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High color stability and CRI (> 80) fluorescent white organic light-emitting diode based pure emission of exciplexes by employing merely complementary colors†

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High color stability and CRI (> 80) pure exciplex WOLEDs with merely complementary colors of orange- and blue-exciplexes are realized with the application of spacers. The WOLEDs with a three spacer structure achieve a maximum current efficiency, power efficiency and external quantum efficiency (EQE) of 16.2 cd A⁻¹, 11.3 lm W⁻¹ and 7.92%, respectively. Besides, a standard white light point close to Commission Internationale de l'Eclairage (CIE) coordinates of (0.31 ± 0.00, 0.37 ± 0.02) and a very high color rendering index (CRI) of ~83 with two colors emitting are obtained simultaneously. The balanced emission and natural broad emission band of the exciplex are responsible for the stable white light spectra and high CRI. We also find that the location of the spacers, and the amount of them, play a key role in the electroluminescence performance of the WOLEDs and more detailed discussions are given below.

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1. Introduction

White organic light-emitting diodes (WOLEDs) have received much attention due to their great application in solid-state lighting, as they are a natural flat-panel light source and could be rolled onto a flexible substrate. In general, WOLEDs are constructed with a tricolor of red, green and blue or with complementary colors of orange and blue. The tricolor WOLEDs can easily achieve pure white light (~0.33, 0.33) and a high color rendering index (CRI) (> 80), but the complicated multicolor emission (three colors) and low spectral stability are a big challenge.^{1–3} On the other hand, the complementary color WOLEDs have a relatively simple emission (two colors) and stable spectra, however their poor white light purity and low CRI (< 80) limit their development.^{4–6}

For emitting materials based on red, orange, green and blue, the fluorescent, phosphorescent and thermally activated delayed fluorescent (TADF) materials are still the main materials that are applied to WOLEDs.^{7–9} However, there is another kind of emitting mode that can in fact form red, orange, green and blue, which is an exciplex. An exciplex does not emit from one fixed material, but forms from the interfacial charge transfer (CT) between the highest occupied molecular orbital (HOMO) of a donor and the lowest unoccupied molecular orbital (LUMO) of an acceptor.^{10,11} An exciplex gives a new emission peak that appears at a long wavelength compared to the intrinsic donor and acceptor. In recent years, a series of high efficiency, various color exciplexes were exploited and developed rapidly with the rise of the TADF mechanism.^{12–17} More importantly, an exciplex has a very broad emission band, which may be due to the broad energy level distributions of singlet and triplet exciplexes.¹⁸ The broad emission bands of exciplexes provide the possibility of a high CRI in complementary color WOLEDs, which is always low (< 80) in WOLEDs with complementary color fluorescent and phosphorescent materials. The Li group has reported many high CRI WOLED based exciplexes, but the efficiency can't satisfy commercial requirements.^{19,20} Hung *et al.* designed the first high efficiency, tandem full exciplex WOLEDs with a maximum external quantum efficiency (EQE) of 11.6%, but the complicated structure of the tandem and relatively low CRI of 70.6 also aren't acceptable.²¹ Therefore, high efficiency WOLEDs with a high CRI using the simple complementary color model need be studied and developed further.

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In this manuscript, fluorescent WOLEDs with a high color stability of $\pm(0.00, 0.02)$ and a high CRI of 83–84 from 6 V to 9 V were achieved by employing just two complementary colors, orange and blue. The high CRI of 83–84 is one of the highest values in WOLEDs with the complementary color model. A moderate efficiency level was also obtained with a maximum current efficiency (CE), power efficiency (PE) and EQE of 16.2 cd A^{-1} , 11.3 lm W^{-1} and 7.92%, respectively. A standard white light point near to the Commission Internationale de l'Eclairage (CIE) coordinates of (0.32, 0.37) at 9 V was realized simultaneously.

2. Experimental section

All of the OLEDs were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance of $10 \Omega \text{ sq}^{-1}$. 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 $3 \times 10^{-4} \text{ Pa}$) 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 $3 \times 3 \text{ mm}^2$. Photoluminescence (PL) spectra were measured with a FluoroMax-4 fluorescence spectrometer (HORIBA Jobin Yvon). 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. All of the measurements were carried out at room temperature and under ambient conditions without any protective coatings.

