

Carbazole-dibenzothiophene core as a building block of host materials for blue phosphorescent organic light-emitting diodes

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ABSTRACT

A high triplet energy core structure, carbazole-dibenzothiophene, was designed and synthesized as a building block of high triplet energy host materials for blue phosphorescent organic light-emitting diodes. The carbazole and dibenzothiophene were interconnected via 3- position of carbazole and 2- position of dibenzothiophene not to sacrifice the high triplet energy the two units. Three compounds with phenyl, dibenzothiophene and 9-phenylcarbazole functional groups as side groups of the carbazole-dibenzothiophene main structure worked as hosts providing high triplet energy over 2.80 eV and the host with a dibenzothiophene substituent achieved high external quantum efficiency over 20% in blue phosphorescent organic light-emitting diodes.

1. Introduction

Blue phosphorescent organic light-emitting diodes (PhOLEDs) have shown potential as next generation organic light-emitting diodes (OLEDs) for low power-consuming displays because of high external quantum efficiency (EQE) [1–5]. The best EQE of the blue PhOLEDs is more than twice that of the fluorescent OLEDs and reached 30% EQE [6–10]. Therefore, it is being generally accepted that the blue PhOLEDs would be the most promising technology for high EQE blue PhOLEDs.

In the pursuit of the high EQE blue PhOLEDs, host materials played an important role because key requirements of high EQE, carrier balance and exciton harvesting are dictated by the hosts [11,12]. Rational design of the hosts intended to balance carriers and harvest excitons increased the EQE of the blue PhOLEDs and most high EQE data were obtained using bis[2-(4,6-difluorophenyl)pyridinato-C²,N](picolinato) iridium(III) (FIrpic) as a phosphorescent emitter. For instance, a bipolar host combined with the FIrpic emitter allowed high EQE of 27.5% and an exciplex host doped with the FIrpic emitter realized high EQE of 22.1% [13,14]. Although high EQE data were reported in many publications using FIrpic as a blue emitter, the FIrpic emitter showed poor lifetime because of instability [15–18]. Therefore, phenylimidazole ligand based Ir emitters have been used in the blue PhOLEDs in combination with the high triplet energy hosts to study lifetime of the blue PhOLEDs [19–23]. However, the phenylimidazole ligand based Ir emitters are quite different from FIrpic in that they are hole trapping type emitter and FIrpic is an electron trapping type emitter. The dissimilar carrier trapping effect in the light-emitting layer required different hosts to optimize the EQE of the blue PhOLEDs. In the case of

FIrpic, bipolar or electron transport type hosts were successful for high EQE, whereas hole transport type hosts were effective to reach high EQE in the case of phenylimidazole based triplet emitters [24]. For example, carbazole or dimethylacridine derived hosts were targeted for high EQE in the phenylimidazole based blue PhOLEDs. However, only several high triplet energy hosts are available for application in the phenylimidazole based Ir emitters, and much more hosts are required to develop high performance blue PhOLEDs.

In this paper, new hosts with a high triplet energy backbone structure, carbazole-dibenzothiophene, were developed as hosts for phenylimidazole ligand based triplet emitters. The backbone structure itself was a high triplet energy core and realized high triplet energy over 2.80 eV. Additionally, the carbazole-dibenzothiophene derived hosts accomplished high EQE over 20% in the blue PhOLEDs doped with a phenylimidazole derived triplet emitter.

