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Luminance materials containing carbazole and triphenylamine exhibiting high hole-transporting properties

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ABSTRACT

Two novel 2-substitutd-8-hydroxyquinoline derivatives were designed and synthesized. Their luminance properties and carrier transporting abilities were studied when integrated in four different organic light-emitting device structures. The four devices are structured as following—device A: indium tin oxide glass substrate (ITO)/2-TNATA/NPB/(2-[2-(9-ethyl-9H-carbazol-2-yl)-vinyl]-quinolato zinc (1) and 2-[2-(4-diphenylamino-phenyl)-vinyl]-quinolato zinc (2))/Alq₃/LiF/aluminum (Al); device B: ITO/2-TNATA/NPB/1 or 2/LiF/Al; device C: ITO/2-TNATA/1 or 2/Alq₃/LiF/Al; device D: ITO/2-TNATA/1 or 2/Alq₃/LiF/Al. In the device A, the maximum brightness of compound 1 is 5857 cd m⁻² at 11 V with the luminance efficiency 1.84 cd A⁻¹. For 2 it is 6047 cd m⁻² at 10 V with an efficiency of 2.22 cd A⁻¹. In the device B, the maximum brightness of 1 is 745 cd m⁻² at 11 V and efficiency of 1.99 and 1.71 cd A⁻¹. In the device D, the maximum brightness and efficiency of 1 are 8512 cd m⁻² at 13 V and 3.10 cd A⁻¹. and they are 9818 cd m⁻² at 13 V and 0.42 cd A⁻¹, for 2. Our results also show that both 1 and 2 are good bipolar and bifunctional molecules with high hole-transporting and luminance properties.

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1. Introduction

During the past decade, organic light-emitting diodes (OLED) have been one of the most intensively explored topics in chemistry and applied physics owing to their wide applications in full-color displays with color filters, backlighting for liquid crystal displays, and general lighting sources [1–7]. While a large number of organic materials exhibit high fluorescence quantum efficiencies and bright in the visible region, there are some drawbacks such as low stability, monocolor emission and high driving voltage associated with these materials. Therefore, they are not suitable yet for use in large area flat-panel displays. To overcome these problems and improve properties of the electroluminescence (EL) devices, it is highly desirable to design and synthesize new dipolar and multifunctional mate-

rials. The most popular way to improve device performance is to use good emitters with bipolar charge transport property, which can maintain well the carrier balance. However, it remains a huge challenge in designing such molecules.

Carbazole and triphenylamine (TPA) derivatives have been extensively studied and developed as hole-transporting materials because of their high hole mobilities [8-11]. The triarylamine-containing compounds such as NPB (N,N')bis(naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine) [12] TPD (N,N'-di-m-tolyl-N,N'-diphenyl-1,10-biphenyl-4,40and diamine) [13] exhibit excellent hole-transporting properties, they generally produce amorphous films with poor morphological stability. Many compounds containing carbazole units are used as effective hole-transporting materials due to the nitrogen atom in the carbazole ring bestows an electron-donating ability, and they possess better morphological stability and thermal durability [14,15]. Following Tang's research work [16], some other metal complexes containing 8-hydroxyquinoline derivatives have been extensively used as emitters in OLED [17-20].

In this paper, we report the synthesis and device fabrication involving 2-substituted 8-hydroxy-quinoline metal derivatives containing carbazole and triphenylamine units (E)-2-(2-(9-ethyl-9H-carbazol-3-yl)vinylquinolato-zinc (**1**), and (E)-2-(4-(diphenyl-



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Scheme 1. The structures of (E)-2-(2-(9-ethyl-9H-carbazol-3-yl)vinylquinolin-zinc (1) and (E)-2-(4-(diphenyl-amino)styryl)quinolin-zinc (2).

amino)styryl)quinolato-zinc (**2**) (Scheme 1). Synthesis of **1** and **2** has been reported elsewhere [21,22]. The combination of 2-substituted 8-hydroxyquinoline with triphenylamine and carbazole is expected to lead to new dipolar and multifunctional materials with excellent performance. Optical and thermal properties of the compounds have been studied. Electroluminescent devices based on these compounds as emission layer (E), hole-transporting layers (HTL) or E&HTL have been fabricated and characterized. The effect of different TPA and carbazole groups on the hole-transport and emission performance of the corresponding compounds has been examined.

2. Experimental details

2.1. Optical properties of the compounds

Photoluminescence of the compounds were measured in solid state using a HITACHIF-2500 Fluorophotometer. Thermogravimetric analysis (TGA) was carried out on a Thermal Analyzer (TGA2050, TA Instruments, USA). The decay curves were measured by a FLS920-Combined fluorescence lifetime and Steady State Spectrometer (EDINBURGH INSTRUMENTS).

2.2. Devices fabrication

The ITO-coated glass substrate was first immersed sequentially in ultrasonic baths of acetone, alcohol and deionized water for 10 min, respectively, and then dried in an oven. The resistance of a sheet ITO is 50 Ω/\Box . The devices were fabricated in a multi-source organic molecule gas deposition system. There are different materials in every source, and the temperature of every source can be controlled independently. Different organic materials were deposited on the ITO-coated glass substrate according to the designed structure, LiF buffer layer and Al were deposited as a co-cathode under a pressure of 5×10^{-4} Pa. Electroluminescent spectra and commission international De L' Eclairage (CIE) coordination of these devices were measured by a PR655 spectra scan spectrometer. The luminance-current-voltage characteristics were recorded simultaneously with the measurement of the EL spectra by combining the spectrometer through a Keithly model 2400 programmable voltage-current source. The layer thicknesses of the deposited materials were monitored in situ using a model FTM-V oscillating quartz thickness monitor made in Shanghai, China. All the measurements were carried out at room temperature under ambient conditions.

