



Thermally activated delayed-fluorescence organic light-emitting diodes based on exciplex emitter with high efficiency and low roll-off

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ABSTRACT

Highly efficient thermally activated delayed fluorescence (TADF) organic light-emitting diodes (OLEDs) based on exciplex are demonstrated in a blended system with commercially available 1,1-bis((di-4-tolylamino)phenyl)cyclohexane (TAPC) and 2,4,6-tris(biphenyl-3-yl)-1,3,5-triazine (T2T). By well adjusting the ratio between these two materials, the optimized device shows a low turn-on voltage of 2.4 V and a high external quantum efficiency (EQE) of 11.6%. More importantly, the device retains an EQE of 9.4% even at a high luminescence of 1000 cd/m². The low efficiency roll-off is attributed to the small singlet-triplet splitting and the short of the delayed fluorescence lifetime. Both EQE and efficiency roll-off are ones of the best performance among the reported TADF OLEDs based on exciplex.

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1. Introduction

Organic light-emitting diodes (OLEDs) have attracted tremendous attention due to their promise value of application in flat panel displays and solid-state [1–3]. The internal quantum (IQE) of OLEDs based on fluorescence is limited to 25%, because it can only use singlet excitons for light emission. The IQE of phosphorescent devices can reach to 100%, because they can harvest both the singlet and the triplet excitons [2,4]. However, most of these materials are hard for practical applications due to their high cost and instability. In the past few years, a lot of efforts have been made to highly emissive thermally activated delayed fluorescence (TADF) materials due to their excellent performance in OLEDs [5–13]. With small singlet-triplet splittings (ΔE_{ST}), they can enable high efficiency by reverse intersystem crossing (RISC) from non-radiative triplet state (T_1) to radiative singlet state (S_1). Therefore, OLEDs based on TADF mechanism can also achieve 100% IQE in theory. Normally, pure organic TADF materials are composed of electron donor and

acceptor components in a molecule, leading to an intramolecular charge transfer to realize a small ΔE_{ST} [7–13]. But it is nontrivial to design a new molecule to attain efficient TADF [14–16]. In addition to intramolecular TADF materials, a promise method to achieve TADF OLEDs is to form exciplex via intermolecular charge transfer between physically electronic donor and acceptor molecules [17–25]. However, it is difficult to select donor and acceptor molecules in conventional materials to achieve devices based on exciplex with excellent properties. At present, there are few exciplex OLEDs which can exhibit external quantum efficiencies (EQEs) higher than 10% [20–25], and more worryingly is that most of these devices suffer a serious efficiency roll-off at high luminescence, which limits them for practical applications.

2,4,6-tris(biphenyl-3-yl)-1,3,5-triazine (T2T) is an excellent commercially available compound and has been used as the host for phosphorescent OLEDs [26] and electron transporting material for TADF OLEDs [14]. In one of our previous works, T2T has been used as the acceptor of a TADF OLED and a blue emission is demonstrated [27]. This device brings an EQE of 4.4%. Higher EQEs can be obtained for the TADF OLEDs with T2T analogues as the acceptors, such as 3P-T2T [19,28], Tm3PyBPZ [23], DPTDCz [22], PO-T2T [21,29], and CNT2T [20]. However, these materials are the

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modification of the molecular structure of T2T and usually commercially unavailable. In this communication, we report a highly efficient OLED based on exciplex with a blended structure containing commercially available 1,1-bis((di-4-tolylamino)phenyl)cyclohexane (TAPC) and T2T as the donor and acceptor, respectively. By optimizing the weight ratio of donor and acceptor materials, the device exhibits a low turn-on voltage of 2.4 V, high current efficiency of 40.4 cd/A, power efficiency of 42.2 lm/W, and EQE of 11.6%. Such an EQE is one of the highest values among the reported exciplex-type TADF OLEDs. More importantly, this device shows a low efficiency roll-off at high luminescence.

