



Tailoring electronic structure of organic host for high-performance phosphorescent organic light-emitting diodes



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ABSTRACT

We investigated highly efficient phosphorescent organic light-emitting diodes (PHOLEDs) based on three novel 1,3,5-triazine derivatives as the host materials and two kinds of iridium complexes as the guests, respectively. For comparison, the devices using a common phosphorescent host 4,4'-N,N'-dicarbazolebiphenyl (CBP) have also been fabricated. Results show that the devices using 9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-9H-carbazole (PTC) and 4-(4,6-diphenoxy-1,3,5-triazin-2-yl)-N,N-diphenylaniline (POTA) as host have better performance than that of CBP. In comparison with the PHOLEDs based on CBP host, PTC- and POTA-based PHOLEDs show significantly lower driving voltages and higher power efficiencies. The high bipolar carrier mobility of the host is found to be critical to this kind of doping system, which would balance the injection of both carriers and improve efficiency.

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1. Introduction

Organic light-emitting diodes (OLEDs) possess high potential in the development of full-color flat-panel displays and lighting [1–6]. In this regard, fabrication of OLEDs with higher power efficiency becomes essential. Phosphorescent organic light-emitting diodes (PHOLEDs) have attracted considerable attention as they can harvest both singlet and triplet excitons and achieve, in theory, 100% internal quantum efficiency, corresponding to a four-fold improvement in efficiency relative to that achievable in single-harvesting fluorescent OLEDs [7–13]. In general phosphorescence emitters are doped into a host [14–16], and the dopant molecules usually get excited through direct charge trapping or energy transfer from the host

[17,18]. However, for most of PHOLEDs, the device quantum efficiency drops rapidly with increasing current density, as triplet excitons relax more slowly and the emission inevitably reaches saturation through a quenching mechanism involving triplet–triplet annihilation [19]. In order to alleviate the problem, the host materials are significant to possess high bipolar carrier mobility and suitable highest occupied molecular orbital (HOMO)/lowest unoccupied molecular orbital (LUMO) levels that match the neighboring layers [20,21]. To achieve this goal, tremendous effort has been devoted. In general, the bipolar host materials possess electron-transporting groups (electron acceptors) and hole-transporting units (electron donors) [22]. The donor–acceptor interaction of bipolar host materials is very critical, which have an influence on triplet energies. In addition, the HOMO–LUMO energy band gap (E_g) and triplet level (E_T) of the host materials should be higher than those of phosphorescent dopants,

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which can prevent reverse energy transfer from the guest back to the host. Nevertheless, a host material with a high E_T often has a wide E_g as a result of its typically large singlet–triplet energy difference (ΔE_{ST}) [23]. The host material with large E_g usually leads to direct charge trapping within the emitting layer (EML), which will finally produce poor conductivity in host material and high operation voltages in phosphorescent devices as result [24]. Host materials with narrow E_g are reported to have better performances [25–27]. In this case, design of bipolar host material with high E_T and narrow E_g is important.

In this study, we report a new series of 1,3,5-triazine derivatives, 2,4,6-tris(4-(9H-carbazol-9-yl)phenyl)-1,3,5-triazine (TCT), 9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-9H-carbazole (PTC) and 4-(4,6-diphenoxy-1,3,5-triazin-2-yl)-N,N-diphenylaniline (POTA), which contain 1,3,5-triazine as the electron-accepting core and carbazole-containing electron acceptors. The three compounds were used as hosts for green and red iridium complex guests for highly efficient OLEDs. Since Baldo et al. developed the concept of PHOLEDs [28], 4,4'-N,N'-dicarbazolebiphenyl (CBP) has been used widely as a host material for PHOLEDs because of its suitable E_T and good hole-transporting ability [29–32]. We compared the electroluminescent (EL) properties of OLEDs based on the three 1,3,5-triazine derivatives with the devices using CBP as the host. We also investigated the effect of structural change on the performance of host: dopant system.

2. Experimental

2.1. General information

NMR spectra were determined on a Bruker Advance-400 spectrometer with chemical shifts reported as ppm. Mass spectrum data were obtained with a Finnigan 4021C GC-MS spectrometer. Phosphorescence spectra at 77 K were recorded with a Hitachi fluorescence spectrometer F-4600. The film UV–vis absorption spectra were performed with Lambda 750, PerkinElmer. Ultraviolet photoemission spectroscopy (UPS) was measured in a Kratos AXIS Ultra DLD ultrahigh vacuum surface analysis system. UPS analysis was performed with an unfiltered He I (~21.2 eV) gas discharge lamp and a total instrumental energy resolution of 100 meV. The Fermi level (E_F) is referred to as the zero binding energy (BE) in the UPS spectra.

