Optical Materials 112 (2021) 110779

Contents lists available at ScienceDirect

Optical Materials

journal homepage: http://www.elsevier.com/locate/optmat

Research Article

The exploration of acceptor ratio in thermally activated delayed fluorescent donor for the effect on exciplex OLED

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ARTICLE INFO

Keywords:
OLED
Exciplex
Mixed ratio
Spectral move

ABSTRACT

The effect of PO-T2T acceptor ratio in thermally activated delayed fluorescent donor of DMAC-DPS on exciplex organic light-emitting diodes (OLED) performance is explored herein. As the ratio decrease of PO-T2T acceptor, we found an obvious blue shift of electroluminescence (EL) spectra from 536 nm to 496 nm, while the EL efficiency maintain a high level with maximum current efficiency of 25.7–38.1 cd/A, power efficiency of 24.2–39.5 l m/W and external quantum efficiency (EQE) of 10.4–12.1%, respectively. That means a wide spectral move range of 40 nm without damaging device EQE in exciplex OLED could be realized. The strong interaction between exciplex induced from charge separation is responsible for the blue shift as acceptor ratio reduce.

1. Introduction

Exciplex emission received much more attention due to its special emission mode that formed from the intermolecular charge transfer of donor and acceptor. And exciplex emission is derived from the charge recombination between the holes on highest occupied molecular orbital (HOMO) energy level of donor and electrons on lowest unoccupied molecular orbital (LUMO) energy level of acceptor. While high efficiency exciplex organic light-emitting diodes (OLED) is exploited by Adachi group in 2012 through the discovery of thermally activated delayed fluorescent (TADF) mechanism with suitable selection of donor and acceptor materials [1,2]. TADF mechanism is the triplet exciton reverse intersystem crossing (RISC) from triplet state energy level to singlet state energy level and contributes to more radiative transition singlet exciton to improve OLED efficiency [3–7]. Spatially separated intermolecular HOMO and LUMO between donor and acceptor results in the small singlet-triplet state energy level gap (ΔE_{ST}) and ensure the efficient RISC process. Therefore, excipelx conducted natural small ΔE_{ST} for highly efficient triplet exciton RISC. After that, a series of high efficiency exciplex OLED with blue, green, yellow and white emission were reported [8–14].

On one hand, the selection of suitable donor/acceptor materials is a key factor for high efficiency exciplex OLED, on the other hand, the mixed ratio of donor/acceptor materials is also very important. In generally, the donor is hole transport material and acceptor is electron transport material and the donor/acceptor ratio is 1:1 in the most exciplex. However, some literature reported that the TADF emitter could also act as donor or acceptor to form exciplex, and the ratio could be also adjusted to balance charge transport and recombination. Zhang et al. studied the exciplex emission by employing TADF emitter of bis [4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (DMAC-DPS) as donor or acceptor with 1:1 ratio, which demonstrated the efficient role to act donor/acceptor [15]. Liu et al. developed high efficiency exciplex OLED with maximum external quantum efficiency (EQE) of 17.8% through two efficient RISC process by utilizing TADF emitter of 6-(9,9-dimethylacridin-10(9H)-yl)-3-methyl-1H-isochromen-1-one (MAC) as donor and different ratio (1,3,5-triazine-2,4,6-triyl)tris (benzene-3,1-diyl) tris (diphenylphosphine oxide) (PO-T2T) as acceptor [16]. Zhao et al. also fabricated the yellow-green exciplex OLED with 9.7% EQE by the suitable selection TADF donor emitter of bis(3-(9,9-dimethyl-9,10-dihydroacridine)phenyl)sulfone (mSOAD) with acceptor of PO-T2T under 1:1 ratio [17]. The reason of TADF emitter could play the role of donor/acceptor is the existence of donor/acceptor-unit in the molecular structure. Therefore, the intermolecular charge transfer between

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https://doi.org/10.1016/j.optmat.2020.110779
Received 21 November 2020; Received in revised form 7 December 2020; Accepted 22 December 2020
Available online 30 December 2020

donor/acceptor-unit of TADF molecule and other donor/acceptor materials contributes to the formation of exciplex. And the RISC process of TADF molecule also enhances the harvest of triplet exciton for higher efficiency exciplex. Although the mixed ratio between donor and acceptor could be changed to modulate the exciplex performance, but the more detailed ratio optimization and regulation is rare to study, especially the acceptor small ratio in TADF donor.

