

Supporting Information

Triplets harvesting by multi-process energy transfer in fluorescent organic light-emitting diodes

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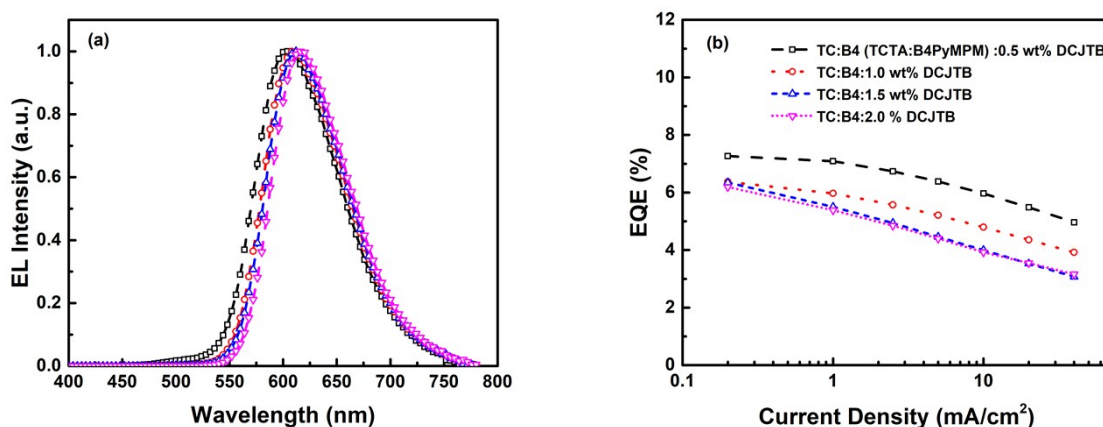


Figure. S1 Electroluminescent (EL) spectra and external quantum efficiencies (EQEs) vs. current density of conventional exciplex-guest FOLEDs. (ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: B4PYMPPM: x wt% DCJTB (30 nm)/B4PYMPPM (55 nm)/Liq (2 nm)/Al (110 nm))

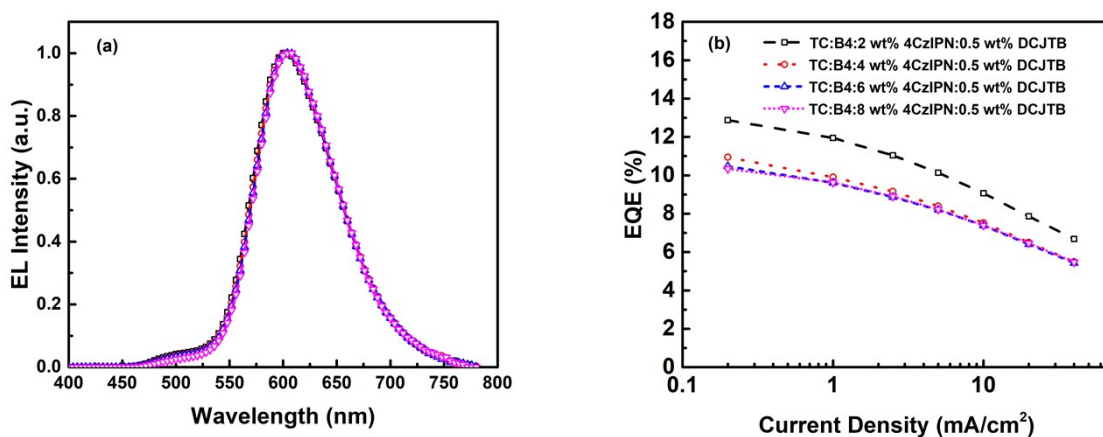


Figure. S2 EL spectra and EQEs vs. current density of novel-designed TADF-sensitized FOLEDs using 4CzIPN as energy transfer assisted TADF molecules. (ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: B4PYMPPM: x wt% 4CzIPN: 0.5 wt% DCJTB (30 nm)/B4PYMPPM (55 nm)/Liq (2 nm)/Al (110 nm))

