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# Highly efficient tandem full exciplex orange and warm white OLEDs based on thermally activated delayed fluorescence mechanism

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ABSTRACT

In this manuscript, we demonstrated highly efficient tandem full exciplex orange and warm white organic light emitting diodes (OLEDs) by using 2,4,6-tris(3-(1H-pyrazol-1-yl)phenyl)-1,3,5-triazine (3P-T2T):cesium carbonate ($Cs_2CO_3$)/Al/molybdenum trioxide ($MoO_3$) as the charge generation layer. The maximum current efficiency and external quantum efficiency (EQE) of the optimized tandem orange OLEDs are 38.1 cd/A and 14.4%, respectively. We then utilized two primary colors of blue and orange exciplex emitter to achieve highly efficient white emission. The tandem WOLEDs made by stacking two single color OLEDs in series demonstrate a maximum EQE of 9.17%. Meanwhile, the spectra are almost the same under various voltages, with the Commission Internationale de l'Eclairage coordinates of (0.41, 0.44), which change only (±0.003, ±0.002) from 6 V to 14 V. We prove that the highly efficient tandem full exciplex orange and white OLEDs are attributed to the thermally activated delayed fluorescence by efficient reverse intersystem crossing. Balanced carriers injection and recombination are responsible for the stable spectra of the white OLEDs.



## 1. Introduction

It is well known that many fluorescent and phosphorescent organic light emitting diodes (OLEDs) have attracted tremendous attention due to their promising applications in flat-panel displays and solid state lighting, etc. However, although the phosphorescent OLEDs can provide a high internal quantum efficiency (IQE) of 100%, but the practical applications of such phosphorescent OLEDs would suffer from rather high cost and a serious efficiency roll-off at high current density [1,2]. The fluorescent OLEDs have advantage of high reliability and low cost, but the limit with a maximum external quantum efficiency (EQE) of 5% is an inevitable drawback, which is because of the non radiative transition of 75% triplet excitons [3,4]. In view of above disadvantages in fluorescent and phosphorescent OLEDs, thermally activated delay fluorescence (TADF) have been successfully applied to high efficient OLEDs with various organic emitters in recent years [5–8]. The research of white OLEDs based on the TADF has also been undergoing [9–11]. Particularly, the attainable 100% IQE of OLEDs based on TADF mechanism has received considerable attention. TADF is characterized by small energy difference ($\Delta E_{(S-T)}$) between the singlet ($S_1$) and triplet ($T_1$) excited states, which result in an efficient

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reverse intersystem crossing (RISC). Because of the RISC efficiency could reach up to 100%, so an IQE of 100% via TADF can be theoretically realized. Exciplex is a charge transfer state formed between the donor and acceptor. Both of the highest occupied molecular orbital (HOMO) of donor ($HOMO_{donor}$) and the lowest unoccupied molecular orbital (LUMO) of acceptor ($LUMO_{acceptor}$) contribute to exciplex formation by the exchange energy. The exciplex could exhibit either positive or negative $\Delta E_{(S-T)}$ [12–14]. The efficiency of RISC could be enhanced through a small $\Delta E_{(S-T)}$, so the EQE of exciplex OLEDs can be further improved. Many papers had report highly efficient exciplex OLEDs with above 5% EQE in terms of RISC processes, but the reports had mainly concentrated on single color OLEDs, especially green emissive exciplex OLEDs [15–17].

In generally, white OLEDs (WOLEDs) could be obtained by either blending three (blue, green and red) primary colors or two (blue and orange) complementary colors from different emitters. The former WOLEDs present the imperfections of cockamamie fabrication processes and color instability with increasing bias voltage. Thus, the WOLEDs comprised of two complementary colors seemingly possess more dominance because of the color stability and the simple fabrication processes. Because of this, orange emission becomes an important complementary color in latter WOLEDs. Orange emission can be achieved by synthesizing a single orange emitting material or mixing green and red emission. In fact, there is another method to earn efficient orange emission, that is, orange exciplex. Orange exciplex can be obtained by simply combining suitable donor and acceptor materials. Moreover, now almost all the highly efficient WOLEDs that composed of two colors are achieved by a blue fluorescent and an orange phosphorescent emitting system or blue and orange full phosphorescent emitting components. In other words, the highly efficient WOLEDs composed of two colors cannot be achieved without orange phosphorescent material. Just as described above, the exciplex OLEDs based on TADF could offer high electroluminescence (EL) efficiency. If we can achieve highly efficient orange and blue exciplex OLEDs and the two emitters could be utilized as two units of the tandem OLEDs, then the highly efficient WOLEDs could be demonstrated.