Therefore, in this work, we studied the effect of acceptor ratio in TADF donor on exciplex OLED performance. A highly efficient blue TADF emitter of DMAC-DPS is chosen as donor material and PO-T2T as the acceptor material to form the exciplex. Firstly, different mixed ratios OLED based exciplex emission of 1:9, 3:7, 5:5, 7:3 and 9:1 between DMAC-DPS and PO-T2T were fabricated and researched. Further, the lower ratio PO-T2T acceptor of 5% and 1% in DMAC-DPS donor were also exploited. As a result, the device efficiency of ~10% EQE remains almost the same with little change, while an obvious blue shift was observed in the electroluminescence (EL) spectra as the decrease of acceptor ratio of PO-T2T.

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 MoO_3 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 mm^2. 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 discussion

A highly efficient TADF emitter of DMAC-DPS was selected as donor materials and PO-T2T based a phosphine oxide as acceptor material, respectively. The exciplex characteristics had been proved in previous literature reports from photoluminescence (PL) and EL between DMAC-DPS and PO-T2T, which demonstrated a highly efficient exciplex emission with TADF behavior [15,18]. So here we focused on the effect of different mixed ratio on device performances. Firstly, we designed a common exciplex OLED with the structure as follows: ITO/MoO_3 (3 nm)/mCP (25 nm)/DMAC-DPS: PO-T2T (x: y) (20 nm)/TPBi (40 nm)/LiF (1 nm)/Al, where m-bis(N-carbazolyl)benzene (mCP) and 1,3, 5-tris(N-phenyl-benzimidazol-2-yl) benzene (TPBi) are hole transport layer and electron transport layer, respectively. ITO/MoO_3 and LiF/Al are the composite electrodes. The interlayer composed of DMAC-DPS: PO-T2T (x: y) with various mixed ratio is the exciplex emitting layer (EML), and five different mixed ratios of 1:9, 3:7, 5:5, 7:3 and 9:1 are optimized. The molecular structure of organic materials used in this work and corresponding device structure are exhibited in Fig. 1.

The EL performances of the exciplex OLED with different donor/acceptor mixed ratio are showed in Fig. 2. An obvious difference of the current density-voltage-luminance (J-V-L) characteristic between 1:9, 3:7 and 5:5, 7:3, 9:1 could be observed from Fig. 2a. That is, a high operation voltage and low current density are obtained with high PO-T2T ratio (1:9, 3:7). While the operation voltage reduces largely combine with increased current density as the decrease of PO-T2T ratio from 5:5 to 9:1. The acceptor material of PO-T2T conducts a deep HOMO energy level of 7.5 eV, which has strong hole block ability [19,20]. So the high ratio of PO-T2T would prevent the injection of hole into exciplex EML seriously, which lead to the high operation voltage and low current density. To the contrary, the hole injection ability enhances gradually as the decrease of PO-T2T, which lower the recombination barrier and enhance the hole transport. Besides the J-V-L curves, efficiency curves of current efficiency, power efficiency and EQE are also displayed in Fig. 2. Although the J-V-L curves exhibit some difference, the current efficiency and EQE present the similar efficiency level with maximum current efficiency of 33.7–38.1 cd/A and maximum EQE of 10.5–12.1%, which derived from the directly proportional relationship between luminance and current density. While the current efficiency and EQE are calculated mainly from these two factors, so the similar current efficiency and EQE are obtained. However, the higher power efficiency level of 33.7–39.5 l m/W with low ratio PO-T2T (5:5, 7:3 and 9:1) than 24.2–25.3 l m/W with high ratio PO-T2T (1:9 and 3:7) are achieved due to the higher operation voltage in high ratio PO-T2T exciplex OLED. As a whole, high efficiency pure exciplex OLED with maximum current efficiency ~35 cd/A and EQE >10% are realized with various TADF donor/acceptor mixed ratio. The high efficiency of exciplex OLED based DMAC-DPS: PO-T2T is originated from the efficient harvest of triplet exciton through RISC process due to the small singlet-triplet state energy level gap. Thus, the improved radiative transition singlet exciton ratio results to the high exciplex OLED efficiency.

