



4, 6-Bis[3-(dibenzothiophen-2-yl)phenyl] pyrimidine bipolar host for bright, efficient and low efficiency roll-off phosphorescent organic light-emitting devices



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ABSTRACT

A bipolar host 4, 6-Bis[3-(dibenzothiophen-2-yl)phenyl] pyrimidine (DBTPhPm) with small singlet-triplet splitting has been synthesized and confirmed through a series of photophysical and electrochemical properties. Monochromatic phosphorescent organic light-emitting devices (PHOLEDs) based on different hosts [(4,4'-N,N'-dicarbazole) biphenyl, 2,7-bis (diphenylphosphoryl)-9-[4-(N,Ndiphenylamino) phenyl]-9-phenylfluorene, (3,3'-bicarbazole) phenyl and DBTPhPm] and dopants are fabricated. Compared to other hosts, the DBTPhPm-based PHOLEDs exhibited high brightness, high efficiency and low efficiency roll-off. The maximum power efficiency of the DBTPhPm-based red (R), green (G), blue (B), yellow (Y), and orange (O) PHOLEDs are 12.2, 47.2, 17.6, 42.6 and 15.1 lm/W, respectively. The current efficiency roll-off of the R, G, B, Y, and O PHOLEDs are 29.8%, 8.6%, 18.2%, 5.9%, and 22.4% from the maximum current efficiency to the high brightness of 5000 cd/m². The detailed working mechanism of the DBTPhPm-based device is discussed.

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1. Introduction

Phosphorescent organic light-emitting devices (PHOLEDs) have attracted much attention in recent years because their theoretical internal quantum efficiency can be, in principle, three times higher than that of the conventional fluorescent based organic light-emitting devices (OLEDs) through utilizing both singlet and triplet excitons for the light emission, and tremendous efforts have been made in the development of highly efficient PHOLEDs [1–7]. In PHOLEDs, phosphorescent materials are often doped into a suitable host material, which plays an indispensable role in energy transfer and carrier transport, in order to reduce aggregation quenching and triplet-triplet annihilation of guest triplet emitters [8,9]. Therefore, the development of high-performance host materials is extremely essential for PHOLEDs. As an efficient host material, HOMO-LUMO (HOMO: highest occupied molecular

orbital; LUMO: lowest unoccupied molecular orbital) energy band gap (E_g) and triplet energy level (T_1) of the host material must be higher than those of the guest to facilitate energy transfer from the host to guest and to prohibit reverse energy transfer from the guest back to host, and the energy level of the host material must be matched with neighboring layers for both effective carrier injection and carrier confinement [6,10–13]. Besides, a good carrier transport property is expected to restrict the recombination of carriers in the emitting layer (EML) and thus reduce the efficiency roll-off. Traditional host materials usually have good transporting properties for only a single type of carriers. For instance, N, N'-dicarbazolyl-3,5-benzene (mCP) exhibits only good hole-transporting properties, while 3-(4-biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole (TAZ) possesses good electron-transporting abilities [6,11,14,15]. These unbalanced carrier-transporting properties are negative to the turn-on voltage and the efficiency roll-off [11,15–17]. Hence, bipolar host materials that can balance carrier-transporting properties become the focus of attention recently [18–28]. Hsu et al. reported a bipolar host material 2,7-bis (diphenylphosphoryl)-9-[4-(N,Ndiphenylamino) phenyl]-9-

