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Articles

High T_g Triphenylamine-Based Starburst Hole-Transporting Material for Organic Light-Emitting Devices

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This paper reports the synthesis of a new starburst molecule, 4,4′,4″-tris[(2,3,4,5-tetraphenyl)phenyl]phenylamine (TTPPPA) and its application as a hole-transporting material in organic light-emitting devices (OLEDs). Although TTPPPA has almost the same ionization potential as 1,4-bis(1-naphthylphenylamino)biphenyl (NPB), the TTPPPA-based device of ITO/TTPPPA/Alq_3/LiF/Al yields much better efficiency of 5.3 cd/A and 4.3 lm/W than the standard ITO/NPB/Alq_3/LiF/Al device (3.0 cd/A and 2.9 lm/W). The remarkable performance enhancement is attributed to a better balance of hole and electron injection in the TTPPPA-based device. Further, TTPPPA has a much higher glass-transition temperature (T_g, 202 °C) than NPB (T_g, 98 °C), suggesting that TTPPPA can be an alternative material to NPB especially for high-temperature applications of OLEDs and other organic electronic devices.

Introduction

Organic light-emitting devices (OLEDs) have attracted much attention because of their applications in full-color flat-panel displays.[1–3] Since the pioneering work of small-molecular OLEDs by Tang and Van Slyke[4] and polymeric OLEDs by Friend[5] et al., much effort has been devoted to the development of electroluminescent materials with high luminescent efficiency, good stabilities, and good film-forming properties.[6–10] Considerable progress has been achieved in both device fabrication techniques and material development in the past decade.[11] Maintaining the charge balance in an OLED is an important issue for achieving good device performance. It is well-known that hole drift mobility of hole-transporting material (HTM) is generally higher than electron mobility of electron-transporting material (ETM). For example, hole mobility in 1,4-bis(1-naphthylphenylamino)biphenyl (NPB) is about 2 orders of magnitude higher than electron mobility in tris-(8-hydroxyquinoline)aluminum (Alq_3). Therefore, to optimize the electroluminescence (EL) efficiency, it is essential to balance the injection of holes and electrons into the emitting layer of OLEDs. Balanced injection can be facilitated via enhancement of electron mobility and/or a judicial reduction of hole mobility. To improve electron injection and transport, an electron-injecting layer such as LiF[12,13] has been adopted to insert between the cathode and the electron-transporting layer (ETL) or a novel ETM such as AlOq[14] has been used. However, ETM with a high electron mobility is relatively scarce compared to efficient ETM. In terms of hole injection and transport, there are generally two strategies to reduce hole mobility, that is, by doping hole-transporting layer (HTL) with hole-trapping material[15] or by inserting a buffer layer at the hole-injecting contact.[16] Both strategies inevitably complicate device fabrication and/or lead to higher operating voltage and thus lower power efficiency.[17]

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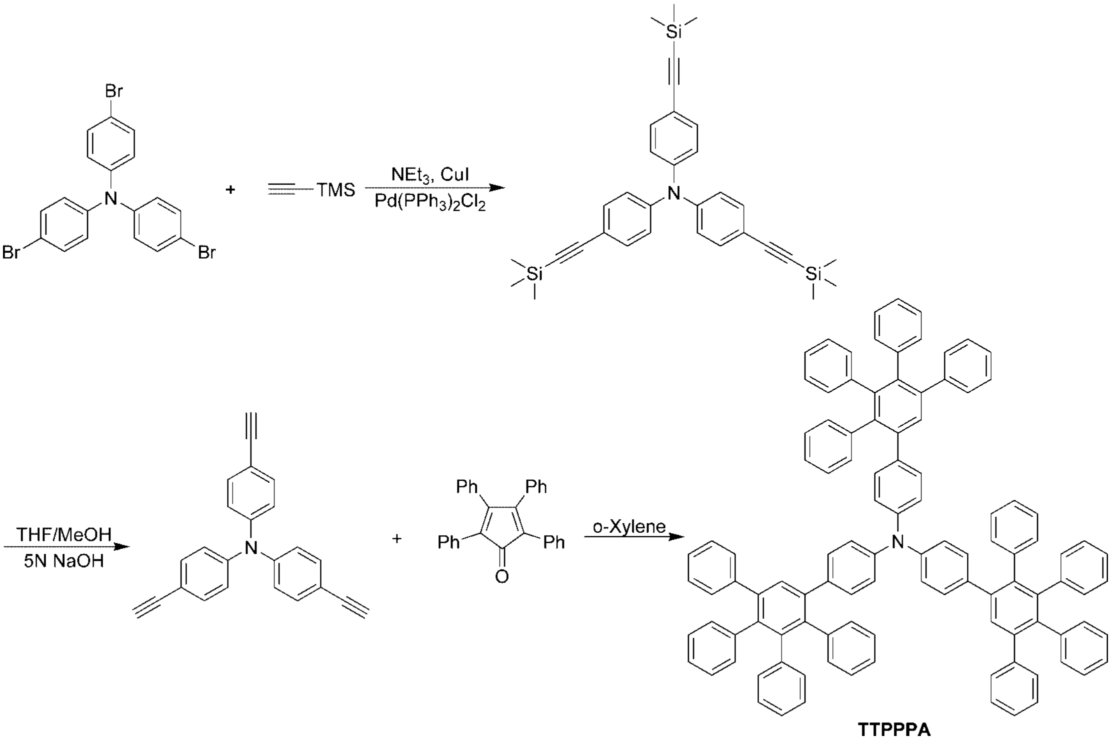
Scheme 1. Molecular Structure and Synthetic Procedure of TTPPPA