The device EL spectra with different mixed ratio are showed in Fig. 3. To our surprise, the EL spectra present obvious blue shift with low ratio

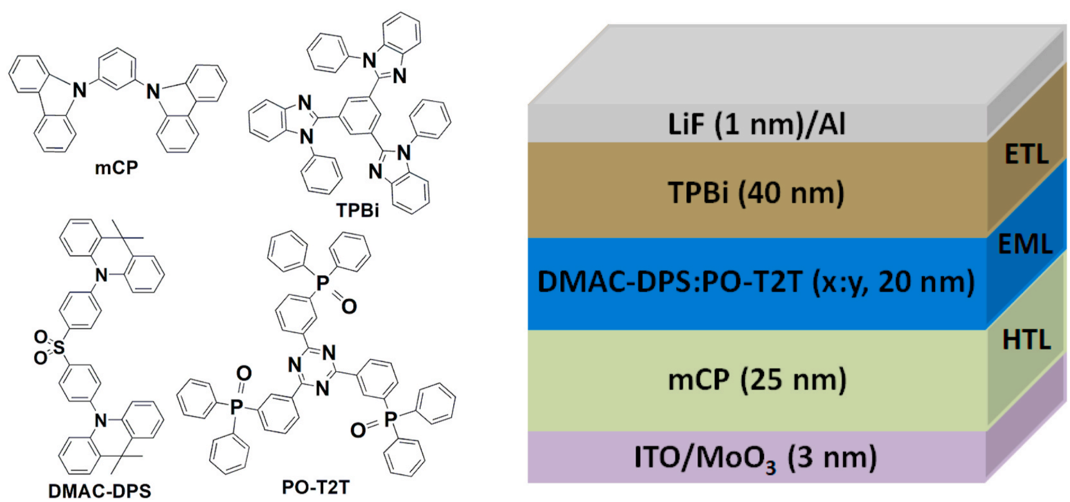

Fig. 1. The organic materials molecular structure and device structure used in this work.

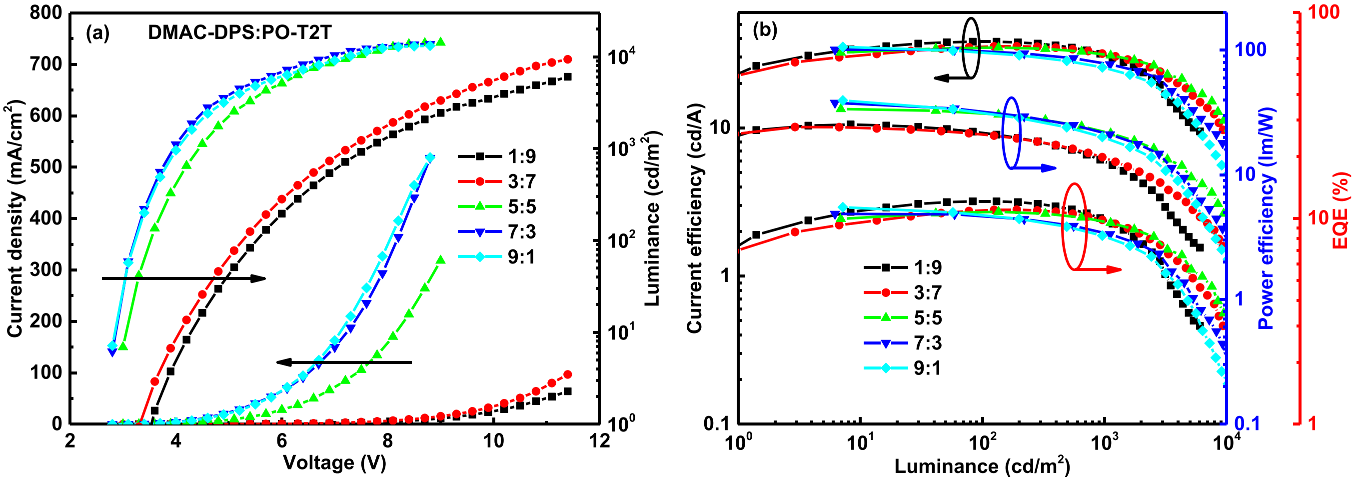

Fig. 2. The EL performance of exciplex OLED based DMAC-DPS: PO-T2T with different mixed ratio of 1:9, 3:7, 5:5, 7:3 and 9:1. (a) Current density-voltage-luminance curves. (b) Current efficiency-luminance-power efficiency/EQE curves.