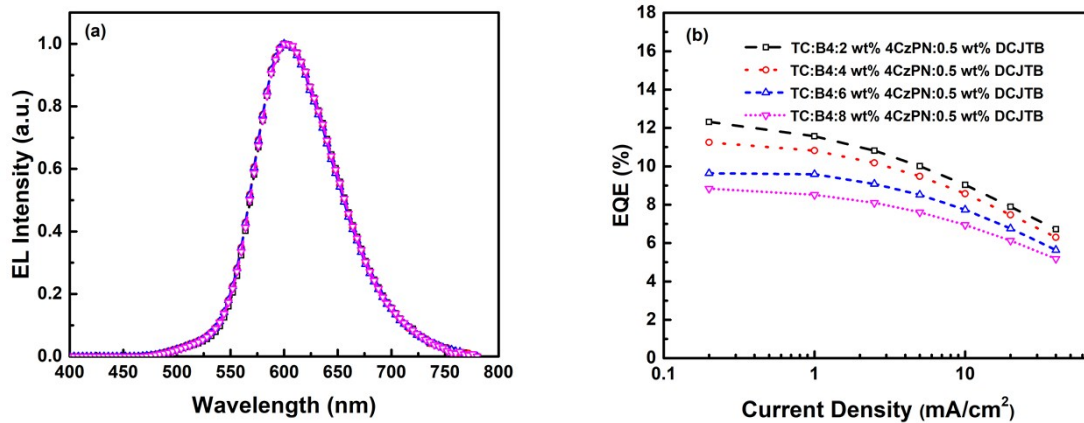


Figure. S3 EL spectra and EQEs vs. current density of novel-designed TADF-sensitized FOLEDs using 4CzPN as energy transfer assisted TADF molecules.
 (ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: B4PYMPM: x wt% 4CzPN: 0.5 wt% DCJTJB (30 nm)/B4PYMPM (55 nm)/Liq (2 nm)/Al (110 nm))

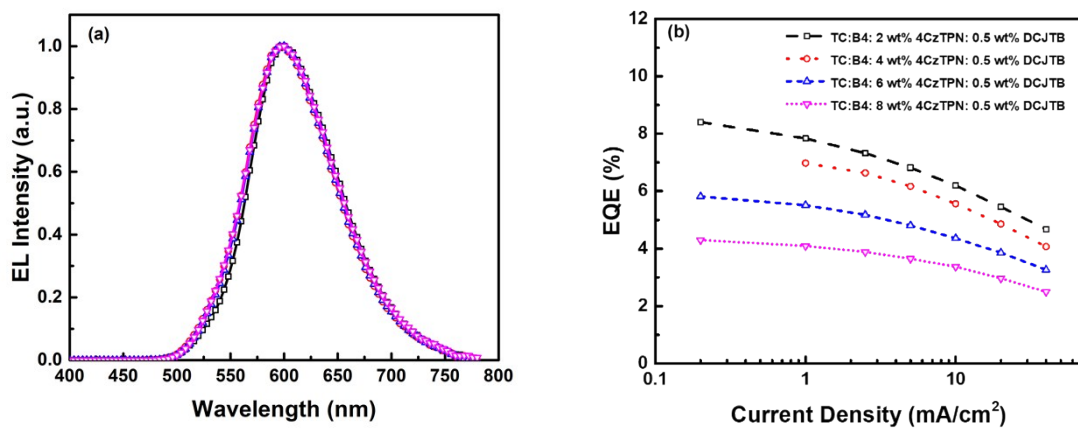


Figure. S4 EL spectra and EQEs vs. current density of novel-designed TADF-sensitized FOLEDs using 4CzTPN as energy transfer assisted TADF molecules.
 (ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: B4PYMPM: x wt% 4CzTPN: 0.5 wt% DCJTJB (30 nm)/B4PYMPM (55 nm)/Liq (2 nm)/Al (110 nm))

