



A strong hole transport type host material for high quantum efficiency blue phosphorescent organic light-emitting diodes



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ABSTRACT

A strong hole transport type host material derived from bicarbazole and ditolylamine, 4,4'-(9H,9'H-[2,3'-bicarbazole]-9,9'-diyl)bis(N,N-di-p-tolylaniline) (BCTA), was synthesized as the host material of blue emitting tris[1-(2,4-diisopropylidibenzo[b,d]furan-3-yl)-2-phenylimidazole] (Ir(dbi)₃) triplet emitter. The strong hole transport character of BCTA compensated hole trapping effect of Ir(dbi)₃ and optimized charge balance in the emitting layer, delivering 17.8% external quantum efficiency in the blue phosphorescent organic light-emitting diodes.

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Introduction

In general, phosphorescent organic light-emitting diodes (PHOLEDs) have host and dopant materials in the emitting layer and the two materials play a decisive role of achieving high external quantum efficiency (EQE) [1,2]. Dopant materials exhibiting high photoluminescence (PL) quantum yield can increase the EQE of the PHOLEDs and host materials harvesting triplet excitons without quenching or loss process can enhance the EQE of the PHOLEDs.

There are several kinds of phosphorescent dopant materials and one of the most popular dopant materials is imidazole derived Ir dopant material [3–6]. The imidazole derived dopant material is particularly useful in blue PHOLEDs because less aromatic and electron deficient character of the imidazole moiety can increase the triplet energy of the Ir triplet emitter. Furthermore, the imidazole type blue triplet emitters are more stable than other blue triplet emitters, and can extend the lifetime of the blue PHOLEDs [7–10]. Therefore, the imidazole type triplet emitters are being widely used as blue triplet emitters in the blue PHOLEDs.

Although the imidazole based triplet emitters have the merits of blue emission and stable operation, they have the highest occupied molecular orbital (HOMO) and the lowest unoccupied

molecular orbital (LUMO) much shallower than those of conventional high triplet energy host materials, which makes it difficult to develop host materials for the imidazole based triplet emitters [11,12]. In order to align the HOMO/LUMO levels of the host materials with those of the imidazole type dopant materials, the host materials should be designed to have very shallow HOMO/LUMO levels. However, common host materials cannot satisfy the HOMO/LUMO requirement, which motivated us to design and synthesize high triplet energy host materials with the shallow HOMO/LUMO levels.

In this paper, we report a strong hole transport type host material, 4,4'-(9H,9'H-[2,3'-bicarbazole]-9,9'-diyl)bis(N,N-di-p-tolylaniline) (BCTA), designed for a tris[1-(2,4-diisopropylidibenzo[b,d]furan-3-yl)-2-phenylimidazole] (Ir(dbi)₃) blue triplet emitter. The BCTA host material was evaluated in the blue PHOLEDs doped with the Ir(dbi)₃ emitter, and demonstrated high quantum efficiency of 17.8%.

Experimental

General information

2-Bromo-9H-carbazole, cesium carbonate, copper iodide and 1,10-phenanthroline purchased from Sigma Aldrich. Co., were used without further purification. 3-(4,4,5,5-tetramethyl-1,3,2-dioxabolan-2-yl)-9H-carbazole and tetrakis(triphenylphosphine)palladium(0) purchased from P&H tech. Co., was used without further

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purification. Tetrahydrofuran, dimethylformamide and potassium carbonate were products of Duksan Sci. Co.

Synthesis

9H,9'H-2,3'-bicarbazole

2-Bromo-9-carbazole (1 g, 4.06 mmol) and 3-(4,4,5,5-tetramethyl-1,3,2-dioxaboroan-2-yl)-9H-carbazole (1.43 g, 4.87 mmol) were dissolved in 200 ml of tetrahydrofuran and the solution was stirred under Ar followed by addition of 2 M potassium carbonate (2.76 g) solution in 100 ml of distilled water. Tetrakis(triphenylphosphine)palladium(0) (0.14 g, 0.12 mmol) was added and the mixture was refluxed for 12 h. After cooling, the mixture was extracted with methylene chloride and distilled water twice. The organic material was separated and dried over magnesium sulfate and purified by column chromatography to yield 9H,9'H-2,3'-bicarbazole (0.5 g, 38%) as a white solid.

$^1\text{H NMR}$ (400 MHz, DMSO): δ = 11.26 (s, 1H), 11.22 (s, 1H), 8.47 (s, 1H), 8.21 (d, 1H, J = 7.2 Hz), 8.15 (d, 1H, J = 8 Hz), 8.09 (d, 1H, J = 7.2 Hz), 7.67–7.56 (m, 2H), 7.57–7.35 (m, 6H), 7.17–7.12 (m, 2H).

4,4'-(9H,9'H-[2,3'-bicarbazole]-9,9'-diyl)bis(N,N-di-p-tolylaniline) (BCTA)

9H,9'H-2,3'-bicarbazole (0.3 g, 0.90 mmol), 4-bromo-N,N-di-p-tolylaniline (0.9 g, 2.70 mmol), copper iodate (0.09 g, 0.45 mmol), cesium carbonate (1.2 g, 3.61 mmol) and 1,10-phenanthroline (0.25 g, 1.35 mmol) were dissolved in 30 ml of dimethyl formamide and refluxed with N_2 gas for 12 h. After cooling, the mixture was extracted with methylene chloride and distilled water twice. The organic material was separated and dried over magnesium sulfate and purified by column chromatography to yield BCTA (0.2 g, 32%) as a white solid.

