



Bipolar host materials with carbazole and dipyrindylamine groups showing high triplet energy for blue phosphorescent organic light emitting diodes



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ABSTRACT

A series of new bipolar host materials with carbazole and different number of dipyrindyl amine groups were synthesized for blue phosphorescent organic light-emitting diodes (PHOLEDs). Especially, we applied dipyrindyl amine to control the polarity as well as electron transporting property of the carbazole based host materials for comparison with the properties of 1,3-di(9H-carbazol-9-yl)benzene (mCP). As a result, all the carbazole based host materials with dipyrindyl amine moieties showed very high triplet energies (>2.9 eV) as well as wide band gap (>3.5 eV). Thus, the blue phosphorescent devices employing those materials with bis(4,6-difluorophenyl pyridinato-*N,C2*)picolinato-iridium (III) (Flrpic) as a dopant and we obtained moderately high external quantum efficiency (EQE) up to 21.6% with *N*-(3,5-di(9H-carbazol-9-yl)phenyl)-*N*-(pyridin-2-yl)pyridin-2-amine (**DCPPy**).

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1. Introduction

Organic light emitting diodes (OLEDs) have received considerable attention due to their superior device properties which are perfectly suitable for large area displays and solid-state lightings. However, OLEDs are still struggling against liquid crystal displays (LCDs) due to a short lifetime as well as a low efficiency of blue emitting materials. Hence, materials for blue phosphorescent organic light-emitting diodes (PHOLEDs) has become very important to overcome such critical issues [1–10]. Nevertheless, it is still being required to understand premise condition for materials design of blue phosphorescent emitters because there has been no great success to synthesize the materials showing moderately long lifetime behavior for last decade. Indeed many research groups are focusing on the development into a suitable host materials for blue PHOLEDs. Especially, they are trying their best to avoid side effects such as triplet-triplet annihilation (TTA), triplet-polaron annihilation (TPA), and many other types of self-quenching processes

[11,12].

In principle, blue host materials are required to have a large band gap (E_g) above 3.5 eV as well as a large triplet energy (T_1) level greater than 2.9 eV to prevent the back energy transfer from dopant to host materials (T_1 of dopant > 2.7 eV) [13,14]. Besides, they should have moderate hole as well as electron charge transport behavior and thermal stability for stable device performance [15–20]. The most common approach is to use a carbazole core moiety and an electron accepting heteroaromatic ring moiety with limited conjugation length to increase the band gap as well as T_1 level for blue phosphorescent devices [21,22]. At the beginning, the most representative material with carbazole groups was 1,3-di(9H-carbazol-9-yl)benzene (mCP) reported by Forrest [23]. Especially, a mCP was a very suitable host material for blue PHOLEDs because it has moderately high triplet energy and large band gap about 2.9 eV and 3.5 eV, respectively. Unfortunately, the devices prepared by using this material show very short lifetime presumably due to its low glass transition temperature (T_g) (about 60 °C) which may cause molecular aggregation or crystallization from joule heating of the devices [24]. To overcome such an issue, it is necessary to synthesize new host materials with increased T_g for blue phosphorescent devices.

In this paper, we report three different types of bipolar host

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materials with high triplet energy ($T_1 > 2.9$ eV) by connecting carbazole and different number of dipyridyl amine groups to the core phenyl linker for improvement of their hole as well as electron transport properties. As a result, we obtained moderately high external quantum efficiency (EQE) up to 21.6% with *N*-(3,5-di(9*H*-carbazol-9-yl)phenyl)-*N*-(pyridin-2-yl)pyridin-2-amine (DCPPy) device.

