



High-performance red organic light-emitting devices based on an exciplex system with thermally activated delayed fluorescence characteristic



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ABSTRACT

To realize high-performance red organic light-emitting diodes (OLEDs), a novel exciplex system Tris-PCz:B4PyPPM with thermally activated delayed fluorescence (TADF) characteristic was developed as the host of both red fluorescent and phosphorescent emitters. Due to the blue-green PL spectrum and suitable triplet energy level of 2.53 eV, the exciplex system matches the requirements of both red fluorescent and phosphorescent dopants. With an optimized mixing molar ratio of 5:5, Tris-PCz:B4PyPPM exciplex system shows outstanding host performance in both the red fluorescent and phosphorescent devices. The red fluorescent device based on DCJTb exhibits a low turn-on voltage of 2.3 V and an extremely high maximum external quantum efficiency (EQE) of 9.3%. And the red phosphorescent device with the Ir(MDQ)₂acac as the emitter realizes a maximum EQE of 20.3%. These remarkable results prove the feasibility of TADF exciplex systems simultaneously as the hosts of both high-performance fluorescent and phosphorescent red devices.

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1. Introduction

Highly efficient red organic light-emitting diodes (OLEDs) always attract continuous focus for the essential requirement in flat-panel displays and solid-state lightings [1–6]. Over the past two decades, lots of efforts have been made to improve the performance of red OLEDs. Due to the narrow bandgaps, concentration quenching effect is easily happened for red organic emitters [7]. Therefore, to reduce the energy loss, no matter red fluorescent or phosphorescent emitters, most of them are doped in suitable host materials to realize high efficiencies in red OLEDs. For red fluorophors, evident overlap between the photoluminescence (PL) spectrum of host and the absorption spectrum of dopant is the essential for complete singlet excitons transfer. Thus, green or yellow emitters, like tris(8-hydroxy-quinolinato)aluminium (Alq₃),

which have suitable emission spectra, are usually used as the host materials for red fluorescent emitters [8–10]. Correspondingly, red phosphors are usually doped in host materials with high triplet energy levels, like 4,4'-Bis(carbazol-9-yl)biphenyl (CBP) and 4,4',4''-Tris(carbazol-9-yl)triphenylamine (TCTA), to ensure triplet excitons fully harvesting on the dopant [11,12]. With these conventional hosts, the external quantum efficiencies (EQEs) of the red OLEDs based on red fluorophors or phosphors have already closed to the theoretical limits of 5% for the fluorescent devices and 20% for the phosphorescent devices [13,14]. However, as a result of the unsuitable frontier orbital energy levels and poor carriers mobilities of the conventional host materials, these red devices suffer the problems of complex devices structures, and high turn-on voltages (defined as the driving voltage at a brightness of 1 cd m⁻²) [2,15]. Although novel bipolar host materials are widely investigated to address the weaknesses of conventional host materials in recent years, the researchers mainly focused on the hosts of green and blue phosphorescent devices [16,17]. Moreover, due to the different requirements of the fluorescent and phosphorescent hosts, the phosphorescent host materials are usually inappropriate for

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fluorescent emitters. Thus, it is still highly required to development the high-performance hosts of red OLEDs.

Recently, the exciplex systems with thermally activated delayed fluorescence (TADF) characteristics draw great attention for their excellent performance as either the emitters or hosts in the OLEDs [18]. Especially as the new class of the host, TADF exciplex systems exhibit much superior properties than the conventional host materials and recently developed bipolar host materials. Formed between an electron-donor molecule (D) and an electron-acceptor molecule (A), which usually have hole- and electron-transporting properties respectively, exciplex system can also possess bipolar property, and even reach a charge balance by adjusting mixing ratio of D and A [19]. Due to the TADF characteristic, exciplex systems should have extremely small singlet–triplet splittings (ΔE_{ST}). The energy loss from singlet state (S_1) to triplet state (T_1) could be minimized when they are used as the phosphorescent hosts, improving the efficiencies of the devices [20]. Besides, the TADF exciplex systems enable highly efficient up-conversion from T_1 to S_1 via reverse intersystem crossing (RISC) process, leading the fluorescent OLED based on these novel hosts to break the fluorescent theoretical limit of 5% [21]. Furthermore, due to the extremely small ΔE_{ST} , the TADF exciplex systems can simultaneously match the requirements of the fluorescent and phosphorescent hosts, becoming the hosts of both fluorescent and phosphorescent devices. Therefore, the TADF exciplex systems are the most appropriate candidates as the high-performance hosts of both red fluorescent and phosphorescent OLEDs.