$^1\text{H NMR}$ (400 MHz, DMSO): δ = 8.56 (s, 1H), 8.33 (t, 2H, J = 7.6 Hz), 8.25 (d, 1H, J = 8 Hz), 7.74 (d, 1H, J = 8.8 Hz), 7.68 (d, 1H, J = 7.6 Hz), 7.64 (s, 1H), 7.51 (d, 1H, J = 8 Hz), 7.45–7.37 (m, 7H), 7.30–7.26 (m, 2H), 7.16 (t, 8H, J = 8.2 Hz), 7.10–7.05 (m, 12H), 2.28 (s, 6H), 2.26 (s, 6H).

$^{13}\text{C NMR}$ (125 MHz, CDCl_3): δ = 147.07, 144.38, 141.08, 140.93, 139.82, 193.50, 133.01, 132.97, 130.24, 127.66, 127.52, 125.08, 124.95, 121.79, 121.56, 110.14, 20.42.

Device fabrication and measurements

The blue PHOLEDs possessing the BCTA and $\text{Ir}(\text{dbi})_3$ had a device structure of indium tin oxide (ITO) (120 nm)/PEDOT:PSS

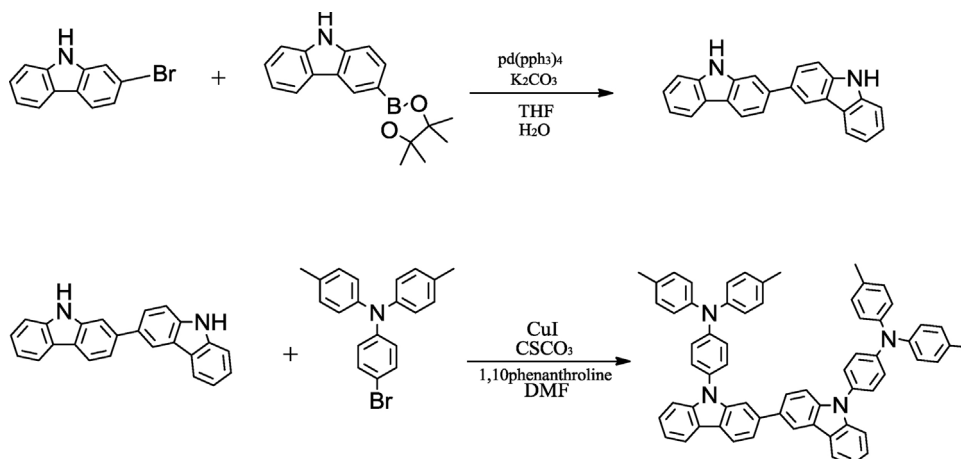
(60 nm)/TAPC (20 nm)/mCP (10 nm)/BCTA: $\text{Ir}(\text{dbi})_3$ (25 nm)/TSPO1 (5 nm)/TPBI (30 nm)/LiF (1 nm)/Al (200 nm). PEDOT:PSS is poly(3,4-ethylenedioxythiophene):polystyrenesulfonate and is a hole injection material. TAPC and mCP are 4,4'-cyclohexylidene-bis[N,N-bis(4-methylphenyl)aniline] and N,N'-dicarbazolyl-1,3-benzene, respectively and are hole transport materials. TSPO1 and TPBI are diphenylphosphine oxide-4-(triphenylsilyl)-phenyl and 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole), and are electron transport materials. Three devices with different $\text{Ir}(\text{dbi})_3$ doping concentrations from 5% to 15% were fabricated to test the BCTA host materials. Detailed device preparation methods were reported in other work. Luminance (L), current density (J) and voltage (V) data were gathered by sweeping the voltage using encapsulated devices. Light-emitting characteristics were measured using CS 2000 spectroradiometer and electrical characteristics were measured using Keithley 2400 source meter.

Results and discussion

The BCTA host material with a bicarbazole backbone structure and a ditolylamine substituent was designed to shift the HOMO and LUMO levels close to those of $\text{Ir}(\text{dbi})_3$ which have the HOMO/LUMO levels of -5.76 eV/ -2.47 eV [3]. The bicarbazole backbone structure is a highly electron rich moiety shallowing the HOMO/LUMO levels and addition of the ditolylamine substituent to the bicarbazole backbone further renders the molecule to show intensified electron rich character. Therefore, the BCTA host material can be suitable as the host of $\text{Ir}(\text{dbi})_3$.

The synthesis of BCTA was simply carried out by CuI mediated coupling reaction between 9H,9'H-2,3'-bicarbazole and 4-bromo-N,N-di-p-tolylaniline at a synthetic yield of 32% as described in Scheme 1. A white powder was produced from the reaction after purification using a vacuum train sublimator. Identification of the produced compound was performed using ^1H nuclear magnetic resonance (NMR) spectrometer, ^{13}C NMR and mass spectrometer. Purity level of purified BCTA was above 99% by analysing the compound with high performance liquid chromatography.

Main design strategy of BCTA was to control the HOMO/LUMO levels of the host to be aligned with those of $\text{Ir}(\text{dbi})_3$, so the HOMO and LUMO of BCTA were investigated by molecular orbital calculation. A basis set of B3LYP 6-31G implemented in Gaussian 09 software was used to calculate the molecular orbital. The HOMO and LUMO of BCTA are described in Fig. 1. The HOMO localization on ditolylamine and LUMO localization on bicarbazole were observed as can be anticipated from electron rich character of bicarbazole and ditolylamine. The HOMO distribution



Scheme 1. Synthetic scheme of BCTA.

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