3. Results and discussion

PO-T2T, a phosphine oxide based material, is an excellent acceptor material with a high electron mobility ($1.7\text{--}4.4 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)

and high triplet energy ($E_T = 2.99 \text{ eV}$).²¹ To achieve highly efficient pure exciplex WOLEDs, we selected di-[4-(*N,N*-ditolylamino)-phenyl]cyclohexane (TAPC) and *m*-bis(*N*-carbazolyl)-benzene (*m*CP) as the donors, together with PO-T2T as the acceptor, to form the orange- and blue-excplexes, respectively. Fig. 1 shows the molecular structures and the PL spectra of the donor, the acceptor and the *m*CP:PO-T2T and TAPC:PO-T2T mixed films (1:1). From the PL spectra of each mixed film, we only observe one emission peak, which is fixed at 468 nm and 580 nm, respectively. The emission peaks are not the intrinsic peaks of *m*CP, TAPC and PO-T2T, but a new peak and the peak value energy is similar to the energy gap between $\text{HOMO}_{\text{donor}}$ and $\text{LUMO}_{\text{acceptor}}$. This demonstrates that the formation of the exciplex is between the donor and acceptor, in which *m*CP:PO-T2T is the blue-excplex and TAPC:PO-T2T is the orange-excplex. The spectral full width at half maximum (FWHM) of the orange- and blue-excplexes reaches 75 nm and 125 nm, respectively. For comparison, we report the FWHM values of typical blue, green, orange and red phosphorescent Ir-complex materials (FIrpic: 63 nm; Ir(ppy)₃: 82 nm; Ir(bt)₂acac: 85 nm; Ir(pq)₂acac: 88 nm) here. It is obvious that the FWHM of the exciplex is much larger than that of the Ir-complex and the value of the orange-excplex even reaches 125 nm. Such broad emission bands of exciplexes have a tremendous potential to realize a high CRI, and before constructing the WOLEDs with the exciplexes, the individual orange- and blue exciplex EL performances were evaluated. The orange exciplex device of TAPC:PO-T2T achieved moderate performances with a maximum CE, PE and EQE of 11.8 cd A^{-1} , 11.8 lm W^{-1} and 5.1%, respectively, which is shown in Fig. S1 (ESI†). The blue exciplex device of *m*CP:PO-T2T exhibited high EL performances with a maximum CE, PE and EQE of 15.5 cd A^{-1} , 18.4 lm W^{-1} and 8.0%, respectively, which had been reported by Hung *et al.*²¹ So we considered high CRI and high efficiency WOLEDs based on the two kinds of exciplex that could be realized and we also fabricated a series of complementary color devices to achieve the pure exciplex WOLEDs with a high CRI.

Firstly, we design the simple bilayer structure as follows: ITO/MoO₃ (3 nm)/TAPC (25 nm)/orange-excplex (15 nm)/blue-excplex

