

High performance fluorescent and phosphorescent organic light-emitting diodes based on a charge-transfer-featured host material



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ABSTRACT

Fluorescent and phosphorescent organic light-emitting diodes (OLEDs) were fabricated using a charge-transfer-featured compound, 6-{3,5-bis-[9-(4-t-butylphenyl)-9H-carbazol-3-yl]-phenoxy}-2-(4-t-butylphenyl)-benzo[de]isoquinoline-1,3-dione, as a host, and the electroluminescent (EL) characteristics of these two kinds of OLEDs were systematically studied. According to the photoluminescent quantum yields (η_{pl}), it was found that the external quantum efficiencies of both fluorescent and phosphorescent OLEDs were exceeding their theoretical limits. Based on the analysis of EL characteristics, the high device performance of fluorescent and phosphorescent OLEDs was attributed to both efficient energy transfer and triplet energy up-conversion, while direct exciton formation was also involved in phosphorescent OLEDs. In addition, the host film possessed high thermal and morphological stabilities due to the attachment of steric bulks on host molecule, resulting in the high doping concentration for both fluorescent and phosphorescent dyes. These results indicated that charge-transfer-featured material could be the promising host for both fluorescent and phosphorescent OLEDs.

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1. Introduction

Since the ground-breaking demonstration of organic light-emitting diodes (OLEDs) by Tang and VanSlyke in 1987 [1], there has been a significant progress in developing highly efficient OLEDs because of the enormous potential in flat panel display and solid-state lighting. During the early days in the 1990s, fluorescent OLEDs have been widely studied, they possess the merits of fast response, low roll-off and long operational lifetime, which are highly required in the aspect of display applications. However, only the excited singlets are emissive in fluorescent dyes, making them not favorable in realizing OLEDs with maximized internal quantum efficiency (IQE). To this end, Forrest and Thompson's groups developed phosphorescent dyes to take advantage of both singlet and triplet excitons, and therefore can, have a 100% IQE, in theory [2]. Noteworthy, both fluorescent and phosphorescent dyes suffer from concentration quenching effect [3], which requires them to be doped as guests into host matrices. However, with raising the doping concentration of the guest material, a significant reduction

of OLED performance will be inevitable. Therefore, it is an important issue to design a proper host material, in which dye molecules can be dispersed uniformly in the host matrix [4–6].

As for the conventional wide band-gap (E_g) hosts, a drawback is that the charge carrier injection barrier is high, resulting in an increase of driving voltage in OLEDs. Therefore, to maintain high triplet energy of the host while reduce the E_g , which approximately corresponds to the excited singlet energy, i.e., a smaller singlet-triplet energy gap (ΔE_{ST}), can facilitate the injection of charge carriers. More importantly, a small ΔE_{ST} is very beneficial to realize efficient up-conversion from triplet to singlet excited states, which has widely been focused on to break the 25% theoretical IQE limit of fluorescent OLEDs [7,8]. Considering the challenge in designing efficient host materials with small ΔE_{ST} , alternatively, the use of an exciplex host material is a promising strategy. The exciplex is well known as a charge-transfer (CT) state formed between electron-donors and electron-acceptors under photo- and electrical excitation [9,10]. The small orbital overlap between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) is responsible for a very small ΔE_{ST} [8].

Recently, several studies have shown that high performance phosphorescent OLEDs can be achieved by using exciplex-forming

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hosts [11–15]. Park et al. have reported highly efficient green phosphorescent OLEDs through the energy transfer from an exciplex co-host to a green phosphorescent emitter [12]. Another recent breakthrough is the realization of high performance blue phosphorescent OLEDs using energy transfer from the exciplex between di-[4-(N,N-ditoly-amino)-phenyl]cyclohexane and 5',-5'''-sulfonyl-di-1,1':3',1''-terphenyl at the interface [14]. However, there still exist some problems ahead by using exciplex hosts. For example, the use of exciplex co-host suffers from the typical phase separation problem, which is detrimental to the long-term operation of OLEDs. Meanwhile, the interfacial exciplex brings an extra organic/organic junction, leading to a more complex device configuration.

Recently, a CT-featured complex, 6-[3,5-bis-[9-(4-t-butylphenyl)-9H-carbazol-3-yl]-phenoxy]-2-(4-t-butylphenyl)-benzo[de]isoquinoline-1,3-dione (CzPhONI) [16], which possessed small ΔE_{ST} resulting from the intramolecular CT state, was realized. It was used as the host of a fluorescent dye, and the results showed that the external quantum efficiency (EQE) of the device exceeded the theoretical limit, ascribed to a triplet-fusion up-conversion mechanism. However, the potential of CzPhONI host to be applied to other fluorescent dyes has not been extended. Moreover, to the best of our knowledge, the research on the utilization of a single CT-featured material as the host of phosphorescent dyes has not been carried out so far.

