



Research Article

Efficient triplet harvest for orange-red and white OLEDs based exciplex host with different donor/acceptor ratios

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ABSTRACT

Highly efficient orange-red and white organic light-emitting diodes (OLEDs) based exciplex host with different donor/acceptor ratios and doping concentrations are fabricated for efficient triplet excitons harvest. As a result, the maximum efficiency is achieved at 5:5 ratios exciplex host in orange-red OLEDs with current efficiency, power efficiency and external quantum efficiency (EQE) of 37.0 cd/A, 37.3 lm/W and 18.3%, respectively. Furthermore, the white OLEDs under two different doping concentrations give warm white (0.48, 0.41) and cool white (0.28, 0.37) emission with maximum current efficiencies, power efficiencies and EQEs of 38.3/37.6 cd/A, 41.3/43.7 lm/W and 17.8/16.6%, respectively. We found that the triplet exciton harvest on exciplex host through reverse intersystem crossing process and efficient energy transfer are responsible for the high device efficiency.

1. Introduction

Exciplex organic light-emitting diodes (OLEDs) received more and more attention since the thermally activated delayed fluorescent (TADF) mechanism was discovered by Adachi group in 2012 [1–3]. TADF is the singlet exciton radiative transition that from triplet exciton reverse intersystem crossing (RISC) process due to the small singlet-triplet state energy level splitting (ΔE_{ST}). Exciplex that formed from intermolecular charge transfer conducts spatially separated highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), which present natural small ΔE_{ST} for high triplet exciton harvest and utilization. Therefore, a series of high efficiency exciplex OLEDs with blue, green and yellow emission were explored by suitable donor/acceptor materials selection [4–8]. In generally, the donor materials are hole transport materials and acceptor materials are electron transport materials, respectively. And the mixed ratio of donor/acceptor materials is 1:1 in most of exciplex OLEDs.

Furthermore, mixed exciplex could be applied as host to sensitize dopant for high efficiency OLEDs due to the excellent charge transport bipolarity, efficient triplet harvest and energy transfer property [9,10]. The mixed hole and electron transport materials make the exciplex layer

exhibit high hole and electron transport ability, which could improve charge recombination efficiency, extend exciton formation zone and reduce exciton concentration. High efficiency exciplex presents efficient TADF behavior, which could improve triplet harvest for high exciton utilization. The highly efficient triplet exciton RISC process could also enhance the long range Förster energy transfer between singlet state energy level of host and dopant. These outstanding characteristics guarantee the efficient application of exciplex in host role and the dopants of traditional fluorescent, phosphorescent and TADF emitter all could be employed in exciplex host to achieve highly efficient dopant emission. Kim et al. reported a series of high efficiency blue, green, orange and white OLEDs by utilizing phosphorescent emitter as dopant and exciplex as host since 2013 [11–15]. Traditional fluorescent emitter of 4-(dicyanomethylene)-2-t-butyl-6-(1,1,7,7-tetra-methyljulolidyl-9-enyl)-4H-pyran (DCJTb) could be also doped into exciplex host to break 10% external quantum efficiency (EQE) [16]. While TADF emitter of 9-[4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-N,N,N',N'-tetraphenyl-9H-carbazole-3,6-diamine (DACT-II) acts as dopant to apply in exciplex host could reach to 34.2% EQE through two RISC processes of TADF emitter and TADF exciplex host [17]. Thus, the high efficiency OLEDs could be realized in exciplex host by efficient triplet exciton harvest.

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However, most of donor/acceptor ratio in exciplex host reported to now is 5:5, the different mixed ratio donor/acceptor in exciplex host is very rare. Besides, the effect of small ratio donor or acceptor in exciplex host is also need be explored to extend more application to develop OLEDs based exciplex further.