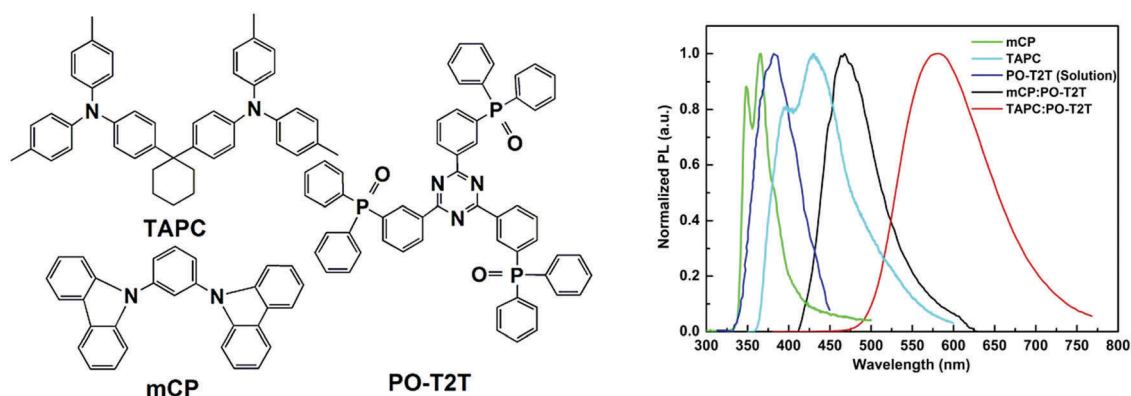


Fig. 1 The molecular structure of donor/acceptor materials and the PL spectra of *m*CP, TAPC, PO-T2T (solution) and *m*CP:PO-T2T and TAPC:PO-T2T mixed films.

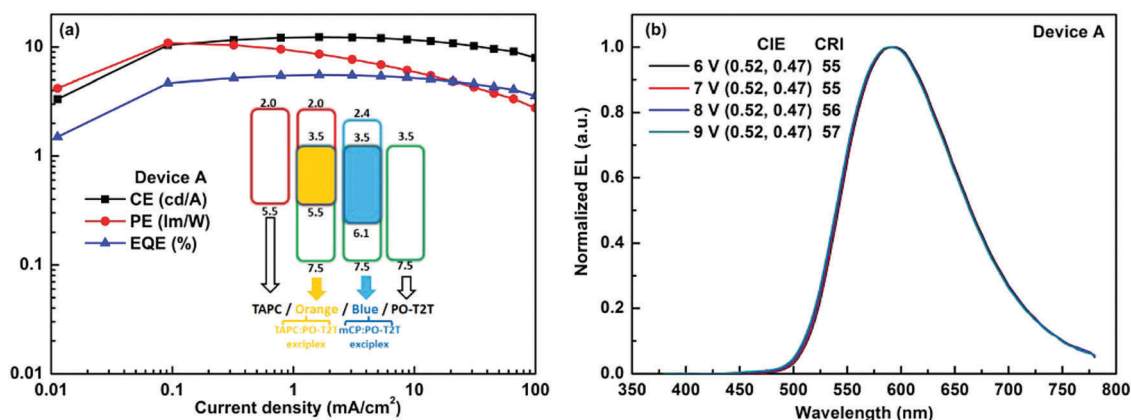


Fig. 2 The EL performance of Device A. (a) CE, PE and EQE–current density curves. Inset: The energy level schematic diagram of the device. The numbers indicate the LUMO and HOMO relative to a vacuum (units in eV). (b) EL spectra with CIE and the CRI under different voltages.

(15 nm)/PO-T2T (40 nm)/LiF (1 nm)/Al, which is defined as Device A. The orange- and blue-excimer emitting layer (EML) of 15 nm is referred to as TAPC:PO-T2T and mCP:PO-T2T mixed film, respectively, and the mixed ratio of the donor/acceptor of the exciplexes is 1 : 1. The pure TAPC and PO-T2T film is acting as the hole transport layer (HTL) and electron transport layer (ETL), respectively. MoO₃ and LiF are the modified layers of the electrodes. Fig. 2 shows the EL performance of Device A. The device exhibits an efficiency with a maximum CE, PE and EQE of 12.3 cd A⁻¹, 10.9 lm W⁻¹ and 5.53%, respectively. But, to our disappointment, the device displays only pure emission of an orange-excimer fixed at ~590 nm and the blue-excimer is not observed in the EL spectra. We believe that the completed energy transfer happens from the blue-excimer to the orange-excimer due to the two adjacent EMLs. Therefore, the high energy excitons of the blue-excimer are quenched by the low energy orange-excimer, which results in pure emission of the orange-excimer.