Therefore, in this work the effect of a novel CT-featured host material of CzPhONI on the performance of several typical fluorescent and phosphorescent OLEDs was studied in detail. It was found that both efficient energy transfer and triplet energy up-conversion were involved in the emission mechanisms of CzPhONI-based fluorescent and phosphorescent OLEDs, while the characteristics of single-carrier devices proved that charge trapping followed by direct exciton formation in phosphorescent OLEDs. Despite the small ΔE_{ST} , CzPhONI exhibits another two features: (i) great thermal stability; (ii) capability of inhibiting the aggregation of guest emitters thus leads to high doping levels, which was elucidated through characterizing the morphology of emissive layers (EML) by atomic force microscopy (AFM).

2. Experimental

Indium tin oxide (ITO) coated glass substrates with a sheet resistance of $15 \Omega/\text{sq}$ were cleaned in detergent water, acetone, deionized water and isopropyl alcohol by using an ultrasonic bath for 30 min, respectively. Afterwards, the substrates were dried with nitrogen gas flow and then, they were treated with O_2 plasma in a vacuum chamber under a pressure of 25 Pa for 5 min, to clean

the surface and enhance hole injection ability of ITO. Organic and metallic layers were subsequently deposited in separate chambers under a pressure of 3×10^{-4} and 3×10^{-3} Pa, respectively, without breaking the vacuum. Deposition rates and thin film thicknesses were monitored in situ using a quartz crystal oscillator. Luminance–current density–voltage (L – J – V) curves were measured using Keithley 4200 source meter and an ST-86LA luminance meter. Electroluminescence (EL) spectra and the Commission Internationale de l'Eclairage (CIE) coordinates of the devices were recorded with an OPT-2000 spectrophotometer. The EQEs of the OLEDs were calculated on the basis of the theory reported in the literature [17]. All measurements were performed in air under ambient condition without encapsulation.

The photoluminescent (PL) spectra of the organic films were recorded with an OPT-2000 spectrophotometer at an excitation wavelength of 365 nm. The absorption spectra of the organic films were measured with a SHIMATZU UV-1700 UV–Vis spectrophotometer. The absolute PL quantum yields (PLQYs) of the film samples were determined with an Edinburgh FLS980 spectrometer. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed on TGA Q500 and DSC Q100 instruments under nitrogen atmosphere at a heating rate of $10^\circ\text{C}/\text{min}$. The morphologies of organic films prepared by thermal evaporation were characterized by AFM (MFP-3D-BIO, Asylum Research) in tapping mode.

3. Results and discussion

3.1. PL emission property of CzPhONI and energy transfer between host and guest

The PL spectra of CzPhONI doped in poly(methyl methacrylate) (PMMA) are recorded, as shown in Fig. 1(a). When the doping concentration is as low as 0.5 wt.%, CzPhONI is dispersed molecularly in PMMA, showing a peak at ~ 422 nm. In this case, only the monomer emission can be observed. When the doping concentration is raised to 5 wt.%, a broadened PL spectrum with a red-shifted peak at ~ 434 nm is observed, indicating the formation of CT state. In the solid film state, where CzPhONI molecules are tightly packed, the interactions become stronger, leading to the more bathochromic spectrum with an emission peak at ~ 493 nm. More detailed investigations have shown that CzPhONI exhibits a twisted intramolecular CT character, and it has been reported elsewhere [16]. The density functional theory calculations show that the overlap between HOMO and LUMO is zero. The HOMO of CzPhONI is merely located on the dicarbazylphenyl moiety, while the LUMO is only distributed on the naphthalimide unit. Therefore, the excited state

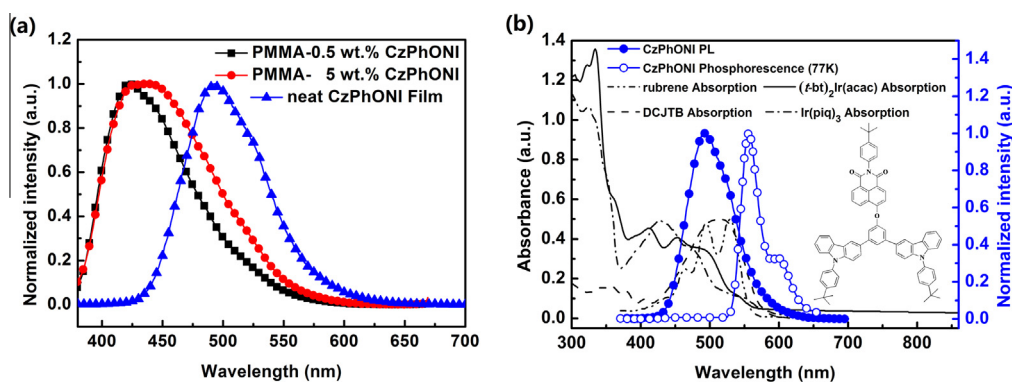


Fig. 1. (a) PL spectra of thin film samples of CzPhONI. (b) Normalized UV–Vis absorption spectra of rubrene, DCJTb, $(t\text{-}bt)_2\text{Ir}(\text{acac})$ and $\text{Ir}(\text{piq})_3$, along with the PL and phosphorescence emission spectra of CzPhONI in solid film state. Inset: molecular structure of CzPhONI.

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