In this work, we fabricated four different donor/acceptor ratios exciplex host to sensitize orange-red phosphorescent dopant of Iridium (III) bis(2-phenylquinoline) acetylacetonate (Ir (pq)₂acac). As a result, the 5:5 ratio achieved the best electroluminescence (EL) performance with maximum current efficiency, power efficiency and EQE of 37.0 cd/A, 37.3 lm/W and 18.3%, respectively. While other OLEDs with mixed ratios of 7:3, 8:2 and 9:1 also obtained a high efficiency level with maximum EQEs of 17.3%, 17.4% and 16.5%, respectively. Further, highly efficient warm and cool white OLEDs with the same exciplex host but smaller acceptor ratio of 1% were realized. The maximum current efficiency, power efficiency and EQE of 38.3 cd/A, 41.3 lm/W and 17.8% in warm white OLEDs and 37.6 cd/A, 43.7 lm/W and 16.6% in cool white OLEDs were realized. Our results demonstrated that the acceptor ratio had little effect on exciplex host to achieve highly efficient OLEDs and the white OLEDs could be also realized with small acceptor ratio and low doping concentration.

2. Experimental section

Indium tin oxide (ITO) coated glass substrates were cleaned routinely and treated with ultraviolet-ozone for 15 min before loading into a high vacuum deposition chamber ($\sim 3 \times 10^{-4}$ Pa). The organic materials were purchased commercially without further purification. And the organic layers were deposited at a rate of 1.0 Å/s, inorganic layers of MoO₃ and LiF at the deposition rate of 0.1 Å/s. Al cathode was deposited in the end with a shadow mask, which defined the device active area of 3×3 mm². EL spectra were measured with OPT-2000 spectrophotometer. The electrical characteristics of the OLEDs were measured with a Keithley model 2400 power supply combined with a ST-900 M spot photometer and were recorded simultaneously. EQE was calculated from the current density, luminance and spectra data. All measurements were carried out at room temperature and under ambient conditions without any protective coatings.

3. Results and discussions

The exciplex host is selected as bis [4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone:(1,3,5-triazine-2,4,6-triyl)tris (benzene-3,1-diyl)tris (diphenylphosphine oxide) (DMAC-DPS:PO-T2T), which is a highly efficient exciplex with high singlet and triplet energy level to act host to sensitize orange-red dopant of Ir (pq)₂acac [18,19]. The photoluminescence (PL) behaviors presented that exciplex could well formed between DMAC-DPS and PO-T2T with high photoluminescence quantum yield (PLQY) over 40%, and the exciplex exhibited highly efficient

TADF characteristics through transient PL decay measurement [19]. Herein, we more focus on the EL behaviors of the host role of exciplex. So the device structure designed as follows: ITO/MoO₃ (3 nm)/mCP (25 nm)/DMAC-DPS:PO-T2T (x:y): 2% Ir (pq)₂acac (20 nm)/TPBi (40 nm)/LiF (1 nm)/Al (100 nm). m-bis(N-carbazolyl)benzene (mCP) and 1,3,5-tris(N-phenyl-benzimidazol-2-yl)benzene (TPBi) are the hole and electron transport layers, respectively. ITO/MoO₃ and LiF/Al are the role of composite anode and composite cathode, respectively. DMAC-DPS:PO-T2T (x:y): 2% Ir (pq)₂acac is the emitting layer (EML) with exciplex host conducted different donor/acceptor ratios to sensitize dopant. 2% concentration of Ir (pq)₂acac could ensure the complete energy transfer to achieve pure dopant emission. The corresponding molecular structures and device energy level diagram are showed in Fig. 1. The donor/acceptor ratios (x:y) between DMAC-DPS and PO-T2T are used as 5:5, 7:3, 8:2 and 9:1, that means the donor of DMAC-DPS molecule is the majority due to its excellent carrier transport bipolarity [20,21], which could make the efficient charge injection, transport and recombination. On the contrary, the ratios of donor/acceptor lower than 50%, that is PO-T2T is the majority, would block the charge injection and transport seriously, which results from the high hole injection barrier and poor charge transport ability of PO-T2T. Therefore, the donor/acceptor ratios of 5:5, 7:3, 8:2 and 9:1 are conducted in this work.

