

Stable Amorphous Bis(diarylamino)biphenyl Derivatives as Hole-Transporting Materials in OLEDs

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Stable bis(diarylamino)biphenyl derivatives N^4, N^4 -di(biphenyl-4-yl)- N^4, N^4 -bis(2-methylbiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-Ph) and N^4, N^4 -bis(2',4'-difluoro-2-methylbiphenyl-4-yl)- N^4, N^4 -bis(2',4'-difluorobiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-(2,4)-F) were synthesized and characterized. High thermal stability ($T_d \geq 480^\circ\text{C}$ and $T_g \geq 108^\circ\text{C}$) in combination with reversible oxidation process render the both promising candidates as hole-transporting materials for organic light-emitting devices. The device with structure of ITO/TPD-Ph/Alq₃/LiF/Al presented the highest current efficiency of 4.2 cd A⁻¹ and good operational stability even under the high current density of 350 mA cm⁻², which outperformed TPD or NPB-based devices.

Keywords: bis(diarylamino)biphenyl, hole-transporting materials, properties, efficiency

1. INTRODUCTION

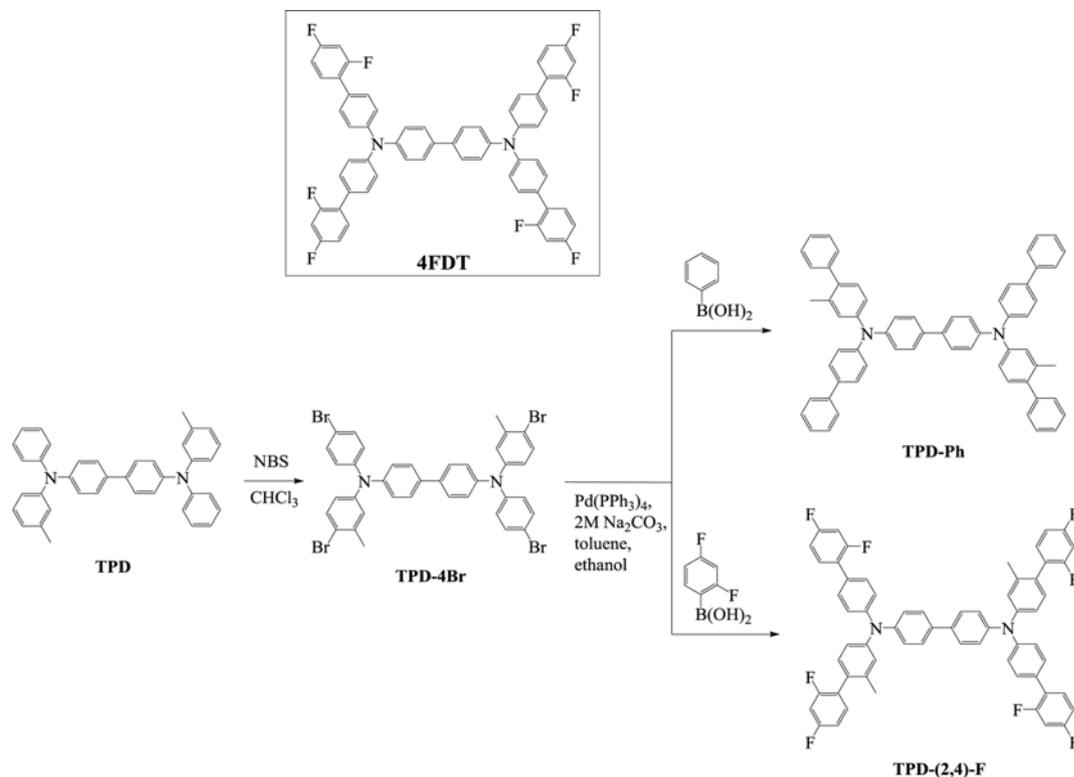
Triarylamine-based derivatives were well-known as effective hole-transporting materials (HTMs) because of their high hole mobilities in organic-based optoelectronic devices such as organic light-emitting devices (OLEDs), organic solar cells (OSCs), or organic thin film transistors (OTFTs).^[1-4] Besides the required high hole-mobility, hole-transport layers (HTLs) in OLEDs should be amorphous and thermally stable, with no absorption in the visible range, so that light output can be effective. The triarylamine-containing compounds such as N, N' -bis(tolyl)- N, N' -diphenyl-1,1'-biphenyl-4,4'-diamine (TPD) and 4,4',4''-tris-(N -3-methylphenyl- N -phenylamino)-triphenylamine (m -MTDATA) exhibit excellent hole-transporting properties, offering devices with excellent electroluminescent efficiencies and low turn-on voltages.^[5-10] However, they generally produce amorphous films with poor morphological and thermal stability due to the low glass-transition temperatures (T_g), which are 65°C and 75°C for TPD and m -MTDATA, respectively.^[11] Additionally the films prepared by sublimation tend to crystallize over time, even at room temperature, which is detrimental to the device performance^[12]. Although the most widely used HTM, N, N' -di-(1-naphthalenyl)- N, N' -diphenyl-4,4'-diamine (NPB), possesses high T_g (95°C), the amorphous