In this work, we developed a novel TADF exciplex system formed with D of 9,9',9''-triphenyl-9H,9'H,9''H-3,3',6',3'''-tercarbazole (Tris-PCz) and A of 4,6-Bis(3,5-di(pyridin-4-yl)phenyl)-2-phenylpyrimidine (B4PyPPM) as the host of both high-performance red fluorescent and phosphorescent OLEDs. In photophysical property study, the extremely small ΔE_{ST} of 0.01 eV ensures the TADF characteristic for Tris-PCz:B4PyPPM exciplex system. And the exciplex system also exhibits a suitable blue-green PL spectrum for red fluorescent emitters and high enough triplet energy level of 2.53 eV for red phosphorescent dopants. With an optimized mixing molar ratio of 5:5, Tris-PCz:B4PyPPM shows outstanding host performance in both the red fluorescent and phosphorescent devices. The fluorescent device based on traditional red emitter DCJTb (4-dicyanomethylene-2-*t*-butyl-6-1,1,7,7-tetramethyljulolidyl-9-enyl-4H-pyran) exhibits a low turn-on voltage of 2.3 V and high efficiencies of 16.5 cd A⁻¹, 22.5 lm W⁻¹ and 9.3% for maximum current efficiency (CE), power efficiency (PE), and EQE, respectively. And the red phosphorescent device with the Ir(MDQ)₂acac (bis(2-methyldibenzof[h]quinoxaline) (acetylacetonate)iridium (III)) as the emitter respectively shows the maximum CE, PE and EQE of 33.7 cd A⁻¹, 37.7 lm W⁻¹, and 20.3%.

These remarkable results prove the feasibility of TADF exciplex systems simultaneously as the hosts of both high-performance fluorescent and phosphorescent devices.

2. Experimental

All the materials were used in the experiments as received from commercial suppliers without further purification. OLEDs were fabricated in the vacuum deposition chamber with a base pressure of about 2×10^{-6} Torr, where organic materials were deposited on the indium tin oxide (ITO) glass substrates with a sheet resistance of 30 Ω per square. In first, the glass substrates were cleaned carefully with acetone, isopropyl alcohol and deionized water in order. Then, the glass substrates were transferred to the ovens for draught at 120 °C for about 2 h and treated with UV-ozone for about 30 min. At last, the glass substrates were transferred to the vacuum deposition chamber for deposition. The injecting layer MoO₃ and LiF were both deposited at rates of 0.1 Å s⁻¹; organic layers and cathode Al were deposited at the rates of 1 and 5 Å s⁻¹, respectively. The evaporation rates of the film of all the materials were measured by quartz crystals, which were controlled by multiple sensor system in the evaporation device. And the molar ratio was calculated from molecular weights and evaporation rates. Electroluminescence (EL) spectra, luminance and commission internationale de l'eclairage (CIE) coordinates were surveyed by a Spectrascan PR655 photometer and the current–voltage characteristics were surveyed by a Keithley 2400 sourcemeter under ambient atmosphere. EQE was calculated from the current density, luminance, and EL spectrum, according to a Lambertian distribution.

3. Results and discussion

Fig. 1 shows the molecular structures of Tris-PCz and B4PyPPM, which are respectively used as the D and A to form the exciplex system [22,23]. As shown in Fig. 2a, the Tris-PCz:B4PyPPM co-deposited film (molar ratio is 5:5) exhibits nearly same absorption behavior to those of Tris-PCz and B4PyPPM, suggesting no formation of new ground-state transition in the mixed film. While in the PL spectra, the mixed film exhibits a strong blue-green emission with a peak at 505 nm, which is significantly red-shifted compared with those of the constituting molecules. These results prove the formation of exciplex in the mixed films of Tris-PCz:B4PyPPM. Furthermore, the board blue-green emission of Tris-PCz:B4PyPPM ensure its capability as the host of the red fluorescent emitters. Fig. 2b shows the fluorescence and phosphorescence spectra of Tris-PCz:B4PyPPM at 77 K. Based on the spectra, the S_1 and T_1 energy levels of Tris-PCz:B4PyPPM are calculated to be 2.54 and 2.53 eV (fluorescence and phosphorescence peaks are at 488 and

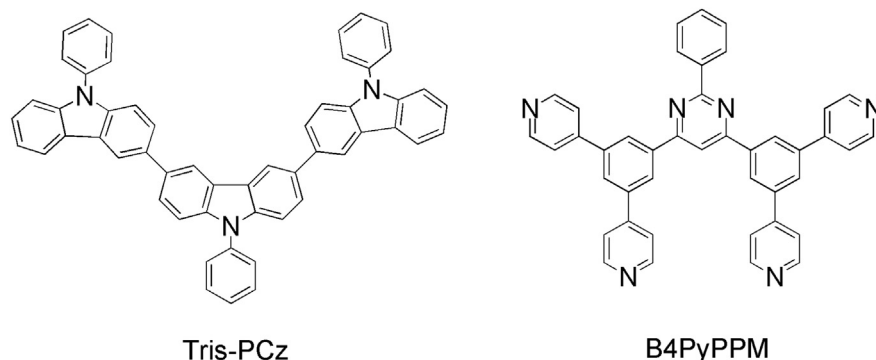


Fig. 1. Molecular structures of Tris-PCz and B4PyPPM.

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