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# High luminance/efficiency monochrome and white organic light emitting diodes based pure exciplex emission

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ABSTRACT

In this work, high luminance and high efficiency monochrome and white organic light emitting diodes (OLED) based pure exciplex emission were realized by the application of spacer. First, monochrome exicplex OLED based blue (26DCzPPy:PO-T2T) and yellow (DMAC-MPM:PO-T2T) emission were studied. Then the pure exciplex WOLED based blue and yellow exciplex complementary colors were designed with three spacer materials of DMAC-MPM, 26DCzPPy and DPEPO, respectively. At last, orange exciplex of TAPC:PO-T2T was applied to replace DMAC-MPM:PO-T2T with 26DCzPPy as spacer to construct pure exciplex WOLED. As results, all the pure exciplex WOLED of W1∼W4 achieved a high maximum luminance of ∼10000 cd $m^{-2}$, current efficiency of ∼20 cd $A^{-1}$, power efficiency of ∼20 lm $W^{-1}$ and external quantum efficiency of ∼8%, respectively. As far as we know from literature review, our WOLED with the maximum luminance and efficiency were one of the best among the pure exciplex emission WOLED. The high performances were derived from the high efficiency monochrome exciplex and spacer selection and application.

## 1. Introduction

Organic light emitting diodes (OLED) received more and more attention due to its application in panel display and solid state lighting. From the view of materials, the emitters used in OLED could be divided three types in general: traditional fluorescent, phosphorescent and thermally activated delayed fluorescence (TADF) [1–3], which all are the monomolecular emissions. Among them, the phosphorescent and TADF emitter could achieve 100% internal quantum efficiency (IQE) due to the triplet exciton utilization [4,5]. Besides, one kind of bimolecular emission based charge transfer (CT) between donor and acceptor materials is also become more and more important, which is called exciplex emission [6,7].

The exciplex is formed from the intermolecular charge transfer excited state between highest occupied molecular orbital (HOMO) energy level of donor materials and lowest unoccupied molecular orbital (LUMO) energy level of acceptor materials [8,9], which exhibit intrinsic small $\Delta E_{ST}$ due to the spatially separated HOMO and LUMO in two different molecules of donor and acceptor [10], respectively. So, similar to the TADF emitter, exciplex could also achieve the triplet excitons harvest through reverse intersystem crossing (RISC) process [11–13] and improve the luminous efficiency, even 100% IQE. A series of highly efficient exciplex OLED based blue, green and yellow emission were also reported. And with the development of monochrome exciplex OLED, white OLED (WOLED) with pure exciplex emission also become an important issue. WOLED could be realized with three primary colors of blue, green and red, or two complementary colors of blue and yellow/orange [14–18]. Exciplex emission conducted a broad spectrum in

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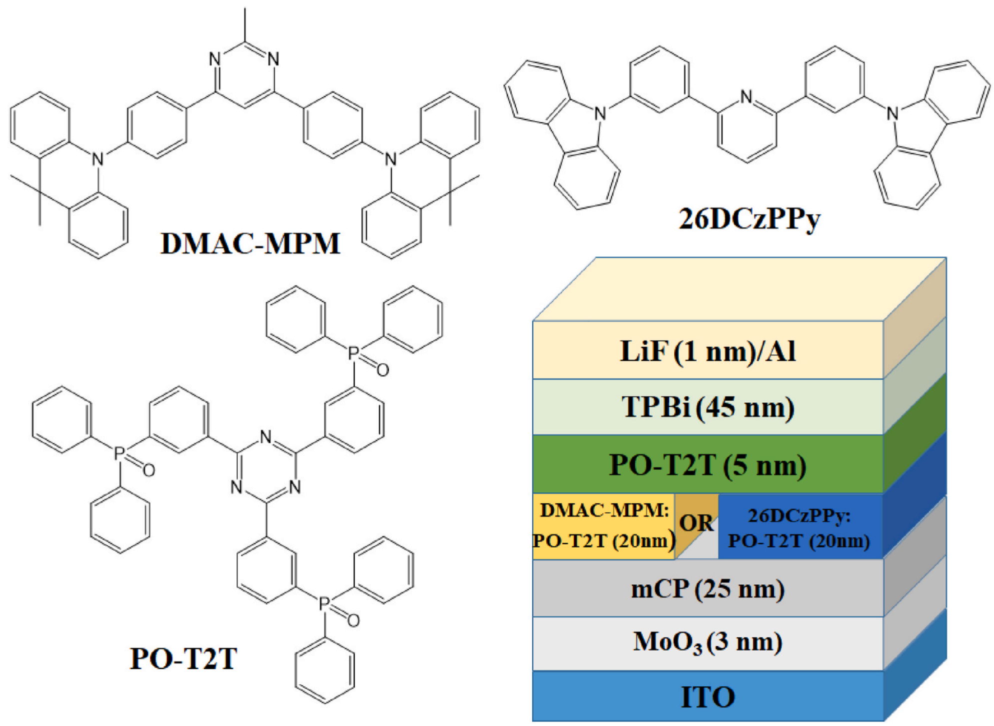


