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Efficient hole-transporter for phosphorescent organic light emitting diodes with a simple molecular structure



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ABSTRACT

N,N-diphenyl-4-(quinolin-8-yl)aniline (SQTPA), which composes a triphenylamine group and a quinoline group, has been synthesized and employed as a hole-transporter in phosphorescent OLEDs. It has been proved that SQTPA has efficient hole-transport property with a hole-mobility of 3.60×10^{-5} cm²/V s at the electric field of 800 (V/cm)^{1/2}, which is higher than that of NPB (1.93×10^{-5} cm²/V s). Blue, orange and green phosphorescent OLEDs have been fabricated based on Firpic, Ir(2-phq)₃, Ir(ppy)₃ with typical structures by using SQTPA as the hole-transporter. The SQTPA-based devices show maximum external quantum efficiencies and power efficiencies of 17.5%, 32.5 lm/W for blue, 12.3%, 20.5 lm/W for orange and 20.3%, 64.5 lm/W for green. The performances of SQTPA-based devices are much better than that of NPB-based phosphorescent OLEDs with similar structures. Thought of its very simple molecular structure and easy synthetic route, SQTPA should be an efficient hole-transporter for phosphorescent OLEDs.

1. Introduction

Phosphorescent OLEDs have been shown to harvest 100% of the excitions generated by electrical injection, corresponding to a four times increment in efficiency compared to that obtained in singletharvesting fluorescent OLEDs [1-3]. Thus, OLEDs have attracted a great deal of attention because of their huge applications in fullcolor flat-panel displays and solid-state lightings [4,5]. Although considerable progresses have been made in OLEDs, there are still two key focuses. One concern is to improve the performance especially the efficiency and the lifetime of the OLEDs. The other issue is to achieve a vast reduction in the cost of the OLEDs, which includes the cost on the fabrication process and the organic materials. To achieve highly-efficient OLEDs, maintaining a good balance between electron and hole currents in OLEDs is an important factor [6]. Various strategies have been developed to meet the requirement of charge balance, which includes the employment of highly efficient charge transporters and the introduction of bipolar or multi-functional molecules [7-12]. Efficient charge transporters including hole transporters and electron transporters are the most important materials to achieve high performance OLEDs. Pyridinebased electron transporters, such as TmPyPB, etc., developed by Kido group [13-16], have been widely used to largely improve the performance of the phosphorescent OLEDs. Triphenylamine-

* Corresponding author. E-mail address: silutao@uestc.edu.cn (S. Tao). based derivatives have been widely used in OLEDs for their relatively high hole-transporting property and appropriate highest occupied molecular orbital level. The triphenylamine-based hole transporters, such as TAPC, are significant to increase the efficiency of phosphorescent OLEDs [17–20]. Bipolar materials combining with a hole transporting unit and an electron transporting unit have been widely investigated in OLEDs in these years [21–25]. It has been reported that some bipolar materials have exhibited with a high hole transporting property, which should be used as potential good hole-transporters [23–25].

Moreover, the molecular structures and the synthetic routes of the charge transporters should be simple as much as possible, which can reduce the cost of OLEDs massively due to a great quantity of use in the fabrication process. Thus, it is important to design and synthesize efficient charge transporters with very simple molecular structure, which can bring high performance and reduce the cost in phosphorescent OLEDs.

In this paper, N,N-diphenyl-4-(quinolin-8-yl)aniline (SQTPA), which composes a triphenylamine group and a quinoline group, has been synthesized and characterized. It has been proved that SQTPA has efficient hole-transport property with a hole-mobility of 3.60×10^{-5} cm²/V s at the electric field of $800 (V/cm)^{1/2}$, meanwhile the hole-mobility of NPB is 1.93×10^{-5} cm²/V s. Blue, orange and green phosphorescent OLEDs have been fabricated based on Flrpic, Ir(2-phq)₃, Ir(ppy)₃ with typical device structures by using SQTPA as the hole-transporter. The devices show maximum external quantum efficiencies and power efficiencies of 17.5%, 32.5 lm/







W for blue, 12.3%, 20.5 lm/W for orange and 20.3%, 64.5 lm/W for green, respectively. The results of SQTPA-based devices are much better (35% higher for blue, 37% higher for orange and 46% higher for green) than that of NPB-based phosphorescent OLEDs with similar device structures. Thought of its very simple molecular structure and easy synthetic route, SQTPA should be an efficient hole-transporter.