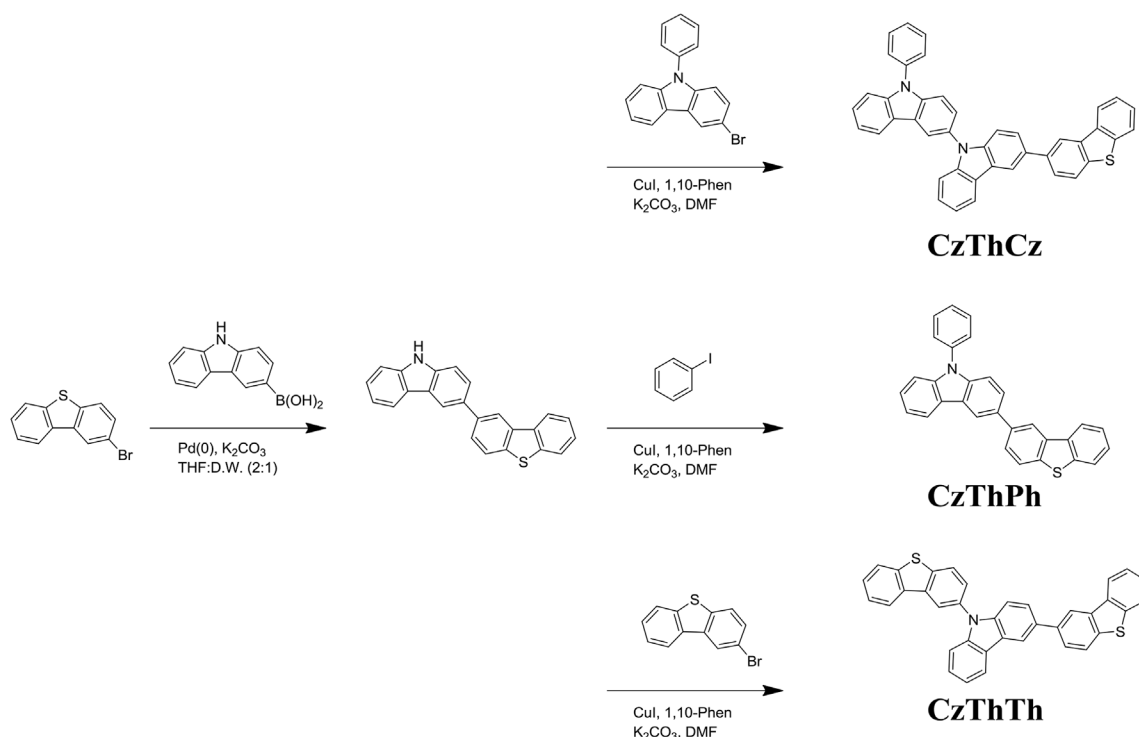
2. Experimental

2.1. General information

2-Bromodibenzo[*b,d*]thiophene, (9*H*-carbazol-3-yl)boronic acid, 3-bromo-9-phenyl-9*H*-carbazole and tetrakis(triphenylphosphane)palladium(0) (P&H tech Co.), *N,N*-dimethylformamide (DMF), potassium carbonate and magnesium sulfate (Duksan Sci. Co.), copper iodide, 1,10-phenanthroline and dimethylsulfoxide-*d*₆ (Sigma-Aldrich Co. LLC.), iodobenzene (Alfa aesar Co.), tetrahydrofuran (Samchun pure chemical Co. Ltd) were purchased and used as received. The analytic instruments used in this work are provided in our previous report [25]

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Scheme 1. Synthetic scheme of CzThCz, CzThPh and CzThTh.

2.2. Synthesis

Synthetic scheme of the CzThCz, CzThPh and CzThTh compounds are described in Scheme 1.

2.3. 3-(Dibenzo[b,d]thiophen-2-yl)-9H-carbazole

2-Bromodibenzo[b,d]thiophene (5.00 g, 1.90 mmol) and (9H-carbazol-3-yl)boronic acid (4.81 g, 2.28 mmol) were dissolved in 40 ml of tetrahydrofuran. Then, potassium carbonate (7.88 g, 5.70 mmol) aqueous solution (20 ml) was poured into the tetrahydrofuran solution, which was refluxed for 3 h with stirring. The solution was cooled down, and extracted with methylene chloride and distilled water. The obtained organic phase was dehydrated using magnesium sulfate, and further purified with silica column chromatography with a mixed eluent of methylene chloride and *n*-hexane.

Yield 78.3% (5.20 g), ^1H NMR (500 MHz, $\text{DMSO-}d_6$): δ 11.36 (s, 1H), 8.78 (s, 1H), 8.64 (s, 1H), 8.26 (d, 1H, $J = 8.0$ Hz), 8.11 (d, 1H, $J = 8.5$ Hz), 8.05 (t, 1H, $J = 8.0$ Hz), 7.95 (d, 1H, $J = 8.0$ Hz), 7.90 (d, 1H, $J = 8.0$ Hz), 7.61 (d, 1H, $J = 8.5$ Hz), 7.57–7.51 (m, 3H), 7.42 (t, 1H, $J = 7.5$ Hz), 7.21 (t, 1H, $J = 7.5$ Hz), MS (APCI) m/z 350.2 $[(M + H)^+]$.

2.4. 3'-(Dibenzo[b,d]thiophen-2-yl)-9-phenyl-9H-3,9'-bicarbazole (CzThCz)

3-(Dibenzo[b,d]thiophen-2-yl)-9H-carbazole (1.50 g, 0.43 mmol), 3-bromo-9-phenyl-9H-carbazole (1.66 g, 0.51 mmol), copper iodide (0.49 g, 0.25 mmol), 1,10-phenanthroline (0.46 g, 0.25 mmol) and potassium carbonate (1.78 g, 1.29 mmol) were dissolved in 15 ml of DMF and refluxed for 10 h with stirring. The reaction mixture was extracted with methylene chloride/distilled water after the reaction was cooled down, and obtained organic phase was dehydrated using magnesium sulfate. A white solid was obtained by purification with column chromatography with an eluent of methylene chloride and hexane, and further purified using vacuum sublimation.