**Fig. 1.** The molecular structure of DMAC-MPM, 26DCzPPy, PO-T2T and device structure of monochrome exciplex OLED.

naturally, which could realize a high color quality white emission with two complementary colors easily [19–22]. And almost all the reported WOLED based pure exciplex emission adopted the two complementary colors. Hung et al. [23] and Zhao et al. [24] designed the high efficiency full exciplex WOLED with maximum external quantum efficiency (EQE) of 11.6% and 9.17%, respectively, but the complicated tandem structure was adopted. Guo et al. proposed the all exciplex WOLED by employing an interface-free sandwich light-emitting unit [25], while the maximum luminance of <3000 cd m$^{-2}$ and maximum device EQE of <5% need be improved further. Xu et al. fabricated the pure exciplex WOLED with a non-doped quantum well-like structure to obtain the maximum EQE of 6.72% [26], but the device present a low luminance of ~1000 cd m$^{-2}$. On the whole, the WOLED based pure exciplex emission conducted a low luminance and efficiency, and the performance need be enhanced

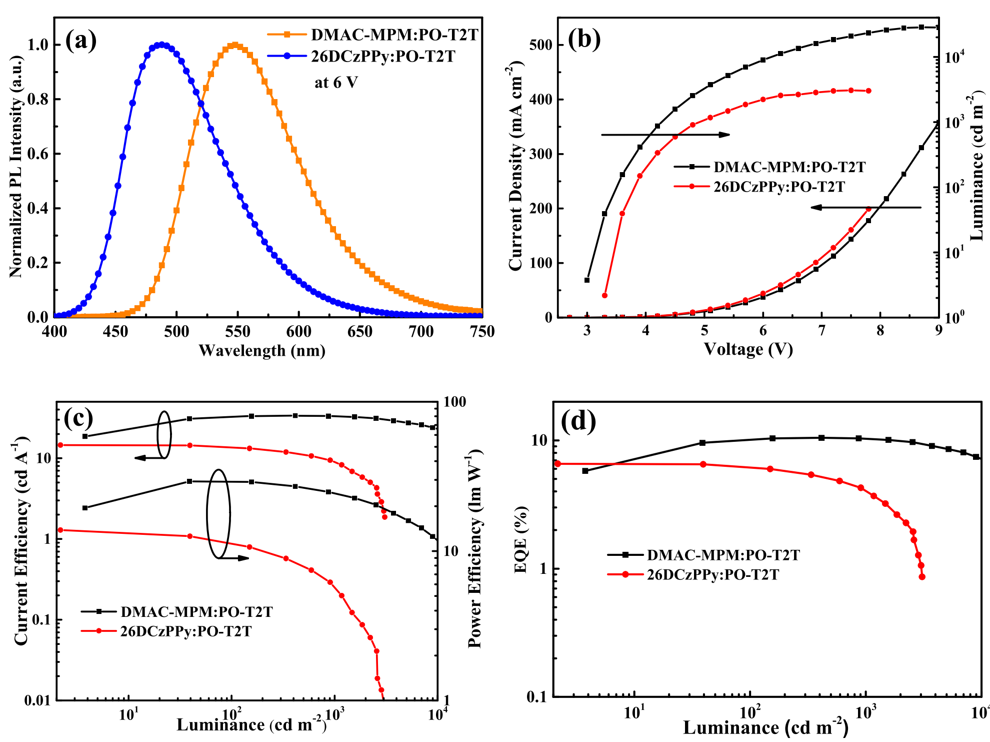


**Fig. 2.** The EL performances of monochrome exciplex OLED. (a) EL spectra. (b) Current density-voltage-luminance curves. (c) Current efficiency-luminance-power efficiency curves. (d) EQE-luminance curves.

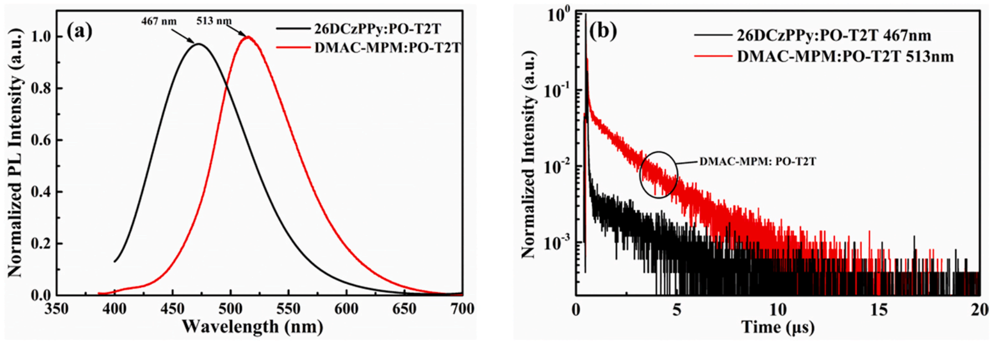


**Fig. 3.** The PL spectra and transient PL decay characteristics of the films of 26DCzPPy:PO-T2T and DMAC-MPM:PO-T2T. (a) Steady state PL spectra. (b) Transient PL decay curves. (Blue observed at 467 nm and yellow observed at 513 nm).

further.

