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Highly efficient white organic light-emitting diodes based on balanced bipolar-transporting blue hybridized local charge transfer fluorophores†

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Fluorescent/phosphorescent hybrid white organic light-emitting diodes (WOLEDs) that combine the blue fluorophore with long-wavelength emissive phosphors have attracted a great deal of attention in the fields of solid-state lighting and full-color display. However, the efficiency and its roll-off at high luminance are still unsatisfactory, resulting from serious exciton-quenching involving long-lived triplet excitons. In this work, we have developed a high-performance hybrid WOLED by integrating a perfect arrangement of emitting layers with a high and balanced hole- and electron-transporting hybridized local charge transfer (HLCT) blue fluorophore. The resulting two-color WOLED with the CIE coordinates of (0.39, 0.43) achieves a high current efficiency of 78.8 cd A^{-1} and a maximum external quantum efficiency (EQE_{max}) of 27.1%. More importantly, the value of EQE is maintained as high as 23.8% at 1000 cd m^{-2}, showing low efficiency roll-off. This efficiency is one of the best results of WOLEDs achieved using blue HLCT materials.

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1. Introduction

The white organic light emitting diode (WOLED) has a particularly high application potential in the field of solid-state lighting and full-color display. This is due to its excellent advantages of being light weight, being thin, having low power consumption, and uniform illumination.[1,2] Generally, according to the difference of used emitting materials, the devices can be divided into all-phosphorescent, all-fluorescent and fluorescent/phosphorescent (F/P) hybrid WOLEDs. Although all-phosphorescent, the all-fluorescent devices based on thermally activated delayed fluorescence (TADF) materials can harvest both singlet and triplet excitons effectively, realizing high external quantum efficiency (EQE) exceeding 20%, and the poor device stability restricts their practical applications, resulting from the lack of stable and efficient blue phosphorescent and TADF materials. In addition, there are usually serious efficiency roll-off events at high luminance in these two WOLEDs due to the triplet-involved quenching processes. In comparison, the F/P hybrid WOLEDs combining the long-term stability blue fluorophore with high efficiency green/red or yellow phosphors are regarded as the most ideal and feasible candidates for the applications of display and lighting technologies in the industrial community.[1–5]

In order to achieve high efficiency in F/P hybrid WOLEDs, many efforts have been made to restrain the loss of triplet excitons from blue fluorophores, including the development of new blue fluorophores and architectural design of an emitting-layer (EML). For example, Wang *et al.* designed highly efficient WOLEDs with three-EML structures using hybridized local charge transfer (HLCT) blue materials as the host, as well as an emitter, achieving the maximum EQE and power efficiency of 25.4% and 70 lm W^{-1}, respectively.[6] Ma and co-workers combined a triplet–triplet annihilation (TTA) blue fluorophore with the use of an interlayer between the blue EML and phosphorescent EML, realizing the EQEs of 20.80% and 19.32% at the practical luminance of 1000 and 5000 cd m^{-2}, respectively.[7] Duan *et al.* developed a simplified single-EML hybrid-WOLEDs based on a blue TADF material, realizing the highest forward-viewing EQE of 20.8% by regulating the doping concentration of yellow phosphor.[8] Based on the blue aggregation-induced emission (AIE) fluorophores, Ma *et al.* reported on a three-color WOLED with a high color rendering index (CRI) of over 80 and good color stability by accurately managing the arrangement of EMLs,[9] while Zhao and co-workers realized a two-color WOLED with a total EQE of 28%

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at 1000 cd m^{-2} by optimizing the co-doping ratio of interlayer.[10] These facts illustrate that the development of high-performance hybrid WOLEDs not only is related to the EML architecture, but also is heavily influenced by the properties of blue fluorophores.[11–13,30,31]

Recently, OLED materials based on the HLCT mechanism have developed rapidly. Compared with TADF materials with the spatially separated distribution of frontier molecular orbitals, most HLCT materials exhibit a mixture of local excitation (LE) and charge transfer (CT) in emission, which allows the coexistence of high fluorescence quantum efficiency and high reverse intersystem crossing (RISC) rate from the high-lying triplet (T_1) to singlet (S_1) states. It is instrumental in solving the efficiency roll-off problem that is induced by long-loved triplet excitons in the non-doped OLEDs. At present, some monochromatic OLEDs based on HLCT materials with high EQE and very small efficiency roll-off have been reported.[14–21] However, there are few reports about simple and efficient hybrid WOLEDs based on the HLCT materials, and their efficiency roll-off at high luminance needs to be further improved.

In this work, we have achieved super-efficient hybrid WOLEDs using blue HLCT molecules as both host and emitter. By finely regulating the arrangement of EMLs, the hybrid WOLED realizes the high electro-generated excitons utilization, as well as small efficiency roll-off at high luminance. As a result, the excellent electroluminescence (EL) efficiencies of 78.8 cd A^{-1}, 53.8 lm W^{-1} and 27.1% are achieved simultaneously, respectively. The EQE remains as high as 23.8% at the practical luminance level of 1000 cd m^{-2}. To the best of our knowledge, this is one of the highest values achieved in the WOLEDs based on HLCT material.