In this manuscript, we selected appropriate donor and acceptor from the commercial available hole and electron transport materials according to the offset between $HOMO_{donor}$ and $LUMO_{acceptor}$ and carrier mobility. The donor and acceptor of blue exciplex are 4,4′,4″-tri(N-carbazolyl)triphen-ylamine (TCTA) and 4,7-diphenyl-1,10-phenanthroline (Bphen), while 1,1-bis((di-4-tolylamino)phenyl)cyclohexane (TAPC) and 2,4,6-tris(3-(1H-pyrazol-1-yl)phenyl)-1,3,5-triazine (3P-T2T) are serviced as the donor and acceptor components of the orange exciplex, respectively. Although the blue exciplex of TCTA:Bphen have been reported, but there were no the research about its delayed fluorescent characteristics [18]. Hung et al. reported a highly efficient yellow exciplex with TCTA:3P-T2T [19]. But actually, the EL spectrum was yellow-green with the peak value at ~544 nm, which could not well play the role of complementary color in white emission. In order to achieve orange emission, we choose TAPC as the donor, which has a relative shallow HOMO energy level (5.5 eV) compared to TCTA (5.8 eV). Firstly, we verified that both orange and blue exciplex were based on TADF mechanism by time resolved photoluminescence (PL) spectra and transient PL decay measurement. Then the efficacy of the charge generation layer (CGL) was proved by its application in tandem orange exciplex OLEDs. At last, the optimal tandem WOLEDs with orange and blue exciplex as the two sub-units offer a maximum current efficiency and EQE of 25.4 cd/A and 9.17%, respectively; at the same time, the WOLEDs exhibit stable Commission Internationale de l'Eclairage (CIE) coordinates of (0.41 ± 0.003, 0.44 ± 0.002). Thus, to date, such a high EQE full exciplex warm white OLEDs based on TADF mechanism was demonstrated firstly.

## 2. Experiments methods

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 (~$3 \times 10^{-4}$ Pa). The organic materials were procured commercially without further purification. Steady-state PL and EL spectra were measured with F7000 and OPT-2000 spectrophotometer, respectively. Time resolved PL spectra and transient PL decay were measured with the combination of Nd-YAG laser (pulse width of 10 ns, repetition frequency of 10 Hz), spectrograph (HJY, Triax 550) and oscilloscope (Tektronix, TDS3052). The samples were loaded in a cryostat and temperature was cooled down from room temperature to 14 K. The electrical characteristics of the OLEDs were measured with a Keithley model 2400 power supply combined with a ST-900M spot photometer and were recorded simultaneously with measurements. EQE was calculated from the current density–voltage–luminance curve and spectrum data. All measurements were carried out at room temperature and under ambient conditions without any protective coatings.

## 3. Results and discussion

The PL spectra of the neat TCTA, TAPC, Bphen and 3P-T2T as well as the co-deposited films of TCTA:50 mol%Bphen and TAPC:50 mol%3P-T2T are displayed in Fig. 1. The PL peaks of TCTA:Bphen and TAPC:3P-T2T are red-shifted to 464 nm (2.67 eV) and 544 nm (2.28 eV) compared to each neat donor and acceptor components, respectively. Both the exciplex peak energies are close to the gap between the $HOMO_{donor}$ and $LUMO_{acceptor}$, indicating that the mixed films of TCTA:Bphen and TAPC:3P-T2T form the blue and orange exciplex in the excited state, respectively.