In order to suppress the energy transfer process from the blue-excimer to the orange-excimer, we insert a thin spacer of bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO) with an optimal thickness of 3 nm at the interface. The device structure is as follows: ITO/MoO₃ (3 nm)/TAPC (25 nm)/orange-excimer (15 nm)/DPEPO (3 nm)/blue-excimer (15 nm)/PO-T2T (40 nm)/LiF (1 nm)/Al, which is defined as Device B. The thin spacer of DPEPO between the orange-excimer and the blue-excimer acts as an exciton block layer (EBL) because of the ultra-high triplet energy ($E_T = 2.98$ eV).²² Fig. 3 shows the EL performance of Device B. As is expected, the device successfully achieves a white light emission and the maximum CE, PE and EQE are 9.3 cd A⁻¹, 8.9 lm W⁻¹ and 4.50%, respectively. The energy transfer is suppressed efficiently and a blue-excimer emission peak fixed at ~470 nm appears due to the introduction of DPEPO. However, the white light spectra change greatly and the intensity of the blue-excimer reduces, while the orange-excimer enhances with the rise in voltage. This demonstrates the exciton recombination zone shift to the orange-excimer at high voltage. DPEPO is an electron transport material with hole and electron mobilities of 1.4×10^{-9} and 7.0×10^{-8} cm² V⁻¹ s⁻¹,

respectively.²² Under the electrical excitation, the electrons and holes would overcome the barrier and go across the spacer to the orange-excimer and blue-excimer EML, respectively. Although the energy barriers for electrons and holes are large (0.9 eV for electrons; 1.3 eV for holes), the thin spacer of DPEPO (3 nm) could also guarantee the penetration of the electrons and holes. With the increase of voltage, the electrons reach the orange-excimer easier than the holes with a relatively small energy level barrier between the blue-excimer and DPEPO (0.9 eV). However, the holes would accumulate partly at the interface of the orange-excimer and DPEPO because of the large barrier of 1.3 eV. Thereby, more and more charge would recombine at the orange-excimer EML as the entrance for the electrons goes across the spacer, which results in the stronger orange and weaker blue exciplexes with the enhancement of voltages.

Next we insert another thin spacer of DPEPO into the interface of the HTL and the orange-excimer based on Device B. The device structure is as follows: ITO/MoO₃ (3 nm)/TAPC (25 nm)/DPEPO (3 nm)/orange-excimer (15 nm)/DPEPO (3 nm)/blue-excimer (15 nm)/PO-T2T (40 nm)/LiF (1 nm)/Al, which is defined as Device C. Fig. 4 shows the EL performance of Device C. Device C exhibits a similar efficiency level and white light spectra to Device B, and the maximum CE, PE and EQE are 8.4 cd A⁻¹, 7.6 lm W⁻¹ and 3.90%, respectively. But the white light spectra show more stability than those of Device B and the blue-excimer intensity change is small with the rise in voltage. We attribute the stable spectra to the efficient blocking of hole injection from the HTL to the orange-excimer EML (a large hole barrier of 1.3 eV at the TAPC/DPEPO interface). The vast holes are retained by the EML, which contributes to stabilizing the exciton recombination zone and to balancing the charge recombination, but also would damage the emitting efficiency in turn. The relatively low EL efficiency and maximum luminance (~1300 cd m⁻²) of Device C also confirm the viewpoint discussed above.²³

As mentioned above, the introduction of thin spacers at the interface of the HTL and orange-excimer could stabilize the white light spectra due to the efficient blocking of large amounts of hole injection from the HTL to the EML. Then, we consider