The EL performance of OLEDs with EML of DMAC-DPS:PO-T2T (x:y): 2% Ir (pq)₂acac are displayed in Fig. 2. Under the different donor/acceptor ratios of 5:5, 7:3, 8:2 and 9:1, all four devices exhibit high EL performances with turn-on voltage of ~ 3 V, maximum luminance of 30,000–45000 cd/m² and intrinsic orange-red emission of Ir (pq)₂acac, which indicate the efficient charge injection, transport, recombination and complete energy transfer from exciplex host to dopant [22]. From the energy level diagram shown in Fig. 1b, we can see that there have no the energy level barrier between transport layer and EML. In our device, the donor of DMAC-DPS is the majority molecule, so the charge injection and transport is dependent on DMAC-DPS. The HOMO energy level of mCP and DMAC-DPS is 6.1 eV and 5.9 eV [23,24], while the LUMO energy level of TPBi and DMAC-DPS is 2.9 eV and 2.7 eV [24,25], respectively. Therefore, the carrier injection barrier from mCP and TPBi to DMAC-DPS is zero, which results to highly efficient charge injection. The bipolarity of DMAC-DPS could transport hole and electron in EML efficiently, which gives the efficient recombination for high EL efficiency. In the four different ratios OLEDs with exciplex host, 5:5 ratio present the maximum efficiency with current efficiency, power efficiency and EQE of 37.0 cd/A, 37.3 lm/W and 18.3%, respectively. While the moderate EQEs of 17.3%, 17.4% and 16.5% are also obtained under other ratios of 7:3, 8:2 and 9:1, respectively. That means the exciplex host with different donor/acceptor ratios have little effect on the device performance and high EQEs of 16.5%–18.3% are realized with various ratios. In the previous reports, the exciplex of DMAC-DPS:PO-T2T shows efficient TADF characteristics [19], which could harvest triplet excitons

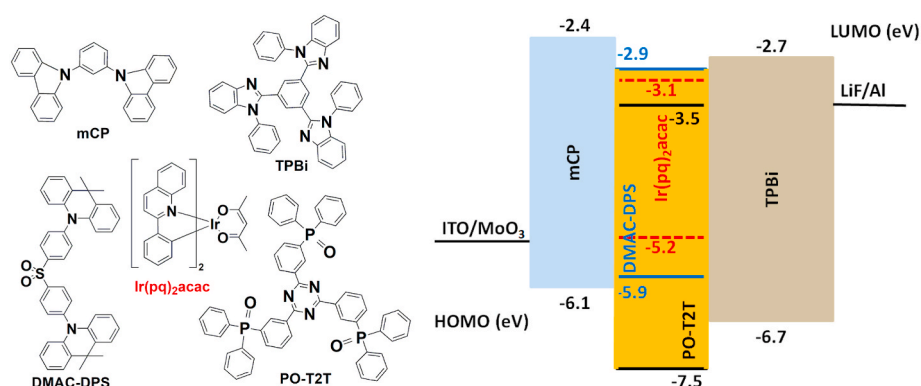


Fig. 1. The molecular structure of organic materials used in this work and device energy level diagram.