In this work, high luminance and efficiency two complementary colors WOLED based pure exciplex emission were obtained by inserting different spacer layer materials into the middle of two colors. The blue exciplex is 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy):2,4,6-Tris[3-(diphenylphosphinyl)phenyl]-1,3,5-triazine (PO-T2T), 2-methyl-4, 6-bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]pyrimidine (DMAC-MPM):PO-T2T and 4,4′-cyclohexylidenebis[N,N-bis(p-tolyl)aniline] (TAPC):PO-T2T are the yellow and orange exciplex, respectively. The materials of DMAC-MPM, 26DCzPPy and bis[2-(diphenylphosphino) phenyl]ether oxide (DPEPO) are applied as the spacer, respectively. As results, a series of high luminance and efficiency WOLED were realized with maximum luminance of ~10000 cd $m^{-2}$, current efficiency (CE) of ~20 cd $A^{-1}$, power efficiency (PE) of ~20 lm $W^{-1}$ and EQE of ~8%, respectively. The intrinsic highly efficient monochrome exciplex emission and middle spacer layer modulation are responsible for the high performance pure exciplex WOLED.

## 2. Experimental section

All of the OLED were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω $sq^{-1}$. The ITO substrates were cleaned first with acetone, deionized water and acetone and then treated with ultraviolet-ozone for 15 min, then the ITO substrates were loaded into a high vacuum chamber (approximately $3 \times 10^{-4}$ Pa) for subsequent deposition. After finishing the deposition of the organic layers, an Al cathode was deposited in the end with a shadow mask, which defined the device area as $3 \times 3$ $mm^2$. Photoluminescence (PL) spectra were measured with a FluoroMax-4 fluorescence spectrometer (HORIBA Jobin Yvon). The transient PL decay profiles of the films were recorded using an Edinburgh Instrument FLS980 spectrometer equipped with an EPL-375 ps pulsed diode laser under nitrogen atmosphere. Electroluminescence (EL) spectra were measured through a PR-655 spectra scan spectrometer with computer control. The current-voltage-luminance curves were measured by a measuring system of a Keithley 2400 power supply combined with a BM-7A luminance colorimeter. The EQE was calculated from the current density-voltage-luminance curve and EL spectra data. All of the organic materials were procured commercially and used without further purification.

## 3. Results and discussions

### 3.1. OLED based monochrome exciplex

To construct the high performance WOLED based pure exciplex emission, the monochrome exciplex OLED performance need be studied firstly. According to the HOMO/LUMO energy level and singlet/triplet energy of donor/acceptor, novel DMAC-MPM and 26DCzPPy were selected as donor materials and PO-T2T was played the role of acceptor material, respectively [27–30]. The PL spectra of donor, acceptor and mixed film showed in Fig. S1 also confirm the formation of blue and yellow exciplex. The molecular structure of DMAC-MPM, 26DCzPPy, PO-T2T and device structure designed of monochrome exciplex OLED were showed in Fig. 1 m-bis(N-carbazolyl)benzene (mCP) and 1,3,5-Tris (1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi) are hole and electron transport layer, respectively, PO-T2T with 5 nm is the buffer layer. DMAC-MPM:PO-T2T and 26DCzPPy:PO-T2T are the monochrome exciplex emitting layers.

**Table 1**
Summary of lifetime parameters for yellow and blue exciplex.

| Exciplex | $\tau_p$ [a] (ns) | $\varphi_p$ [b] (%) | $\tau_d$ [c] (μs) | $\varphi_d$ [d] (%) |
|---|---|---|---|---|
| DMAC-MPM:PO-T2T | 63.2 | 6.9 | 1.85 | 93.1 |
| 26DCzPPy:PO-T2T | 36.3 | 11.2 | 2.48 | 88.8 |

[a] Prompt fluorescent lifetime.
[b] Prompt fluorescent proportion.
[c] Delayed fluorescent lifetime.
[d] Delayed fluorescent proportion.