2. Results and discussion

In F/P hybrid WOLEDs, blue fluorophores have a significant influence on the EL performance. An excellent blue fluorophores should provide the following features: i) higher T_1 energy level than that of the used phosphors, which can prevent the reverse energy transfer from the phosphor to the fluorophore, as well as avoid the use of an interlayer that may lead to several disadvantages;[22] ii) appropriate highest occupied molecular orbitals (HOMO) and lowest unoccupied molecular orbitals (LUMO) energy levels, which is useful to realize effective holes and electron injection, and reduce carrier accumulation in the interface between the EML and neighbouring layers;[23] iii) excellent bipolar-transporting properties, which can broaden the exciton recombination zone and suppress exciton annihilation; iv) high solid-state photoluminescence quantum yield (PLQY), which can ensure the reliable blue emission of WOLEDs; v) excellent thermal property, which favors the improvement of the device stability. In recent years, our group has been devoting a lot of effort towards the design and synthesis of red and blue HLCT materials, and their applications in OLEDs.[14,16,17,21] According to the requirements mentioned above of blue fluorophores, a very promising HLCT material *N,N*-diphenyl-4-(7-(4-(5-phenyl-1,3,4-oxadiazol-2-yl)phenyl)9,9-dipropyl-9-*H*-fluoren-2-ylaniline (TPACFOXZ) was selected. As shown in Fig. 1a, it showed fantastic characteristics with the glass transition and decomposition temperatures of 107 and 450 °C, PLQY of 73% in neat film, HOMO/LUMO energy levels of −5.38/−2.62 eV, T_1 of 2.3 eV and balanced bipolar-transporting feature (hole and electron mobilities of 5.60×10^{-5} cm^2 V^{-1} s^{-1} and 6.60×10^{-5} cm^2 V^{-1} s^{-1} under the electrical field of 5×10^5 V cm^{-1}). Furthermore, it achieved a high EQE of 9.8% and stable blue emission with the CIE coordinates of (0.15, 0.19) in the non-doped OLEDs, which is one of the best performances in the blue non-doped HLCT-OLEDs.

Before fabricating the hybrid WOLEDs, we firstly investigated the application potential of TPACFOXZ as a host for the phosphorescent dopant. According to the energy level matching principle, a typical yellow phosphorescent material [2-(thieno[3,2-c]pyridin-4-yl)phenyl] iridium (III) (PO-01) with the T_1 of 2.2 eV, and HOMO/LUMO energy levels of −5.4/−2.8 eV was elected. It can be seen from Fig. 1b that there exists a large overlap between the absorption spectrum of PO-01 and the emission spectrum of TPACFOXZ, illustrating that the Förster resonant energy transfer (FRET) process from TPACFOXZ to PO-01 should be possible and efficient. Furthermore, the T_1 of TPACFOXZ (2.3 eV) is higher than that of PO-01, which can effectively suppress the inversive energy transfer from PO-01 to TPACFOXZ, guaranteeing the full utilization

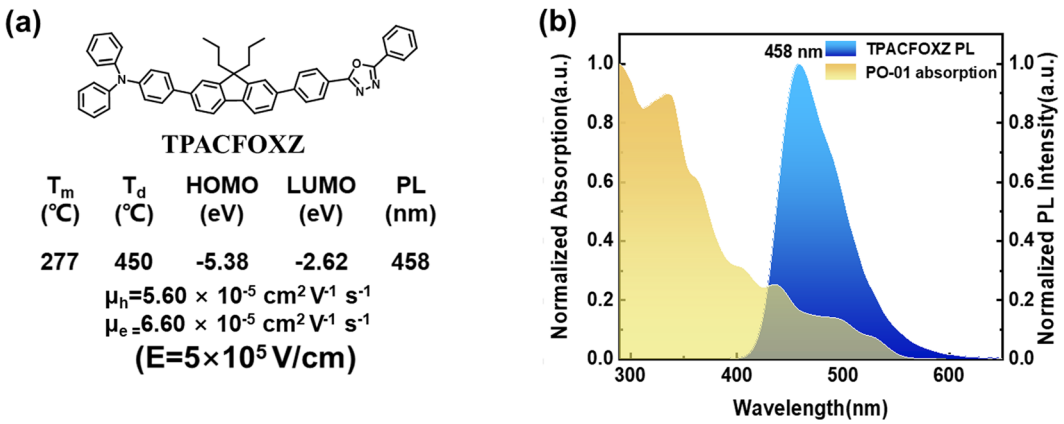

Fig. 1 (a) Schematic diagram of the molecular structure and properties of TPACFOXZ. (b) The normalized absorption spectrum of PO-01 and the emission spectrum of TPACFOXZ.