molecular materials with high T_g are highly desirable in OLED applications.

It has been suggested that the bigger size of organic π -electron systems is an efficient method to improve the T_g , which facilitates the formation of thermally and morphologically stable amorphous films.^[13,14] In our previous research,^[15] we designed and synthesized a triphenylamine dimer with four 2,4-difluorophenyl peripheral moieties, namely N^4, N^4, N^4, N^4 -tetrakis-(2',4'-difluoro-biphenyl-4-yl)-biphenyl-4,4'-diamine (4FDT, Scheme 1). We found that 4FDT exhibits high T_g (113°C) and other intriguing properties, which make it promising for hole-injecting/hole-transporting material in OLEDs. Moreover, the asymmetry of conformation in triarylamine compounds prevents the crystallization of the low molecular mass compounds, thus resulting in higher thermal stability as compared to symmetric derivatives.^[16,17]

In this paper, we have synthesized bis(diarylamino)biphenyl derivatives, namely, N^4, N^4 -di(biphenyl-4-yl)- N^4, N^4 -bis(2-methylbiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-Ph) and N^4, N^4 -bis(2',4'-difluoro-2-methylbiphenyl-4-yl)- N^4, N^4 -bis(2',4'-difluorobiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-(2,4)-F), in which the phenyl/2,4-difluorophenyl moieties were covalently incorporated to the TPD core to achieve high- T_g active materials. The compounds were found to form easily stable amorphous films by either vacuum deposition or spin-coating and to function as HTMs in OLEDs. The OLED employing TPD-Ph as the HTM showed the highest current efficiency of 4.2 cd A⁻¹, and good operational

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Scheme 1. Synthesis of compounds TPD-Ph and TPD-(2,4)-F and chemical structure of 4FDT.

stability even under the high current density of 350 mA cm^{-2} .

2. EXPERIMENTAL PROCEDURE

The manipulation involving air-sensitive reagents was performed under an inert atmosphere of dry nitrogen. TPD was prepared by the reported and well characterized method in literature.^[6,18] The absorption spectra were obtained using a Hitachi UV 3010 spectrophotometer. The photoluminescence (PL) spectra were collected by a Horiba Jobin Yvon Fluoromax-4 spectrophotometer. Decomposition temperatures (T_d) were obtained by TG209C thermal gravimetric analysis (TGA) with a heating rate of $20^\circ\text{C min}^{-1}$ and glass transition temperatures (T_g) were determined with a differential scanning calorimeter (DSC, TA instruments DSC200PC) at a heating rate of $10^\circ\text{C min}^{-1}$ under a N₂ atmosphere. Cyclic voltammetry was performed using a Princeton Applied Research model 273A potentiostat at a scan rate of 100 mV s^{-1} . All experiments were carried out in a three-electrode compartment cell with a Pt-sheet counter electrode, a glassy carbon working electrode and a Pt-wire reference electrode. The supporting electrolyte used was 0.1 M tetrabutylammonium hexafluorophosphate ([Bu₄N]PF₆) solution in dichloromethane. The cell containing the solution of the sample (1 mM) and the supporting electrolyte was purged with nitrogen gas thoroughly before scanning for its

oxidation and reduction properties. Ferrocene was used for potential calibration in each measurement. All the potentials were reported relative to ferrocene-ferrocenium (Fc/Fc⁺) couple, whose oxidation potential was +0.19 V relative to the reference electrode. The oxidation and reduction potentials were determined by taking the average of the anodic and cathodic peak potentials. The HOMO and LUMO values were estimated by using the following general equation: $E_{\text{HOMO}} = -(qE_{\text{ox}} + 4.8) \text{ eV}$; $E_{\text{LUMO}} = E_{\text{HOMO}} + E_{\text{g}}^{\text{opt}}$, which were calculated with the internal standard ferrocene value of -4.8 eV with respect to the vacuum level.^[19]

