



A facile way to synthesize high-triplet-energy hosts for blue phosphorescent organic light-emitting diodes with high glass transition temperature and low driving voltage



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ABSTRACT

Two novel bipolar host materials were designed and synthesized, by incorporating α -carboline and carbazole to spiro-acridine-fluorene skeleton. As expected, both materials exhibited high triplet energy over 2.8 eV, indicating they could act as suitable blue hosts in phosphorescent organic light-emitting diodes. By changing the appending group from carbazole to α -carboline, the thermal property and molecular bipolarity were improved. When they were applied as hosts for sky-blue emitter iridium(III) bis[4,6-difluorophenyl]pyridinato-N, C2'] picolinate (Flrpic), good performance of 35.5 cd A⁻¹, 35.8 lm W⁻¹ and 29.5 cd A⁻¹, 24.5 lm W⁻¹ was achieved for α -carboline derivative and carbazole derivative, respectively.

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1. Introduction

Phosphorescent organic light-emitting diodes (PHOLEDs) have made a big breakthrough on OLED technology due to the theoretical 100% internal quantum efficiency [1]. In recent years, considerable attentions have been paid to PHOLEDs, but the development of highly efficient PHOLEDs still remains a challenge, especially for blue PHOLEDs [2–4]. This is because the blue phosphors have the highest energies among Red–Green–Blue colors, and hence they need host materials with the widest energy gap to prevent possible energy back-transfer [5]. This host–guest (phosphor) system is indispensable for typical PHOLEDs [6,7]. By this way, the triplet excitations will not be self-quenched through triplet–triplet annihilation (TTA) process at high concentration [8]. Besides the aforementioned wide-energy-gap, the blue hosts also should possess good carrier transporting properties as well as high thermal and morphological stabilities [9].

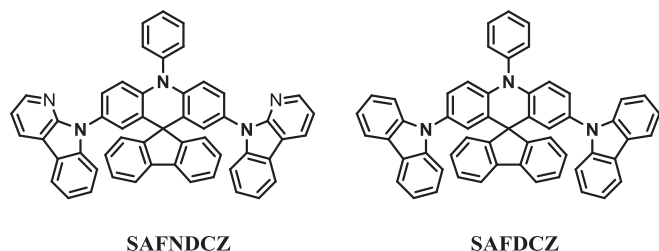
A cruciform spirocyclic aromatic system is an effective concept to improve thermal and morphological stabilities of organic materials [10]. For example, spiro-bifluorene (SF) and spiro-acridine-fluorene (SAF) are constructed and used as building blocks for exploring new host materials [11–16]. The latter SAF contains an electron-donating nitrogen atom and leads to its hole-transport property [17]. This advantage makes it more remarkable as compared to SF in recent research. However, the derivatives based on SAF are primarily from the fluorene part of the spiro-system to date; what's more, the derivatization must be carefully designed to avoid reducing the triplet energy of the resulted host in case it fails for blue emitters [18–21].

In this article, we dissected the SAF block and found that the acridine part would be another alternative to derivatize for wide-energy-gap blue hosts. In this regard, two new host materials named 2,7-di(9H-carbazol-9-yl)-10-phenyl-10H-spiro[acridine-9,9'-fluorene] (SAFDCZ), 10-phenyl-2, and 7-bis(9H-pyrido[2,3-b]indol-9-yl)-10H-spiro[acridine-9,9'-fluorene] (SAFNDCZ) were designed (Scheme 1), in which two carbazole groups or two α -carboline groups were appended on the para-positions of acridine moiety [22]. With this method, we could avoid the fussy steps to prepare the 3-halogenated fluorene and also get the high triplet energy host materials for blue dopants [11]. On the other hands, the

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Scheme 1. Chemical structures of **SAFNDCZ** and **SAFDCZ**.

change from carbazole to α -carboline will present their difference in material properties and performance. To prove whether the change is efficient, the thermal, photophysical and electrochemical properties of **SAFNDCZ** and **SAFDCZ** are fully investigated. And the device performance of these two hosts doped with sky-blue phosphor iridium(III) bis[4,6-difluorophenyl]pyridinato-N, C^{2'}]picolinate (Flrpic) in PHOLED is characterized.

