



A host material with a small singlet–triplet exchange energy for phosphorescent organic light-emitting diodes: Guest, host, and exciplex emission

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ARTICLE INFO

Article history:

Received 24 March 2012

Received in revised form 26 May 2012

Accepted 8 June 2012

Available online 21 June 2012

Keywords:

Host material

Singlet–triplet exchange energy

Phosphorescent

Organic light-emitting diode

Exciplex

ABSTRACT

A host material containing a triazine core and three phenylcarbazole arms, called 2,4,6-tris(3-(carbazol-9-yl)phenyl)-triazine (TCPZ), was developed for phosphorescent organic light-emitting diodes (OLEDs). Ultra-low driving voltages were achieved by utilizing TCPZ as the host due to its decreased singlet–triplet exchange energy (ΔE_{ST}) and low-lying lowest unoccupied molecular orbital (LUMO) energy level. Interaction between the RGB triplet emitters and TCPZ were studied in both photoluminescent and electroluminescent processes. Transient photoluminescence (PL) measurement of the co-deposited film of *fac*-tris(2-phenylpyridine) iridium ($\text{Ir}(\text{PPy})_3$):TCPZ exhibits a shoulder at 565 nm whose lifetime is about two times longer than that of the $\text{Ir}(\text{PPy})_3$ triplet excitons and can be attributed to the triplet exciplex formed between $\text{Ir}(\text{PPy})_3$ and TCPZ. Such exciplex was also found for the green phosphorescent OLED, giving the most efficient phosphorescent OLED with triplet exciplex emission hitherto. Different from the PL process, a broad featureless band with a maximum at 535 nm was found for the OLED based on an EML of iridium(III) bis(4,6-(di-fluorophenyl)pyridinato- N,C^2)picolinate (Flrpic):TCPZ, which can be attributed to the emission from the singlet excited state of TCPZ formed by direct hole-electron recombination. A multi-emitting-layer white OLED was also fabricated by utilizing Flrpic and tris(1-phenylisoquinolinolato- C^2,N)iridium(III) ($\text{Ir}(\text{piq})_3$) as the complementary triplet emitters and TCPZ as the host. Different from most of ever reported white OLEDs fabricated with blue/red complementary triplet emitters that exhibit color rendering index (CRI) lower than 70, a high CRI of 82 is achieved due to the combination of blue and red phosphorescence emissions from Flrpic and $\text{Ir}(\text{piq})_3$, and the emerging green fluorescence emission from TCPZ.

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1. Introduction

Since the development on phosphorescent transition metal complexes by Forrest and Thompson's groups, nearly

100% internal quantum efficiency of electroluminescence has been realized by harvesting both electro-generated singlet and triplet excitons by fast intersystem crossing for phosphorescence emission from their triplet states [1]. For those phosphorescent organic light-emitting diodes (OLEDs), phosphorescent emitters are normally doped into an appropriate host material to avoid self-quenching. As thus, it is essential that the triplet energy (E_T) of the host is higher than that of the emitter in order

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to achieve an efficient energy transfer from the host to the guest and also to prevent energy back transfer from the guest to the host [2]. In addition, an optimized carrier recombination in the emitting layer (EML) is also indispensable to give a high efficiency. In general, the electron mobility of many hosts is much lower than the hole mobility because of the fact that they mainly consist of strong electron donors like aromatic amines or carbazoles [3]. Besides the building blocks of electron donors like aromatic amines or carbazoles, introduction of electron deficient heterocycles, like pyridine [4], oxadiazole [5], and phenanthroline [6], was proven to be an effective route to give host materials with improved bipolarity. Triazine has an electron affinity larger than those of other typical electron deficient heterocycles (e.g., pyridine, pyrimidine), and its derivatives are known to be good electron transport materials in OLEDs [7]. Besides, some triazine derivatives were also reported as electron-transport type hosts. Adachi and co-workers established a series of 1,3,5-triazine derivatives as hosts for the green phosphor of *fac*-tris(2-phenylpyridine) iridium ($\text{Ir}(\text{PPy})_3$), resulting in an external quantum efficiency (η_{ext}) of 10.2% and a power efficiency (η_{p}) of 14 lm W^{-1} [8]. Wong and co-workers demonstrated a series of 1,3,5-triazine derivatives as hosts for the green phosphors of $\text{Ir}(\text{PPy})_3$ and $(\text{PPy})_2\text{Ir}(\text{acac})$, providing maximum η_{ext} and η_{p} of 17.5% and 59.0 lm W^{-1} [9]. Strohriegl and co-workers demonstrated a series of donor substituted 1,3,5-triazine derivatives as hosts for the blue phosphor of iridium(III) bis(4,6-(di-fluorophenyl)pyridinato- N, C^2')picolinate (Flrpic), yielding a maximum current efficiency up to 21 cd A^{-1} [3]. Our previous reports demonstrated that the energy difference (ΔE_{ST}) between the singlet and triplet emission transitions decreases with introducing electron deficient heterocycles combined with electron donors like carbazole, giving host materials as attractive candidates to reduce the driving voltage of OLEDs [10]. It is anticipated that the bipolar host materials consisting of electron acceptor like triazine should also possess a small ΔE_{ST} to give a reduced driving voltage of OLEDs. However, the introduction of both the electron donor and acceptor to the host material may also lead to an intramolecular charge transfer, resulting in the reduction of the energy band-gap (E_{g}) of the molecule, and the effect may be much more serious for a triazine-containing bipolar host material because of its higher electron affinity than most of other electron deficient heterocycles. Moreover, as a host material co-deposited with triplet emitters in phosphorescent OLEDs, the interaction between the host material and the triplet emitters and thus the origin of the emitting state are critical factors for the device performance. To date, few works reported on such an interaction, especially for blue, green, and red phosphorescent emitters, which is of importance to realize highly efficient phosphorescent OLEDs for applications to full color flat-panel displays and lighting.