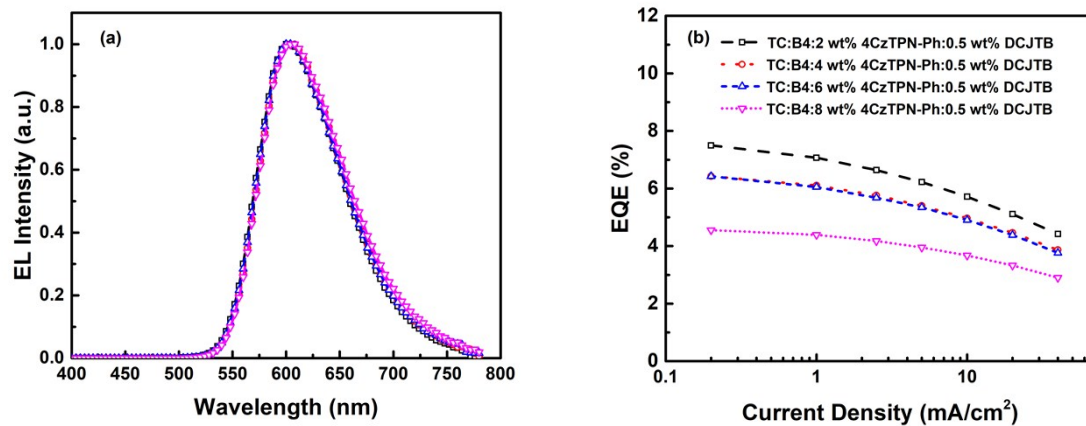


Figure. S5 EL spectra and EQEs vs. current density of novel-designed TADF-sensitized FOLEDs using 4CzTPN-Ph as energy transfer assisted TADF molecules.

(ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: B4PYMPPM: x wt% 4CzTPN-Ph: 0.5 wt% DCJTb (30 nm)/B4PYMPPM (55 nm)/Liq (2 nm)/Al (110 nm))

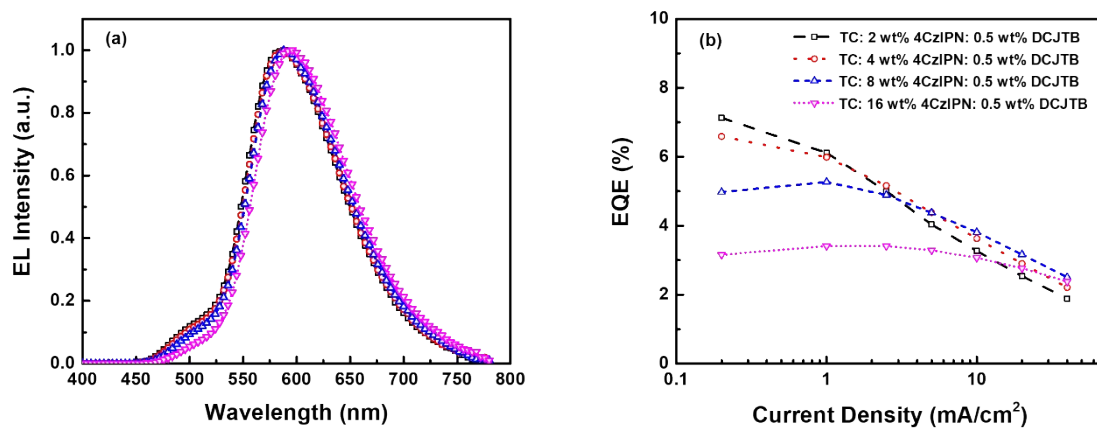


Figure. S6 EL spectra and EQEs vs current density of conventional host-energy transfer assisted TADF molecules-guest FOLEDs.

(ITO/HAT-CN (10 nm)/TAPC (55 nm)/TCTA (10 nm)/TCTA: x wt% 4CzIPN:0.5 wt% DCJTb (30 nm)/B4PYMPPM (55 nm)/Liq (2 nm)/Al (110 nm))

Table S1. The photoluminescence quantum yields (PLQYs) of emitting layers.

Emitting layer	Photoluminescence quantum yield (PLQY)
TCTA:B4PyMPM:0.5 wt% DCJTB	55%
TCTA: 4 wt% 4CzIPN: 0.5 wt% DCJTB	48%
TCTA:B4PyMPM:2 wt% 4CzIPN: 0.5 wt% DCJTB	65%
TCTA:B4PyMPM:2 wt% 4CzPN: 0.5 wt% DCJTB	63%
TCTA:B4PyMPM:2 wt% 4CzTPN: 0.5 wt% DCJTB	57%
TCTA:B4PyMPM:2 wt% 4CzTPN-Ph: 0.5 wt% DCJTB	54%

(The PLQYs were tested by HORIBA, Fluorolog-3.)

Table S2. The basic photophysical data of used materials in this study.