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phenylfluorene (POAPF), and the corresponding blue monochromatic OLEDs own a rather low turn-on voltage of 2.5 V due to the balance of carriers in the EML [24]. Zheng et al. applied a series of new synthesis host materials to blue monochromatic OLEDs, and obtained a low voltage of 2.6 V and a gentle efficiency roll-off. The device using 9,9-bis(9-methylcarbazol-3-yl)-4,5-diazafluorene (MCAF) as the host has a maximum current efficiency (CE) of 32.2 cd/A and a maximum power efficiency (PE) of 31.3 lm/W, and still keep a high CE of 27.6 cd/A and a high PE of 14.5 lm/W at the brightness of 10000 cd/m² [29]. Furthermore, Son et al. achieved bipolar host materials 4-(*N*-a-carbolinyl)-4'-(*N*-carbazolyl) triphenylamine (ADCTA), 4,4'-di(*N*-a-carbolinyl)-4''-(*N*-carbazolyl) triphenylamine (DACTA) and 4,4',4''-(*N*-acarbolinyl) triphenylamine (TATA) with the nature of small singlet-triplet energy gap (ΔE_{S-T} : the difference between singlet and triplet energy levels) so that the dopant may avoid being deep trap sites due to the small singlet to triplet splitting energy (0.4 eV) [30,31].

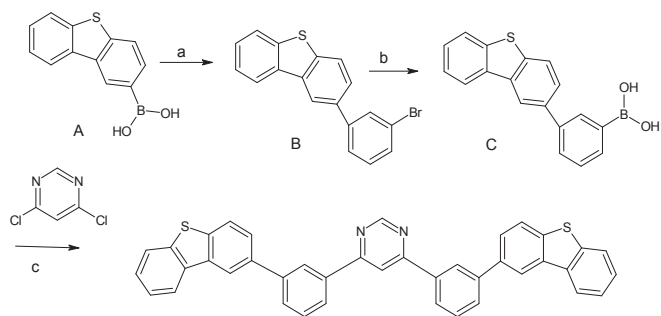
In this paper, a bipolar host material 4,6-Bis[3-(dibenzothiophen-2-yl)phenyl] pyrimidine (DBTPhPm) is reported and a series of low-voltage, highly efficient, and low efficiency roll-off DBTPhPm-based PHOLEDs are designed. Iridium (III) Bis [1-(3,5-dimethylphenyl)-7-methylisoquinoline] (acetylacetonate) [Ir(pmi-q)₂(acac)], tris(2-phenylpyridine) iridium [Ir(ppy)₃], [(bis[2-(4,6-difluorophenyl)pyridyl-N,C^{2'}]] iridium (III) [Firpic], Iridium(III)bis-(2-methyldibenzo-[f,h] quinoxaline) (acetylacetonate) [Ir(MD-Q)₂(acac)] and Iridium(III) bis(4-phenylthieno [3,2-c] pyridinato-N,C^{2'}) acetylacetonate [PO-01] are used as the red, green, blue, orange, and yellow dopants. For comparison, device using (4,4'-*N,N'*-dicarbazole) biphenyl (CBP), POAPF and (3,3'-bicarbazole) phenyl (BCZph) [32] as host were also fabricated. The results indicated that DBTPhPm is an efficient host for most common phosphorescent dopants.

2. Experimental

UV–visible absorption and photoluminescence (PL) studies at room temperature were carried out using U3010 spectrometer (Hitachi, Japan) and F-7000 FL spectrophotometer, respectively. The phosphorescence spectrum was recorded from the delayed emission of DBTPhPm at 77 K. The cyclic voltammogram (CV) experiments were performed using a BAS 100 W instrument at room temperature in CH₂Cl₂ solutions at a scan rate of 100 mV/s. All devices were fabricated on glasses substrates covered by conducting indium tin oxide (ITO). The substrates were cleaned in Decon 90 and deionized water, dried in the oven and then treated in plasma for about 5 min. Finally organic layers and cathode materials were sequentially deposited on the substrates without breaking vacuum ($\sim 5.0 \times 10^{-4}$ Pa). A shadow mask was used to define the cathode and to make four 10 mm² devices on each substrate. Current-Voltage-Luminance (I–V–L) characteristics of unpackaged devices were measured with a Keithley 2400 Source Meter and a Minolta Luminance Meter LS-110. The spectra of the devices were measured with Ocean Optics Maya 2000-PRO spectrometer.