In order to verify that both blue and orange exciplex contain the contribution of delayed fluorescence, the time resolved PL spectra at 300 K and the transient PL decay feature at various temperatures for TCTA:Bphen and TAPC:3P-T2T co-deposited films were determined, as shown in Figs. 2 and 3, respectively. In Fig. 2, we see that both the blue (464 nm) and orange (544 nm) exciplex emissions are ranging from a short delay time of 100 ns to a long

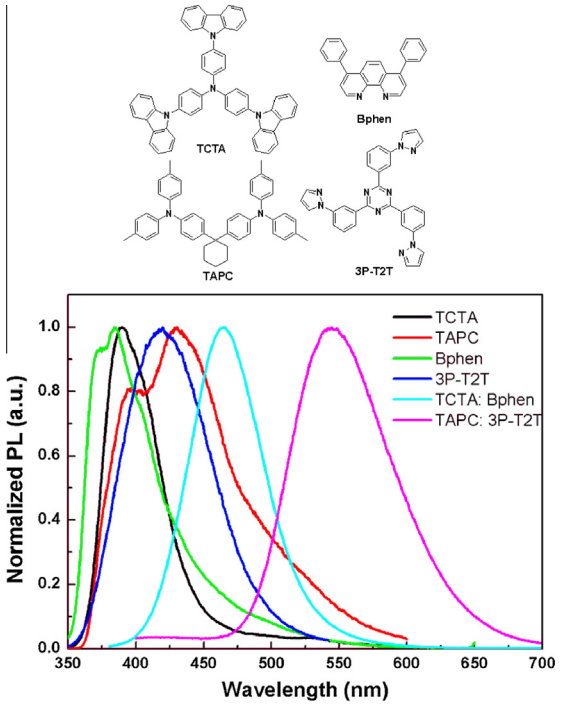


**Fig. 1.** The chemical structure of organic materials and PL spectra of the films of TCTA, Bphen, TAPC, 3P-T2T, TCTA:Bphen and TAPC:3P-T2T.

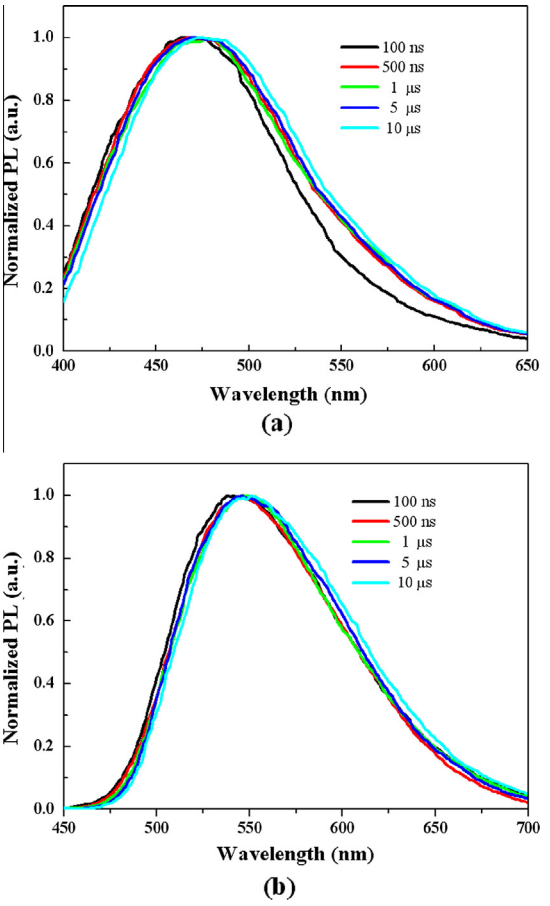


**Fig. 2.** Normalized time resolved PL spectra of the mixed films at different delay times at 300 K. (a) TCTA:Bphen mixed films. (b) TAPC:3P-T2T mixed films.

delay time of 10 μs, demonstrating that exciplex emissions are based on TADF mechanism. It is interesting that with the increase of the decay time, the PL spectra are slightly red-shifted, which is possibly ascribed to the existence of a broad energy level distribution of the exciplex [13].

From Fig. 3, we observe that the transient PL decay curve from the two co-deposited films of TCTA:Bphen and TAPC:3P-T2T exhibit the similar characteristics. That is, both the PL intensity and excited state lifetime increase with the temperature rising from 14 K to 300 K. It demonstrates the efficient RISC from triplet to single level by thermally activated. All the decay curves are best fitted by two-exponential decay kinetics. The PL decays of the blue and orange exciplex fluorescent emissions can be divided into prompt components of 79.1 ns and 82.9 ns and the delayed components of 1.12 μs and 2.92 μs at 300 K, respectively. Thus, we confirm that the blue and orange exciplex emissions should be resulted from efficient RISC processes from triplet to single excited states. In terms of the time resolved spectra and the transient PL decay characteristics, a $\Delta E_{(S-T)}$ of almost zero could be expected so that the RISC processes could take place easily [20].

