

Bipolar host

ABSTRACT

A δ -carboline derived compound, 5-(3'-(9-carbazolyl)-[1,1'-biphenyl]-3-yl)pyrido[3,2-b]indole, was synthesized as a high triplet energy bipolar host material for blue phosphorescent organic light-emitting diodes and it was compared with α -carboline derived host material with the same backbone structure. The δ -carboline derived host material showed better electron transport properties than the host with α -carboline due to better electron accepting properties. Therefore, the new host material reduced driving voltage and increased the power efficiency of blue phosphorescent organic light-emitting diodes compared to a standard host with α -carboline moiety. A high external quantum efficiency of 25.3% and a high power efficiency of 36.4 lm/W were achieved in the blue phosphorescent organic light-emitting diodes.

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1. Introduction

Host materials are critical to the quantum efficiency of phosphorescent organic light-emitting diodes (PHOLEDs) because they play a role of balancing holes and electrons, managing energy transfer, and controlling charge injection in the emitting layer. High triplet energy host materials are required for the energy transfer and bipolar host materials are necessary for the good charge balance and charge injection. Therefore, high triplet energy bipolar host materials have been actively studied for application in PHOLEDs [1–7].

The bipolar host materials can be developed by combining a hole transport unit and an electron transport unit, or using a bipolar charge transport unit. Typically, the hole transport and electron transport units were used in the molecular structure for the bipolar transport properties [1–7], but bipolar units can also be effectively used as the building unit of the high triplet energy bipolar host materials [8–13]. Recently, carboline moieties are being investigated as the bipolar transport type building block of the bipolar host materials [8–13]. Several carboline based host materials have been synthesized and could improve the quantum efficiency of blue PHOLEDs due to high triplet energy and bipolar charge transport properties. However, most carboline based host materials were derived from α -carboline [11–13] and little

research was carried out about the bipolar host materials derived from carboline unit with different nitrogen orientations [10].

In this work, a carboline based bipolar host material derived from δ -carboline, 5-(3'-(9-carbazolyl)-[1,1'-biphenyl]-3-yl)pyrido[3,2-b]indole (CzBPDCb), was synthesized and it was compared with 9-(3'-(9-carbazolyl)-[1,1'-biphenyl]-3-yl)-pyrido[2,3-b]indole (CzBPCb) derived from α -carboline. It was demonstrated that the δ -carboline unit enhanced electron transport properties of the host materials and reduced driving voltage of the device while keeping the high quantum efficiency of the device compared to α -carboline unit.

2. Experimental

2.1. General information

2-Bromopyridin-3-amine, iodobenzene, diiodobenzene, and 1,8-diazabicyclo[5,4,0]undec-7-ene (DBU) (TCI Chem. Co.) were used without further purification. Sodium *tert*-butoxide, *tert*-butyl alcohol, 2-(dicyclohexylphosphine)biphenyl, copper iodide (CuI), *trans*-1,2-diaminocyclohexane (1,2-TCH) (Aldrich Chem. Co.), potassium carbonate (K₂CO₃), *N,N*-dimethylacetamide (DMAC), 1,4-dioxane (Duksan Sci. Co.), palladium acetate, 4,5-bis(diphenylphosphino)-9,9-dimethylxanthene(xantphos), (3-(9H-carbazol-9-yl)phenyl)boronic acid, tetrakis(triphenylphosphine) palladium(0) (Pd(PPh₃)₄) (P&H tech Co.), and potassium phosphate (K₃PO₄) (Daejung Sci. Co.) were also used as received. General analysis method of the synthesized compounds is described in previous work [14].

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2.2. Synthesis

Synthetic scheme of CZBPDCB is described in Scheme 1.

2.3. 2-Bromo-N-phenylpyridin-3-amine (1)

2-Bromo-N-phenylpyridin-3-amine was synthesized according to the method described in the literature [15]. 2-Bromopyridin-3-amine (1.75 g, 10.12 mmol) was used instead of 3-bromopyridin-2-amine.