Synthesis of *N,N'*-bis(4-bromo-3-methylphenyl)-*N,N'*-bis(4-bromo-phenyl)-biphenyl-4,4'-diamine (TPD-4Br). To a 100 mL round-bottom flask were added TPD (2.58 g, 5 mmol) and NBS (3.56 g, 20 mmol). Chloroform (30 mL) was added, and the solution was stirred at room temperature (RT) for 1 h. At this stage 12.5 mL of acetic acid were added, and the solution was stirred for 6.5 h at RT. The product was extracted with diethyl ether, and washed with water and brine twice in sequence. The organic layer was dried over anhydrous sodium sulfate. After solvent evaporation, the crude product was recrystallized from chloroform and hexane to afford white solid (3.41 g, 82%). ¹H NMR (CDCl₃, 400 MHz): δ 2.30 (s, 6H), 6.84 - 6.86 (m, 2H), 6.96 - 6.99 (m, 6H), 7.10 (d, $J = 8.4 \text{ Hz}$, 4H), 7.32 - 7.40 (m, 6H), 7.42 - 7.46 (m, 4H). Anal. Calcd for C₃₈H₂₈N₂Br₄: C, 55.06%; H, 3.12%; N, 11.82%.

3.39%; N, 3.37%. Found: C, 54.79%; H, 3.21%; N, 2.65%.

Synthesis of N^4, N^4 -di(biphenyl-4-yl)- N^4, N^4 -bis(2-methylbiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-Ph). THF (15 mL) and 2.0 M aqueous solution of K_2CO_3 (15 mL) were added to a flask containing TPD-4Br (0.58 g, 0.7 mmol), phenylboronic acid (0.41 g, 3.4 mmol) and $Pd(PPh_3)_4$ (0.06 g, 0.05 mmol) under nitrogen. The reaction mixture was heated to reflux and maintained at this temperature for 24 h. When the reaction was completed (inspected by thin-layer chromatography), water was added to quench the reaction. Then, the products were extracted with CH_2Cl_2 . The organic portion was washed with brine, dried over anhydrous $MgSO_4$, and concentrated by evaporating off the solvent for further purification by column chromatography on silica gel in solvent as mobile phase. TPD-Ph was obtained as a white powder. Yield: 83% (0.48 g). 1H NMR ($CDCl_3$, 400 MHz): δ 2.18 (s, 6H), 7.29 - 7.37 (m, 16H), 7.40 - 7.46 (m, 12H), 7.53 (d, $J = 7.2$ Hz, 8H), 7.60 (d, $J = 7.6$ Hz, 8H). Anal. Calcd for $C_{62}H_{48}N_2$: C, 90.70%; H, 5.89%; N, 3.41%. Found: C, 88.84%; H, 4.32%; N, 4.45%. MS: m/z 820.3811 [M^+] (Calcd: 821.0580).

Synthesis of N^4, N^4 -bis(2',4'-difluoro-2-methylbiphenyl-4-yl)- N^4, N^4 -bis(2',4'-difluorobiphenyl-4-yl)biphenyl-4,4'-diamine (TPD-(2,4)-F). The experimental procedure for TPD-(2,4)-F was similar to that for the synthesis of TPD-Ph described above. TPD-(2,4)-F was obtained as a white powder. Yield: 86% (0.83 g). 1H NMR ($CDCl_3$, 400 MHz): δ 2.12 (s, 6H), 6.88 - 6.97 (m, 12H), 7.01 - 7.11 (m, 8H), 7.35 - 7.36 (m, 2H), 7.44 (d, $J = 6.8$ Hz, 8H), 7.52-7.56 (m, 4H). Anal. Calcd for $C_{62}H_{40}N_2F_8$: C, 77.17%; H, 4.18%; N, 2.90%. Found: C, 76.41%; H, 3%; N, 3.05%. MS: m/z 964.3020 [M^+] (Calcd: 964.9817).