Yield 45.4% (1.15 g), T_g 137.5 °C ^1H NMR (500 MHz, $\text{DMSO-}d_6$): δ

8.82 (s, 2H), 8.59–8.56 (m, 2H), 8.43 (d, 1H, $J = 7.5$ Hz), 8.36 (d, 1H, $J = 8.0$ Hz), 8.11 (d, 1H, $J = 8.0$ Hz), 8.04 (d, 1H, $J = 9.0$ Hz), 7.96 (d, 1H, $J = 8.5$ Hz), 7.92 (d, 1H, $J = 8.5$ Hz), 7.74–7.73 (m, 4H), 7.63–7.44 (m, 9H), 7.40 (d, 1H, $J = 8.5$ Hz), 7.36–7.32 (m, 2H). ^{13}C NMR (125 MHz, $\text{DMSO-}d_6$): δ 141.6, 140.9, 140.7, 139.3, 139.1, 137.8, 136.9, 136.6, 135.8, 135.2, 131.9, 130.3, 129.2, 127.9, 127.1, 126.9, 126.8, 126.4, 126.1, 125.5, 125.3, 124.7, 123.9, 123.3, 123.3, 123.1, 122.7, 122.4, 122.4, 121.2, 120.8, 120.4, 119.9, 119.8, 119.5, 118.9, 111.0, 110.1, 109.9, 109.8. MS (FAB-HR) m/z 590.1815 $[(M + H)^+]$.

2.5. 3-(Dibenzo[b,d]thiophen-2-yl)-9-phenyl-9H-carbazole (CzThPh)

Using the same synthetic procedure of CzThCz, iodobenzene (1.05 g, 0.51 mmol) was used as reagent instead of 3-bromo-9-phenyl-9H-carbazole. A white product was obtained after vacuum train sublimation.

Yield 68.4% (1.25 g), T_g 77.6 °C ^1H NMR (500 MHz, $\text{DMSO-}d_6$): δ 8.80 (d, 2H, $J = 10.0$ Hz), 8.56 (d, 1H, $J = 9.0$ Hz), 8.40 (d, 1H, $J = 8.0$ Hz), 8.11 (d, 1H, $J = 8.5$ Hz), 8.04 (d, 1H, $J = 9.0$ Hz), 7.95 (d, 1H, $J = 8.5$ Hz), 7.92 (d, 1H, $J = 9.0$ Hz), 7.72–7.65 (m, 4H), 7.57–7.45 (m, 5H), 7.41 (d, 1H, $J = 8.0$ Hz), 7.34 (t, 1H, $J = 7.75$ Hz). ^{13}C NMR (125 MHz, $\text{DMSO-}d_6$): δ 140.6, 139.6, 139.1, 137.7, 136.9, 136.8, 135.8, 135.2, 132.2, 130.2, 127.7, 127.1, 126.6, 126.5, 126.1, 125.5, 124.7, 123.5, 123.3, 123.1, 122.9, 122.4, 120.8, 120.2, 119.8, 118.9, 110.0, 109.7. MS (FAB-HR) m/z 425.1235 $[(M + H)^+]$.

2.6. 3,9-Bis(dibenzo[b,d]thiophen-2-yl)-9H-carbazole (CzThTh)

Using the same synthetic procedure with CzThCz, 2-bromodibenzo[b,d]thiophene (1.36 g, 0.51 mmol) was used as reagent instead of 3-bromo-9-phenyl-9H-carbazole. A white product was obtained after vacuum train sublimation.

Yield 50.0% (1.14 g), T_g 124.2 °C, ^1H NMR (500 MHz, $\text{DMSO-}d_6$): δ 8.82 (s, 2H), 8.72 (s, 1H), 8.57 (d, 1H, $J = 9.0$ Hz), 8.48 (d, 1H, $J = 8.0$ Hz), 8.43 (d, 1H, $J = 7.5$ Hz), 8.33 (d, 1H, $J = 8.5$ Hz), 8.11 (d, 2H, $J = 8.5$ Hz), 8.04 (d, 1H, $J = 9.5$ Hz), 7.96 (d, 1H, $J = 8.5$ Hz), 7.92 (d, 1H, $J = 8.5$ Hz), 7.76 (d, 1H, $J = 8.5$ Hz), 7.59–7.45 (m, 7H), 7.36 (t, 1H, $J = 7.0$ Hz), ^{13}C NMR (125 MHz, $\text{DMSO-}d_6$): δ 141.2, 140.2,

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