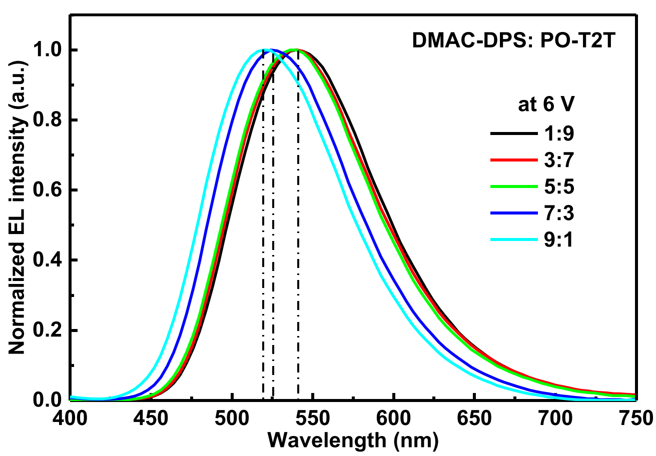

Fig. 3. Normalized EL spectra at 6 V of exciplex OLED with different DMAC-DPS: PO-T2T mixed ratios.

PO-T2T. Under the mixed ratio of 1:9, 3:7 and 5:5, the exciplex OLED exhibit same EL spectra with emission peak of 536 nm. While as the ratio reduces of PO-T2T acceptor, the EL spectra move to short wavelength direction and emission peak fixes on 525 nm and 520 nm under the ratio of 7:3 and 9:1, respectively. That is the blue shift happens when the ratio of PO-T2T is lower than DMAC-DPS, and the lower of PO-T2T ratio, the more blue shift wavelength. According to previous report, TADF donor of DMAC-DPS exhibited highly efficient blue emitter with emission peak of 470 nm [21], while the exciplex formed between DMAC-DPS and PO-T2T conducted a green emission with peak at 536 nm [15]. So we consider if the blue shift happens with the overlap of pure DMAC-DPS emitter emission and DMAC-DPS: PO-T2T exciplex emission due to the decrease of PO-T2T ratio. To clarify the idea, we study the full width at half maximum (FWHM) characteristics of EL spectra. The FWHM of pure exciplex emission spectra under 1:9, 3:7 and 5:5 keeps the same of 102 nm, while the FWHMs of 100 nm under 7:3 ratio and 99 nm under 9:1 ratio are also obtained. Almost the same FWHM value of ~100 nm under the five different mixed ratios verifies the simple spectra blue shift rather than the overlap of DMAC-DPS emitter and exciplex emission, because the FWHM would be extended if the overlap occurs between two adjacent peak emissions. Besides, the shape of EL spectra also presents one emission peak without any shoulder or swelling, even at 470 nm, where is the intrinsic emission location of DMAC-DPS. So the EL spectra of DMAC-DPS: PO-T2T exciplex OLED exhibits pure exciplex emission with obvious blue shift property as the decrease of PO-T2T ratio from 5:5 to 7:3 and 9:1, which could exclude the influence of intrinsic emission of DMAC-DPS emitter. Besides, the EL spectra under different voltages present the same emission peak and curves without any change, which also prove the pure exciplex emission in our OLED.

Based on the discussions above, we try to reduce the PO-T2T ratio further. Therefore, two lower ratio exciplex OLED with PO-T2T concentrations of 5% and 1% were fabricated. The EL performance of spectra and efficiency are showed in Fig. 4. Interesting, the exciplex OLED with low concentrations PO-T2T also conducts the exciplex emission with one peak and smooth curves, which demonstrate the exciplex could be formed under extremely low acceptor ratio. From Fig. 4a of EL spectra, we also observe the continued blue shift with emission peak fixing at 512 nm and 496 nm under 5% and 1% doping concentration of PO-T2T, respectively. The peak of 496 nm even enters into blue emission peak zone. The FWHM is also studied to confirm the simple spectral move and the value is 99 nm with 5% PO-T2T and 100 nm with 1% PO-T2T, respectively. Hence, the exciplex of DMAC-DPS: PO-T2T in this work could be formed under high and low acceptor ratio of 1:9, 3:7, 5:5, 7:3, 9:1, 5% and 1%, while the continuously moved spectra toward to short wavelength direction is also realized. The emission peak could shift from 536 nm to 496 nm as the decrease of acceptor ratio from 50% to 1%.

The efficiencies and J-V-L curves are displayed in Fig. 4b. The maximum current efficiency, power efficiency and EQE are 32.9/25.7 cd/A, 38.3/24.4 l m/W and 11.2/10.4% with the PO-T2T concentrations of 5% and 1%. Although the obvious blue shift appears due to different PO-T2T concentration, the EQE still maintain a high efficiency level of above 10%. That means we can achieve a largely spectral move range of 40 nm without damaging the device efficiency level. The J-V-L curves embed in Fig. 4b also exhibit the excellent device performance of low turn-on voltage (<3 V) and high maximum luminance (>10^4 cd/m^2). The EL performance of all exciplex OLED in this work are listed in Table 1. The PL behavior with different donor/acceptor ratio also exhibited the similar characteristics with EL spectra [18], so we focus on the electrical-excitation situation to explore the effect on spectra and device efficiency.