On the basis of above description, we confirm that both the blue and orange exciplexes include the contribution of TADF through the efficient RISC processes. Then the two exciplexes are serviced as the sub-units to design full exciplex tandem WOLEDs. However, in order to demonstrate preferably the highly efficient WOLEDs with tandem structure, the design of CGL is quite crucial. The CGL usually consists of an n-type layer and a p-type layer. The tandem OLEDs with the excellent CGL could realize a high luminance and efficiency at a low current density. In this paper, we selected previous reported ETL:$Cs_2CO_3$ and $MoO_3$ as the n-type and p-type layers, respectively [21]. The devices with only CGL composed of ITO/Bphen (30 nm)/3P-T2T:$Cs_2CO_3$ (10 nm)/Al ($x$ nm)/$MoO_3$ (5 nm)/TAPC (30 nm)/Al ($x$ = 0 or 1) were fabricated. Note that the current density–voltage curve of the device with 1 nm thick Al is better than the device without Al layer, as described in Fig. 4. This is because the charge separation and electron injection to 3P-T2T:$Cs_2CO_3$ layer could be accelerated by the middle Al layer. The presence of 1 nm Al layer in CGL decreases dramatically the operation voltage because of the reduced voltage drop across the CGL [22,23]. The excellent charge separation and electron injection of CGL gives a good foundation for achieving highly efficient tandem WOLEDs.

In order to further verify that 3P-T2T:$Cs_2CO_3$/Al/$MoO_3$ possesses excellent charge generation capability, the tandem orange exciplex OLEDs was fabricated (see Fig. 5(a)). The EL parameters with current density, luminance, current efficiency, EQE and EL spectra of single and tandem OLEDs are shown in Fig. 5. From Fig. 5(b), we can see that the voltage and luminance of the tandem doubled compared to single OLEDs. At the current density of 20 mA/cm$^2$, the voltage and luminance of single and tandem

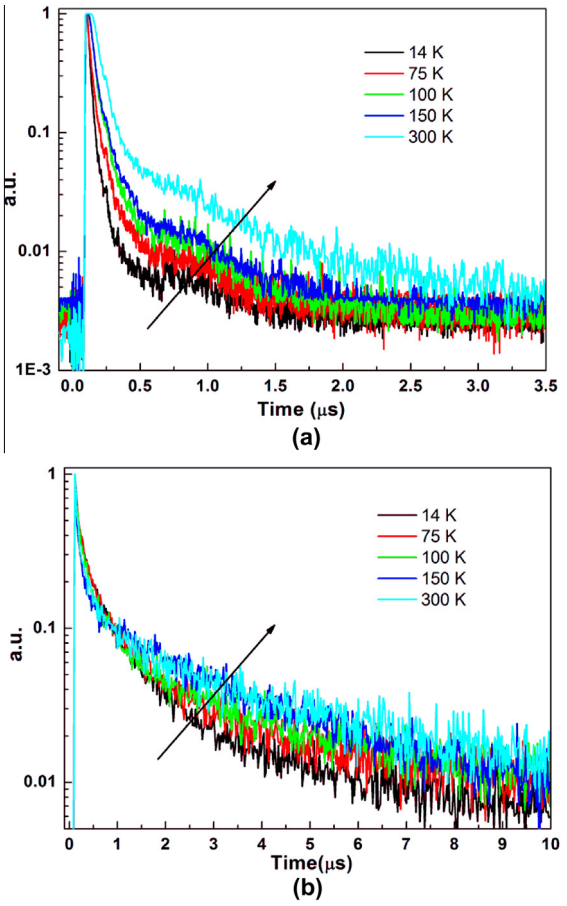


**Fig. 3.** The transient PL decayed curves of the mixed films under different temperatures. (Instrument response of 10 ns) (a) TCTA:Bphen mixed films at 464 nm. (b) TAPC:3P-T2T mixed films at 544 nm.