2. Experimental detail

The OLEDs were optimized with a configuration of ITO/MoO₃ (3 nm)/TAPC (25 nm)/TAPC:T2T (15 nm)/T2T (5 nm)/Bphen (30 nm)/LiF (1 nm)/Al (100 nm). The molecular structures and energy levels of the materials are shown in Fig. 1. TAPC and Bphen were used as the hole-transporting layer and electron-transporting layer, respectively. The hole mobility of TAPC is 1.0×10^{-2} cm²/V s at $\sim 10^5$ V/cm, and electron of Bphen is 5.2×10^{-4} cm²/V s at $\sim 10^5$ V/cm [30,31]. In consideration of their mobilities, an exciton-blocking layer of T2T was inserted to confine excitons in the emitting layer. ITO coated glasses were pre-cleaned and treated with ultraviolet-ozone for 15 min before fabrication of OLEDs. All layers were thermally evaporated in a vacuum chamber with a base pressure of about 4×10^{-4} Pa. All organic materials were obtained without further purification. Absorption spectra were recorded on a Shimadzu UV-3101PC spectrophotometer. Steady-state photoluminescence (PL) and electroluminescence (EL) spectra were measured with a Shimadzu F7000 and OPT-2000 spectrophotometers, respectively. Transient PL decay was measured with a combination of Nd-YAG laser (pulse width of 10 ns, repetition frequency of 10 Hz), spectrograph (HJY, Triax 550) and oscilloscope (Tektronix, TDS3052B). The electrical properties of devices were measured with a Keithley 2400 source meter under ambient atmosphere without any protective coatings. EQE was calculated from the current density, luminance, and EL spectrum, assuming a Lambertian distribution.

3. Results and discussion

Fig. 2 shows the absorption and PL spectra of TAPC, T2T, and TAPC:T2T (4:6) films. The PL peak of TAPC is 442 nm, while it is

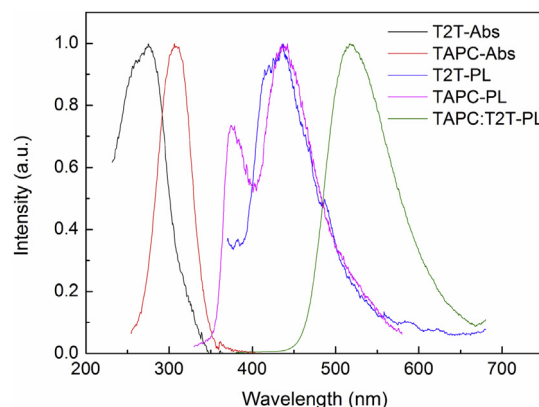


Fig. 2. Absorption and PL spectra of TAPC, T2T, and TAPC:T2T (4:6).

437 nm for T2T. However, the PL of TAPC:T2T film is broad with a peak of 517 nm, which is significantly red-shifted relative to those of the constituting molecules. The PL of the TAPC:T2T film corresponding to a bandgap of 2.4 eV, which is close to the difference of 2.6 eV between the highest occupied molecular orbital (HOMO) energy level of TAPC and the lowest unoccupied molecular orbital (LUMO) energy level of T2T. Thus this emission can be attributed to the exciplex formed between TAPC and T2T after excitation. The fluorescence quantum yields (Φ_{PL}) of this TAPC:T2T system is 0.69 measured with an integrating sphere. Such a high Φ_{PL} suggests the device with this blended film may get a high efficiency.

The delayed fluorescence character of this exciplex can be confirmed by the transition PL decay of the TAPC:T2T (4:6) film. Fig. 3 shows the transition PL decay at 517 nm with 310 nm excitation at 300 K. The decay has two decay components and can be fitted by the formula as follows:

$$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \quad (1)$$

The calculated lifetimes of prompt and delayed components are 15 ns and 1.09 μ s, respectively. Thus the prompt (Φ_{PF}) and delayed (Φ_{DF}) fluorescence components of Φ_{PL} are 0.06 and 0.63, respectively [8]. This suggests that most of the fluorescence is contributed from the delayed component.

The delayed fluorescence can be found with the mechanisms of triplet-triplet annihilation [32] and TADF [9]. To distinguish these effects, ΔE_{ST} is calculated. The ΔE_{ST} can be calculated with the equation [25]:

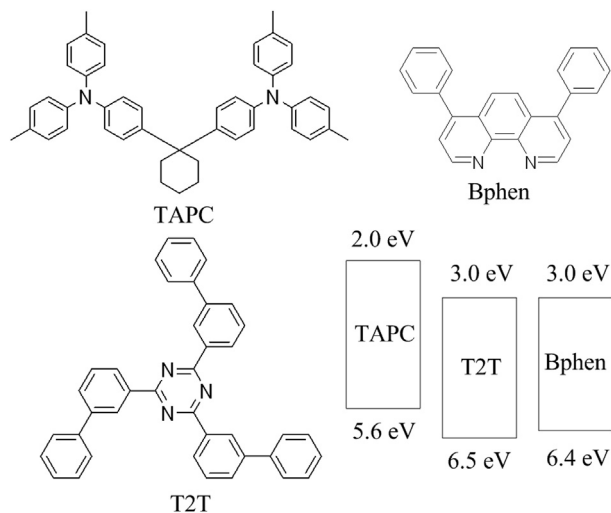


Fig. 1. Molecular structures and energy levels of TAPC, Bphen, and T2T.