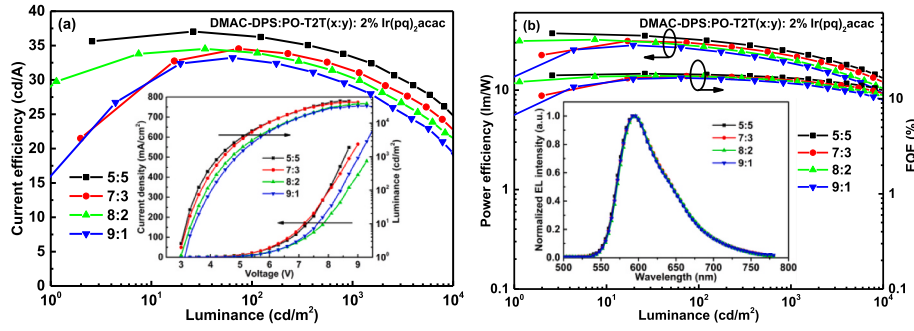


Fig. 2. The EL performances of OLEDs with EML of DMAC-DPS:PO-T2T (x:y): 2% Ir (pq)₂acac. (a) Current efficiency-luminance curves. Inset is the current density-voltage-luminance curves. (b) Power efficiency-luminance-EQE curves. Inset is the EL spectra under different donor/acceptor ratios.

by RISC process. So the high EQE in this work demonstrate the triplet excitons could be also collected under low ratio acceptor of PO-T2T for efficient energy transfer.

Furthermore, we found that the small ratio acceptor of PO-T2T with 5% and 1% in DAMC-DPS donor is also efficient to form exciplex in our previous work, high EQE of >10% could be realized in pure exciplex emission of DMAC-DPS:PO-T2T with different donor/acceptor ratios and the emission peak could fix on 496 nm of blue emission with 1% PO-T2T in DMAC-DPS [26]. So here we design the EML of DMAC-DPS: 1% PO-T2T: x% Ir (pq)₂acac to fabricate white OLEDs to explore the application of small ratio acceptor exciplex host on white OLEDs. The EL efficiency curves of white OLEDs with 1.0% and 0.5% Ir (pq)₂acac dopant are showed in Fig. 3. The white OLEDs with 1.0% Ir (pq)₂acac exhibits high efficiency with maximum current efficiency, power efficiency and EQE of 38.3 cd/A, 41.3 lm/W and 17.8%, respectively. While maximum current efficiency, power efficiency and EQE of 37.6 cd/A, 43.7 lm/W and 16.6%, respectively, are also obtained in the white OLEDs with 0.5% Ir (pq)₂acac. Meantime, a low turn-on voltage of ~2.5 V and high luminance of ~20,000 cd/m² are also achieved in the two white OLEDs. The almost same current density-voltage curves showed in inset of Fig. 1a demonstrates the energy transfer is the main emission mechanism rather than direct charge trapping in the white OLEDs [27–29]. Besides, the low dopant concentration of 1.0% and 0.5% also make the trapping difficult to happen [30]. The detailed energy transfer and emission process would be described in the next section. And the EL performances of all the OLEDs in this paper are summarized in Table 1.

The normalized EL spectra of white OLEDs with 1.0% and 0.5% concentration are displayed in Fig. 4. Both of the two white OLEDs exhibit two emission peaks with blue peak of exciplex host of DMAC-DPS:PO-T2T and orange-red peak of dopant of Ir (pq)₂acac without other emission behavior. More important, warm white and cool white emission is realized in the two white OLEDs, respectively. The concentration of 1.0% white OLEDs shows the warm white emission of stronger orange-red emission with Commission Internationale de l'Eclairage (CIE) coordinates of (0.48, 0.41) at 6 V. While cool white emission of

Table 1

The list of orange-red and white OLEDs performance under different donor/acceptor ratios and concentrations in this paper.

DMAC-DPS:PO-T2T (D:A = x: y): z% Ir (pq) ₂ acac (dopant)	CE _{max} ^a (cd/A)	PE _{max} ^b (lm/W)	EQE _{max} ^c (%)	CIE at 6 V
D:A (5:5): 2% dopant	37.0	37.3	18.3	(0.59, 0.40)
D:A (7:3): 2% dopant	34.5	31.2	17.3	(0.59, 0.40)
D:A (8:2): 2% dopant	34.5	32.1	17.4	(0.59, 0.40)
D:A (9:1): 2% dopant	33.2	28.2	16.5	(0.59, 0.40)
D:1% A: 1% dopant	38.3	41.3	17.8	(0.48, 0.41)
D:1% A: 0.5% dopant	37.6	43.7	16.6	(0.28, 0.37)

^a Maximum current efficiency.