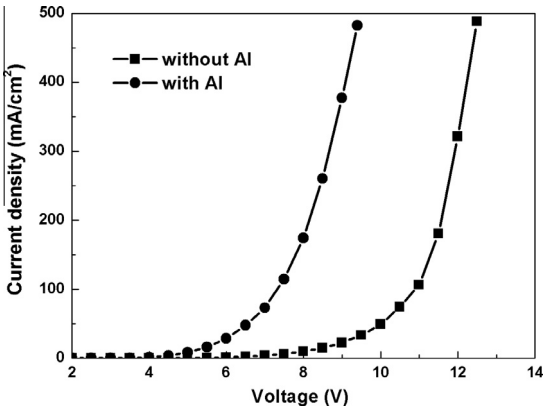


**Fig. 4.** The current density–voltage characteristic curves of the CGL-only device.

OLEDs are 4.3 V, 2460 cd/m$^2$ and 8.4 V, 5451 cd/m$^2$, respectively. The optimal tandem orange exciplex OLEDs offers a maximum current efficiency of 38.1 cd/A and a peak EQE of 14.4%, respectively. To our best knowledge, the EQE of the tandem orange exciplex OLEDs is the highest value in the single color exciplex OLEDs. At the current density of 20 mA/cm$^2$, the tandem device gives a current efficiency and EQE of 27.4 cd/A and 10.3%, while the single device offers 12.6 cd/A and 4.78%, respectively. It exhibits clearly the twofold current efficiency and EQE at the same current density. Table 1 summarizes the data of all the devices in this paper. In terms of above mentioned data of orange exciplex OLEDs, we conclude that 3P-T2T:$Cs_2CO_3$/Al/$MoO_3$ is an efficient CGL. The electrons and holes could generate and separate efficiently at the CGL and then inject into the two adjacent electron and hole transport layers, respectively.

The PL spectrum of the co-deposited film of TAPC:3P-T2T and the EL spectra of the single and tandem orange OLEDs are depicted in Fig. 6. Differing from the PL spectrum with the emission peak at 544 nm, the EL spectrum is shifted to a longer wavelength and fix on 570 nm. The phenomenon was different from the report of Hung et al., which showed the same emission peak of PL and EL [19]. We consider that the occurrence of spectral red-shift can be explained by the different excited mechanism between the PL and EL processes, as described below. Under electrical excitation, the excitons are produced from the recombination between electron on $LUMO_{acceptor}$ and hole on $HOMO_{donor}$, respectively [24,25]. While under light excitation, the electron on $HOMO_{donor}$ is excited firstly into its LUMO and then the electron transferred into $LUMO_{acceptor}$. The electron on $LUMO_{acceptor}$ and hole leaving at $HOMO_{donor}$ are coupled by coulomb interactions and then the coupled electron and hole are relaxed to the ground state [26,27]. Thus, we speculate the difference between the emission peak of PL and EL could be carried out even though both of the emissions of PL and EL are derived from the radiative relaxation from the excited state to ground state of exciplex. We also note that the spectral full width at half maximum (FWHM) of the tandem OLEDs is 105 nm, which is narrowed slightly by 15 nm compared to the single device with a FWHM of 120 nm. The phenomenon of spectral narrow arises from a stronger microcavity effect of tandem device than the single device.

In order to design full exciplex WOLEDs, blue exciplex device with the structure of ITO/$MoO_3$ (3 nm)/TCTA (15 nm)/TCTA:Bphen (12 nm)/Bphen (30 nm)/LiF/Al was also investigated except for above orange exciplex device, the main performances are also shown in Table 1. The blue exciplex OLEDs shows a relative low EQE of 2.65%, but it still behaves the delayed fluorescence emission based on the argument above. Finally, in the light of above investigation on orange and blue exciplex OLEDs and the special design of CGL, an optimized tandem full exciplex WOLEDs was demonstrated successfully. Fig. 7 depicts the EL performances and the insert of Fig. 7(b) denotes the device structure of the WOLEDs. We obtain a warm white emission and the WOLEDs provides a maximum current efficiency of 25.4 cd/A; even at a luminance of 1000 cd/m$^2$, the current efficiency also remains 23.9 cd/A. More important, a peak EQE of 9.17% is harvested; to our best knowledge, this is the highest EQE based on full exciplex warm white OLEDs. It is significant that so high EQE is still kept 8.63% at 1000 cd/m$^2$, which could satisfy the requirement