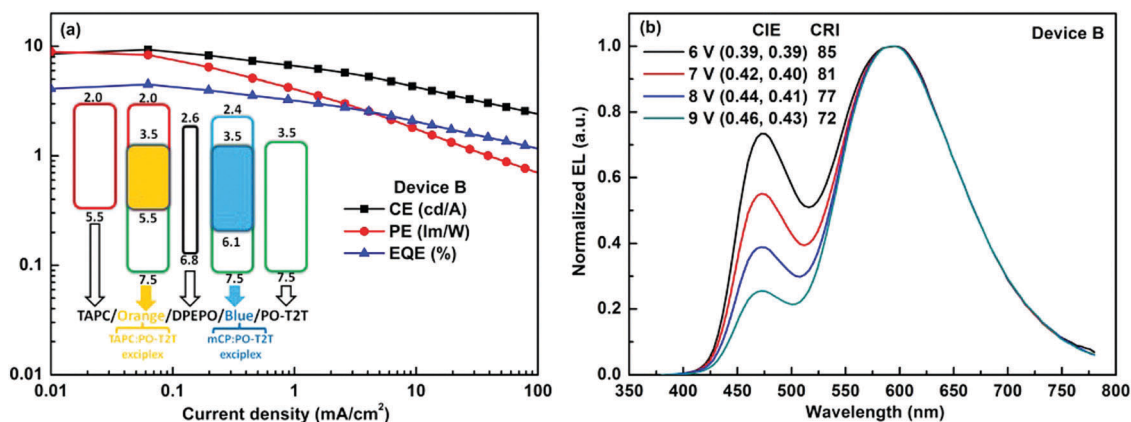


Fig. 3 The EL performance of Device B. (a) CE, PE and EQE–current density curves. Inset: The energy level schematic diagram of the device. The numbers indicate the LUMO and HOMO relative to a vacuum (units in eV). (b) EL spectra with CIE and the CRI under different voltages.

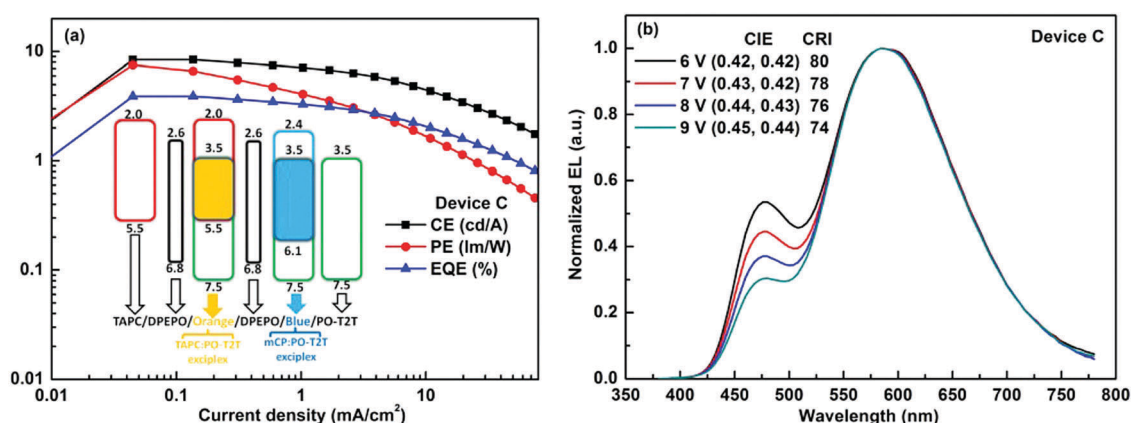


Fig. 4 The EL performance of Device C. (a) CE, PE and EQE–current density curves. Inset: The energy level schematic diagram of the device. The numbers indicate the LUMO and HOMO relative to a vacuum (units in eV). (b) EL spectra with CIE and the CRI under different voltages.

another device structure in which the spacer is inserted into the interface of the blue-excimer and ETL based Device B. So the new structure is as follows: ITO/MoO₃ (3 nm)/TAPC (25 nm)/orange-excimer (15 nm)/DPEPO (3 nm)/blue-excimer (15 nm)/DPEPO (3 nm)/PO-T2T (40 nm)/LiF (1 nm)/Al, which is defined

as Device D. Fig. 5 shows the EL performance of Device D. The device achieves a high efficiency with a maximum CE, PE and EQE of 16.8 cd A⁻¹, 13.2 lm W⁻¹ and 8.59%, respectively. Besides, almost the same white light spectra under different voltages are realized simultaneously, with stable CIE coordinates