2. Experimental

2.1. Synthesis of new host material

2.1.1. Instruments

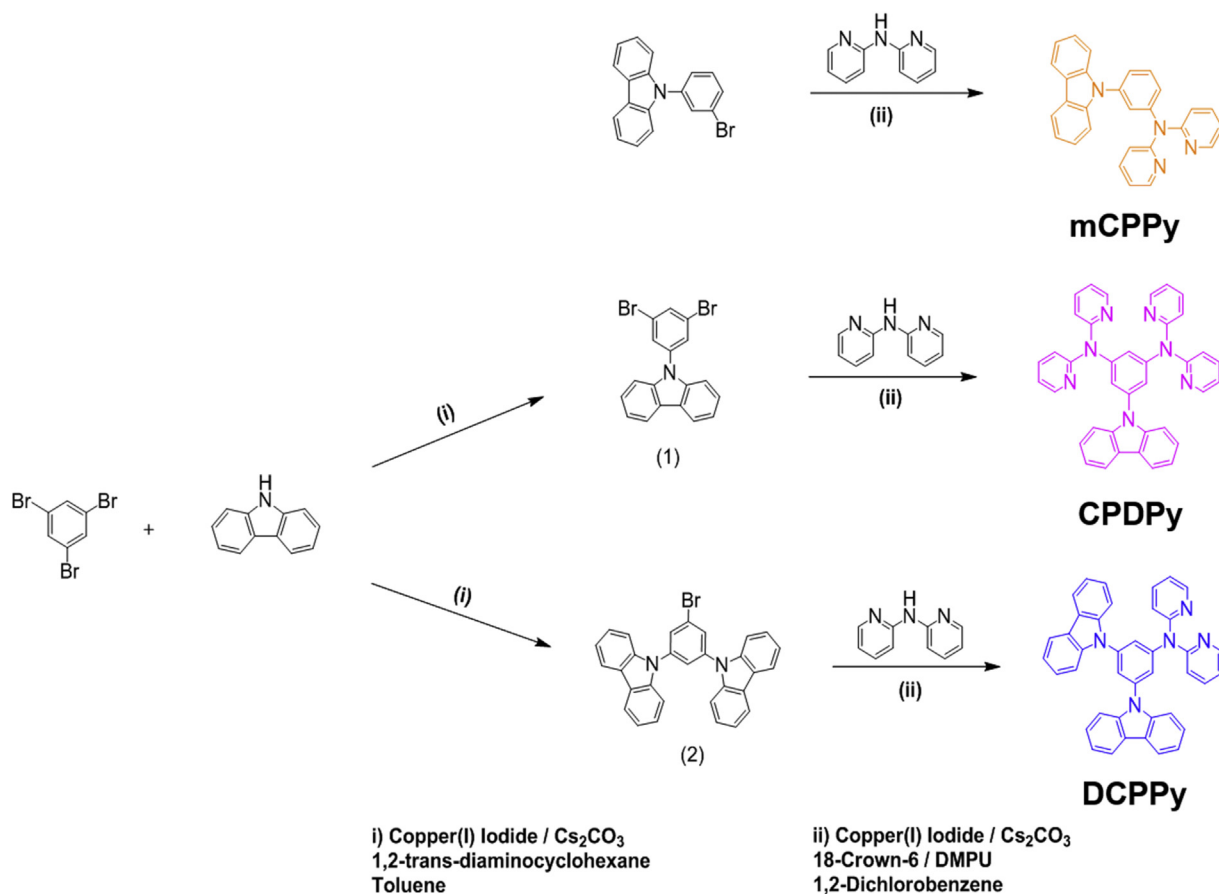
^1H NMR and ^{13}C NMR spectra were recorded on a Bruker AVANCE 400FT-NMR(400 Hz). The absorption spectra were obtained by ultraviolet–visible (UV–vis) spectrophotometer (HEWLETT PACKARD 8453 UV–vis spectrophotometer). Photoluminescence (PL) spectra were collected by PERKIN ELMER LS 50B luminescence spectrometer. The differential scanning calorimetry (DSC) was performed on TA instruments and thermogravimetric analysis (TGA) using TA INSTRUMENT TA4100. High resolution mass spectra were recorded using a HR-ESI MS SYNAPU G2 (Waters, U.K.) HOMO (Highest Occupied Molecular Orbital) was measured with a photoelectron spectrometer (AC2, Hitachi High Tech).

2.1.2. Synthetic details

2.1.2.1. Synthesis of *N*-(3-(9*H*-carbazol-9-yl)phenyl)-*N*-(pyridin-2-yl)pyridin-2-amine (mCPPy). In a Schlenk flask, 9-(3-bromophenyl)-

9*H*-carbazole (5 g, 10.3 mmol), 2,2'-dipyridylamine (1.9 g, 11.3 mmol), copper(I) iodide (0.2 g, 1.0 mmol), 18-Crown-6 (0.09 g, 0.34 mmol), cesium carbonate (7.2 g, 20.5 mmol) was dissolved in 1,2-dichlorobenzene (72 ml). And 1,3-Dimethyl-3,4,5,6-tetrahydro-2(1*H*)-pyrimidinone (DMPU) (0.25 mL) was put into reactor, then keep heating at 170 °C for 18 h under nitrogen. After cooling to room temperature, the mixture was poured into a cold hexane. The precipitate was filtered and washed with hexane. The solid was purified with column chromatography [n-Hexane: Ethylacetate = 1: 5 (vol.: vol.)] to give yellowish powder in 38% of yield. After primary purification, *N*-(3-(9*H*-carbazol-9-yl)phenyl)-*N*-(pyridin-2-yl)pyridin-2-amine (mCPPy) was obtained as a white powder by train sublimation. (m.p. = 132 °C) ^1H NMR (400 MHz, CD_2Cl_2) δ (ppm) 8.35 (s, 2H) 8.10–8.12 (d, $J = 8.0$, 2H) 7.62–7.65 (t, $J = 8.0$, 2H) 7.56–7.60 (t, $J = 8.0$, 1H) 7.50–7.52 (d, $J = 8.0$, 2H) 7.39–7.43 (t, $J = 8.0$, 3H) 7.36 (s, 1H) 7.25–7.29 (t, $J = 8.0$, 2H) 7.21–7.23 (d, $J = 8.0$, 1H) 7.11–7.12 (d, $J = 8.0$, 2H) 7.00–7.02 (d, $J = 8.0$, 1H) 6.99–7.00 (d, $J = 8.0$, 1H) ^{13}C NMR (400 MHz, CD_2Cl_2) 117.7, 119.1, 120.4, 120.5, 123.1, 123.7, 125.21, 125.25, 126.3, 130.8, 138.0, 138.8, 140.8, 147.1, 148.9, 158.3 ppm $[\text{M}+\text{H}]^+$: m/z calcd for $\text{C}_{28}\text{H}_{21}\text{N}_4$, 413.1766 found 413.1768.

2.1.2.2. Synthesis of 9,9'-(5-bromo-1,3-phenylene)bis(9*H*-carbazole) (1). In a flask a mixture of 1,3,5-tribromobenzene (10 g, 31.8 mmol), carbazole (10.7 g, 64.2 mmol), copper(I) iodide (2.4 g, 12.7 mmol), Cesium carbonate (16.8 g, 47.7 mmol) in Toluene (96 ml) was stirred. And trans-1,2-diaminocyclohexane (1.9 ml, 15.9 mmol) was put into reactor, then keep it heating at 80 °C under N_2 atmosphere for 24 h. The resulting mixture was cooled to room temperature and



Scheme 1. Synthesis of new blue host materials.

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