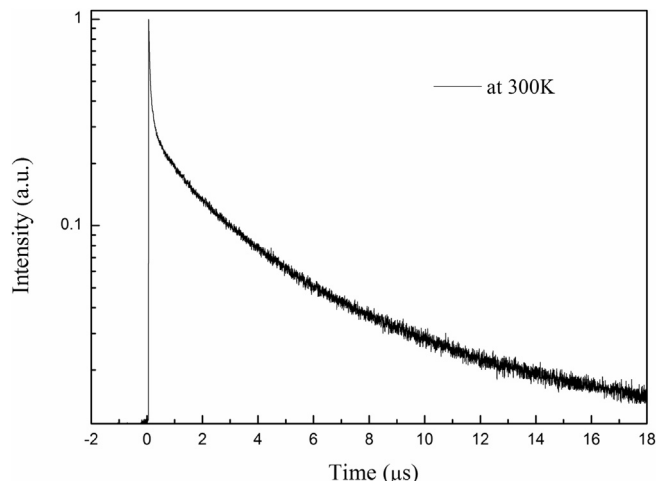


Fig. 3. PL decay curves of the TAPC:T2T (4:6) film at 300 K.

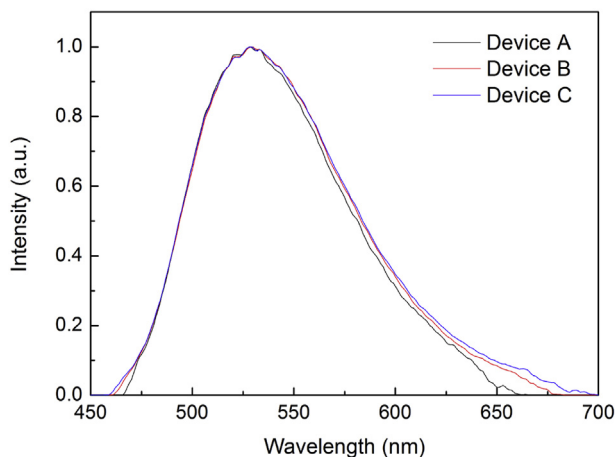


Fig. 4. EL spectra of the devices at 4 V.

$$\Delta E_{ST} = RT \ln(K_{eq}/3) \quad (2)$$

where R is the ideal gas constant, T is the absolute temperature, K_{eq} is the equilibrium constant, and the factor of 3 stands for the triplet degenerate states. K_{eq} can be obtained by the ratio of A_1 and A_2 in equation (1) and is deduced to be 6.3 at 300 K. Thus ΔE_{ST} is calculated to be 19.1 meV. Such a low ΔE_{ST} ensures a high RISC rate (k_{RISC}) in the TAPC:T2T system. Thus the delayed fluorescence can be primarily assigned to the TADF, and the prompt component can be attributed to the radiation of the excitons formed directly on the S_1 , while the delayed one to the RISC from T_1 to S_1 of the exciplex.

Table 1
Parameters of devices with different weight ratios.

	Weight ratio of TAPC:T2T	Maximum luminance (cd/m ²)	EQE _{max} (%)	EQE at 1000 cd/m ² (%)
Device A	3:7	8787	8.5	6.1
Device B	4:6	13476	11.6	9.4
Device C	5:5	10436	8.6	6.0

Then OLEDs with this TAPC:T2T layer are fabricated. The weight ratio of TAPC:T2T is increased from 3:7 to 4:6 and 5:5, corresponding to Device A, B, and C, respectively. Fig. 4 shows the EL spectra of these devices at 4 V. All the three devices exhibit the same EL spectrum with the peak at 528 nm, indicating that the weight ratio has no effect on the EL spectrum. Comparing with PL spectrum, the EL spectra are red-shifted for about 10 nm, and these emissions can also be assigned to the TAPC:T2T exciplex.

Fig. 5 presents the performance of the devices, and the parameters derived from these curves are given in Table 1. Device B with a TAPC:T2T ratio of 4:6 exhibits the highest current density and luminescence at the same voltage among the three devices, indicating that a more balanced hole and electron injections in this device. As a result, Device B shows the highest current efficiency, power efficiency, and EQE of 40.4 cd/A, 42.2 lm/W, and 11.6%, respectively. Such an EQE is one of the highest values among the reported TADF OLEDs based exciplexes [20–25]. It should be noted that Device B shows a low turn on voltage (V_{on}) (corresponding to a luminescence of 1 cd/m²) of 2.4 V. Such a V_{on} is consistent to the theoretical minimum value defined by corresponding emission peak (528 nm) photo energy ($h\nu = 2.35$ eV) divided by the electron charge (e).