Other than high device efficiency, thermal and morphological stability of organic EL materials is important for OLEDs. For example, HTL having a high glass-transition temperature (T_g) is one of the requirements to realize high-stability OLEDs. Shirota et al. and Schmidt et al.[18–23] have reported several families of high T_g starburst aromatic amines are useful HTMs for OLED applications. Certainly, a useful HTM not only should have a high T_g but also a low barrier for hole injection from the anode. Therefore, it is important to increase T_g without compromising other important properties of HTM. Li et al.[24] reported a carbazole derivative TECEB as an HTM which has a relatively high T_g of 130 °C. It has hole mobility, energy levels, and performance in double-layer HTL/Alq$_3$ devices similar to those of NPB. Furthermore, as charge-transporting materials in OLEDs, they should have high optical transparency that allows the passage of light emitted from the device to ensure a high light collecting efficiency.[16] Therefore, a morphologically stable amorphous HTL having a moderate hole mobility to match the electron mobility of ETL, a low ionization potential (IP) for efficient hole injection, a high optical transparency to visible light, and good film-forming property are attributes for realizing highly efficient and stable OLEDs.

In this paper, we report the synthesis of a new starburst triphenylamine derivative 4,4′,4″-tris[(2,3,4,5-tetraphenyl)phenyl]phenylamine (TTPPPA) as shown in Scheme 1 and its application as an HTM in an Alq$_3$-emitting OLED. Although TTPPPA is comparable to NPB in terms of the highest occupied molecular orbital (HOMO) energy level, the TTPPPA-based device shows remarkably enhanced device efficiency over the corresponding NPB device. In addition, the T_g of TTPPPA is much higher than that of NPB, suggesting TTPPPA can be an alternative material to NPB especially in OLEDs and other devices for high-temperature operations. The mechanisms for enhancement in brightness and efficiency are discussed.

Experimental Section

Materials and Instruments. All the reagents and solvents used for the synthesis of TTPPPA were purchased from Aldrich and Acros companies and used without further purification. Alq$_3$ and NPB for EL device fabrication were purchased and purified by train sublimation. ^{1}H NMR spectra were recorded with a Bruker DPX-400 spectrometer (300 MHz). Elemental analyses (EA) were performed by the Flash EA 1112 method. Photoluminescence (PL) and absorption spectra of TTPPPA in solution and in films were recorded with a Perkin-Elmer LS50 fluorescence spectrometer and a Perkin-Elmer Lambda 2S UV–visible spectrophotometer, respectively. T_g and decomposition temperature (T_d) were measured determined by differential scanning calorimetry (DSC) with a Perkin-Elmer DSC7 and by thermogravimetric analysis (TGA) with a Dupont Instruments 983 Dynamic Mechanical Analyzer under N_2 atmosphere, respectively.

Preparation of 4,4′,4″-Tris[(trimethylsilylethynyl)phenyl] phenylamine. To a Schlenk tube containing Pd(PPh$_3$)$_2$Cl$_2$ (630 mg, 0.9 mmol), CuI (45 mg, 0.45 mmol), and 4,4′,4″-tribromophenylamine (7.23 g, 15 mmol) in NEt$_3$ (60 mL) was added trimethylsilylacetylene (6.63 g, 67.5 mmol). The mixture was frozen and thawed three times. Then it was heated to reflux in an oil bath for 48 h. The mixture was rinsed with ether and filtered; the solvent

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was then removed by rotary evaporation. Silica gel column chromatography (hexane) gives a pale yellow solid (6.05 g, yield 76%). MS: *m/z* 533. ^{1}H NMR (300 MHz, CD_2Cl_2) δ [ppm]: 0.19 (s, 27H), 6.98 (d, 6H, $J = 4.5$ Hz), 7.34 (d, 6H, $J = 4.5$ Hz).