2. Experiment

2.1. Chemical and instrument

All powders and solvents were used in this reaction as received from commercial sources without further purification. The final product was demonstrated with ¹H NMR spectra, elemental analysis (EA) and Mass Spectrometry (MS). The glass transition temperature (Tg) was measured with differential scanning calorimetry (DSC) by using Perkin-Elmerpyris DSC6 operated at a heating rate of 10 °C min⁻¹. The decomposition temperature (Td) was measured by thermo gravimetric analysis (TGA) which was performed on a TA SDT Q600 instrument at a heating rate of 10 °C min⁻¹ under nitrogen flowed. Optical absorption and emission spectra were recorded with a Hitachi U-3010 UV-vis spectrophotometer and a Hitachi F-4500 fluorescence spectrophotometer, respectively. Highest occupied molecular orbital (HOMO) energy level was confirmed via ultraviolet photoelectron spectroscopy (UPS). Lowest unoccupied molecular orbital (LUMO) energy level was calculated by the difference valves between the HOMO energy level and energy gap (Eg) obtained from the optical absorption edge.

2.2. Materials synthesis

4-(8-Quinolyl)-triphenylamine (SQTPA). 4-Bromotriphenylamine (2 mmol), 8-quinoline boronic acid (2.5 mmol), Pd(PPh₃)₄ (0.5 mmol), aqueous Na₂CO₃ (2.0 M, 20 ml), ethanol (15 ml), and toluene (20 ml) were mixed in a flask. The mixture was degassed and the reaction was refluxed at 100 °C for 24 h under nitrogen. After being cooled, the solvent was evaporated under vacuum and the product was extracted with dichloromethane (CH₂Cl₂). The CH₂Cl₂ solution was washed with water and dried with MgSO₄. Evaporation of the solvent, followed by column chromatography on silica gel (petroleum ether/CH₂Cl₂) yielded white powder. Yield: 65%. 1H nuclear magnetic resonance: (CDCl₃, 300 MHz) δ: 8.97-8.96 (m, 1H), 8.21-8.18 (d, 1H), 7.80-7.74 (m, 2H), 7.64 (d, 2H), 7.59 (d, 1H), 7.43-7.40 (m, 1H), 7.30-7.26 (m, 4H), 7.21-7.17 (m, 6H), 7.03 (d, 2H); Mass Spectrometry (ESI+): m/z 372.2 (M+); elemental analysis: Anal. Calcd. For C₂₇H₂₀N₂ C: 87.07%, H: 5.41%, N: 7.52%. Found: C: 87.10%, H: 5.40%, N 7.32%.

2.3. Device fabrication and measurement

Indium tin oxide (ITO)-coated glass substrates with a sheet resistance of 15 Ω per square were used as the starting substrates. Before device fabrication, the ITO glass substrates were pretreatment carefully by washing with isopropyl alcohol and deionized water, dried in an oven at 120 °C over 2 h and then treated with ultraviolet-ozone for 30 min before loading in the vacuum deposition chamber. All organic layers were deposited on the ITO glass substrates with a rate of $1-2 \text{ Å s}^{-1}$ under a base pressure 5×10^{-4} Pa. The electron-injecting layer LiF and cathode Al were deposited with a rate of 0.1 Å s^{-1} and 10 Å s^{-1} , respectively. EL spectra, CIE coordinates, and current density-voltage-luminance (*J*-*V*-*L*) characteristics were evaluated with a Spectrascan PR655 photometer and a computer-controlled Keithley model 2400 source meter under ambient atmosphere.

3. Results and discussions

The synthetic route of SQTPA is shown in Fig. 1. SQTPA can be obtained by a one-step Suzuki coupling reaction with a high yield of 65%. The molecule structure of SQTPA was confirmed with ¹H NMR spectrometry, EA and MS. The thermal stability was determined by thermo gravimetric analysis (TGA), showing a thermal decomposition temperature (Td, corresponding to 5% weight loss) of 248 °C. By differential scanning calorimetry (DSC), no distinct glass transition was observed. The highest occupied molecular orbital (HOMO) energy level of SQTPA was measured to be 5.5 eV by ultraviolet photoelectron spectroscopy (UPS), and the lowest unoccupied molecular orbital (LUMO) was 2.5 eV.

Fig. 2 shows the room-temperature UV–vis absorption, photoluminescence (PL) spectra in a dilute dichloromethane (CH₂Cl₂) solution and phosphorescence spectra in dilute 2-methyl-tetrahydrofuran (2-MeTHF) at 77 K of SQTPA, respectively. The room-temperature (RT) short-wavelength absorptions peaked at 301 nm in dilute CH₂Cl₂ solution can be ascribed to the π – π * transitions from triphenylamine unit [26–28]. The singlet state (E_S) and triplet state (E_T) energy of SQTPA were calculated to be 2.54 eV and 2.28 eV, respectively.