The devices were fabricated by conventional vacuum deposition of the organic layers, LiF and Al cathode onto an ITO-coated glass substrate under a base pressure lower than

1×10^{-3} Pa. The thickness of each layer was determined by a quartz thickness monitor. The effective size of the OLED was 14 mm^2 . The hole-only devices with structure ITO/TPD-Ph or TPD-(2,4)-F (100 nm)/Ag (80 nm) and the typical two-layer OLEDs with the structure of ITO/TPD-Ph or TPD-(2,4)-F (60 nm)/Alq₃ (60 nm)/LiF (1 nm)/Al (70 nm) were fabricated. Here, Alq₃ (tris(8-hydroxyquinoline)aluminum) served as an electron transporting and light-emitting layer. For comparison, NPB and TPD-based devices with a similar device structure were also fabricated under similar conditions. The voltage-current density (V - J), voltage-brightness (V - L) and current density-current efficiency (J - η) curves of devices were measured with a computer-controlled Keithley 2602 Source-Meter under ambient condition.

3. RESULTS AND DISCUSSION

The chemical structures and the synthetic routes of compounds TPD-Ph and TPD-(2,4)-F were outlined in Scheme 1. Bromination of TPD by means of *N*-bromosuccinimide in glacial acetic acid at ambient temperature gave TPD-4Br in 82% yield.^[15] TPD-Ph and TPD-(2,4)-F were readily synthesized using Suzuki-Miyaura coupling by reacting phenylboronic acid or 2,4-difluorophenylboronic acid with TPD-4Br.^[20] All compounds were purified by column chromatography on silica gel with yields ranging from 83% to 86%, and the molecular structures were confirmed by 1H nuclear magnetic resonance (H NMR), high resolution mass spectrometry (MS), and element analysis. The results are in good agreement with proposed structures. Theoretical calculations on the electronic states of TPD-Ph and TPD-(2,4)-F were carried out at the DFT//B3LYP/6-31G level in the Gaussian 03 program. As illustrated in Fig. 1, the HOMOs of TPD-Ph and TPD-(2,4)-F are almost all localized on the TPD moieties. The LUMO of TPD-Ph mostly resides on the

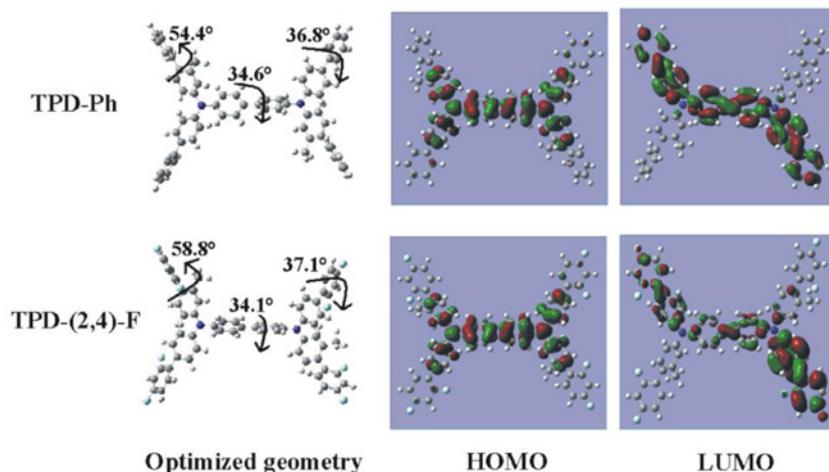


Fig. 1. The optimized geometries and the molecular orbital surfaces of the HOMOs and LUMOs for TPD-Ph and TPD-(2,4)-F obtained at the B3LYP/6-31G level.