The EL performance of spectra, current density-voltage-luminance and efficiencies were exhibited in Fig. 2. The exciplex of DMAC-MPM: PO-T2T presented a yellow emission with peak at 548 nm, while 26DCzPPy:PO-T2T showed a blue emission with peak at 488 nm. The interfacial exciplex of mCP/PO-T2T was hard to form due to the sufficient mixture of 26DCzPPy and PO-T2T in emitting layer (EML) with 1:1 ratio. So the WOLED could be achieved by constructing the two complementary colors with DMAC-MPM:PO-T2T and 26DCzPPy:PO-T2T. The high maximum luminance of 3000 cd $m^{-2}$ with blue exciplex and 28000 cd $m^{-2}$ with yellow exciplex indicated the high luminance pure exciplex WOLED could be also obtained. Besides, the yellow exciplex OLED exhibited a high efficiencies with maximum CE, PE and EQE of 33.7 cd $A^{-1}$, 29.3 lm $W^{-1}$ and 10.5%, respectively. The maximum CE, PE and EQE of 14.5 cd $A^{-1}$, 13.8 lm $W^{-1}$ and 6.6%, respectively, were earned in blue exciplex OLED. The high efficiency of monochrome exciplx OLED also guarantees the realization of highly efficient pure exciplex emission WOLED.

The steady state PL spectra and transient PL decay behavior were presented in Fig. 3 to explore the luminous mechanism of blue and yellow exciplex based 26DCzPPy:PO-T2T and DMAC-MPM:PO-T2T, respectively. Similar to the EL spectra, the PL spectra also exhibited one peak with pure exciplex emission. Furthermore, the transient PL decay curves of blue and yellow exciplex conducted two lifetimes with prompt lifetime and delayed lifetime, which presented a delayed fluorescent emission. The transient PL decay lifetime parameters were summarized in Table 1. Thus, the efficient collection and utilization to triplet excitons are responsible for the high efficiency of exciplex OLED [31].

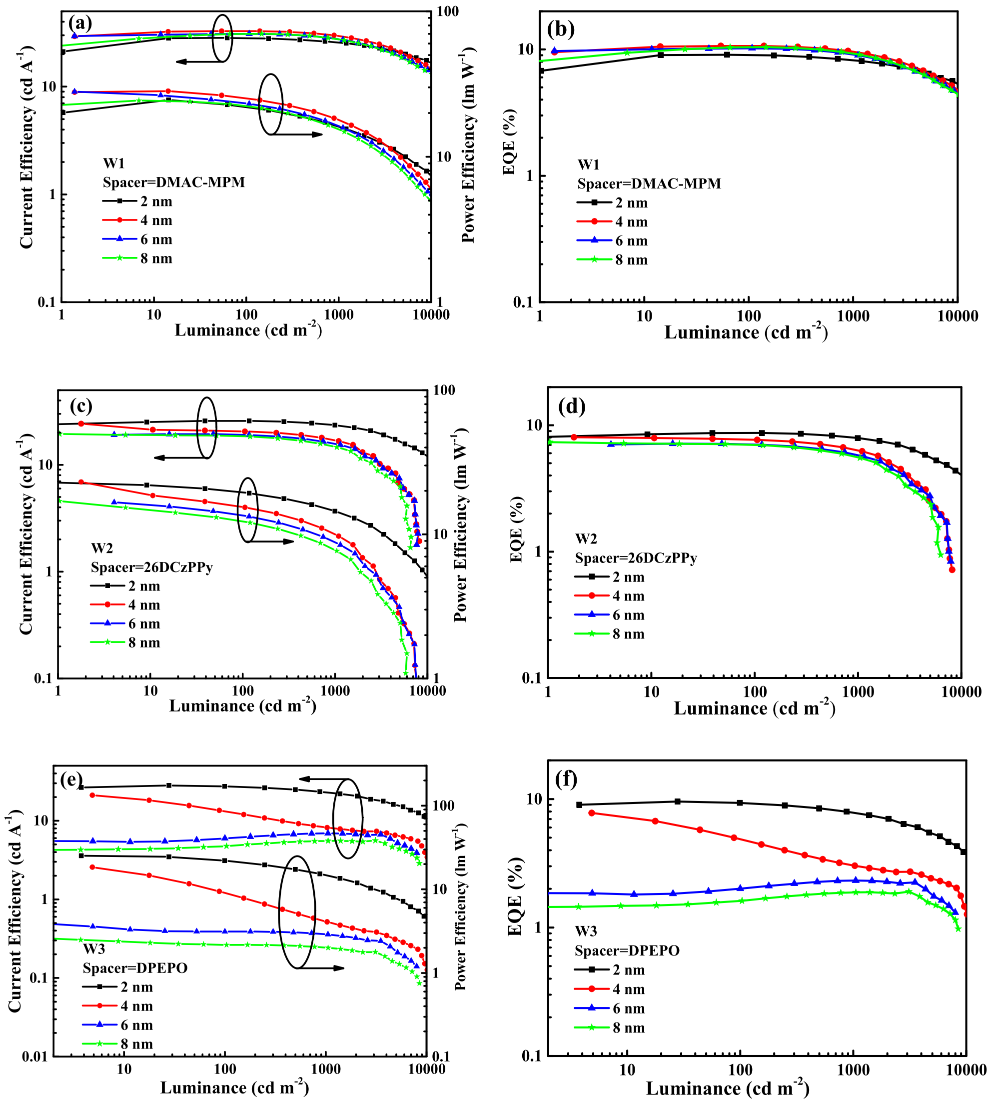


**Fig. 4.** The EL efficiencies of pure exciplex WOLED based DMAC-MPM (W1), 26DCzPPy (W2) and DPEPO (W3) as spacer, respectively. (a) Current efficiency-luminance-power efficiency curves of W1. (b) EQE-luminance curves of W1. (c) Current efficiency-luminance-power efficiency curves of W2. (d) EQE-luminance curves of W2. (e) Current efficiency-luminance-power efficiency curves of W3. (f) EQE-luminance curves of W3.