2. Experimental

2.1. Chemicals and instruments

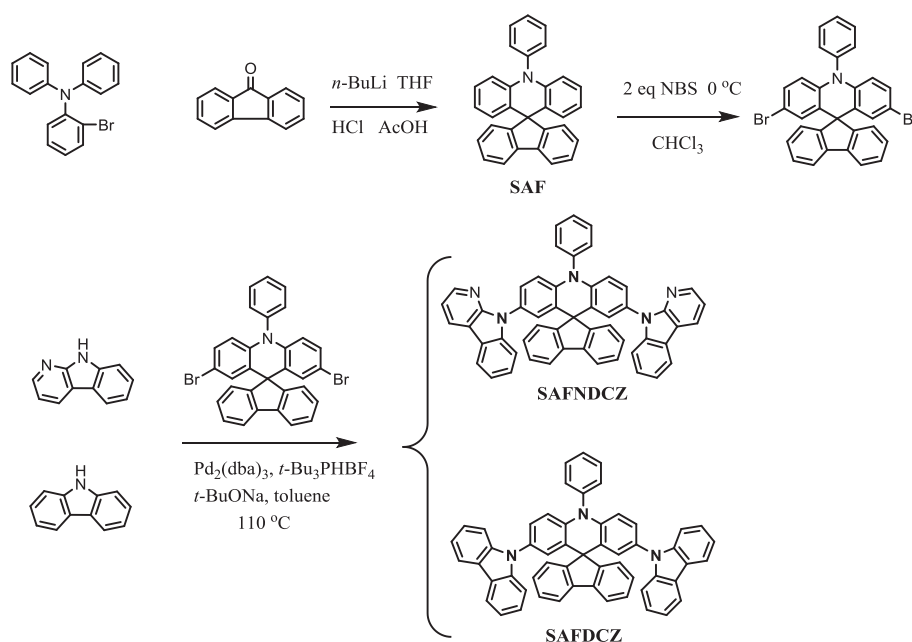
All chemicals and reagents were used as received from commercial sources without further purification. THF and toluene used in synthetic routes were purified by PURE SOLV (Innovative Technology) purification system. ¹H NMR and ¹³C NMR spectra were recorded on a Bruker 400 spectrometer at room temperature. Mass spectra were recorded on a Thermo ISQ mass spectrometer using a direct exposure probe. UV–vis absorption spectra were recorded on a Perkin Elmer Lambda 750 spectrophotometer. PL spectra and phosphorescent spectra were recorded on a Hitachi F-4600 fluorescence spectrophotometer. Differential scanning calorimetry (DSC) was performed on a TA DSC 2010 unit at a heating rate of 10 °C min⁻¹ under nitrogen. The glass transition temperatures (*T*_g) were determined from the

second heating scan. Thermogravimetric analysis (TGA) was performed on a TA SDT 2960 instrument at a heating rate of 10 °C min⁻¹ under nitrogen, temperature at 5% weight loss was used as the decomposition temperature (*T*_d).

Electrochemical measurements were made using a CHI600 voltammetric analyzer. A conventional three-electrode configuration consisting of a platinum working electrode, a Pt-wire counter electrode, and an Ag/AgCl reference electrode was used. The solvent in all measurements was CH₂Cl₂, and the supporting electrolyte was 0.1 M [Bu₄N]PF₆. Ferrocene was added as a calibrant after each set of measurements, and all potentials reported were quoted with reference to the ferrocene-ferrocenium (Fc/Fc⁺) couple at a scan rate of 100 mV s⁻¹. Theoretical calculations based on density functional theory (DFT) approach at the B3LYP level were performed with the use of Gaussian 09 program.

2.2. Preparation of 10-phenyl-10H-spiro(acridine-9,9-fluorene) (SAF)

2-Bromotriphenylamine (3.24 g, 10 mmol) was dissolved in 80 mL THF in a 200 mL Schlenk tube under argon. After the solution was cooled to -78 °C, *n*-butyl lithium (4.38 mL, 10.5 mmol) was added dropwise via a syringe. The resulting mixture was allowed to stir for 1 h at -78 °C, and then fluorene (1.8 g, 10 mL) in 80 mL THF was added over a period of 0.5 h. After 1 h reaction at -78 °C, the mixture was gradually warmed up to room temperature overnight. 5 mL water was added to the mixture and THF was evaporated under reduced pressure. The resulting solid was dissolved in 80 mL dichloromethane and washed with water (3 × 50 mL). Then the organic layer was separated, dried over sodium sulfate, filtered and evaporated, resulting in 4.51 g light yellow solid, which was directly used in the next reaction without further purification. The crude product was dissolved in 45 mL acetic acid and 4.5 mL hydrochloric acid (36%). After refluxed for 4 h under stirring, the reaction was cooled to room temperature, filtered and washed with petroleum ether. The resulting solid was further purified by column chromatography using petroleum ether as eluent to afford a white powder (3.52 g, 86.4%).



Scheme 2. Synthetic routes of **SAFNDCZ** and **SAFDCZ**.

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