Yield 50%, $^1\text{H NMR}$ (400 MHz, DMSO): δ 7.88 (d, 1H, $J=4.40$ Hz), 7.68 (s, 1H), 7.52 (d, 1H, $J=7.60$ Hz), 7.30–7.25 (m, 3H), 7.12 (d, 2H, $J=8.40$ Hz), 6.97 (t, 1H, $J=7.80$ Hz) MS (ESI) m/z 248 [(M+H) $^+$].

2.4. 5H-pyrido[3,2-b]indole(δ -carboline) (2)

5H-pyrido[3,2-b]indole(δ -carboline) was prepared according to the synthetic method reported earlier [16].

Yield 62%, $^1\text{H NMR}$ (400 MHz, DMSO): δ 11.45 (s, 1H), 8.44 (d, 1H, $J=4.80$ Hz), 8.18 (d, 1H, $J=8.00$ Hz), 7.88 (d, 1H, $J=8.00$ Hz), 7.56 (d, 1H, $J=8.40$ Hz), 7.50 (t, 1H, $J=4.80$ Hz), 7.38 (t, 1H, $J=6.40$ Hz), 7.24 (t, 1H, $J=7.40$ Hz) MS (ESI) m/z 169 [(M+H) $^+$].

2.5. 9-(3'-Iodo-[1,1'-biphenyl]-3-yl)carbazole (3)

(3-(Carbazol-9-yl)phenyl)boronic acid (2.84 g, 9.9 mmol), diiodobenzene (14 g, 49.49 mmol), and K_2CO_3 (2.74 g, 19.8 mmol) were dissolved in tetrahydrofuran (150 ml), methanol (50 ml), and distilled water (20 ml) under bubbling with nitrogen for 30 min. $\text{Pd}(\text{PPh}_3)_4$ (1.15 g, 0.99 mmol) was added to the solution and the resulting solution was refluxed for 24 h under a nitrogen atmosphere. The mixture was diluted with ethyl acetate and washed with distilled water. The organic layer was dried over anhydrous MgSO_4 and evaporated *in vacuo* to give a crude product. The extract was evaporated to dryness affording a white solid, which was further purified by column chromatography using methylene chloride/hexane as an eluent.

Yield 13.6%, $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 8.16 (d, 2H, $J=7.60$ Hz), 7.99 (t, 1H, $J=1.80$ Hz), 7.75–7.56 (m, 6H), 7.46–7.40 (m, 4H), 7.31 (t, 2H, $J=8.00$ Hz), 7.19 (t, 1H, $J=7.80$ Hz) MS (FAB) m/z 445 [(M) $^+$].

2.6. 5-(3'-(Carbazol-9-yl)-[1,1'-biphenyl]-3-yl)pyrido[3,2-b]indole (CzBPDCb) (4)

9-(3'-Iodo-[1,1'-biphenyl]-3-yl)carbazole (0.6 g, 1.35 mmol), δ -carboline (0.27 g, 1.62 mmol), CuI (0.13 g, 0.68 mmol), and K_3PO_4 (0.57 g, 2.7 mmol) were dissolved in 1,4-dioxane (30 ml) under a nitrogen atmosphere. The reaction mixture was stirred for

30 min and *trans*-1,2-diaminocyclohexane (0.08 ml, 0.68 mmol) was added to the solution followed by reflux overnight. After cooling to room temperature, the mixture was filtered, diluted with methylene chloride, and washed with distilled water. The organic layer was dried over anhydrous MgSO_4 and evaporated *in vacuo*. The crude product was purified by column chromatography using methylene chloride/n-hexane as an eluent. A white powder was obtained after vacuum train sublimation.