To investigate the hole-transporting property of SQTPA, holeonly device was fabricated with the structure of ITO/MoO₃ (10 nm)/SOTPA (80 nm)/MoO₃ (10 nm)/Al. MoO₃ serves as hole injecting layer (HIL) and electron blocking layer (EBL). For comparison. NPB (4,4'-bis(N-(1-naphthyl)-N-phenylamino)biphenyl) based hole-only device was also fabricated with the structure of ITO/MoO₃ (10 nm)/NPB (80 nm)/MoO₃ (10 nm)/Al. In the devices, most of carriers can be restrained in the organic layer due to the large energy gap between the LUMO of MoO_3 (2.3 eV) and the work function of Al cathode (4.3 eV). Holes can be injected from the anode to the organic layer, thus the hole-mobility of SQTPA and NPB can be obtained. Fig. 3 (insert) describes the current density versus voltage (J-V) curves of the devices. Remarkable higher hole-current density in the SQTPA-based device is obvious to demonstrate that SQTPA is capable of good hole-transporting



Fig. 1. The synthetic route of SQTPA.



Fig. 2. Absorption and photoluminescence properties of SQTPA.

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Fig. 3. Mobility character at different electric field and current density versus voltage (inset) of hole-only devices.



Fig. 4. Normalized EL spectra at 1000 cd/m² of orange OLEDs with SQTPA and NPB.

property, which should be attributed to the much higher electrondonating effect of the triphenylamine moiety.

To further certificate the hole-mobility, SCLC method is used for the hole-only device [29]. The formula can be described as follows:

$$J = \frac{9}{8}\varepsilon_r\varepsilon_0\mu\frac{V^2}{L^3} \tag{1}$$

and

$$\mu = \mu_0 \exp\left(\beta \sqrt{\frac{V}{L}}\right) \tag{2}$$

where ε_r is relative permittivity (≈ 3.00), ε_0 is vacuum permittivity ($\approx 8.85 \times 10^{-14}$ F cm⁻¹), μ is the carrier mobility determined by SCLC measurements, *L* is the cathode–anode distance (100 nm for the device), μ_0 is zero-field mobility and β is a coefficient that is proportional to the Poole–Frenkel factor [30]. The value of μ_0 and β are calculated to be 2.88×10^{-6} cm²/V s, 2.82×10^{-3} (V cm⁻¹)^{-1/2} for SQTPA. By using the obtained values, the field-dependent holes mobility can be obtained by Eq. (2). The characters of hole-mobility against electric field ($E^{1/2}$) were shown in Fig. 3. The hole mobility of SQTPA and NPB are 3.60×10^{-5} cm²/V s and 1.93×10^{-5} cm²/V s at high electric field of 800 (V/cm)^{1/2}, respectively. These results imply that the hole-transporting property of SQTPA is better than that of the widely used hole-transporter NPB at the same electric field.

Thought of the good hole-transporting property and the triplet energy (2.28 eV) of SQTPA, phosphorescent orange OLED using SQTPA as a hole transporter was fabricated with a structure of ITO/MoO₃ (1 nm)/SQTPA (40 nm)/CBP: 4%Ir(2-phq)₃ (30 nm)/TPBi (40 nm)/LiF (1 nm)/Al. ITO and Al are anode and cathode, MoO₃ and LiF function as hole injecting layer (HIL) and electron injecting layer (EIL), respectively. 4,4'-Bis(N-carbazolyl)-1,1'-biphenyl (CBP) is the host of EML; 1,3,5-tri(1-phenyl-1H-benzo[d]imidazol-2-yl) phenyl (TPBi) is electron transporting layer (ETL) which can confine the triplet excitions in EML; In the device, tris(2phenylquinoline) iridium(III) (Ir(2-phq)₃) acts as phosphorescent orange dopant. The doping concentration of Ir(2-phq)₃ was exactly maintained at 4%. For comparison, a control device was also prepared using the widely used hole-transporter NPB (4,4'-bis(N-(1naphthyl)-N-phenylamino)biphenyl) with the same structure.

Fig. 4 shows the EL spectra of the orange OLEDs based on SQTPA and NPB. At 1000 cd/m^2 , the devices both show orange emission with an emission peak at 588 nm. The CIE coordinates are (0.56, 0.43) for the SQTPA-based device, (0.57, 0.43) for the NPB-based device. There is no redundant emission from the host or transport layers, suggesting that the device structure with rational design can confine excitions within the EML effectively and effective energy transfer from host to dopant occurred.