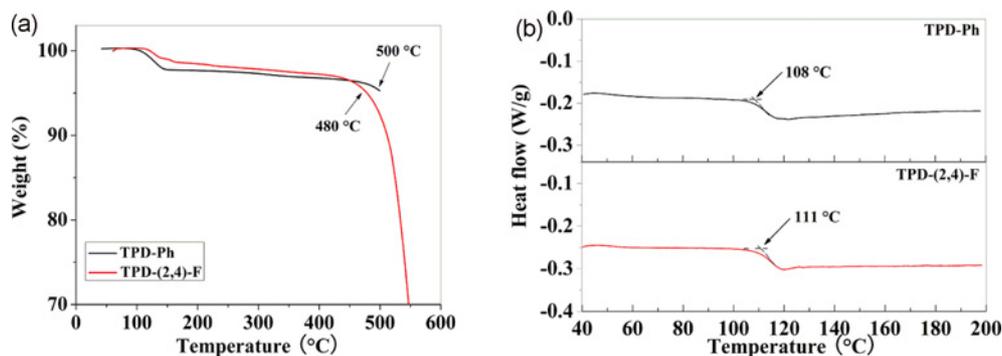


Fig. 2. TGA (a) and DSC (b) measurements for TPD-Ph and TPD-(2,4)-F; (DSC measurements: 2nd scan after N₂ treatment, 10°C min⁻¹, under N₂).

Table 1. Optical, thermal, and electrochemical properties of the compounds.

Compound	$\lambda_{\max}^{\text{Abs}}$ soln ^a /film ^b (nm)	$\lambda_{\max}^{\text{PL}}$ soln ^a /film ^b (nm)	$\Phi_{\text{PL}}^{\text{c}}$ soln	E_{ox}^{d} (V)	HOMO/LUMO _{exp} (E_{g}) (eV)	HOMO/LUMO _{cal} ($\Delta E_{\text{HOMO-LUMO}}$) (eV)	$T_{\text{m}}/T_{\text{g}}/T_{\text{d}}$ (°C)
TPD-Ph	345/344	421/419	0.23	0.36, 0.60	-5.16/-2.10 (3.06)	-5.04/-0.97 (4.07)	NA/108/500
TPD-(2,4)-F	337/338	414/413	0.54	0.38, 0.59	-5.18/-2.10 (3.08)	-5.26/-1.22 (4.04)	169/111/480

^aMeasured in CH₂Cl₂.

^bMeasured in film.

^cDetermined in CH₂Cl₂ using quinine sulfate ($\Phi_{\text{PL}} = 0.56$ in 1.0 M H₂SO₄ solution) as standard.

^dMeasured versus Fc/Fc⁺ in CH₂Cl₂.

biphenyl core and two diagonal phenyl/difluorophenyl arms of the molecule while that of TPD-(2,4)-F mostly resides on one substituted biphenyl arm and have a little on another. The difference may depend on the electron-withdrawing ability of the peripheral 2,4-difluorophenyl groups in TPD-(2,4)-F. Besides, from the optimized geometry of TPD-Ph and TPD-(2,4)-F it can be seen that the two adjacent phenyl rings at the ends and the cores of the molecules are twisted, which makes the molecular structures non-planar, thus facilitating the formation of an amorphous film and reducing the intermolecular π - π stacking and/or interaction. The calculated HOMO energy levels of TPD-Ph and TPD-(2,4)-F are -5.04 eV and -5.26 eV, respectively, as listed in Table 1.

Since the material with a high T_{g} can increase the thermal and operation stability of OLED, we have covalently incorporated the TPD and phenyl/2,4-difluorophenyl moieties into the high- T_{g} active materials. The thermal stabilities of the synthesized materials were measured by TGA and DSC, as shown in Fig. 2. The decomposition temperature (T_{d}) values for TPD-Ph and TPD-(2,4)-F were at 480°C and 500°C, respectively. The T_{d} was higher for the non-fluorinated analogue by about 20°C. The glass transition temperatures (T_{g}) of the two compounds were nearly the same, 108°C and 111°C for TPD-Ph and TPD-(2,4)-F, respectively, which are higher than those of the most commonly used hole transport materials TPD, *m*-MTDATA and NPB.^[11] The increase in

the molecular weight along with the incorporation of the rigid biphenyl moieties in TPD-Ph and TPD-(2,4)-F is suggested to be responsible for the increase in T_{g} for both compounds relative to the parent molecule TPD.^[13]