### 3.2. WOLED based complementary colors exciplex

In this section, pure exciplex WOLED based the blue and yellow exciplex mentioned above were constructed by the application of spacer. Here, three spacer materials of DMAC-MPM, 26DCzPPy and DPEPO were inserted into the middle of blue and yellow exciplex EML, respectively. The structure of pure exciplex WOLED designed as follows: ITO/$MoO_3$ (3 nm)/mCP (25 nm)/DMAC-MPM:PO-T2T (20 nm)/Spacer (x nm)/26DCzPPy:PO-T2T (20 nm)/PO-T2T (5 nm)/TPBi (45 nm)/LiF (1 nm)/Al (100 nm), the spacer thickness of x = 2, 4, 6 and 8, respectively. And the WOLED with DMAC-MPM, 26DCzPPy and DPEPO as the spacer materials were defined as W1, W2 and W3, respectively. As results, the efficiency curves of CE, PE and EQE of the pure exciplex WOLED were showed in Fig. 4.

Almost the same device performances were achieved in W1 with different thickness spacer of DMAC-MPM and the CEs, PEs and EQEs were 28.3–32.8 cd $A^{-1}$, 24.4–28.3 lm $W^{-1}$ and 9.1–10.7%, respectively. Meantime, a high maximum luminance of ~20000 cd $m^{-2}$ was also obtained in W1. But it's pity that the EL spectra of W1 exhibited a single peak under the different thickness spacer of 2, 4, 6 and 8 nm, which were showed in Fig. 5a. The single peak was marked as the emission of DMAC-MPM:PO-T2T exciplex, which means the exciton recombination zone was located at yellow exciplex EML regardless of the spacer thickness of DMAC-MPM. Actually, DMAC-MPM is a highly efficient blue emitter with TADF behavior, so the bipolar charge transport characteristics is owned, which would present well electron transport ability even at 8 nm thickness [24]. Therefore, the W1 with different thickness spacer still exhibited the main yellow exciplex of DMAC-MPM:PO-T2T, and the spectra also keep the same under different voltages, which were showed in Fig. S2. When 26DCzPPy with the same thickness of 2, 4, 6 and 8 nm as the spacer was inserted into W2, the blue exciplex emission appeared and an obvious blue emission could be found under 4–8 nm thickness in Fig. 5b. We consider the strong hole transport ability of 26DCzPPy block the sectional electron transport to yellow exciplex EML, which result to the blue exciplex emission of 26DCzPPy:PO-T2T [29,30]. Therefore, W2 showed a white emission with Commission Internationale de l'Eclairage (CIE) coordinate near (0.28, 0.44). Meantime, a high luminance above 6000 cd $m^{-2}$ was achieved in W2, and the maximum CEs, PEs and EQEs under the different thickness spacer of 26DCzPPy were 19.4–25.8 cd $A^{-1}$, 16.7–23.1 lm $W^{-1}$ and 7.1–8.7%, respectively. However, the thin thickness of 2 nm can't block electron and exciton into yellow exciplex EML efficiently, which result to the weak blue exciplex emission. The