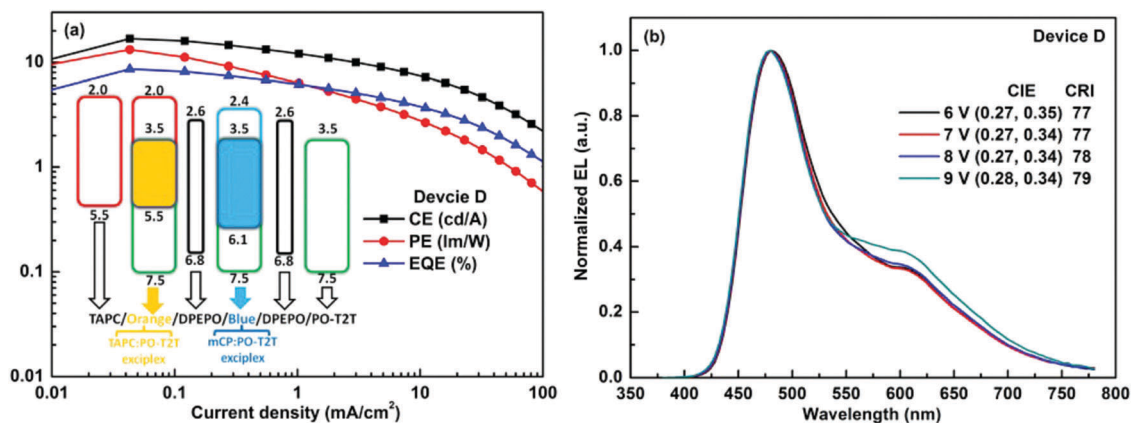


Fig. 5 The EL performance of Device D. (a) CE, PE and EQE–current density curves. Inset: The energy level schematic diagram of the device. The numbers indicate the LUMO and HOMO relative to a vacuum (units in eV). (b) EL spectra with CIE and the CRI under different voltages.

of $(0.27 \pm 0.01, 0.34 \pm 0.01)$ and a CRI of 78 ± 1 from 6 V to 9 V. We can see that the shape of the EL spectra is very different from that of Device C, in which the intensity of the blue-excimer is higher than that of the orange-excimer. The insertion of DPEPO between PO-T2T and the blue-excimer EML based Device B further decreases the entrance of electrons to the orange-excimer EML due to the large energy level barrier of 0.9 eV, which results in the enormous reduction of the orange-excimer. In contrast, the decelerated electron injection and transport due to the addition of spacers also balances the electron and hole transport and their recombination. Meanwhile, the high triplet energy of the spacers also efficiently confines the blue and orange excitons in each EML, which improves the device efficiency.²⁴ Therefore the balanced transport, recombination and efficient confinement are responsible for the high efficiency and stable spectra.

Despite Device D achieving a considerable EL performance with high efficiency and stable white light spectra, the CRI of 78 ± 1 can't satisfy practical application. Therefore, we design the final devices based on the discussion above and the structure as follows: ITO/MoO₃ (3 nm)/TAPC (25 nm)/DPEPO (3 nm)/orange-excimer (15 nm)/DPEPO (3 nm)/blue-excimer (15 nm)/DPEPO (3 nm)/PO-T2T (40 nm)/LiF (1 nm)/Al, which is defined as Device E. A thin spacer of DPEPO is also inserted into the interface of the HTL and the orange-excimer based Device D. That is, three thin spacers of DPEPO exist in Device E, at the interfaces of HTL/orange-excimer, orange-excimer/blue-excimer and blue-excimer/ETL. The orange- and blue-excimer EMLs are confined by DPEPO spacers in Device E. To our surprise, Device E exhibits an excellent EL performance, which is shown in Fig. 6. The maximum CE, PE and EQE are 16.2 cd A^{-1} , 11.3 lm W^{-1} and 7.92%, respectively. At the same time, the standard white light point near to the CIE coordinates of (0.31, 0.37) and the high CRI of ~ 83 , along with the high spectral stability under different voltages, are realized. In particular, the high CRIs of ~ 83 from 6 V to 9 V are much higher than those of the WOLEDs reported previously with the complementary color model. The high CRI values are benefiting from the broad

emission band of the excimer and the strong emission of the blue- and orange-excimeres. The excimer forms from the CT between HOMO_{donor} and LUMO_{acceptor} as mentioned earlier. From the energy level schematic diagram inserted in Fig. 6a, the orange- and blue-excimeres are well confined in the energy level of DPEPO (orange-excimer: 5.5 eV HOMO and 3.5 LUMO; blue-excimer: 6.1 eV HOMO and 3.5 LUMO; DPEPO: 6.8 eV HOMO and 2.6 eV LUMO²⁵), which improve the efficiency of exciton recombination. On the other hand, the existence of spacers balances the emission of blue- and orange-excimeres further²⁶ and achieves a complete confinement of excitons in each EML on account of the high triplet energy of the spacer. In summary, the carriers balancing transport and efficient confinement for the carriers and excitons due to the suitable spacer and the natural broad emission band of the excimer are the reasons for the high efficiency/stability/CRI WOLEDs.