Preparation of 4,4′,4″-tris[(2,3,4,5-tetraphenyl)phenyl]phenylamine (TTPPPA). To a solution of 4, 4′, 4″-tris [(trimethylsilylethynyl) phenyl] phenylamine (1.20 g, 2.25 mmol) in a mixture of THF (20 mL) and methanol (10 mL) was added 2 mL NaOH aqueous solution (5 N). The reaction mixture was stirred for 1 h at room temperature and then ethyl acetate (50 mL) was added. The mixture was washed with water and brine and dried with anhydrous sodium sulfate. After removal of the solvent, the residue was refluxed with tetraphenylcyclopentadienone (2.84 g, 7.39 mmol) in 50 mL of *o*-xylene under an argon atmosphere, and the resultant mixture was heated for 18 h at 170 °C (oil bath temperature). After the mixture was cooled to room temperature, ethanol (75 mL) was added. The precipitate was filtered, washed with ethanol (300 mL), and dried in vacuum. Silica gel column chromatography (dichloromethane/petroleum ether, 1:3) affords a white solid (2.80 g, 90%). ^{1}H NMR (300 MHz, CD_2Cl_2) δ [ppm]: 6.56 (d, 6H), 6.80–7.00 (m, 51H), 7.16 (m, 15H), 7.53 (s, 3H). MS: *m/z* 1386. Anal. Calcd for $C_{108}H_{75}N$: C, 93.54; H, 5.45; N, 1.01%. Found: C, 93.83; H, 5.52; N, 0.92%.

Fabrication and Characterization of OLEDs. An OLED was fabricated by vacuum deposition with a configuration of indium tin oxide (ITO) (30 Ω/□)/TTPPPA (70 nm)/Alq_3 (60 nm)/LiF(0.8 nm)/Al(100 nm). TTPPPA was used as the HTM, and Alq_3 was used as the electron-transporting and emitting material. Before loading into a deposition chamber, the ITO substrate was cleaned with detergent and deionized water for at least three times, then dried in an oven at 120 °C for about 2 h, and finally treated with UV ozone for 25 min. The device was fabricated by evaporating organic layers at a rate of 0.1–0.3 nm/s onto the ITO substrate sequentially at a pressure below 5×10^{-6} mbar. Onto the Alq_3 layer, a 0.8 nm-thick LiF was deposited as an electron injecting layer. Al (100 nm) was then deposited at a rate of 0.6 nm/s as the cathode. For comparison, a standard device with the same configuration but with TTPPPA replaced by NPB was also prepared under identical conditions. EL spectra and current–voltage–luminescence characteristics of the devices were measured with a Spectrascan PR 650 photometer and a computer-controlled DC power supply under ambient conditions. The emission area of the devices was 0.1 cm^2 as determined by the overlapping area of the anode and the cathode.

Results and Discussion

Design and Synthesis of TTPPPA. The general principles for designing morphologically stable molecule have been discussed by Shirota et al.,[18–20,25–31] Wirth,[32] and Naito and Miura.[33,34] A nonplanar molecular structure inhibiting easy packing and crystallization of molecules and an increased number of conformers in the molecule are the main premises in the design and synthesis of molecular glasses. Glass formation is enhanced by incorporation of bulky substituents. The corporation of bulky substituents also hinders translational, rotational, and vibrational motions of molecules, leading to an increase in T_g. Recently, a new type of fully conjugated polyphenylphenyl (tetraphenylphenyl and pentaphenylphenyl) dendron has been successfully incorporated into both small-molecular weight light-emitting materials[35,36] and polymeric light-emitting materials[37,38] through simple synthetic procedures.[39] Tetraphenylphenyl dendron has been proven to be effective molecular construction blocks to inhibit the formation of molecular aggregation or π–π stacking in the solid state.[37,38] The nonplanar dendron also renders the molecule with good thermal stability (high T_g) and film forming property.[36] As a result of the electron-donating nature of nitrogen atoms, triarylamines typically act as HTMs in various applications like xerography, light-emitting diodes, solar cells, photorefractive systems, and so forth. TTPPPA is a typical π-electron starburst derivative of triphenylamine, which includes three tetraphenylphenyl bulky groups. Therefore, it is expected to have a high T_g, good thermal stability, and good film-forming ability. TTPPPA was synthesized following the synthetic procedures outlined in Scheme 1. Palladium-catalyzed Sonogashira coupling reaction between the 4,4′,4″-tribromophenylamine and (trimethylsilyl)acetylene was employed to introduce the ethynyl group into the aromatic ring. When a molar number of 4.5 (trimethylsilyl)acetylene was used, good to excellent yield of trisubstituted product could be obtained (yield 76%). Tetraphenylphenyl group was introduced into the molecular backbones through the simple Diels–Alder cycloaddition reaction between the arylethynyl and tetraphenylcyclopentadienone in high yield (90%).