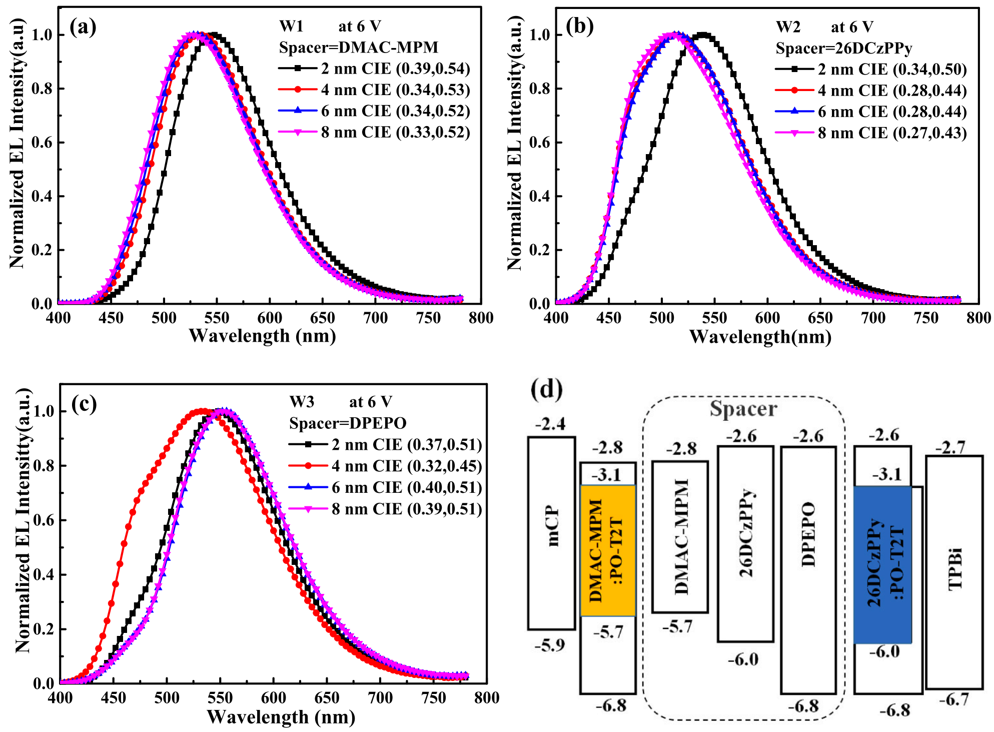


**Fig. 5.** The energy level diagram and EL spectra of pure exciplex WOLED based DMAC-MPM, 26DCzPPy and DPEPO as spacer with different thickness, respectively. (a) EL spectra of W1. (b) EL spectra of W2. (c) EL spectra of W3. (d) Device energy level diagram.

reduced relative intensity of blue exciplex with increased voltage shown in Fig. S3 means that the recombination zone moved to yellow exciplex EML with more charge injection. Another spacer material of DPEPO with the same different thickness was adopted to construct WOLED in W3. Distinguish from the similar device efficiencies in W1 and W2, W3 showed in Fig. 4e–f conducted the large efficiency difference under the spacer thickness of 2, 4, 6 and 8 nm with the maximum CE/PE/EQE of 28.1 cd $A^{-1}$/25.2 lm $W^{-1}$/9.6%, 21.1 cd $A^{-1}$/18.4 lm $W^{-1}$/7.8%, 6.9 cd $A^{-1}$/3.9 lm $W^{-1}$/2.3% and 5.6 cd $A^{-1}$/2.6 lm $W^{-1}$/1.9%, respectively. Combining the EL spectra shown in Fig. 5c, the W3 under 4 nm spacer realized the best performance with maximum EQE of 7.8% and CIE coordinate of (0.32, 0.45) at 6 V. And the spectra showed different change rule with increased spacer thickness compared to W1 and W2, the blue emission was enhanced first and then weakened as the increased thickness in W3. As shown in Fig. 5d of the energy level diagram, DPEPO possessed a wide energy gap with HOMO of 6.8 eV and LUMO of 2.6 eV[32], and a low hole/electron mobility of $1.4 \times 10^{-9}$/$7.0 \times 10^{-8}$ $cm^2 V^{-1} s^{-1}$. Therefore, under a thin DPEPO thickness, the hole could reach to blue exciplex EML through tunneling effect [29] to achieve a blue emission, however, the hole would be blocked completely when the thickness enhanced to 4 and 8 nm due to the too large energy barrier of 1.1 eV ($HOMO_{DPEPO}$:6.8 eV-$HOMO_{DMAC\text{-}MPM}$:5.7 eV) [30,32], which would prevent the hole transport on HOMO energy level. So under a thick DPEPO thickness, the blue exciplex emission decreased sharply, also along with the device maximum luminance and efficiency due to the weakened exciton recombination [33]. The blue exciplex emission relative intensity shown in Fig. S4 also decreased with improved voltage, which results from the injection of electron into yellow exciplex EML to form more excitons with strong yellow exciplex emission.

The current density-voltage-luminance curves of W1∼W3 shown in Fig. S5 demonstrated the thicker spacer would improve the operation voltage and reduce current density. Therefore, the thickness of spacer played an important role on the modulation of device EL performance. As a whole of W1∼W3, W2 with the spacer of 26DCzPPy achieved a high EQE of 7.1–8.7% and relatively stable white spectra under different spacer thickness. In order to obtain a higher color quality WOLED, red shift orange exciplex of TAPC:PO-T2T [34] was applied to replace

