



# Quenching in single emissive white phosphorescent organic light-emitting devices

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## ABSTRACT

To explore energy loss by diffusive triplet excitons in single emissive white phosphorescent organic light-emitting devices, the authors investigated collisional quenching between the electron transport materials 4,7-diphenyl-1,10-phenanthroline (Bphen), 2',2'',2'''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi), or 1,3,5-tri(3-pyrid-3-yl-phenyl)benzene (TmPyPB) and the blue phosphorescent material, 3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) Iridium III (Flrpic) spectroscopically in solution. The luminous efficiency and the external quantum efficiency (EQE) of an emissive white phosphorescent organic light-emitting device, in which TmPyPB acted as the electron transport material, was found to be greater than those of devices prepared using Bphen or TPBi due to the lack of collisional quenching. In addition, it was found that to prevent triplet exciton loss, an ETL material should have a low bimolecular quenching rate constant  $k_q$  of less than  $1.458 \times 10^7 \text{ s}^{-1} \text{ M}^{-1}$ , which is the  $k_q$  of TmPyPB.

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## 1. Introduction

Since Tang and Van Slyke first described bulk heterojunction organic light-emitting devices (OLEDs) [1], OLEDs have attracted significant research attention because of their applications in solid state lighting, and flat-panel displays due to their bright, full color, light emissions, which originate from radiative relaxation of the excited states of  $\pi$ -conjugated molecular systems [2–10]. Recently, much effort has been focused on the enhancement of OLED performance, and phosphorescent organic light-emitting devices (PHOLEDs) are viewed with considerable interest because of the technical advantages offered by their 100% internal quantum efficiencies, which are achieved by the harvesting of singlet and triplet excitons [11–13]. Several methods, such as, multiple emitters in single or multiple emissive layers, tandem stacks with two or three

color combinations, hybrid structures incorporating fluorescent and phosphorescent materials and quantum well structures [14–19], have been used to produce highly efficient white PHOLEDs. However, studies performed to date have largely taken the form of experimental trials, and in order to fabricate high efficiency white PHOLEDs, some complicated designs such as, multiple emissive units, have been employed, but unfortunately, these designs require complex fabrication processes and are often incompatible with the requirements of mass production. On the other hand, mass-production compatible white PHOLEDs with a single emissive layer tend to have relatively limited exciton recombination zones [20]. Single emissive white PHOLEDs using two colors have been studied by many other research groups. The devices based on two colors are relatively easy to realize white emission because there is only energy transfer from one dopant with higher energy levels to the other with lower energy level compared with the device based on three colors. In three colors white PHOLEDs, the additional number of energy transfer should be considered for adjusting balance of blue, green and red emission, respectively. In fact, three colors white PHOLEDs have been widely studied by many research groups but these were doped in multi-layer, hybrid and

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tandem structures [21–28]. In this study, in addition to considering energy transfer mechanism in the single emissive layer, we deeply examined the interaction between the emissive layer and electron transport layer to improve performance of single emissive white PHOLEDs.

Charge transporting organic semiconductors are an important class of materials that play crucial roles in electronic and optoelectronic devices, such as, OLEDs, thin film transistors, and photovoltaic cells. OLEDs with a hole transport layer (HTL) and an electron transport layer (ETL) sandwiched between two electrodes with show low driving voltages and bright emissions are important for full color flat panel display and lighting applications. To maximize the efficiency of white PHOLEDs, it should be determined how to confine triplet excitons in emissive layers (EMLs) without energy loss. Furthermore, host materials with higher triplet energy levels than dopants are required to prevent reverse energy transfer to host materials [29,30]. In addition to triplet exciton confinement by host materials, triplet exciton diffusion into HTLs and ETLs should be considered, since triplet excitons formed by charge carrier recombination are usually formed near HTL/ETL interfaces [31]. Furthermore, triplet exciton loss in EMLs caused by HTL/ETL quenching can decrease the efficiencies of white PHOLEDs.

In this study, we fabricated single emissive white PHOLEDs with three primary colors and observed the behaviors of triplet excitons in their EMLs. Förster energy transfer (FET) [32] between host and phosphorescent dopants, Dexter energy transfer (DET) [33] between phosphorescent dopants in EMLs, and collisional quenching [34] between Flrpic (3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) Iridium III) blue phosphorescent dopant and ETL materials, that is, Bphen (4,7-diphenyl-1,10-phenanthroline, TPBi (2',2'',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole)) or TmPyPB, (1,3,5-tri(3-pyrid-3-yl-phenyl)benzene) were followed spectroscopically in solution.