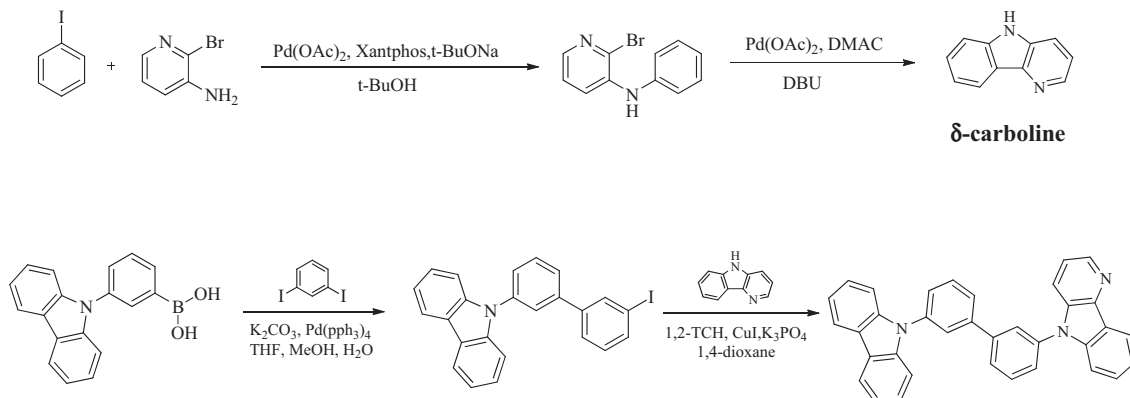
Yield 53%, $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 8.62 (d, 1H, $J=6.00$ Hz), 8.46 (d, 1H, $J=7.60$ Hz), 8.15 (d, 2H, $J=8.00$ Hz), 7.85 (d, 2H, $J=10.00$ Hz), 7.77–7.68 (m, 5H), 7.62–7.56 (m, 2H), 7.53 (d, 2H, $J=6.00$ Hz), 7.47–7.37 (m, 5H), 7.34–7.28 (m, 3H) $^{13}\text{C NMR}$ (400 MHz, CDCl_3): δ 142.75, 142.41, 142.36, 141.88, 141.56, 140.85, 138.57, 137.68, 134.33, 130.70, 130.58, 128.15, 126.63, 126.57, 126.17, 126.14, 126.06, 125.79, 125.48, 123.48, 122.61, 121.01, 120.40, 120.29, 120.12, 116.74, 110.04, 109.70 MS (FAB) m/z 468 [(M+H) $^+$]. Elemental Analysis (calculated for $\text{C}_{35}\text{H}_{23}\text{N}_3$): C, 86.57; H, 4.77; N, 8.65. Found: C, 85.80; H, 4.72; N, 8.65.

2.7. Device fabrication and measurements

Blue PHOLEDs were grown on a 50 nm thick indium tin oxide (ITO) substrate. The ITO substrate was cleaned using deionized water, isopropanol and acetone before device fabrication. After cleaning of the ITO substrate, 60 nm thick polyethylene-3,4-dioxythiophene:poly-styrenesulfonate (PEDOT:PSS) was spin coated on the ITO substrate followed by baking at 120 °C for 10 min in ambient condition and 150 °C for 10 min inside a glove box. The PEDOT:PSS coated substrate was transferred to a vacuum evaporation chamber which has a high vacuum pressure of 7.0×10^{-7} torr. 1,1'-bis(di-4-tolylaminophenyl)cyclohexane (TAPC, 20 nm) and *N,N*-dicarbazolyl-3,5-benzene (mCP, 10 nm) were deposited as a double layer hole transport layer and CzBPDCb:iridium (III) bis[2-(4,6-difluorophenyl)-pyridinato- N,C^2]picolate (Flrpic) (25 nm, 3%) was evaporated on the mCP hole transport layer as an emitting layer. Finally, diphenylphosphine oxide-4-(triphenylsilyl)phenyl (TSPO1, 35 nm), LiF (1 nm), and Al (100 nm) were thermally deposited on the mCP:Flrpic emitting layer as an electron transport layer, an electron injection layer and a cathode. After the cathode deposition, the device was encapsulated in the glove box using a glass cover and a desiccant. Device performances were measured in ambient condition using CS 2000 spectroradiometer (Minolta Co.) and Keithley 2400 source measurement unit.

3. Results and discussion

The CzBPDCb compound was developed as a bipolar type host by merging a hole transport type carbazole and a weak electron



Scheme 1. Synthetic scheme of CzBPDCb.

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