In order to study the material properties of **1** and **2**, every compound was fabricated into four different structures devices. The device structures and related information are outlined in Fig. 1, where ITO glass is transparent anode; N,N'-bis(naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) is HTL or emitting material (EM); N-(naphthalene-2-yl)-N',N'-bis(naphthalene-2-yl(phenyl)amino)phenyl)-N-phenylbenzene-1,4-diamine(2-TNATA) is used as hole-injecting material(HIM); Tris-8-hydroxyl

quinolato-aluminum (Alq₃) is electron-transporting material; $\mathbf{1}$ and $\mathbf{2}$ is HTL or EM in all devices. LiF is the buffer layer.

2.3. Preparation of compounds 1 and 2

The synthetic processes of **1** and **2** were according to the published papers [21,22]. A mixture of 2-methyl-8-hydroxyquinoline (25 mmol), 9-ethyl-9*H*-carbazole-3-carbal-dehyde (25 mmol for **1**) or 4-(diphenylamino)benzaldehyde (25 mmol for **2**) and acetic anhydride (25 mL) were stirred and heated at 125–135 °C for 40 h under nitrogen atmosphere. After cooling, the mixture was poured into ice water (100 mL) and stirred 4–5 h, and a brown black solid was obtained by filtration. Then the solid was dissolved in DMF (30 mL) by heating at 120 °C, aqueous hydrochloric acid (36–38%, 10 mL) was added to the solution, and the mixture was heated at 125–135 °C for 2 h. The precipitated orange solid was filtered off, washed with water and dried in vacuum.

A flask was charged with a mixture of the orange solid (9 mmol) and DMF (25 mL). The mixture was heated under reflux, then triethylamine (2.1 g, \sim 20 mmol) was added, and immediately a clear solution was obtained. After completion of the reaction, the mixture was poured into ice water (100 mL), the obtained yellow solid was filtered off, then washed with water and dried to afford crude ligards of **1** and **2**. The product was purified by silica gel (100–200 mesh) column chromatography using ethyl acetate/petroleum ether as an eluent. Ligard of 1, yield 82.42%. m.p. 140-141 °C; FT-IR (KBr) ν (cm⁻¹): 3392.59, 3037.03, 2972.83, 2923.45, 1618.04, 1593.40, 1561.36, 1507.15, 1489.90, 1477.58, 1332.19, 1245.94, 1231.16, 962.55, 748.17, 587.99; ¹H NMR (DMSO); δ: 9.48 (s, 1H), 8.53 (s, 1H), 8.30 (d,1H, J = 16.08 Hz), 8.27 (d, 1H, J = 8.53 Hz), 8.21 (d, 1H, J = 7.66 Hz), 7.87 (d, 1H, J = 8.55 Hz), 7.79 (d, 1H, J = 8.58 Hz), 7.68 (d, 1H, J = 8.56 Hz), 7.63 (d, 1H, J = 8.56 Hz), 7.50 (d, 1H, J = 16.11 Hz),7.49(t, 1H, J = 8.14 Hz), 7.37(q, 2H, J = 7.18 Hz), 7.26(t, 1H, J = 7.60 Hz),7.10(d, 1H, J = 6.48 Hz), 4.47(q, 2H, J = 7.20 Hz), 1.35(t, 3H, J = 7.15 Hz).EI-Ms *m*/*z* (M+): 364. Anal. Calcd for C₂₅H₂₀N₂O: C 82.42; H 5.49; N 7.69. Found C 82.31; H 5.81; N 7.55; ligard of 2, yield 25.69%. m.p. 152–153 °C; FT-IR: ν (cm⁻¹): 3447.17, 3034.92, 1637.02, 1587.72, 1565.93, 1509.27, 1491.83, 1462.84, 1327.16, 1280, 830, 749, 694, 620; ¹H NMR [(CD₃)₂CO]; δ : 7.03 (d, 2H, J=6.74 Hz), 7.09–7.13 (m, 7H), 7.32–7.42 (m, 6H), 7.36 (d, 1H, J=15.84 Hz), 7.62 (d, 2H, J=8.61 Hz), 7.77 (d, 1H, J=8.60 Hz), 8.01 (d, 1H, J=16.18 Hz), 8.25 (d, 1H, I = 8.32 Hz). ESI-Ms m/z (M+H): 415. Anal. Calcd for C₂₉H₂₂N₂O: C 84.06; H 5.31; N 6.76. Found: C 83.82; H 5.46; N 6.66.

Then, 15 mL zinc acetate solution in anhydrous methanol (0.033 mol/L) was added dropwise to the solution of 1 mmol ligards of **1** and **2** in anhydrous methanol (50 mL) under stirring, and then the mixture was stirred for 24 h. Yellow precipitates were obtained, then filtered off, washed with methanol and DMF 3–5 times, respectively, and dried in vacuum. Compound **1**, yield 87.6%, m.p. \geq 300 °C; elemental analysis: found C 74.31, H 4.94, N 6.73 (%), calculated for (EtCzHQ)₂Zn·H₂O (**1**): C 74.17; H 4.94; N 6.92 (%); IR (KBr) ν (cm⁻¹): 3048, 2969, 2929, 1616, 1586, 1502, 1493, 1477, 1435, 1386, 1297, 1231, 1101, 829, 745, 472; compound **2**, yield 83.5%, IR (KBr) ν (cm⁻¹): 3449, 3191, 1628, 1591, 1281, 1178,830, 748, 697, 614; ele-

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