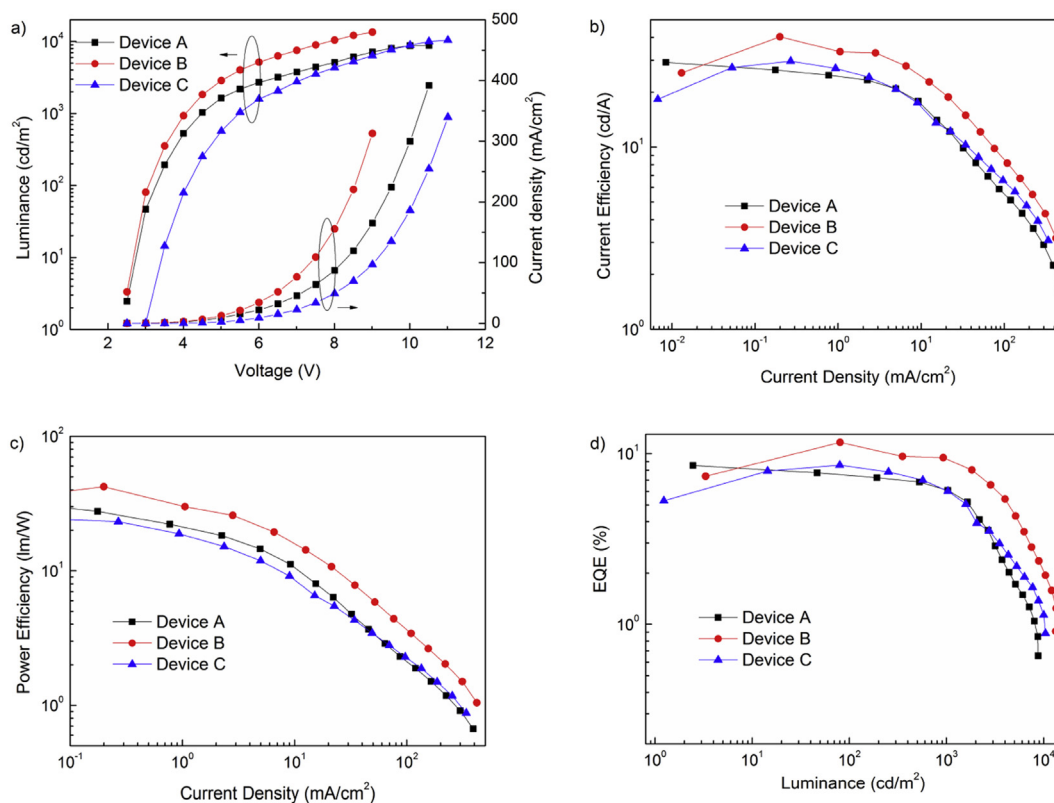


Fig. 5. a) Current density-luminance-voltage plots of devices A–C. b) Current efficiency versus current density of devices A–C. c) Power efficiency versus current density of devices A–C. d) EQE-luminance characteristics of devices A–C.

Table 2
Summary of high-efficiency TADF OLEDs based on exciplex emitters.

Emitter	V _{on} ^a (V)	EQE _{max} (%)	EQE at 1000 cd/m ² (%)	Roll-off ^b (%)	Delayed fluorescence lifetime (μs)	Reference
TAPC:T2T	2.4	11.6	9.4	19.0	1.09	This work
Tris-PCz:CNT2T	2.0	11.9	11.1	6.7	2.85	Ref. [20]
MAC:PO-T2T	2.4	17.8	12.3	30.9	—	Ref. [21]
TAPC:DPTPCz	2.7	15.4	8.3	46.1	1.8	Ref. [22]
TCTA:Tm3PyBPZ	2.4	13.1	8.8	32.8	2.3	Ref. [23]
mCP:HAP-3MF	4.0	11.3	8.0	29.2	1.7	Ref. [24]
m-MTDATA:PPT	—	10.0	4.0	60.0	—	Ref. [25]

^a Turn-on voltage, estimated at the brightness of 1 cd/m².

^b Decrease of EQE from maximum to 1000 cd/m².

Under electrical excitation, the internal quantum efficiency ($\Phi_{\text{EL(int)}}$) of a TADF OLED can be calculated with the formula [7,10]:

$$\Phi_{\text{EL(int)}} = \eta_r(S_1)\Phi_{\text{PF}} + [\eta_r(T_1) + \eta_r(S_1)(1 - \Phi_{\text{PF}})] \frac{\Phi_{\text{DF}}}{(1 - \Phi_{\text{PF}})} \quad (3)$$

where $\eta_r(S_1)$ and $\eta_r(T_1)$ are the branching ratio of singlet and triplet exciton formations, respectively. Thus, $\Phi_{\text{EL(int)}}$ is calculated to be 0.67. Assuming a light out-coupling efficiency of 20%, the maximum EQE of the device is estimated to be 13.4%, which is comparable to the measured EQE.

