

Synthesis of a dibenzothiophene/carboline/carbazole hybrid bipolar host material for green phosphorescent OLEDs



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ABSTRACT

A bipolar host material for green phosphorescent organic light-emitting diodes (OLEDs) was designed and synthesized through a combination of dibenzothiophene, α -carboline, and carbazole (DTCC) moieties. The synthesized host material showed a sufficient HOMO/LUMO bandgap (3.49 eV) and triplet energy (2.67 eV) for green emitting bis[2-(2-pyridinyl-N)phenyl-C] (acetylacetonato) iridium(III) [Ir(ppy)₂(acac)]. From the results of a single charge device for DTCC, the hole current density of DTCC was similar to the electron current density, indicating that DTCC possesses bipolar charge transport properties, confirming its bipolar nature and thus its applicability as the host of PHOLED. Thus, the DTCC host showed efficient energy transfer to the Ir(ppy)₂(acac) dopant in the device. A maximum external quantum efficiency of 18.9% was obtained using DTCC as the host material and the color coordinate of the green PHOLED was (CIE_{x,y} = 0.34, 0.62) at 10% doping concentration.

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1. Introduction

For more than ten years, phosphorescent organic light-emitting diodes (PHOLEDs) have continued to attract intense interest because they effectively harvest electro-generated singlet and triplet excitons to accomplish an internal quantum efficiency of close to 100%, which is considerably superior to the 25% upper limit imposed by the formation of singlet excitons in fluorescence [1,2]. To realize highly efficient PHOLEDs by reducing competitive factors such as concentration quenching and triplet-triplet annihilation, phosphorescent emitters of heavy-metal complexes, the PHOLEDs are usually doped in a suitable host material [3]. Therefore, the synthesis of host materials and dopants is very important for the formation of efficient PHOLEDs. A good host material for a PHOLED emission must have a high triplet energy [4–9], a sufficiently large bandgap for effective energy transfer to the guest, good carrier transport properties for a balanced recombination of carriers in the emitting layer, and energy-level matching with neighboring layers for effective charge injection [10,11]. Besides these electrical properties, good host materials must be morphologically stable during fabrication and operation [12,13]. However, designing a good host material having higher triplet energy levels than those of

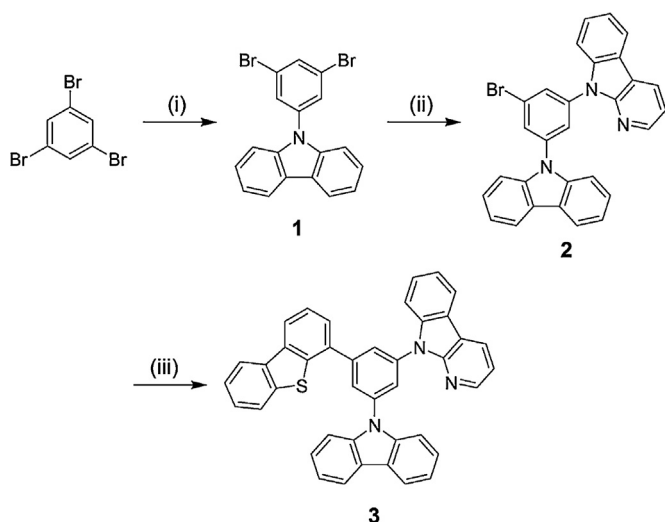
the phosphorescent dopant remains a challenge for materials researchers.

Recently, PHOLED-related research has significantly shifted towards the development of host materials possessing bipolar properties because they can provide more balance in electron and hole fluxes, simplifying the device structure [14]. Up to now, many high triplet energy materials have been synthesized as host materials for PHOLEDs [15]. In general, high triplet moieties such as carbazole [16–19], arylsilane [20,21], fluorene [22], dibenzofuran [23,24], and dibenzothiophene [25–27] have been used in molecular structures as core structures to obtain high triplet energy. Hole transport type cores have been combined with electron transport groups, while electron transport type cores have been modified with hole transport units for bipolar charge transport properties. Carbazole is the most widely used hole transport core for triplet host materials because of its high triplet energy of 3.02 eV; carbazole was also substituted with various electron transport units such as diphenylphosphine oxide [17], oxadiazole [18], and triazole [19] to balance the hole and electron transport properties.

In this work, we report the synthesis, characterization, and PHOLED applications of a new bipolar host material comprising three different chemical units covalently linked to the C1, C3, and C5 position of the benzene core. The carbazole are widely used as hole transporting moieties and α -carboline has an electron transporting characteristic. Furthermore, the dibenzothiophene was usually reported as the electron-donating core structure, but it

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Scheme 1. Reagents and conditions: (i) Carbazole, CuI, K_2CO_3 , 1,10-phenanthroline, DMF, $160^\circ C$ (ii) α -Carboline, CuI, K_3PO_4 , *trans*-1,2-diaminohexane, 1,4-dioxane, $110^\circ C$; (iii) DBT, K_2CO_3 , $Pd(PPh_3)_4$, toluene, reflux.

has been reported that the electron withdrawing sulfur group could make the aromatic ring electron deficient [28–31]. Thus it could be made to act as an electron transporting unit by hybridization with strong electron-rich moiety such as carbazole moiety. For this reason, the dibenzothiophene, α -carboline, and carbazole units were selected to demonstrate that the hybrid host material was suitable to apply to green phosphorescent OLEDs, showing good external quantum efficiency.