^b Maximum power efficiency.

^c Maximum EQE.

stronger blue emission with CIE coordinates of (0.28, 0.37) at 6 V is achieved in the white OLEDs with 0.5% concentration. That means the spectra could be modulated easily by change the doping concentration, which derived from the efficient control of energy transfer between exciplex host and dopant. The blue emission intensity is enhanced with increased voltages, which may be derived from the insufficient energy transfer from exciplex host to dopant, which leads to a mass of excitons produced with increased voltage, could not be transferred to dopant efficiently.

In order to clarify the exciton formation and energy transfer mechanism further, the schematic diagram of energy transfer and emission process are described in Fig. 5. In orange-red OLEDs, the energy transfer and direct charge trapping could be obtained due to the relative high doping concentration of 2%. First, the triplet excitons produced on exciplex host convert into singlet excitons through RISC process due to

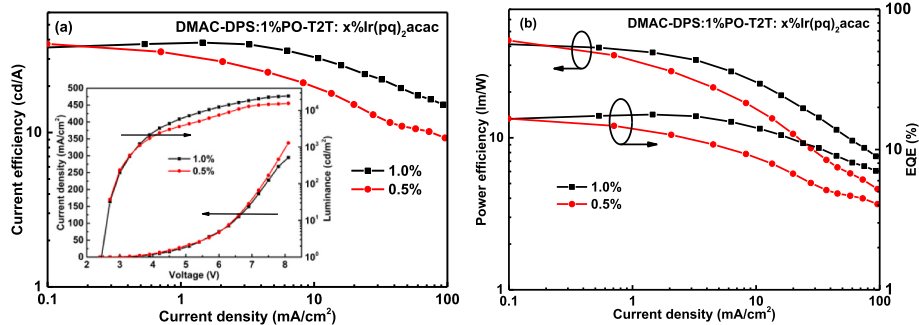


Fig. 3. The EL efficiencies of white OLEDs with EML of DMAC-DPS:1%PO-T2T: x% Ir (pq)₂acac (x = 1.0 and 0.5). (a) Current efficiency-current density curves. Inset is the current density-voltage-luminance curves. (b) Power efficiency-current density-EQE curves.

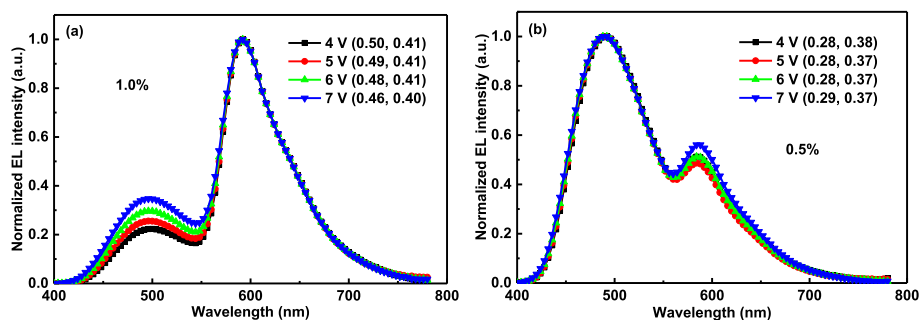


Fig. 4. The normalized EL spectra of white OLEDs under different voltages with 1.0% and 0.5% concentrations. (a) 1.0% concentration. (b) 0.5% concentration.