For display and lighting applications, a high luminescence is generally required. However, the highest EQE is usually obtained at an extremely low luminescence for most of the reported TADF OLEDs based on exciplex. Thus a low efficiency roll-off is highly desired. Table 2 compares the properties of our Device B to other reported state-of-the-art TADF OLEDs based on exciplexes. Most of those devices exhibit an efficiency roll-off higher than 20% at 1000 cd/m² compared to their respective maximums. The lowest efficiency roll-off was recently reported by Hung et al., which is only 6.7% [20]. The EQEs at 1000 cd/m² of our Device B is 9.4%, corresponding to a decrease of 19.0% to its maximum. Such an efficiency roll-off is one of the lowest values among the reported TADF OLEDs based on exciplexes [20–25]. Besides, although the maximum EQE of our device is a little lower, the EQE at 1000 cd/m² is higher than most of the reported devices.

The efficiency roll-off of a TADF OLED is primary attributed to the long delayed fluorescence lifetime which related to its large ΔE_{ST} and to the unbalanced charge carrier injection [11,33,34]. These result in a serious triplet exciton quenching, such as single-triplet exciton annihilation [35], triplet-triplet exciton annihilation [36], and triplet-polaron annihilation [37]. Thus a high k_{RISC} will indicate a low efficiency roll-off of the device. In assuming that the non-radiative rate from T_1 of the exciplex is zero, the inter-system crossing rate (k_{ISC}) and k_{RISC} are described with the following formulas [17,38]:

$$k_{\text{ISC}} = \frac{\Phi_{\text{DF}}}{\Phi_{\text{DF}} + \Phi_{\text{PF}}} \times k_{\text{PF}} \quad (4)$$

$$k_{\text{RISC}} = \frac{k_{\text{DF}}k_{\text{PF}}}{k_{\text{ISC}}} \frac{\Phi_{\text{DF}}}{\Phi_{\text{PF}}} \quad (5)$$

where k_{PF} and k_{DF} are prompt and delayed fluorescence rate constants, respectively. Then k_{ISC} and k_{RISC} of our Device B can be calculated to be $3.65 \times 10^6/\text{s}$ and $6.65 \times 10^6/\text{s}$, respectively. It should be noted that k_{RISC} is a little higher than k_{ISC} , which may be attributed to the assumption that the non-radiative rate from T_1 of the exciplex is zero, and similar results have been found by Ishimatsu et al. [39]. Meanwhile, it can be found that this k_{RISC} is a little higher than that in Tris-PCz:CNT2T device ($2.29 \times 10^6/\text{s}$ calculated from the data provided in their paper) reported by Hung et al. [20],

which can be attributed to the short delayed fluorescence lifetime and small ΔE_{ST} of TAPC:T2T film. Although Tris-PCz:CNT2T has a long delayed fluorescence lifetime, it exhibits a low efficiency roll-off. This can be attributed to the balance between hole mobility of Tris-PCz and electron mobility of CNT2T, which are both in the range of 10^{-5} to 10^{-4} cm²/V s. However, the hole mobility of TAPC (1.0×10^{-2} cm²/V s) are two orders higher than electron mobility of T2T (1.2×10^{-4} cm²/V s) at $\sim 10^5$ V/cm [30,31]. This indicates that charge carrier injection balance not only plays an important role in determining the maximum EQE [20], but also the efficiency roll-off of a TADF OLED [11,33,34].

4. Conclusions

In summary, we demonstrate a highly efficient TADF OLED based on exciplex in the TAPC:T2T mixed system. The optimized device shows a low turn-on voltage of 2.4 V, high current efficiency of 40.4 cd/A, power efficiency of 42.2 lm/W, and EQE of 11.6%. Such an EQE is one of the highest values among the reported TADF OLEDs based on exciplex. More importantly, the device retains an EQE of 9.4% even at a high luminescence of 1000 cd/m², indicating an efficiency roll-off of about 19%. Such an efficiency roll-off is one of the lowest values among the TADF OLEDs based on exciplex. The low efficiency roll-off is attributed to the small ΔE_{ST} and the short delayed fluorescence lifetime. The findings of this work indicate that high performance TADF OLEDs can also be fabricated with commercially available compounds by well designing the device structure.

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