The photophysical properties of TPD-Ph and TPD-(2,4)-F were measured and the UV-visible absorption and photoluminescence (PL) spectra were shown in Fig. 3 and Table 1. The compounds absorb in the UV spectral region below 400 nm with maxima between 337 and 345 nm, which can be attributed to π - π^* transitions of the conjugated aromatic rings. The compound TPD-(2,4)-F shows a negligible blue-shift in the absorption ($\lambda_{\max} = 337$ nm) when compared with the non-fluorinated analogue TPD-Ph ($\lambda_{\max} = 345$ nm), which is due to the incorporation of the fluorine atoms at the *ortho*-positions in the phenyl ring of each arm in TPD-(2,4)-F that slightly decreases the π -conjugation between TPD and 2,4-difluorophenyl units (Fig. 1). In absorption spectra, a blue shift in fluorescence maximum in the title compounds is observed (TPD-Ph: 421 nm, TPD-(2,4)-F: 414 nm) upon replacing phenyl groups with electron-withdrawing 2,4-difluorophenyl groups. The transparent and pinhole-free films were fabricated by spin-coating from a chloroform solution of the compounds on quartz substrates.^[21] There were almost no red-shift in the absorption and emission spectra of the solid film compared to its CH₂Cl₂ solution (Fig. 3). This phenomenon results from the three-dimensional

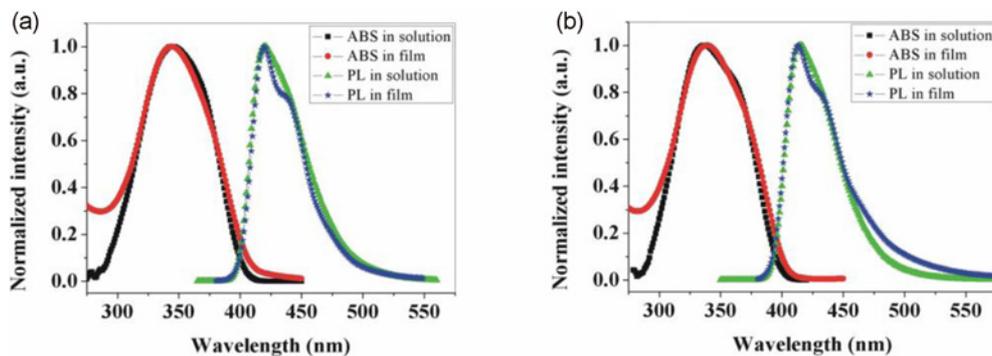


Fig. 3. The absorption and PL spectra of TPD-Ph (a) and TPD-(2,4)-F (b) in CH_2Cl_2 and films.

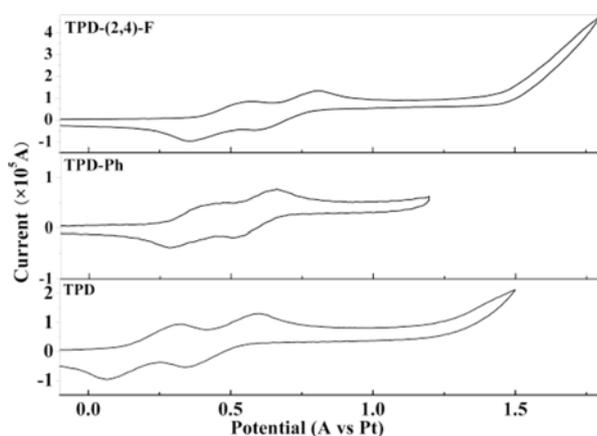


Fig. 4. Traces of cyclic voltammetric measurements of TPD, TPD-Ph and TPD-(2,4)-F, Pt wire used as reference electrode.

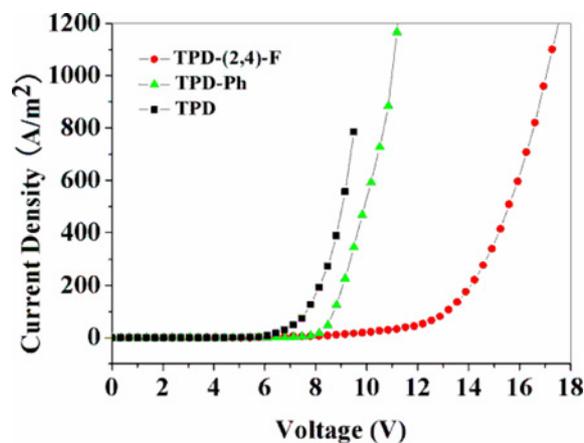


Fig. 5. Current density-voltage for the hole-only devices, ITO/TPD, TPD-Ph or TPD-(2,4)-F (100 nm)/Ag (80 nm).

propeller structure of triarylamine moieties, as well as the twisting conformation between the peripheral phenyl/2,4-difluorophenyl groups and TPD core (see Fig. 1), which is helpful for decreasing intermolecular interactions in the aggregated state. The optical band gaps (E_g) determined from the onset of the film absorption spectra are 3.06 eV and 3.08 eV for TPD-Ph and TPD-(2,4)-F, respectively. The two compounds could be applied to HTL due to lack of absorption in visible region of spectrum.