2.2. Synthesis

Starting chemicals and reagents were commercially available and used without further purification. Compounds TCT, PTC and POTA were synthesized via palladium-catalyzed Suzuki reactions by using chlorine-substituted 1,3,5-triazine units with 4-(9H-carbazol-9-yl)phenylboronic acid or 4-(diphenylamino)phenylboronic acid.

TCT: Toluene (12 ml), ethanol (6 ml), and 2 M aqueous Na_2CO_3 (9 ml) were added to a mixture of 2,4,6-trichloro-1,3,5-triazine (0.368 g, 2 mmol), 4-(9H-carbazol-9-yl)phenylboronic acid (1.894 g, 6.6 mmol), and tetrakis(triphenylphosphine)platinum (0.234 g, 0.2 mmol). The mixture was refluxed for 26 h under a nitrogen atmosphere.

When cooled to room temperature, the mixture was extracted with dichloromethane and dried over Na_2SO_4 . After the solvent had been removed under reduced pressure, the residue was purified by column chromatography on silica gel using dichloromethane–petroleum ether (1:1) as the eluent to give a light yellow solid (0.547 g, 34%). ^1H NMR (400 MHz, DMSO-d_6) δ (ppm) 8.89 (d, $J = 8.6$ Hz, 6H), 8.29 (d, $J = 7.7$ Hz, 6H), 7.94 (d, $J = 8.6$ Hz, 6H), 7.58 (d, $J = 8.2$ Hz, 6H), 7.53–7.45 (m, 6H), 7.35 (t, $J = 7.4$ Hz, 6H). ^{13}C NMR spectra was unavailable due to the poor solubility. MS-MALDI $[\text{M} + \text{H}]^+$: calcd 805.957. Found: 805.032.

PTC: This compound was synthesized according to the synthesis procedure of TCT from the intermediates of 2-chloro-4,6-diphenyl-1,3,5-triazine and 4-(9H-carbazol-9-yl)phenylboronic acid. ^1H NMR (400 MHz, CDCl_3) δ (ppm) 9.03 (d, $J = 8.5$ Hz, 2H), 8.82 (dd, $J = 11.6, 10.0$ Hz, 4H), 8.18 (d, $J = 7.7$ Hz, 2H), 7.83 (d, $J = 8.5$ Hz, 2H), 7.70–7.53 (m, 8H), 7.46 (t, $J = 7.3$ Hz, 2H), 7.34 (t, $J = 7.3$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.48, 170.15, 150.67, 136.70, 129.97, 133.25, 130.22, 129.41, 128.54, 128.38, 127.98, 124.33, 123.42, 120.97, 121.22, 110.41. MS-EI $[\text{M}]^+$: calcd 474.1844. Found: 474.1440.

POTA was synthesized following the reported procedure [33]. The molecular structures of these three compounds are shown in Fig. 1.

2.3. Device fabrication and measurement

Patterned indium tin oxide (ITO, 6 Ω/sq) substrates (Lum Tech), organic (Nichem Fine Tech) and inorganic materials (Alfa Aesar), were commercially purchased. After the routine cleaning and ultraviolet (UV) ozone treatment, the ITO substrates were introduced into a high-vacuum deposition chamber (Trovato MFG, base pressure about 1×10^{-6} Torr) with multiple thermal evaporation sources, where film thickness was monitored *in situ* with a calibrated quartz crystal microbalance. N,N'-diphenyl-N,N'-bis(1-naphthyl)-(1,1'-biphenyl)-4,4'-diamine (NPB), 4,4',4''-tris(N-carbazolyl)-triphenyl amine (TCTA), host-dopant composition, 1,3,5-tris(phenyl-2-benzimidazolyl)-benzene (TPBi), lithium fluoride (LiF), and aluminum (Al) were evaporated on the substrates in turn by vacuum deposition. Phosphorescent bis(2-phenylpyridine)iridium acetylacetonate $[\text{Ir}(\text{ppy})_2(\text{acac})]$ and tris(2-phenylquinoline) iridium(III) $[\text{Ir}(\text{2-phq})_3]$ were used as dopants for green and orange-red emissions, respectively. The comparative devices were fabricated under identical experimental conditions. The EL spectra and the current density–voltage–luminance (J – V – L) characteristics of the corresponding devices were measured simultaneously with a Photo Research PR-655 spectrometer and a computer controlled programmable Keithley model 2400 power source. All measurements were carried out at room temperature under ambient conditions after the devices have been encapsulated in a glove box.

3. Results and discussion

The UV–vis absorption spectra of neat TCT, PTC, and POTA films are shown in Fig. 2(a). From the onsets of the absorption spectra, narrow E_g values are obtained for

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