



Small molecular hole-transporting and emitting materials for hole-only green organic light-emitting devices



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ABSTRACT

Three small molecular hole-transporting and emitting materials with naphthalene bridged triphenylamine or carbazole were synthesized. These compounds exhibited high absolute fluorescence quantum yields (70–88%) at the green light region (540–555 nm), suitable highest occupied molecular orbital levels (−5.26 ~ −5.34 eV) and good hole mobilities ($\sim 10^{-4}$ cm² V^{−1} s^{−1}). The hole-only organic light emitting diodes with as-synthesized hole-transporting and emitting materials showed a tune-on voltage and a maximum brightness of 2.6 V and 2010 cd m^{−2}, respectively. Three stages of voltage-dependent current were observed, which coincide with the characteristic of Ohmic current, Fowler-Nordheim current and space charge limited current. With analysis of above models, overlarge Fowler-Nordheim current and space charge limited current contribute to the low efficiency of the device. All these results indicated that the high hole mobility showed the negative effect on the device efficiency, especially at high operation voltage.

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1. Introduction

The research on multilayer organic light-emitting diodes (OLEDs) is dominant in recent decades for the outstanding performance [1,2]. While, simplifying device structure is still regarded as one of the most important solution for low cost OLEDs [3,4]. This requires that the material of active layer should be multifunctional with charge transfer and light emission. In earlier studies, 8-hydroxyquinoline aluminum (Alq₃) derivatives were applied as emitting and electron-only OLEDs fabricated with vacuum thermal deposition, which showed poor performance of 210 cd m^{−2} at 15 V and the highest current efficiency reached 0.17 cd A^{−1} at 40 cd m^{−2} [5,6]. The solution-processed OLEDs with hole-transporting character mainly refer to electroluminescence polymer, such as poly(*p*-phenylene vinylene) (PPV) [7]. However, the uncertain molecular structure and weight of polymer lead to the complicated synthesis and purification processes. Therefore, the simple-structured small

molecule OLEDs (SMOLEDs) with solution process has become an exciting way for the development of OLEDs [8,9].

As reported, the triphenylamine (TPA) and carbazole (Cz) derivatives have been normally used as small molecular hole-transporting materials (HTMs) [10–14]. Previously, our group have reported a series of TPA based HTMs with outstanding film-forming character [15,16]. In addition, TPA and Cz based materials with π -conjugation bridge showed excellent performance as fluorescence emission materials [17,18], such as 4,4'-di-2-(4-*N,N'*-diphenylamino phenyl vinyl biphenyl (DPAVBi) and 4,4'-bis((*E*)-2-(9-ethyl-9*H*-carbazol-3-yl) vinyl)-1,1'-biphenyl (BCzVBi), which were widely used in multilayer blue OLEDs.

To these viewpoints, three small molecular hole-transporting and emitting materials (HT-EMs) with naphthalene bridged TPA or Cz were synthesized (Molecular structures see Fig. 1, synthesis routes see ESI Scheme S1). These as-synthesized HT-EMs showed hole mobility (μ_h) of 4.03×10^{-4} , 4.22×10^{-4} and 3.24×10^{-4} cm² V^{−1} s^{−1} at the electric field of $\sim 1.5 \times 10^5$ V cm^{−1}, respectively. Fully solution-processed simple-structured OLEDs (Poly(3,4-ethylene-dioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) and poly[(9,9-bis(3'-(*N,N*-dimethylamino)propyl)-

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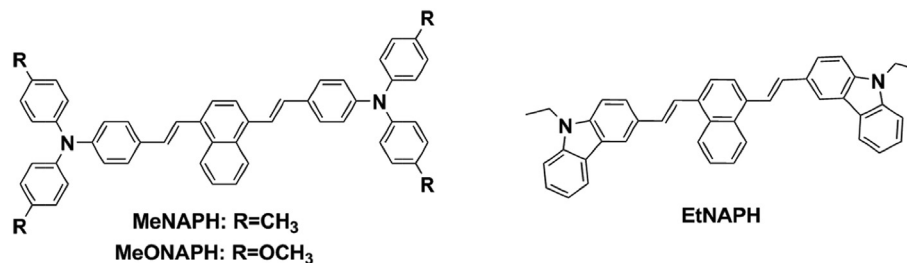


Fig. 1. Molecular structures of as-synthesized HT-EMs.

2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) were applied as the electrode modified materials, HT-EMs as light-emitting materials) were fabricated and showed tune-on voltages, the maximum brightness, maximum luminance efficiency and external quantum efficiency were 2.6 V, 2010 cd m⁻², 0.65 cd A⁻¹ and 0.25%, respectively. In this kind of device, three stages have been observed in the *J-V* curves based on different operation voltage, which showed a feature of Ohmic type current (J_{ohmic}), Fowler-Nordheim type current (J_{FN}) and space charge limited type current (J_{SCLC}). In the second and third stages, the J_{FN} and J_{SCLC} showed a directly proportional to μ_h . In this work, the hole mobility showed an negative effect on the efficiency in simple-structured device. An equilibrium of carrier transfer plays a key role in achieving high performance in simple-structured OLEDs.