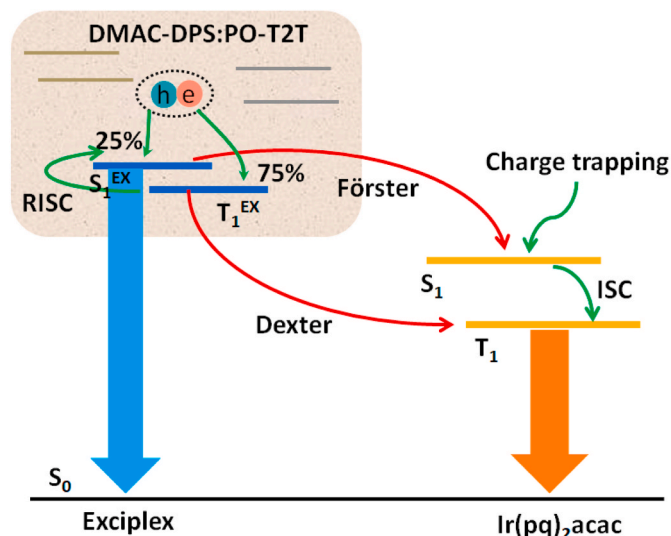


Fig. 5. The schematic diagram of energy transfer and emission process in our OLEDs.

the small singlet-triplet state energy level splitting. Then the long range Förster energy transfer between singlet energy level could transfer the singlet exciton to dopant for phosphorescent radiative transition from triplet energy level by subsequent intersystem crossing (ISC). Besides, the Dexter energy transfer between triplet energy level may be also efficient due to the relative high doping concentration. Therefore, the orange-red emission in monochromatic OLEDs could be originated from the direct charge trapping recombination, long range Förster and short range Dexter energy transfer with the relative high concentration of 2%. While in the white OLEDs, the doping concentration is reduced to 1.0% and 0.5%, so the direct charge trapping recombination and Dexter energy transfer could be restrained efficiently [30]. Hence, the exciplex host triplet exciton RISC and followed by long range Förster energy transfer becomes the main pathway for dopant emission. The realization of white emission is also derived from the low doping concentration for incomplete energy transfer, which acquire the exciplex host blue and dopant orange-red emission simultaneously for white emission. In one word, the triplet exciton RISC process in exciplex host plays a key role for efficient triplet exciton harvest for high OLEDs efficiency.

4. Conclusion

In conclusion, the orange-red and white OLEDs are realized by employing different donor/acceptor ratios exciplex host and doping concentrations. The orange-red OLEDs with various donor/acceptor ratios (9:1, 8:2, 7:3 and 5:5) exciplex host give a high maximum current efficiencies, power efficiencies and EQEs of 33.2–37.0 cd/A, 28.2–37.3 lm/W and 16.5–18.3%, respectively. While the white OLEDs with

different doping concentration (1.0% and 0.5%) also present high EL efficiency with maximum current efficiencies, power efficiencies and EQEs of 37.6–38.3 cd/A, 41.3–43.7 lm/W and 16.6–17.8%, respectively. Besides, the warm white emission with CIE coordinates of (0.48, 0.41) and cool white emission of (0.28, 0.37) are also achieved in the white OLEDs with doping concentration of 1.0% and 0.5%, respectively. The results demonstrate the donor/acceptor ratio of exciplex host have little effect on device performance and triplet exciton RISC process in small acceptor ratio exciplex host also acts as the important function for efficient exciton harvest and high device efficiency. We believe the application of exciplex host would promote the development of OLEDs in more device structure design.

CRedit authorship contribution statement

Qingjiang Ren: Writing – original draft, Data curation, Formal analysis, wrote the manuscript and conducted most of the experiments and data collection analysis. **Yi Zhao:** guided the progress of experiments and manuscript. **Chang Liu:** Formal analysis, took part in the data analysis and discussions. All authors have reviewed the manuscript. **Hongmei Zhan:** Formal analysis, took part in the data analysis and discussions. All authors have reviewed the manuscript. **Yanxiang Cheng:** Formal analysis, took part in the data analysis and discussions. All authors have reviewed the manuscript. **Wenlian Li:** guided the progress of experiments and manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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