Thermal Properties of TTPPPA. Thermal stability of TTPPPA was measured with DSC and TGA analysis. Figure 1 shows the DSC thermograms of TTPPPA measured at a heating rate of 10 °C/min. When TTPPPA was heated and cooled quickly by swirling water, an amorphous glass was spontaneously formed. When the amorphous glass of TTPPPA was heated for the second run, a glass transition was observed obviously at 202 °C, which is taken as the T_g of TTPPPA, and is significantly higher than that of the most commonly used HTM (e.g., T_g of NPB is 98 °C) and comparable to the T_g of m-TDATA ($T_g = 202$ °C).[18] The T_g reading can be repeatedly reproduced following the above procedure. After three such heating cycles, the properties of TTPPPA, including ^{1}H NMR and mass spectra, and photophysical spectra, remain almost the same, indicating that TTPPPA is

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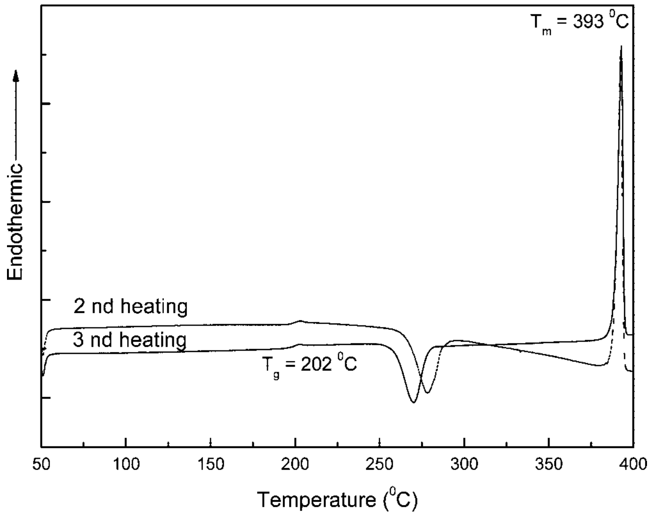

Figure 1. DSC curves of TTPPPA at a scanning rate of 10 °C /min.

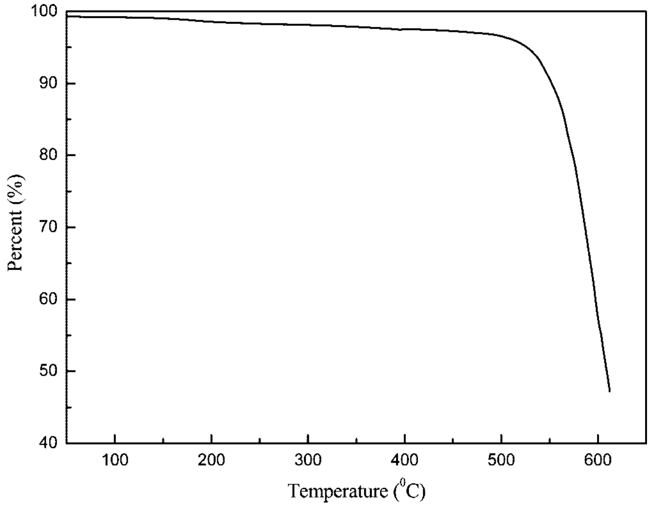

Figure 2. TGA thermogram of TTPPPA at a heating rate of 10 °C /min.

thermally stable. As depicted in Figure 1, TTPPPA has a melting point at 393 °C, which remains fairly constant after several heating cycles. The thermal stability of TTPPPA was further confirmed by the TGA results shown in Figure 2. A weight-loss transition corresponding to the endothermic evaporation was detected at 500 °C. Before and during evaporation, no decomposition was observed.