2. Experimental section

2.1. General information

1,4-naphthalenedicarboxylic acid, poly(3,4-ethylene-dioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) and Aluminum (Al, 99%) were purchased from Tianjin Heowns Biochemical Technology Co. Ltd. (Tianjin, China), other reagents were purchased from Tianjin Guangfu Fine Chemical Research Institute (Tianjin, China). Indium-tin-oxide (ITO) glasses were purchased from South China Xiangcheng Technology Co. Ltd. (Shenzhen, China). Poly[(9,9-bis(3'-(*N,N*-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctyl fluorene)] (PFN) was provided by South China University of Technology. Tetrahydrofuran (THF) was distilled before use. All the other agents were used without further treatments.

As-synthesized HT-EMs were identified by nuclear magnetic resonance (NMR) and mass spectroscopies (MS). The NMR were recorded on a Bruker AVANCE III 400 MHz spectrometer, with the chemical shifts reported in ppm using tetramethylsilane (TMS) as an internal standard. MS was recorded on a LCQ Advantage MAX mass spectrometer. Ultraviolet–visible absorption (UV–Vis) and photoluminescence (PL) spectra were obtained on the Thermo Evolution 300 UV–Visible spectrometer and a Cary Eclipse fluorescence spectrometer, respectively. Fluorescence lifetime was obtained by FluoroLog 3 Steady transient fluorescence test system. Fluorescence quantum yields were obtained on a Varian Cary Eclipse test system. Decomposition temperature (T_d) and glass transition temperature (T_g) were determined by the thermal gravimetric analysis (TGA) and differential scanning calorimeter (DSC) on a TA Q500 thermo gravimetric analysis under nitrogen atmosphere. The photoelectron yield spectroscopy (PYS) was carried out on the Sumitomo PYS-202 ionization energy detection system. The morphology of solid film was characterized by a Rigaku Miniflex 600 X-Ray diffraction (XRD) and Nanosurf AG Easyscan 2 atomic force microscopy (AFM).

Crystal structure of MeNAPH was obtained by Smart Apex X-Ray single crystal diffraction. X-Ray crystallography of MeNAPH were

recorded with the graphine monochromated Mo- $K\alpha$ radiation ($\lambda = 0.71073 \text{ \AA}$) on a Bruker P4 four-circle diffractometer. The structure was solved with a SHELXS-97 program [19]. Non-hydrogen atoms were located in successive difference Fourier syntheses. The final refinement was performed by full matrix least-squares methods with anisotropic parameters for nonhydrogen atoms on F^2 using the SHELXTL and ORTEP software [20]. Crystallographic data reported in this paper had been deposited with the Cambridge Crystal Data Centre and CCDC No. 1413201.

The time-of-flight (TOF) measurement was recorded on a TOF401 (Sumitomo Heavy Industries, Ltd., Japan). Samples were prepared through vacuum deposition with a structure of ITO/HT-EMs (~1 μm)/Al (100 nm) and an active area of $4 \times 4 \text{ mm}^2$.

2.2. Quantum chemical calculation

Quantum chemical calculation was performed on a Gaussian 03 program with the Beck's three-parameter exchange functional and the Lee-Yang-Parr's correlation functional (B3LYP) using 6-31G (d) basis sets [21].

2.3. OLED fabrication and measurements

ITO glasses (sheet resistance: 10 Ω /square) were cleaned with following sequence: acetone, methanol and diluted water, then annealed at 120 °C for 20 min followed by O₂ plasma treatment [22]. An anode modified layer (~20 nm) was achieved by spin-coating the PEDOT: PSS colloidal solution on the ITO side at 3000 rpm, then baked at 120 °C for 30 min. The 20 mg mL⁻¹ concentrated HT-EMs THF solutions were spin-coated on the PEDOT:PSS film at 2000 rpm, and 80 nm thick HT-EMs were achieved after baking at 80 °C for 30 min. The cathode modified layer of PFN (~20 nm) was deposited by spin-coating at 3000 rpm, followed by baking at 80 °C for 20 min. A 100 nm thickness of Al was thermally deposited as a cathode under vacuum at a base pressure $<1 \times 10^{-4} \text{ Pa}$, monitored by the Sycon STM-100 crystal thickness meter. The thicknesses of all spin-coated films were measured by the TencorAlfa Step-500 terrace detector. The active area of the device is $4 \times 4 \text{ mm}^2$, determined by the cross breadth between the cathode (Al) and the anode (ITO). The EL spectra was measured by the SpectrScan PR705 spectra. Steady current-luminance-voltages were measured by the Keithley 236 with silicon photodiode. All tests were performed under air atmosphere at room temperature.

3. Results and discussions

3.1. Molecular structures and energy levels

The as-synthesized HT-EMs were synthesized by Wittig reaction (ESI Scheme S1). The HT-EMs are soluble in common organic solvents such as toluene, tetrahydrofuran, chloroform and chlorobenzene, which ensure they can be deposited with solution

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