In this article, we report on a bipolar host material composed of 1,3,5-triazine as the core and three phenylcarbazoles as the arms, called 2,4,6-tris(3-(carbazol-9-yl)phenyl)-triazine (TCPZ). Its carrier mobility and energy levels, including the highest occupied molecular orbital (HOMO), the lowest unoccupied molecular orbital (LUMO), singlet and triplet energies, and singlet–triplet exchange

energy, were thoroughly characterized. Transient photoluminescence (PL) decays of the co-deposited films with red, green, and blue (RGB) phosphors of tris(1-phenylisoquinolinolato- C^2, N)iridium(III) ($\text{Ir}(\text{piq})_3$), $\text{Ir}(\text{PPy})_3$, and Flrpic were measured for the study of their interaction in photoluminescence process. In addition, phosphorescent OLEDs were also fabricated with those RGB triplet emitters as the guest and TCPZ as the host for the study of their interaction in electroluminescence process. Moreover, a multi-emitting-layer white OLED was fabricated by utilizing Flrpic as a blue emitter and $\text{Ir}(\text{piq})_3$ as a complementary red emitter. In contrast to most of ever reported white OLEDs fabricated with blue/red phosphorescent emitters that give CRI lower than 70, a high CRI of 82 is achieved due to the combination of the phosphorescence emissions from $\text{Ir}(\text{piq})_3$ and Flrpic and the emerging fluorescence emission from TCPZ.

2. Experimental

2.1. General

All the reagents were purchased from Sigma–Aldrich and were used without further purification. 2,4,6-Tris(3-bromophenyl)-triazine (TBrPZ) was synthesized according to the literature procedure [7c]. The developed TCPZ was purified by silica gel chromatography and then repeated thermal gradient vacuum sublimation before characterization and device fabrication. ^1H and ^{13}C NMR spectra were recorded on Varian 500 (500 MHz) spectrometer. Mass spectra were obtained using a JEOL JMS-K9 mass spectrometer. Differential scanning calorimetry (DSC) was performed using a Perkin–Elmer Diamond DSC Pyris instrument under nitrogen atmosphere at a heating rate of 10 $^{\circ}\text{C min}^{-1}$. Thermogravimetric analysis (TGA) was undertaken using a SEIKO EXSTAR 6000 TG/DTA 6200 unit under nitrogen atmosphere at a heating rate of 10 $^{\circ}\text{C min}^{-1}$. UV–Vis absorption spectrum of the neat film was measured with a Shimadzu UV-3150 UV–vis–NIR spectrophotometer. Room temperature steady-state photoluminescent (PL) spectrum of the neat film was obtained with a FluoroMax-2 (Jobin–Yvon–Spex) luminescence spectrometer. Ionization potential was determined by atmospheric ultraviolet photoelectron spectroscopy (Rikken Keiki AC-3). Time-resolved emission spectra of the neat film were obtained at $T = 4.2$ K under excitation by a nitrogen laser ($\lambda = 337$ nm, 50 Hz, 800 ps) combined with a streak scope C4334 (Hamamatsu) and a synchronous delay generator C4792-02 (Hamamatsu). In comparison, transient PL decays and the corresponding simultaneous PL spectra of the phosphorescent emitter-doped films were recorded at room temperature. PL quantum efficiencies of the co-deposited films were measured by using an integrating sphere under nitrogen gas flow at room temperature. Density functional theory (DFT) calculations were performed by using the Gaussian suite of programs (Gaussian 03 W). For the calculation of HOMO and LUMO energy levels, the ground state structures were optimized at the restricted B3LYP/6-31G(d) level, and the single-point energies were calculated at the restricted B3LYP/6-311+G(d,p)

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