Finally, we attempt to explore the spectral move phenomenon due to different PO-T2T acceptor ratio. Under the donor/acceptor ratio of 1:9, 3:7 and 5:5, the spectra keep almost the same, so acceptor molecule of PO-T2T could be regard as majority. Instead, the donor molecule of DMAC-DPS is the majority under the ratio of 7:3, 9:1, 5% and 1% and the schematic diagram is described in Fig. 5. As shown in Fig. 5a, the exciplex formation zone is fixed at the interface of mCP and exciplex

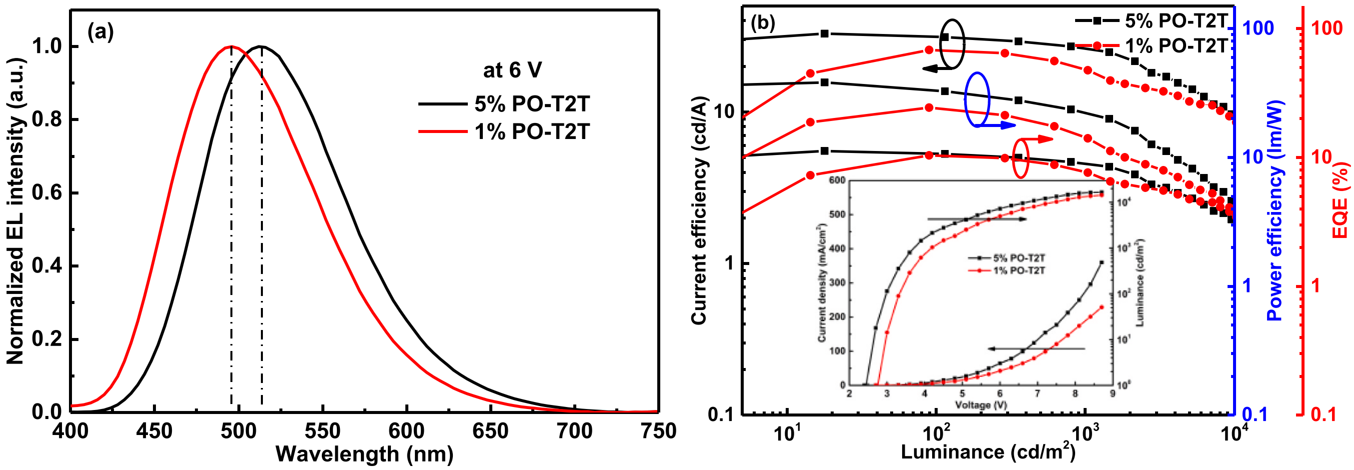

Fig. 4. The EL performance of exciplex OLED based DMAC-DPS: PO-T2T with lower PO-T2T concentrations of 5% and 1%. (a) Normalized EL spectra at 6 V. (b) Current efficiency-luminance-power efficiency/EQE curves. Inset is the J-V-L curves.

Table 1
The list of all exciplex OLED performance under different donor/acceptor ratio.

DMAC-DPS: PO-T2T	CE_{max} [a] (cd/A)	PE_{max} [b] (lm/W)	EQE_{max} [c] (%)	EQE_{1000} [d] (%)	Emission peak (nm)	FWHM (nm)
1:9	38.1	25.3	12.1	9.9	536	102
3:7	35.6	24.2	11.0	9.8	536	102
5:5	34.9	33.7	10.8	9.7	536	102
7:3	33.7	37.8	10.5	8.5	525	100
9:1	35.2	39.5	11.3	8.2	520	99
5% PO-T2T	32.9	38.3	11.2	9.0	512	99
1% PO-T2T	25.7	24.4	10.4	7.8	496	100

[a] Maximum current efficiency.
[b] Maximum power efficiency.
[c] Maximum EQE.
[d] EQE at 1000 cd/m^2.