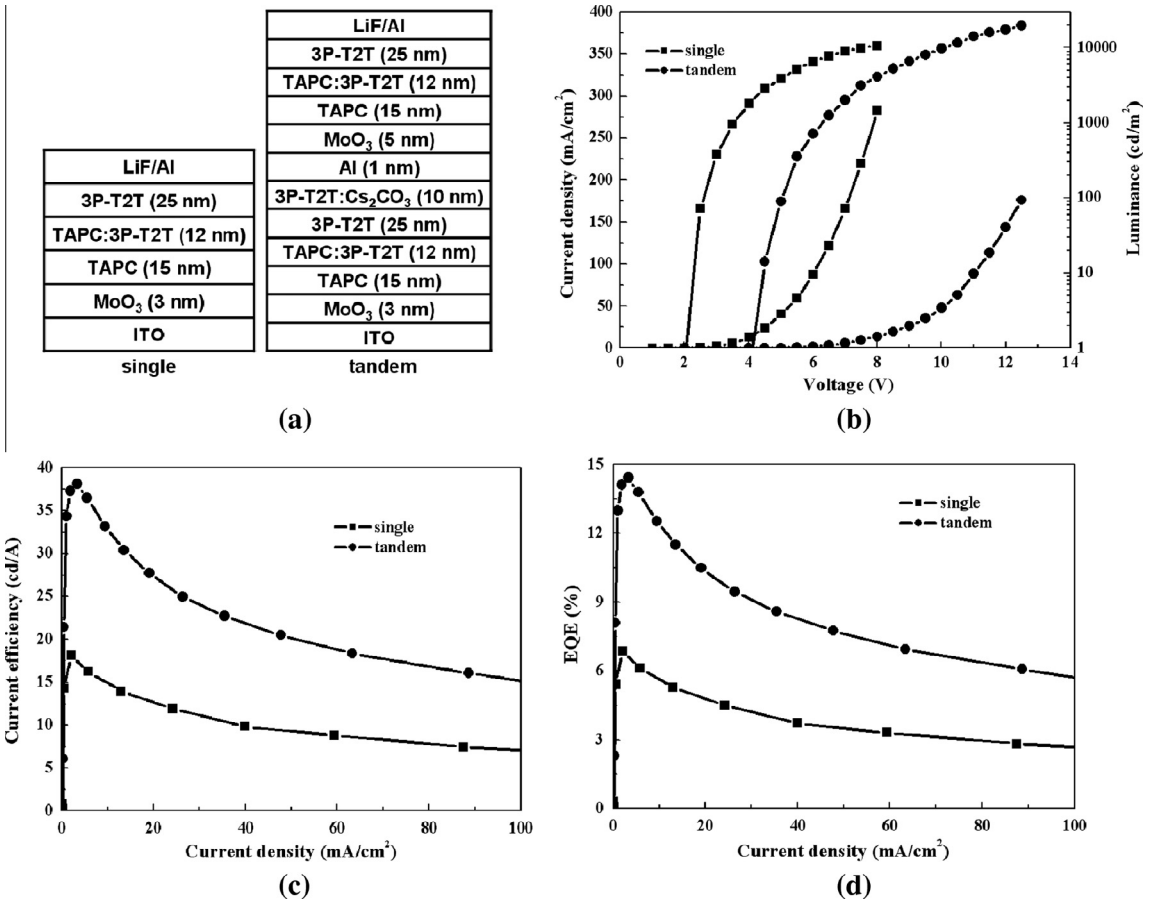


**Fig. 5.** (a) The device structures of the single and tandem OLEDs. (b) Current density–voltage–luminance curve. (c) Current efficiency–current density curve (d) EQE-current density curve.