Finally, we list the EL performance of Device A to Device E in Table 1. Except for Device A, all other devices achieve white light emission and Device E achieves the best performance with a high EQE of 7.92% and a high CRI of ~ 83 . In spite of the CRIs of Device B to Device D being lower than 80, the values of 76–78 are still higher than those of many other WOLEDs reported before with the complementary color model. Table 2 shows the EL performances of representative WOLEDs with only complementary color emission for comparison. The high CRI value in our work is attributed to the intrinsic broad EL spectra with excimer emission. Fig. 7 shows plots of the current density–voltage–luminance curves for all of the devices. It is a pity that the turn-on voltage increases with the addition of spacer number. The large energy barrier between the EML and DPEPO is the reason for the higher turn-on voltage and the charges need a higher voltage to overcome the barrier into the EML. Another pity is the emitting luminance; the luminance of the devices with spacers is far lower than that of the device without a spacer (Device B to Device E vs. Device A). The low luminance is also caused by the introduction of a spacer, which blocks too many charges in the EML and reduces the amount of excitons because of the shallow LUMO, deep HOMO and low

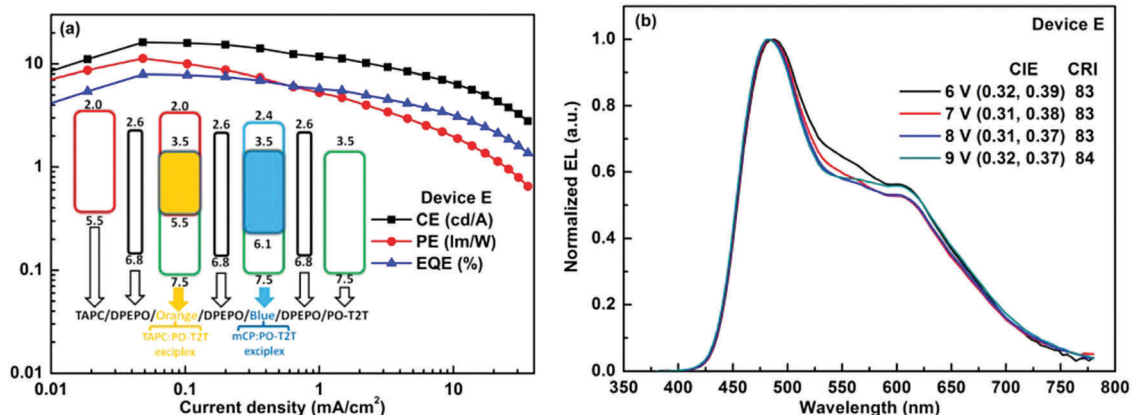


Fig. 6 The EL performance of Device E. (a) CE, PE and EQE–current density curves. Inset: The energy level schematic diagram of the device. The numbers indicate the LUMO and HOMO relative to a vacuum (units in eV). (b) EL spectra with CIE and the CRI under different voltages.