EML due to the hole accumulation, which result from the poor hole transport ability and strong hole block ability of PO-T2T. Thus, the exciplex still maintain a high concentration with the ratio of 1:9 and 3:7 due to the narrow formation zone. However, in Fig. 5b, the exciplex formation zone would extend to whole exciplex EML under low ratio PO-T2T because of the excellent charge transport bipolarity of DMAC-DPS [22,23]. So the exciplex concentration would be diluted and the distance between exciplex would be increased as the ratio decrease of PO-T2T acceptor. Therefore, we consider the spectral move phenomenon could be explained as the effect of polarity of DMAC-DPS: PO-T2T exciplex, which could be regarded as the molecule with a separated charge inside the molecule. And the charge separation induces strong interaction between exciplex, which blue-shifts the emission wavelength according to the decrease of exciplex concentration [18,24]. Furthermore, the universality of spectral move in other exciplex system would be researched in the future.

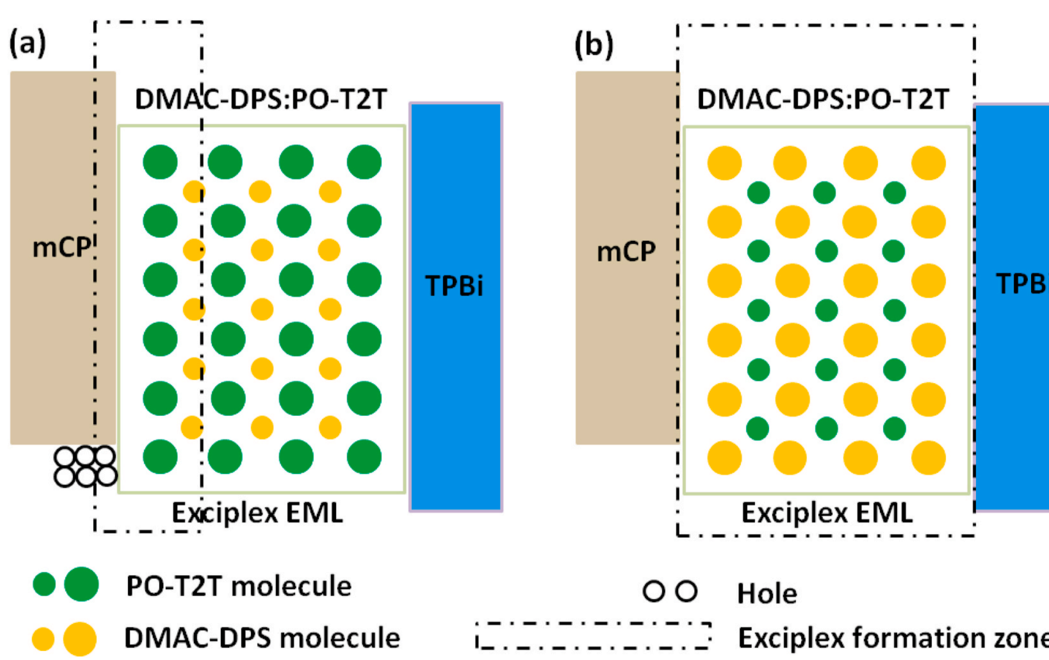

Fig. 5. The schematic diagram of different ratio donor/acceptor molecule in exciplex EML. (a) Acceptor molecule of PO-T2T is the majority. (b) Donor molecule of DMAC-DPS is the majority.

4. Conclusion

In conclusion, the exciplex OLED with a large donor/acceptor (DMAC-DPS: PO-T2T) ratio range of 1:9, 3:7, 5:5, 7:3, 9:1, 5% and 1% are fabricated, and the effect on device performance are studied further. As a result, the EL spectra keep almost the same with emission peak of 536 nm under a high PO-T2T acceptor ratio of 1:9, 3:7 and 5:5. While a large gradually spectral move of 40 nm to short wavelength direction are observed as the decrease of PO-T2T ratio, even shift to 496 nm at the PO-T2T concentration of 1%. Besides, the EL efficiencies are hardly any sacrificed under different donor/acceptor ratio with maximum current efficiency of 25.7–38.1 cd/A, power efficiency of 24.2–39.5 l m/W and EQE of 10.4–12.1%, respectively. We consider the effect of polarity of DMAC-DPS: PO-T2T exciplex is the key and charge separation induces strong interaction between exciplex lead to the spectral move. Therefore, we believe the donor/acceptor ratio, especially the small ratio similarly doping system, could play more important role in the development of exciplex OLED, which could modulate spectra without damaging device efficiency.

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:** took part in the discussions. All authors have reviewed the manuscript. **Hongmei Zhan:** took part in the discussions. All authors have reviewed the manuscript. **Yanxiang Cheng:** took part in the 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.

Acknowledgements

This work was financially supported by National Natural Science Foundation of China (NSFC) (61675089).

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