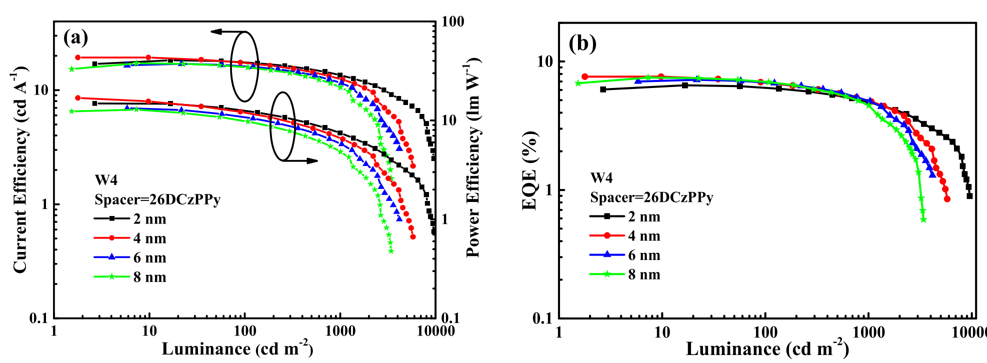


**Fig. 6.** The EL efficiencies of pure exciplex WOLED based TAPC:PO-T2T as orange exciplex, 26DCzPPy as spacer and 26DCzPPy:PO-T2T as blue exciplex (W4). (a) Current efficiency-luminance-power efficiency curves. (b) EQE-luminance curves.

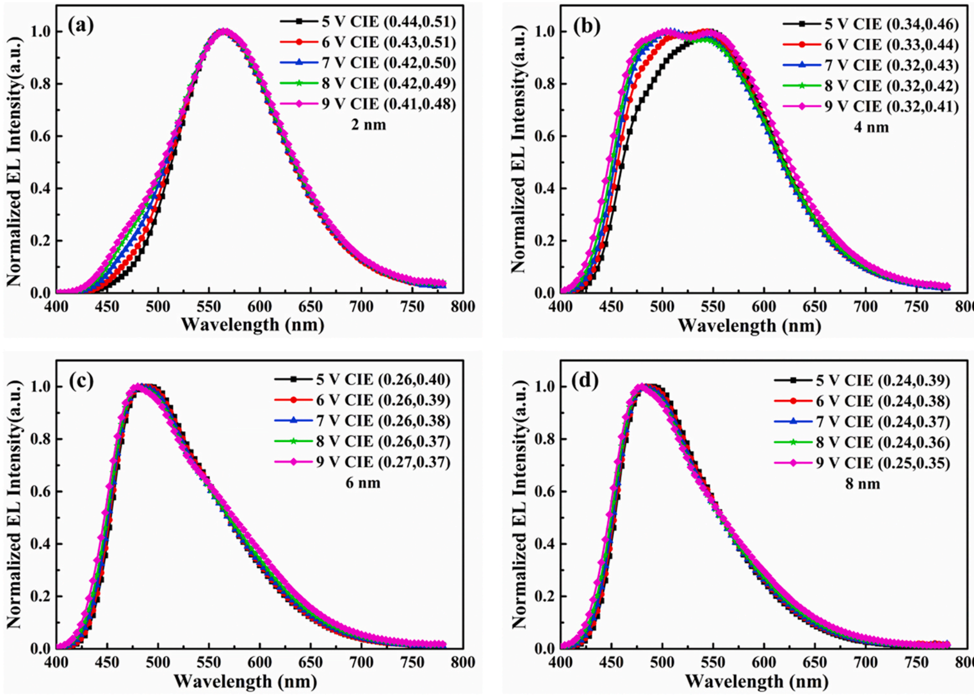


**Fig. 7.** The EL spectra of pure exciplex WOLED based TAPC:PO-T2T as orange exciplex and 26DCzPPy as spacer with different thickness (W4). (a) 2 nm. (b) 4 nm. (c) 6 nm. (d) 8 nm.

**Table 2**
The performances summary of all the OLED used in this work.

| Device structure | S[a] (nm) | $V_{on}$[b] (V) | $L_{max}$[c] (cd $m^{-2}$) | $CE_{max}$[d] (cd $A^{-1}$) | $PE_{max}$[e] (lm $W^{-1}$) | $EQE_{max}$[f] (%) | CIE (at 6 V) |
|---|---|---|---|---|---|---|---|
| O (TAPC:PO-T2T) | ~ | 3.0 | 11000 | 11.8 | 11.8 | 5.1% | (0.49, 0.48) |
| Y (DMAC-MPM:PO-T2T) | ~ | 3.0 | 28610 | 33.7 | 29.3 | 10.5 | (0.39,0.56) |
| B (26DCzPPy:PO-T2T) | ~ | 3.3 | 3070 | 14.5 | 13.8 | 6.6 | (0.20,0.36) |
| W1 (Y:DMAC-MPM:B) | 2 | 3.3 | 24470 | 28.3 | 24.6 | 9.1 | (0.39,0.54) |
| | 4 | 3.3 | 20000 | 32.8 | 28.3 | 10.7 | (0.34,0.53) |
| | 6 | 3.3 | 20160 | 31.0 | 28.0 | 10.2 | (0.34,0.52) |
| | 8 | 3.6 | 17890 | 30.9 | 24.4 | 10.4 | (0.33,0.52) |
| W2 (Y:26DCzPPy:B) | 2 | 3.3 | 17850 | 25.8 | 22.8 | 8.7 | (0.34,0.50) |
| | 4 | 3.3 | 8153 | 24.3 | 23.1 | 7.9 | (0.28,0.44) |
| | 6 | 3.6 | 7903 | 19.4 | 16.7 | 7.1 | (0.28,0.44) |
| | 8 | 3.6 | 6650 | 19.5 | 17.0 | 7.4 | (0.27,0.43) |
| W3 (Y:DPEPO:B) | 2 | 3.3 | 17470 | 28.1 | 25.2 | 9.6 | (0.37,0.51) |
| | 4 | 3.6 | 10180 | 21.1 | 18.4 | 7.8 | (0.32,0.45) |
| | 6 | 4.5 | 7968 | 6.9 | 3.9 | 2.3 | (0.40,0.51) |
| | 8 | 5.1 | 8480 | 5.6 | 2.6 | 1.9 | (0.39,0.51) |
| W4 (O:26DCzPPy:B) | 2 | 3.6 | 9555 | 18.3 | 14.8 | 6.5 | (0.43,0.51) |
| | 4 | 3.6 | 5788 | 19.3 | 16.9 | 7.6 | (0.33,0.44) |
| | 6 | 3.9 | 4166 | 17.0 | 13.3 | 7.2 | (0.26,0.39) |
| | 8 | 3.9 | 3413 | 17.2 | 12.9 | 7.5 | (0.24,0.38) |