Table 1 The summarized EL performance of all of the WOLEDs in this paper

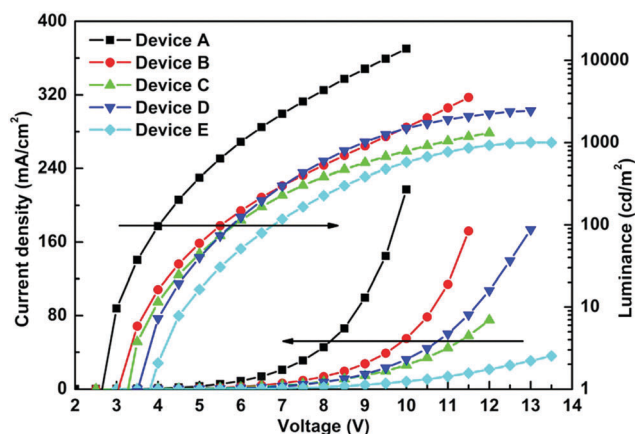
	CE _{Max./500} ^a [cd A ⁻¹]	PE _{Max./500} ^b [lm W ⁻¹]	EQE _{Max./500} ^c [%]	CIE at 8 V	ΔCIE (6–9 V)	CRI (6–9 V)
Device A	12.3/12.2	10.9/7.3	5.53/5.45	(0.52, 0.47)	±(0.00, 0.00)	55–57
Device B	9.3/4.1	8.9/1.6	4.50/1.95	(0.44, 0.41)	±(0.07, 0.04)	85–72
Device C	8.4/4.2	7.6/1.5	3.90/1.95	(0.44, 0.43)	±(0.03, 0.02)	80–74
Device D	16.8/8.7	13.2/3.5	8.59/4.41	(0.27, 0.34)	±(0.01, 0.01)	77–79
Device E	16.2/7.5	11.3/2.5	7.92/3.68	(0.31, 0.37)	±(0.00, 0.02)	83–84

^a Maximum current efficiency, and at 500 cd m⁻² (CE_{Max./500}). ^b Maximum power efficiency, and at 500 cd m⁻² (PE_{Max./500}). ^c Maximum EQE, and at 500 cd m⁻² (EQE_{Max./500}).

Table 2 EL performances of the representative WOLEDs with complementary color emission

WOLEDs	CE [cd A ⁻¹]	PE [lm W ⁻¹]	EQE [%]	CIE	CRI
Fluorescent					
This work	16.2	11.3	7.92	(0.31, 0.37)	84
Ref. 27 ^a	14	9.2	5.6	(0.334, 0.337)	79
Ref. 28 ^b	N/A	11.3	N/A	(0.30, 0.31)	67
Ref. 21 (tandem) ^a	27.7	15.8	11.6	(0.29, 0.35)	70.6
Phosphorescent					
Ref. 29 ^c	N/A	44	25	(0.335, 0.396)	68
Ref. 30 ^a	23.1	15.6	13.5	(0.33, 0.34)	59
Ref. 31 ^b	44.4	45.4	19.1	(0.34, 0.35)	72.2
Ref. 32 ^b	N/A	34	20	(0.44, 0.44)	75
Fluorescent/phosphorescent hybrid					
Ref. 33 ^a	N/A	N/A	16.8	(0.44, 0.38)	74
Ref. 4 ^a	N/A	29.4	15.0	(0.348, 0.422)	63
Ref. 4 ^a	N/A	41.5	19.1	(0.358, 0.430)	54
Ref. 4 ^a	N/A	51.2	20.8	(0.398, 0.456)	48
Ref. 34 ^b	34.59	N/A	N/A	(0.469, 0.419)	45

^a Maximum efficiency. ^b Efficiency at 100 cd m⁻². ^c Efficiency at 1000 cd m⁻².

**Fig. 7** The current density–voltage–luminance curves of all of the OLEDs in this paper.

carrier mobility of the DPEPO spacer. The ultra-low current density with the spacer devices in Fig. 7 also confirms the point. Anyway, the design idea with a complementary color exciplex to achieve a high CRI and stable spectra is feasible and we would improve the luminance and efficiency further in future work by seeking more suitable spacer materials.

4. Conclusion

In conclusion, by tuning the spacer location and amount, high color stability and high CRI (>80) pure exciplex WOLEDs with merely complementary colors are obtained. For Device E with a three spacer structure, the maximum current efficiency, power efficiency and EQE are 16.2 cd A⁻¹, 11.3 lm W⁻¹ and 7.92%, respectively. A high spectral stability with CIE coordinates of (0.31 ± 0.00, 0.37 ± 0.02) and a high CRI of ~83 with only two colors emitting under different voltages are realized simultaneously. The balanced transport, recombination and confinement for carriers and excitons by the suitable application of spacers, along with the natural broad emission band of exciplexes, are responsible for the excellent WOLED performance. We provide a simple way to improve the CRI, and the luminance and efficiency are expected to be improved further by deeper exploration in the future.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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