Fig.5(a) reveals the luminous efficiencies characteristics of both devices. The SQTPA-based device owns maximum efficiencies of 12.3% for external quantum efficiency (EQE), 27.1 cd/A for current efficiency (CE) and 20.5 lm/W for power efficiency (PE). Meanwhile, the orange device containing NPB shows the maximum efficiencies of 10%, 22.9 cd/A, 15 lm/W, respectively. The results show that the power efficiency of SQTPA-base device is 37% higher than



Fig. 5. The current density, luminance versus voltage characteristic and the luminance efficiency properties of orange OLEDs with SQTPA and NPB, respectively.

Table 1							
Performances	summary	of the	devices	in	this	work	

Device	Von (V)	EQE^{a} (%), CE^{b} (cd/A), PE^{c} (lm/W)					
		10 cd m ⁻²	$100 \text{ cd } \text{m}^{-2}$	$1000 \text{ cd } \text{m}^{-2}$	Max	CIE ^d	
01	3.4	7.0, 25.8, 20.3	11.8, 26.8, 19.1	12.1, 26.2, 14.6	12.3, 27.1, 20.5	(0.57, 0.43)	
02	4.0	9.3, 21.6, 14.5	9.9, 22.7, 13.9	8.9, 20.6, 10.0	10.0, 22.9, 15.0	(0.56, 0.43)	
G1	3.0	20.3, 70.0, 61.0	14.6, 52.8, 38.7	10.8, 37.8, 17.6	20.3, 71.9, 64.5	(0.32, 0.62)	
G2	3.2	13.8, 49.3, 43.5	12.4, 45.3, 36.2	11.5, 41.1, 18.1	13.8, 49.3, 44.3	(0.32, 0.62)	
B1	3.6	17.5, 41.8, 32.9	14.4, 34.2, 22.6	12.3, 29.4, 14.6	17.5, 41.5, 32.5	(0.18, 0.40)	
B2	3.8	14.0, 32.2, 24.0	14.4, 33.9, 19.5	11.6, 27.7, 12.1	15.0, 35.3, 24.1	(0.18, 0.40)	

Blue, orange and green OLEDs with SQTPA were named as B1, O1 and G1, respectively. Blue, orange and green OLEDs with NPB were named as B2, O2 and G2, respectively.

^a External quantum efficiency.

^b Current efficiency.

^c Power efficiency.

^d Measured at 1000 cd m^{-2} .



Fig. 6. Normalized EL spectra at 1000 cd/m² of green OLEDs with SQTPA and NPB.

that of NPB-based device. Maintaining a good balance between electron and hole currents in OLEDs is an important factor to achieve highly-efficient OLEDs, which means that the mobility of the hole transporter and the electron transporter should be comparable. Although the hole injection barrier at the ITO/SQTPA and ITO/NPB interfaces are almost the same due to the similar HOMO levels of SQTPA and NPB. The higher hole mobility of SQTPA should enhance the hole injection and the probability of excitions recombination in EML. Thus, a better balance between electrons and holes in the EML was obtained, which induced a higher efficiency. Fig.5(b) describes *J*–*V*–*L* characteristics of two orange devices. It



Fig. 8. Normalized EL spectra at 1000 cd/m^2 of blue OLEDs with SQTPA and NPB.

clearly reveals that the current density of the SQTPA-based device is much higher than that of the NPB-based device. The device with SQTPA exhibits a lower turn-on voltage (at a brightness of 1 cd m^{-2}) of 3.4 V than that of the device with NPB (4.0 V). Thought of the similar HOMO level of NPB and SQTPA, the lower turn-on voltage should be attributed to the higher hole-transporting property of SQTPA than NPB [31–33]. Table 1 summarizes key performance parameters of the devices.

Moreover, green phosphorescent OLEDs with a configure of ITO/ MoO_3 (1 nm)/SQTPA or NPB (40 nm)/CBP: 8%Ir(ppy)₃ (30 nm)/TPBi (40 nm)/LiF (1 nm)/Al were fabricated. The emission layer consists of CBP doped with 8 wt% of the Ir(ppy)₃. Fig. 6 gives the EL spectra



Fig. 7. The luminance efficiency properties curve and the current density, luminance versus voltage characteristic of green OLEDs with SQTPA and NPB, respectively.