[a] Spacer thickness.
[b] Turn-on voltage.
[c] Maximum luminance.
[d] Maximum CE.
[e] Maximum PE.
[f] Maximum EQE.

DMAC-MPM:PO-T2T to design the WOLED and the spacer of 26DCzPPy was adopted simultaneously, which was defined as W4. So the structure of W4 as follows: ITO/$MoO_3$ (3 nm)/mCP (25 nm)/TAPC:PO-T2T (20 nm)/26DCzPPy (x nm)/26DCzPPy:PO-T2T (20 nm)/PO-T2T (5 nm)/TPBi (45 nm)/LiF (1 nm)/Al (100 nm), x = 2, 4, 6 and 8, respectively. Similar to W2, almost the same device efficiencies shown in Fig. 6 were realized under different spacer thickness with maximum CEs, PEs and EQEs of 17.0–19.3 cd $A^{-1}$, 12.9–16.9 lm $W^{-1}$ and 6.5–7.6%, respectively. Among them, the WOLED with spacer thickness of 4 nm achieved the maximum CE, PE and EQE of 19.3 cd $A^{-1}$, 16.9 lm $W^{-1}$ and 7.6%, respectively, which means the optimal spacer thickness was 4 nm.

The EL spectra of W4 with different spacer thickness were displayed in Fig. 7. Under a thin thickness of 2 nm, the main emission peak was orange exciplex and the main emission peak changed to blue exciplex as the thickness increased to 6 and 8 nm. While at the optimal thickness of 4 nm, obvious two emission peaks were obtained and the CIE coordinate fixed at (0.32, 0.43) nearby under various operation voltages of 6–9 V, which was a warm white emission and obtained a higher color quality

WOLED compared to W1~W3. All the OLED performances used in this work were listed in Table 2. Therefore, all the WOLED in this work achieved a high maximum luminance of ~10000 cd $m^{-2}$, CE of ~20 cd $A^{-1}$, PE of ~20 lm $W^{-1}$ and EQE of ~8%, respectively. As far as we know from literature review, our WOLED with the maximum luminance and efficiency were one of the best among the pure exciplex emission WOLED. The high efficiency monochrome exciplex and spacer selection and application were responsible for the high performance pure exciplex WOLED [24]. Although our WOLED based pure exciplex presented a high luminance and efficiency, however, the color rendering index (CRI) was low about ~70. Speak frankly, high CRI (>80) pure exciplex WOLED based complementary color could be realized, which was benefited from the large full width at half maximum (FWHM) of exciplex emission. We would also try to seek or synthesis more suitable donor/acceptor to exploit higher efficiency orang/red exciplex to design high luminance/efficiency/CRI pure exciplex WOLED in the future work.

## 4. Conclusions

In conclusion, highly efficient monochrome and white OLED based pure exciplex were realized by employing complementary color monochrome exciplexes of yellow/orange (DMAC-MPM:PO-T2T/TAPC:PO-T2T) and blue (26DCzPPy:PO-T2T) emission through the application of spacer (DMAC-MPM, 26DCzPPy and DPEPO). As results, all the WOLED achieved a high maximum luminance of ~10000 cd $m^{-2}$, CE of ~20 cd $A^{-1}$, PE of ~20 lm $W^{-1}$ and EQE of ~8%, respectively. The selection and application of high performances monochrome exciplex and spacer play the key role for highly efficient pure exciplex WOLED. Herein, we explored the application of spacer with different thickness to construct high efficiency WOLED based pure exciplex emission. And we believe higher performances pure exciplex WOLED with high luminance (>10000 cd $m^{-2}$)/EQE (>15%)/CRI (>85) would be exploited by obtaining more efficiently orange/red exciplex OLED.

## 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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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.orgel.2022.106528.

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