## 2. Experimental section

White PHOLEDs were fabricated using the device architecture: ITO (150 nm; Indium Tin Oxide)/TAPC (70 nm; 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane)/mCP (N,N'-dicarbazolyl-3,5-benzene): Flrpic–8.0%: Ir(ppy)<sub>3</sub>–0.5% (Tris(2-phenylpyridine)iridium(III)): Ir(piq)<sub>3</sub>–0.5% (Tris(1-phenylisoquinoline)iridium(III)) (30 nm)/three different ETL materials (Bphen, TPBi and TmPyPB) (30 nm)/Liq (2 nm; 8-Hydroxyquinolinolato-lithium)/Al (120 nm; Aluminium). ITO coated glasses with a sheet resistance of ~12 Ω/sq were used as substrate. Line patterns of ITO were formed using a photolithography process. ITO glass substrates were cleaned in an ultrasonic bath in the following order: deionized water, isopropyl alcohol, acetone, deionized water, and isopropyl alcohol, and then treated with O<sub>2</sub> plasma under vacuum conditions (5.0 × 10<sup>-2</sup> Torr) at 50 W for 2 min. All organic materials were deposited by thermal evaporation at a pressure of ~1.0 × 10<sup>-7</sup> Torr. To fabricate white PHOLEDs we used TAPC as the HTL material, mCP as the host material, blue, green, or red phosphorescent Flrpic, Ir(ppy)<sub>3</sub>, or Ir(piq)<sub>3</sub>, respectively, as dopants, Bphen, TPBi, or TmPyPB as the ETL materials, and an electron injection layer of Liq. The Al cathode was formed by thermal evaporation at an evaporation rate of 5.0 Å/s. All fabricated devices had an active area of 3 × 3 mm<sup>2</sup>, as defined by the shadow mask used for cathode deposition. The electro-optical characteristics of the white PHOLEDs were determined using a Keithley 238 LMS PR-650 spectrophotometer, a colorimeter, and a current density-voltage-luminance (J-V-L) system. The emission/absorption spectra of the organic materials were measured in CH<sub>2</sub>Cl<sub>2</sub> at room temperature or at 77 K. Time resolved transient photoluminescence decay at room temperature was conducted using sub-nanosecond optical pulses at an excitation wavelength of

416 nm, that is, at the metal-to-ligand charge transfer (MLCT) of Flrpic in an O<sub>2</sub> free Ar atmosphere. Schematic energy band diagrams of white PHOLEDs containing Bphen, TPBi, or TmPyPB (hereafter referred to as PHOLEDs A, B, or C) and corresponding energy levels of the HOMOs and LUMOs (highest occupied and lowest unoccupied molecular orbitals, respectively) and their molecular structures are shown in Fig. 1 [35–40].

## 3. Results and discussion

This study was undertaken to explore energy loss by diffusive triplet excitons in single emissive white PHOLEDs by investigating collisional quenching between electron transport materials Bphen, TPBi or TmPyPB and Flrpic spectroscopically in solution. We found that the luminous efficiency and external quantum efficiency (EQE) of a single emissive white PHOLED, in which TmPyPB acted as the electron transport material, was higher than those of devices prepared using Bphen or TPBi due to a lack of collisional quenching, and that to prevent triplet exciton loss, the ETL material should have a lower value of bimolecular quenching rate constant  $k_q$  than  $1.458 \times 10^7 \text{ s}^{-1} \text{ M}^{-1}$ , which is the  $k_q$  of TmPyPB.

In this study, we fabricated electron only devices (EODs) A, B and C in order to evaluate electron injection and transport by observing current density-voltage (J-V) characteristics (Table 1). Usually, the J-V characteristics of OLEDs depend critically on electronic states at metal/organic or organic/organic interfaces. The J-V characteristics of A, B and C are shown in Fig. 2. All three EODs exhibited electron transport, but their turn-on voltages increased in the order A → C → B. This phenomenon is described by thermionic current density ( $J_{th}$ ) as defined by the Schottky thermionic emission equation [41]:

$$J_{th} = A^* T^2 \exp\left(-\frac{q\Phi_B}{kT}\right)$$

where Richardson's constant  $A^*$  is:

$$A^* = \frac{4\pi q m^* k}{h^3}$$

and  $q$  = electric charge,  $m^*$  = carrier effective mass,  $k$  = Boltzmann's constant,  $h$  = Planck's constant,  $T$  = temperature (Kelvin), and  $\Phi_B$  = the energy barrier. Based on considerations of  $J_{th}$ , one would expect more charge carrier injection for a lower energy barrier  $\Phi_B$ . As shown in Fig. 1, the expected energy barrier difference  $\Phi_B$  between Bphen electron transport materials and Liq electron injection materials was 0.5 eV, and therefore, 0.8 eV for TPBi electron transport materials, and 0.8 eV for TmPyPB electron transport materials, which suggests Bphen has a lower energy barrier  $\Phi_B$  and a lower turn-on voltage. However, this does not seem to explain the different J-V characteristics of EODs B and C given the same energy barrier  $\Phi_B$  of 0.8 eV. This different J-V characteristics between EODs B and C were caused by an electron mobility difference (the electron mobilities of Bphen, TPBi, and TmPyPB are  $5.2 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at  $E = 5.5 \times 10^5 \text{ V cm}^{-1}$ ,  $3.3 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at  $E = 4.7 \times 10^5 \text{ V cm}^{-1}$  and  $1.0 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at  $E = 6.4 \times 10^5 \text{ V cm}^{-1}$ , respectively [42–44]. This issue will be described later in this study. The J-V characteristics of A, B, and C and their driving voltages are affected to device performance later.

White PHOLEDs A, B, and C were fabricated using Flrpic with Bphen, TPBi, or TmPyPB (Table 1). TAPC was used as the HTL material due to its hole transport ability and high triplet energy levels, and Liq was used as an electron injection layer (EIL) material due to electron injection ability and LUMOs between cathode and ETL. The

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