Fig. 9. The luminance efficiency properties curve and the current density, luminance versus voltage characteristic of blue OLEDs with SQTPA and NPB, respectively.

of the green OLEDs. At 1000 cd/m^2 , the two devices both exhibit a pure green emission from $Ir(ppy)_3$ (peaked at 516 nm) and CIE coordinates of (0.32, 0.62). Fig. 7(a) displays luminous efficiencies characteristics of the green devices. The green device with SQTPA shows EQE, CE and PE of 20.3%, 71.9 cd/A and 64.5 lm/W, which are much higher than that of NPB (13.8%, 49.3 cd/A and 44.3 lm/ W). In this device, the excitions recombination zone should be located near the interface of the emission layer (EML) and electron-transporting layer (ETL) due to the high hole transporting property of SQTPA. The triplet energy of TPBi is high enough to confine the formed triplet excitions at the interface. Thus, although the $E_{\rm T}$ of SOTPA (2.28 eV) is lower than that of $Ir(ppy)_3$ (2.42 eV), the EQE of SQTPA-based device is obtained as high as 20.3%. Some other groups have found similar results in their green phosphorescent devices and blue phosphorescent devices by using a low triplet energy hole transporter [34–35]. Fig. 7(b) gives the *I–V–L* characteristic of the green devices. The turn-on voltage of the SQTPA-based device (3.0 V) is lower than that of NPB-based device (3.2 V).

To further demonstrate the application of SQTPA in phosphorescent OLEDs, blue devices have been fabricated with the configuration of ITO/MoO₃ (1 nm)/SQTPA or NPB (40 nm)/mCP (5 nm)/ SPPO1: 10% FIrpic (30 nm)/SPPO1 (30 nm)/LiF (1 nm)/Al. In the device, bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl)iri dium(III) (FIrpic) is utilized as the blue dopant. Due to the triplet energy is lower than that of FIrpic (SQTPA:2.28 eV; FIrpic:2.68 eV), 1,3-bis(carbazol-9-yl)benzene (mCP) is used as an excition blocking layer (EBL) for its high-lying triplet energy of 2.9 eV, which can confine the triplet excitions in EML effectively; 9,9-spirobifluo ren-2-yl-diphenyl-phosphine oxide (SPPO1) is utilized as the host and the electron transporter in the device due to its high triplet energy (2.9 eV) and good electron-transporting property. The doping concentration of FIrpic in the device is optimized to be 10%.

Fig. 8 shows the EL spectra of the blue devices. At 1000 cd/m^2 , the devices both give blue emission with two peaks centered at 472 and 497 nm, and the same CIE coordinates of (0.18, 0.40). Fig. 9 shows the current efficiency–luminance–power efficiency plots and *J*–*V*–*L* curves of blue devices. The SQTPA-based device demonstrates the performances with a maximum external quantum efficiency (EQE) of 17.5%, a maximum current efficiency (CE) of 41.5 cd/A, and a maximum power efficiency (PE) of 32.5 lm/W (at 8.3 cd/m²). Whereas that of NPB-based device gives corresponding efficiencies of 15%, 35.3 cd/A and 24.1 lm/W. The efficiency of blue device based on SQTPA has dramatically increased (by 36%) compared to that of the blue device based on NPB, which should be partially attributed to the characteristics of hole injection and transportation of HTL material. In addition, the good device structure can confine the excitions in EML effectively, which

is beneficial for the high performance. The turn-on voltage of SQTPA based device is 3.6 V, which is also lower than that of device with NPB (3.8 V).

Owing to the simple molecular structure, easy synthetic route and the excellent performances of the phosphorescent devices using the new hole transporter, it is believed that SQTPA should be an efficient hole-transporter for phosphorescent OLEDs.

4. Conclusion

A simple molecule SQTPA, which composes a triphenylamine group and a quinoline group, has been synthesized and employed as an excellent hole-transporter in phosphorescent OLEDs. The hole-transporting property was examined in simple monochromatic phosphorescent OLEDs. Flrpic, Ir(2-phq)₃, Ir(ppy)₃ were respectively used in blue, orange and green phosphorescent OLEDs. Maximum external quantum efficiencies, current efficiencies and power efficiencies of 17.5%, 41.5 cd/A and 32.5 lm/W for blue, 12.3%, 27.1 cd/A and 20.5 lm/W for orange, 20.3%, 71.9 cd/A and 64.5 lm/W for green were obtained. The performances of SQTPA-based devices are much better than that of NPB-based phosphorescent OLEDs with similar structures. Thought of its very simple molecular structure and easy synthetic route, SQTPA should be an efficient hole-transporter for phosphorescent OLEDs.

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