3. Results and discussion

Scheme 1 depicts the synthetic route and structure of the new host material DBTPhPm. Dibenzothiophene-2-boronic acid was purchased from Bepfarm Chemical (China). The boronic acid C was synthesized according to literature [33]. Other reactants or reagents were used as received. 10 g (67 mmol) 4,6-dichloropyrimidine, 60 g (20 mmol) 3-(dibenzothiophen-2-yl) phenylboronic acid, 80 g (70 mmol) K₂CO₃, 500 ml toluene and 200 ml H₂O were put in a 1 L recovery flask. While the pressure was reduced, the mixture was



Scheme 1. Synthetic routes to DBTPhPm. (a) toluene, H₂O, K₂CO₃, PdCl₂(PPh₃)₂; (b) THF, -78 °C, n-BuLi, B(OPr)₃; (c) toluene, H₂O, K₂CO₃, PdCl₂(PPh₃)₂.

stirred to be degassed. Then 0.5 g PdCl₂(PPh₃)₂ was added in this mixture and the atmosphere was replaced with N₂. The mixture was stirred while reaction container was heated. After heating, water was added to the mixture, and the mixture was filtered to give residue. The obtained solid was washed with dichloromethane and methanol. The obtained solid was recrystallized from toluene to give 25 g white solid (yield 63%). ¹H NMR(CDCl₃, 400 MHz): 7.41–7.51 (m, 4H), 7.58–7.62 (m, 4H), 7.68–7.79 (m, 4H), 8.73 (dt, 2H), 8.18–8.27 (m, 7H), 8.54 (t, 2H), 9.39 (d, 1H). MODI-TOF: 596.76. Anal. calcd for C₄₀H₂₄N₂S₂ (%): C 80.51, H 4.05, N 4.69, S 10.75; Found: C 80.50, H 4.05, N 4.71, S 10.74.

DBTPhPm has a simple molecular structure that having two dibenzothiophene and one pyrimidine moiety. Pyrimidine core structure was designed as the electron transport type core structure with high triplet energy and high rigidity for high glass transition temperature ($T_g = 268.9$ °C). The T_g of DBTPhPm is much higher than that of the CBP(62 °C) [34], POAPF(129 °C) [24] and BCZph(100 °C) [35]. The high T_g would be benefited to the stability of the devices. The dibenzothiophene unit withdraws electron due to S atom with high electronegativity. The ultraviolet–visible (UV–vis) absorption, PL emission and low-temperature photoluminescence (LTPL) emission spectra of DBTPhPm in CH₂Cl₂ solution (1×10^{-5} M) are measured, and Fig. 1 shows the characteristics with an inset figure of the chemical structure of DBTPhPm. Absorption peaks at 336 nm can be attributed to π - π^* transitions of the dibenzothiophene chromophore and π - π^* and n - π^* transitions of central aryls, respectively. The optical energy band gap (E_g) of DBTPhPm is calculated to be 3.69 eV from the onset of the absorption spectrum (336 nm) according to the UV–vis absorption curve. The fluorescence emission peak of DBTPhPm is

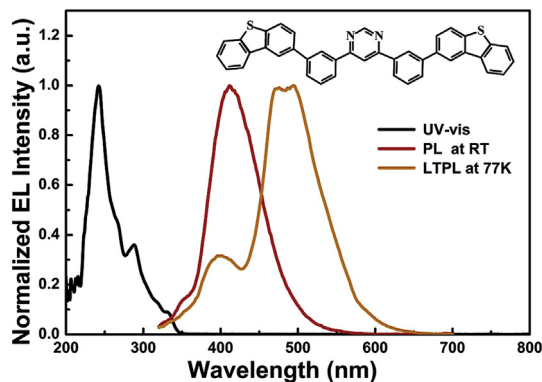


Fig. 1. The UV–vis absorption, photoluminescence (PL) emission (at RT) and low-temperature photoluminescence (LTPL) emission (at 77 K) spectra of DBTPhPm in CH₂Cl₂ solution (1×10^{-5} M).

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