**Table 1**
The EL performances of all the OLEDs in this paper.

| | $V_{on}$[a] (V) | Lum.[b] (cd/m$^2$) | $CE_{max}$[c] (cd/A) | CE[d] (cd/A) | $EQE_{max}$[e] (%) | EQE[f] (%) | $PE_{max}$[g] (lm/W) |
|---|---|---|---|---|---|---|---|
| Single | 2.06 | 2460 | 18.1 | 12.6 | 6.86 | 4.78 | 18.9 |
| Tandem | 4.11 | 5451 | 38.1 | 27.4 | 14.4 | 10.3 | 19.6 |
| Blue | 2.62 | 482 | 3.66 | 2.55 | 2.65 | 1.79 | 3.82 |
| White | 4.64 | 3329 | 25.4 | 16.7 | 9.17 | 6.02 | 11.5 |

[a] Turn-on voltage.
[b] Luminance at 20 mA/cm$^2$.
[c] Maximum current efficiency.
[d] Current efficiency at 20 mA/cm$^2$.
[e] Maximum EQE.
[f] EQE at 20 mA/cm$^2$.
[g] Maximum power efficiency.

of the general lighting applications. The small $\Delta E_{(S-T)}$ of exciplex promote RISC process and the efficient emission of delayed fluorescence is realized further, which should be responsible for the high EQE. The CGL also combine efficiently the sub-unit of blue and orange exciplex by effective generation, separation and injection of carriers. The achievement of such a high EQE is ascribed to the contribution of TADF and efficient CGL. Recently, Hung et al. reported a tandem, all-exciplex WOLEDs [28]. Their WOLEDs realized the maximum efficiencies at a very low luminance and suffered from a very high operation voltage. The high operation voltage means high power cost, which cannot be tolerated for WOLEDs. However, our device achieves the maximum efficiency at a high luminance and has a rather low operation voltage. The low operation voltage in this paper can be attributed to the more efficient CGL and orange emission. Therefore, our WOLEDs are advantage from the power cost and practical application.

Fig. 7(d) shows the EL spectra of the WOLEDs under various voltages. Different from Hung et al. paper that showed

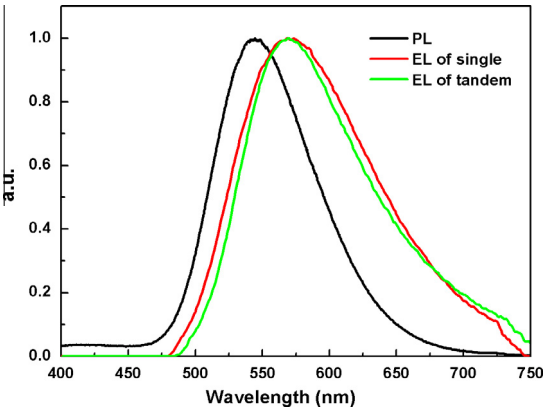


**Fig. 6.** The PL spectrum of TAPC:3P-T2T mixed film and the EL spectra of single and tandem OLEDs.

a cool white emission [28], we earned a warm white OLEDs and the correlated color temperature (CCT) was 3723 K. Because warm white give a sweet, soft and warm feel, so our WOLEDs is more suitable for home lighting compared with the WOLEDs in Ref. [28]. The characteristic emission peaks of two exciplex with the blue sub-band around 451 nm and orange sub-band around 570 nm are observed in the white EL spectra, respectively. It is well known that the excellent spectral stability of WOLEDs depicts very large significance for the application of either displays or solid state lightings. It is surprising that the EL spectra of our WOLEDs are quite stable, i.e., the spectra are almost the same with the rising voltage. The CIE coordinates behave a very small change ranging from (0.416, 0.447) to (0.413, 0.446) with the voltage from 6 V to 14 V. The change of CIE coordinates is only (±0.003, ±0.002). The stable spectra can be attributed to the balanced injection and transport of carriers, which results in an efficient recombination between electron and hole in emitting layer [29].