Photophysical Properties of TTPPPA. Figure 3 shows the absorption and fluorescence spectra of a vacuum-deposited film of TTPPPA on a quartz substrate. The absorption spectrum of the TTPPPA film shows the characteristic absorption peaks of triphenylamine between 300 and 400 nm ($\lambda_{\max} = 350$ nm). TTPPPA hardly absorbs light at wavelengths longer than 400 nm, indicating its high transparency to visible light to ensure a high light collecting efficiency. The PL spectrum of the TTPPPA film is structureless and mirrors the low-energy absorption band. TTPPPA is moderately fluorescent with emission peaking at 410 nm, and the luminescence spectrum is narrow with a full width at half-maximum of 45 nm.

The HOMO energy level of TTPPPA was determined as 5.46 eV by ultraviolet photoelectron spectroscopy (UPS). The HOMO–LUMO (lowest unoccupied molecular orbital) band

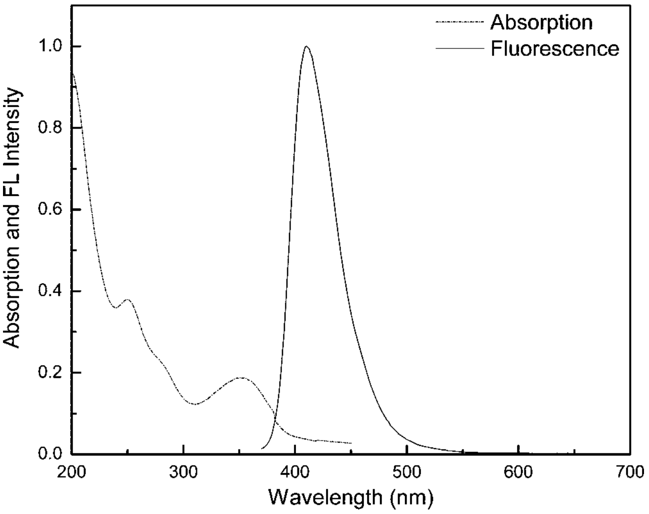

Figure 3. Normalized absorption (dashed line) and fluorescence (solid line) spectra of TTPPPA in pure films on quartz substrate.

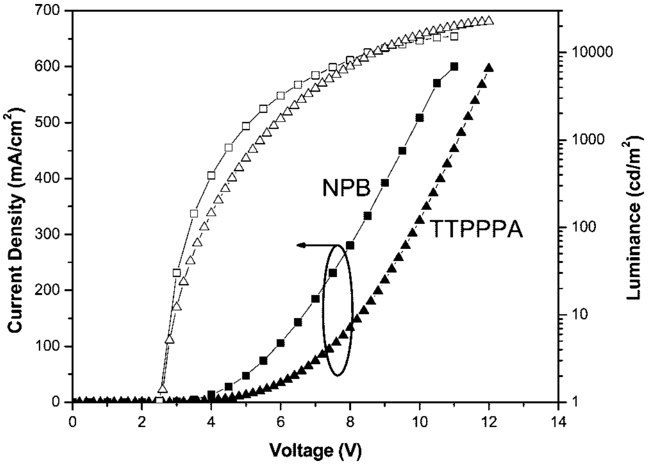

Figure 4. Current density–voltage and EL brightness–voltage characteristics of ITO/TTPPPA/Alq_3/LiF/Al (△) and ITO/NPB/Alq_3/LiF/Al (□) devices.

gap of TTPPPA was determined from the optical absorption spectrum of TTPPPA film on the quartz substrate to be 3.1 eV. The LUMO energy level was calculated to be 2.36 eV by subtraction of the optical band gap from the HOMO energy level. The HOMO and LUMO energy levels of TTPPPA are comparable to those of NPB (HOMO, 5.4 eV; LUMO, 2.3 eV).[40]

EL Performances of TTPPPA-Based Devices. Both the TTPPPA-based device and the NPB-based standard device showed pure green emission from Alq_3(523 nm), suggesting that the hole and electron recombination is located in the Alq_3 layer and TTPPPA acts primarily as a hole transporter without causing exciplex formation at the interface with Alq_3. The current density–voltage and EL brightness–voltage characteristics of the two devices are shown in Figure 4. It shows that current density at the same driving voltage is generally smaller in the TTPPPA-based device. The TTPPPA-based device has a turn-on voltage of 2.54 V (at 1 cd/m^2) and a brightness of 22 660 cd/m^2 at 12 V (which is the upper measuring limit of our instrument), while the standard device