The electrochemical properties of TPD-Ph and TPD-(2,4)-F were estimated by cyclic voltammetry (CV), as shown in Fig. 4 along with the CV of TPD in the same conditions for comparison. During the oxidation scan in dichloromethane, both TPD-Ph and TPD-(2,4)-F showed two sequential one-electron processes, corresponding to removal of two electrons from the TPD unit. A positive change in the first oxidation potentials relative to ferrocene from 0.32 to 0.36 to 0.38 V was observed from TPD to TPD-Ph to TPD-(2,4)-F, corresponding to the HOMO energy levels of -5.12 eV for TPD, -5.16 eV for TPD-Ph and -5.18 eV for TPD-(2,4)-F, respectively. The energy level was lower for the fluorinated analogue TPD-(2,4)-F than that of the parent molecule TPD by about 0.06 eV due to the strongly electron-withdrawing

fluoro substituent on the peripheral phenyls.^[15,22,23] By subtraction of the optical band gap from the HOMO energy level, the LUMO energy levels of TPD-Ph and TPD-(2,4)-F were estimated to be -2.10 eV, as listed in Table 1. The experimental HOMOs were in excellent agreement with the calculated values.

To investigate the hole-transport ability of TPD-Ph and TPD-(2,4)-F, the hole-only devices with structure ITO/TPD, TPD-Ph or TPD-(2,4)-F (100 nm)/Ag (80 nm) were fabricated and characterized. Their current density-voltage characteristics are shown in Fig. 5, it can be seen that the current density at a given applied voltage exhibit $\text{TPD} > \text{TPD-Ph} > \text{TPD-(2,4)-F}$, suggesting their hole mobility decreased in turn. The hole mobility of the 2,4-difluorophenyl-substituted counterpart TPD-(2,4)-F is much lower than that of the parent molecule TPD, which may be attributed to a more twisting conformation between the peripheral 2,4-difluorophenyl-substituted phenyl at the *ortho*-position and TPD core in TPD-(2,4)-F (see Fig. 1). The strong electron-withdrawing fluorinated substituents in the molecule, which significantly lowered the hole mobility.^[15,24] Moreover, the phenyl-substituted counterpart TPD-Ph shows slightly lower hole mobility than the parent molecule TPD.