Molecules	Medium	λ_{PL} (nm)	τ_D (μ s)	ϕ_{PL} (%)	Ref
			Experimental results		
TCTA:B4PyMPM	PL: mixed films (1:1 molar)	510	19	60 ± 2	12
4CzIPN	PL- TCTA film (2 wt%)	520	5.1 (in TOL)	94 ± 2 (in TOL)	12
4CzPN	PL- TCTA film (2 wt%)	525	13.9 (in TOL)	74 ± 3 (in TOL)	12
4CzTPN	PL- TCTA film (2 wt%)	530	1.9 (in TOL)	72 ± 3 (in TOL)	12
4CzTPN-Ph	PL- TCTA film (2 wt%)	570	1.1 (in TOL)	26 ± 1 (in TOL)	12
DCJTB	PL- TC: B4 film (0.5 wt%)	600	-	-	-

(TOL: toluene; Ref: reference in the manuscript)

Calculations and discussions of Dexter energy transfer process

We calculated the Dexter energy transfer radius roughly in the novel TADF-sensitized FOLEDs while using different TADF molecules (4CzIPN, 4CzPN, 4CzTPN and 4CzTPN-Ph) and make a comparison. The specific process of calculations and comparisons is shown as follows:

The Dexter energy transfer radius R_0' can be represented by (J. Chem. Phys. 1965, 43, 1978):

$$R_0' = 7.346c_0^{-1/3} \text{ (angstrom)} \quad (1)$$

In Eq. (1) above, R_0' is defined as the Dexter radius. At this critical transfer distance, the energy transfer rate is equal to the rate of the spontaneous decay of the donor in the absence of acceptors. c_0 is a constant, which is called the critical transfer concentration and follow the relation below (J. Chem. Phys. 1967, 47, 3211):

$$c_0 = k'S^{-3/s} \quad (2)$$

with

$$S = \int_0^{\infty} \frac{I(E)\sigma(E)}{E^{s-2}} dE \quad (3)$$

where k' is a constant independent of species of acceptor. s (≥ 3) is a measure of the range dependence of interactions. $I(E)$ and $\sigma(E)$ are normalized emission spectrum of the donor and absorption spectrum of the acceptor, respectively. S is the overlap integral and defined by Eq. (3) above.

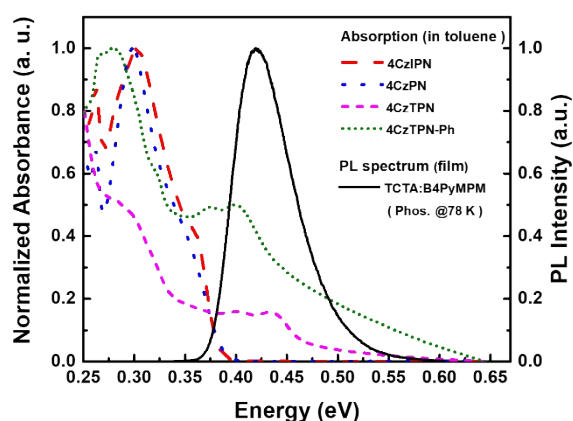


Figure. S7 Phosphorescent spectrum of TCTA: B4PyMPM mixed solid film by 1:1 molar at 78 K and the absorption spectra of four energy transfer assisted TADF materials, 4CzIPN, 4CzPN, 4CzTPN and 4CzTPN-Ph in toluene.

When the Dexter energy transfer is considered in novel double-dopant TADF-sensitized OLEDs, we only need to concern DET2 from T1 of exciplex co-host to that of TADF molecules as shown in Fig. 1(b) of the manuscript. Dexter energy transfer processes from T1 of the co-host to that of the guest (DET1) and from T1 of TADF molecules to that of the guest can be ignored due to low concentration of fluorescent emitters. While using different four TADF molecules (4CzIPN, 4CzPN, 4CzTPN, 4CzTPN-Ph), k' in Eq. (2) above can be the same due to the similar chemical structure of the above four TADF molecules.

Based on Eqs. (1), (2), and (3) and Figure S7 above, it can be concluded that Dexter radius R_0' ($4CzIPN$) $<$ R_0' ($4CzPN$) $<$ R_0' ($4CzTPN$) $<$ R_0' ($4CzTPN-Ph$). While at the same doping concentration of energy transfer assisted TADF sensitizers, Φ_{DET2} ($4CzIPN$) $<$ Φ_{DET2} ($4CzPN$) $<$ Φ_{DET2} ($4CzTPN$) $<$ Φ_{DET2} ($4CzTPN-Ph$).

However, the doping ratio of TADF molecules in the novel TADF-sensitized OLEDs is quite low, so the effect of DET2 process to the maximum EL efficiency is limited in comparison to Förster resonance energy transfer (FRET) processes.