2. Experimental

2.1. General procedure

Chemicals were commercially purchased and used without further purification. Thin layer chromatography (TLC) was conducted on flexible sheets precoated SiO_2 (Baker-flex; IB2-F) and visualized by UV light. Column chromatography was conducted by using silica gel (60–200 mesh; Fisher Scientific Co.). Melting point was determined on melting point apparatus (MEL-TEMP; model 1102D). 1H and ^{13}C NMR spectra were recorded on JEOL JNM-LA400 FT NMR spectrometer using $CDCl_3$, as the solvent, except where noted. Mass spectra were obtained on LC/mass spectrometer (Agilent Technology 6120). Photophysical properties were analyzed by using UV/vis spectrometer (Scinco Co., S-3100) and

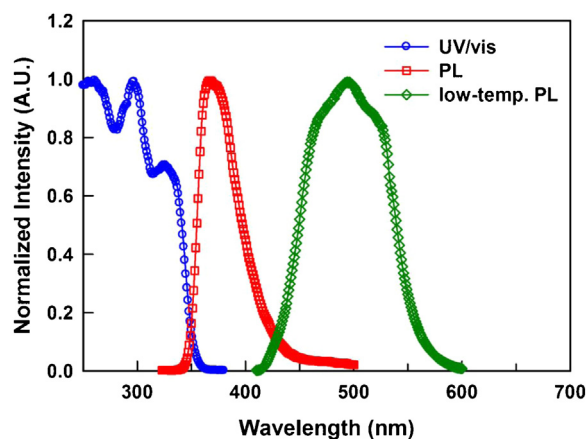


Fig. 2. UV/vis absorption, solution PL, and low-temperature PL spectra of DTCC.

photoluminescence (PL) spectrometer (Hitachi High Technologies; F-7000). The host material was dissolved in THF and 2-methoxyethanol at a concentration of ca. $1.0 \times 10^{-4} M$ for UV/vis and PL measurements, respectively. Triplet energy of the host material was calculated from low temperature PL spectrum measured in the liquid nitrogen (77 K) condition. Cyclic voltammetry (CV) measurement was conducted by using potentiostat (SP-50; Bio-Logic Science Instruments) in electrolyte solution, which is CH_3CN with tetrabutylammonium perchlorate at 0.1 M concentration. Ag was used as the reference electrode and Pt was the counter electrode. The host material was coated on indium-tin-oxide substrate and immersed in the electrolyte solution for measurement. Ferrocene was used as a standard material of the CV measurement. Glass transition temperature and thermal decomposition temperature were measured by using a differential scanning calorimeter (TA, DSC Q2000) and thermogravimetric analyzer (TA, TGA Q50) at a heating rate of $10^\circ C \text{ min}^{-1}$ under a nitrogen atmosphere, respectively.

2.2. Synthesis

2.2.1. 9-(3,5-Dibromophenyl)-9H-carbazole (1)

Carbazole (8.4 g, 50.3 mmol), 1,3,5-tribromobenzene (9.7 g, 30.8 mmol), K_2CO_3 (4.9 g, 35.5 mmol), copper(I) iodide (0.06 g, 0.3 mmol), and 1,10-phenanthroline (0.06 g, 3.3 mmol) were added to 100 mL of dried DMF. The mixture was heated at $160^\circ C$ for 12 h under nitrogen atmosphere. After this mixture cooled to room temperature, the reaction was quenched with 200 mL of H_2O and extracted with 100 mL of dichloromethane three times. Combining

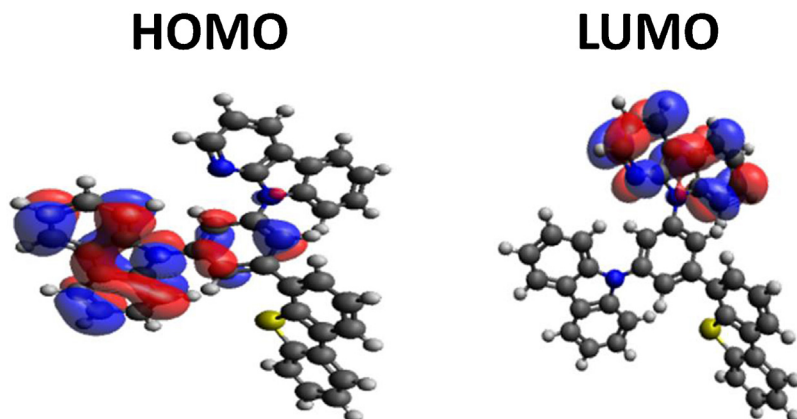


Fig. 1. DFT calculations of HOMO and LUMO distribution for DTCC.

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