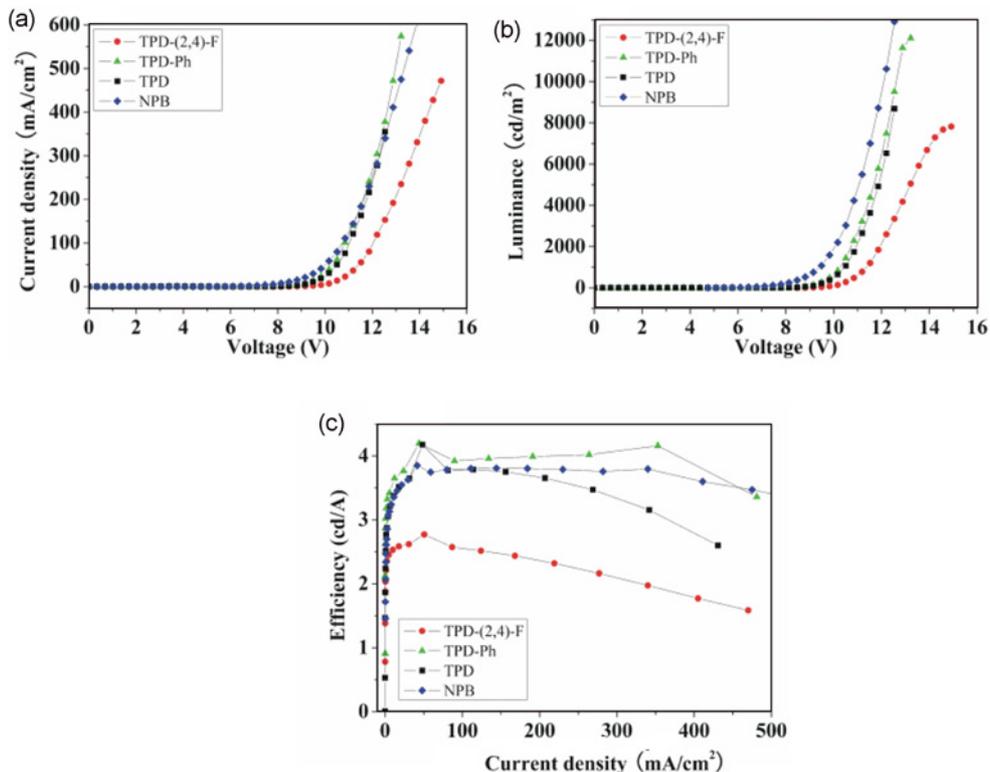


Fig. 6. Current density-voltage curves (a), luminance-voltage curves (b) and efficiency--current density curves (c) for the ITO/NPB, TPD, TPD-Ph or TPD-(2,4)-F (60 nm)/Alq₃ (60 nm)/LiF (1 nm)/Al (70 nm) devices.

Based on the unique properties of TPD-Ph and TPD-(2,4)-F including good thermal stabilities, optical properties, proper HOMO/LUMO energy levels and the hole-transporting nature of bis(diarylamino)biphenyl derivatives, both are expected to be potential hole-transporting materials in OLEDs. Figure 6 shows the current density-voltage (J - V), the luminance-voltage (L - V) and the current density-current efficiency characteristics (J - η) of the two-layer devices ITO/HTL (60 nm)/Alq₃ (60 nm)/LiF (1 nm)/Al (70 nm) (HTL = TPD, NPB, TPD-Ph or TPD-(2,4)-F). It clearly shows that the current density of the TPD-(2,4)-F-based device is much lower than that of the NPB, TPD or TPD-Ph-based device at all voltages (Fig. 6(a)). This further confirms that TPD-(2,4)-F offer the lowest mobility for hole transportation, along with minimal hole injecting properties among these compounds (see Fig. 5). In Fig. 6(b), device employing TPD-Ph as the HTM possessed lower turn-on voltage (defined as the voltage required to give a luminance of 1 cd m⁻²) and higher luminance than those of the device with TPD as the HTM. At 1000 cd m⁻², the driving voltages of NPB, TPD, TPD-Ph and TPD-(2,4)-F-based device are 9.4, 10.2, 10.4 and 11.3 V, respectively. Among the devices, the TPD-Ph-based device shows the best performances, it presents the highest efficiency of 4.2 cd A⁻¹ at a voltage of 5 V, which is higher than that of the device based on NPB as the HTL (3.8 cd/A) (shown in Fig. 6(c)). The best performance of TPD-Ph-

based device is attributed to the balanced carriers injection and transport, due to suitable hole mobility and HOMO energy level of phenyl-substituted analogue. Notably, the efficiency-versus-current density curve in TPD-Ph-based device remained stable until very-high current density (~4.2 cd A⁻¹ over 350 mA cm⁻²). This means that the efficiency does not roll off under high luminance. While the TPD-(2,4)-F-based device shows the poorest performance, its current efficiency is only 2.7 cd A⁻¹, which may be resulted from the lower hole mobility and injection in comparison to TPD (see Fig. 5).

4. CONCLUSIONS

In summary, new stable bis(diarylamino)biphenyl derivatives TPD-Ph and TPD-(2,4)-F, have been designed and synthesized. It has been shown that both the increase of the molecular size of organic π -electron systems and the substitution of the amine with a group bearing asymmetrical conformation facilitate the formation of thermally and morphologically stable amorphous molecular materials. The high T_d ($\geq 480^\circ\text{C}$) and T_g ($\geq 108^\circ\text{C}$) in along with the reversible oxidation process render both promising candidates as HTMs in OLEDs. The TPD-Ph-based double-layer device offered the highest efficiency of 4.2 cd A⁻¹. Additionally, the efficiency-versus-current density curve remained stable until

very-high current density ($\sim 4.2 \text{ cd A}^{-1}$ over 350 mA cm^{-2}), which were superior to those of the TPD or NPB-based device. Owing to the better thermal stability, and the higher performance of TPD-Ph-based device, TPD-Ph is a promising hole-transporting material for application in OLEDs.

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