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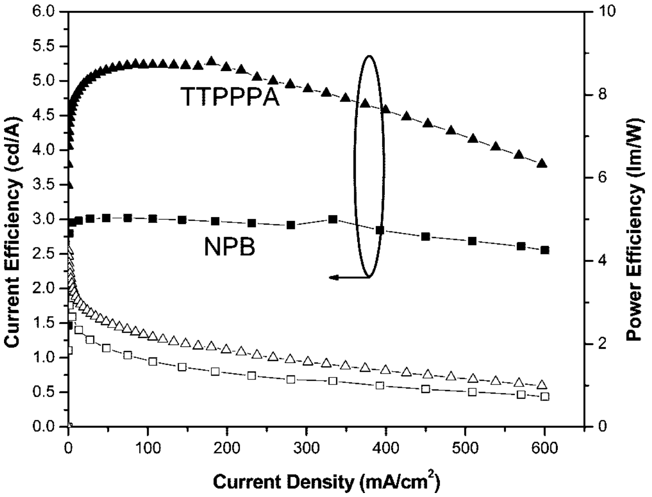

Figure 5. Current efficiency–current density and power efficiency–current density characteristics of ITO/TTPPPA/Alq_3/LiF/Al (△) and ITO/NPB/Alq_3/LiF/Al (□) devices.

has almost the same turn-on voltage of 2.54 V and a maximum luminescence of 15 320 cd/m^2 at 11 V. The superiority in brightness of the TTPPPA-based device to the standard device is much more obvious in the high driving voltage region. At a given voltage, the current density of the TTPPPA-based device is lower than that of the standard device. Figure 5 shows that the TTPPPA-based device gives a maximum current efficiency of 5.3 cd/A, which is substantively higher than that (3.0 cd/A) of the standard device. Although the TTPPPA-based device has slightly higher operating voltages than the standard device, the power efficiency of the former can reach a high value of 4.26 lm/W, which is much higher than that of (2.92 lm/W) of the standard device.

The significant enhancement in the brightness and efficiency of the TTPPPA-based device can be attributed to better balanced charge recombination at the emitting interface. Because the anode, cathode, electron injecting layer, ETL, and the emitter layer in the TTPPPA-based device and the standard device are the same, the current density decrease should be associated with a larger hole injection barrier at the interface between ITO and TTPPPA and/or a reduced hole current through the HTL. Considering the HOMO level of TTPPPA (5.46 eV) is almost the same as that of NPB (5.4 eV), the hole injection barrier difference between the TTPPPA-based device and the standard device is expected to be small. Therefore, the smaller hole mobility in the TTPPPA layer should be the dominant factor that influences the current density–voltage characteristics. The hole-drifting mobility of TTPPPA was determined by transient EL measurements on the TTPPPA single-layer device using our previous method.[41,42] Indeed, the hole mobility of TTPPPA is determined to be 5×10^{-5} cm^2 V^{-1} s^{-1}, which is smaller than that of NPB (10^{-3} cm^2 V^{-1} s^{-1}) but comparable to that of Alq_3 (10^{-5} cm^2 V^{-1} s^{-1}).[43,44] The usage of TTPPPA with a moderate hole mobility would decrease the hole current and thus the excess holes at the emitting interface. This analysis is also supported by the lower current density of the TTPPPA-based device at the same voltage relative to that of the standard device (Figure 4). Consequently, injection of holes and electrons into the emitting layer is better balanced, and thus the brightness and efficiency are significantly enhanced in the TTPPPA-based device. In addition, the good film-forming property of TTPPPA may also contribute to benefit the EL brightness and efficiency and device stability.

To summarize, a new starburst amorphous molecule, TTPPPA, was designed and synthesized for usage as a HTM in OLEDs. The TTPPPA-based OLED device exhibited significantly enhanced EL brightness and better efficiency compared to the standard NPB-based device. The smaller hole mobility in the TTPPPA layer offered a better balance of hole and electron currents and thus enhanced recombination, resulting in the improved brightness and efficiency. Moreover, TTPPPA has a T_g considerably higher than that of NPB, and thus expectedly better morphological stability and thermal durability. On the basis of the merits of TTPPPA including high T_g, good film-forming ability, high transparency to visible light, and moderate hole mobility, TTPPPA should be a promising alternative material to NPB especially for high-temperature applications of OLEDs and other organic electronic devices.

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