## 4. Conclusion

In conclusion, we fabricated successfully the tandem full exciplex orange and warm white OLEDs by utilizing 3P-T2T:$Cs_2CO_3$/Al/$MoO_3$ as the CGL. The tandem orange and warm white OLEDs achieve a maximum EQE of 14.4% and 9.17%, respectively. Such a high efficiency can be explained by the excellent CGL and the presence of

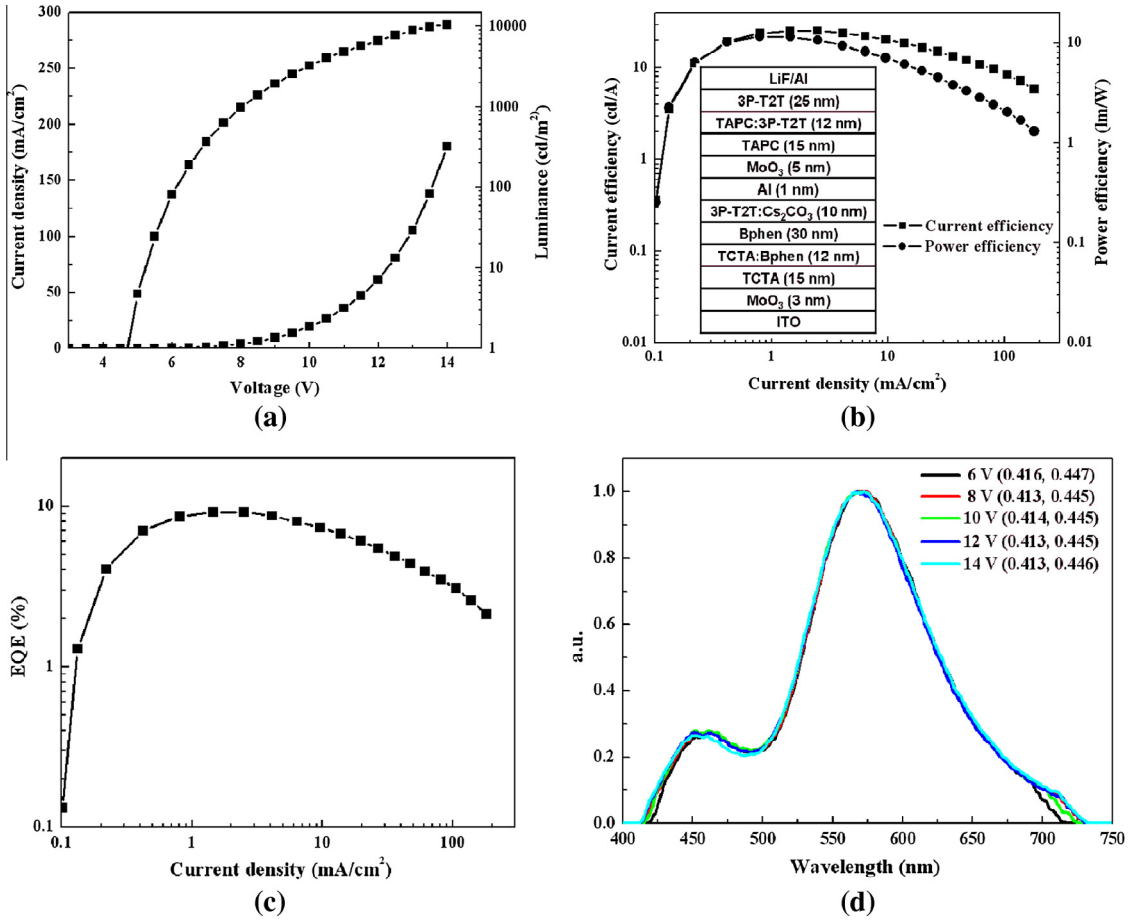


**Fig. 7.** (a) Current density–voltage–luminance curve. (b) Current efficiency and power efficiency curve. (Inset is the device structure of WOLEDs) (c) EQE-current density curve. (d) The EL spectra under various voltages.

TADF in orange and white exciplex emission system. We also obtain very stable warm white EL spectra with CIE coordinates of (0.41 ± 0.003, 0.44 ± 0.002) from 6 V to 14 V. Balanced injection and transport of carriers is responsible for the stable spectra. The full exciplex WOLEDs combine the advantage of fluorescent and phosphorescent OLEDs, that is, low cost, high reliability and efficiency. Besides, such warm white could be used for room lighting because of the more suitable illumination color. We believe that full exciplex white OLEDs will express larger application prospect and higher efficiency can be achieved by screening carefully more appropriate donor and acceptor materials.

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