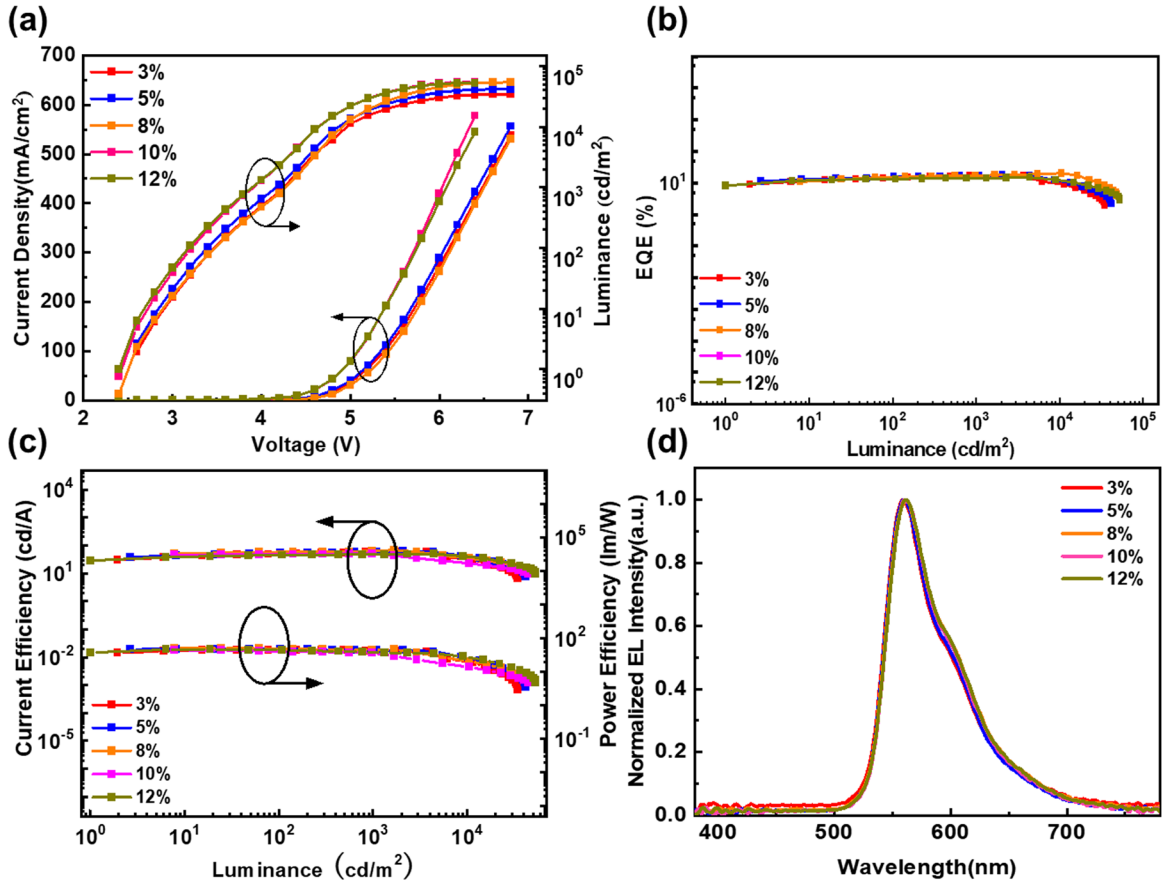

Fig. 2 EL performance of yellow OLEDs with the doping concentrations of 3%, 5%, 8%, 10%, and 12% for the PO-01 materials. (a) Current density-voltage-luminance curves. (b) EQE-luminance curves. (c) Current efficiency-luminance-power efficiency curves. (d) EL spectra at a voltage of 4 V.

of triplet excitons. Thus, the yellow OLEDs with the structure of ITO/PEDOT:PSS (40 nm)/TCTA (20 nm)/TPACFOXZ: PO-01 (xx, 10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm) were fabricated by adjusting the doping concentrations of PO-01 (where xx = 3%, 5%, 8%, 10%, 12%, respectively). Here, indium tin oxide (ITO) and aluminium (Al) served as the anode and cathode, poly(3,4-ethylenedioxythiophene):poly(styren-esulfonate) (PEDOT: PSS) and lithium fluoride (LiF) were used as the hole- and electron-injection layers, and 4,4′,4″-tris(carbazol-9-yl)triphenylamine (TCTA) and 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl)-benzene (TPBi) acted as the hole- and electron-transporting layers, respectively. The energy level diagram and EL performance parameters are shown in Fig. 2 and Fig. S1 (ESI†), and summarized in Table 1. All the devices show low turn-on voltages of 2.4–2.6 V and achieved the luminance of 1000 and 10 000 cd m^{-2} at the voltages of 3.9–4.3 and 4.7–5 V, which can be comparable to that based on *p-i-n* junctions.[24] As the doping concentration of PO-01 increases, the EL performance presents the trend of first increasing and then decreasing. One reason for the phenomenon may be that the Dexter energy transfer (DET) process from TPACFOXZ to PO-01 is not effective enough at low doping concentration, while the concentration quenching effect can be accelerated at high doping concentration. When doping 8% PO-01 into the TPACFOXZ host, the device realizes the best EL performance. The maximum efficiencies of 68.4 cd A^{-1}, 50.9 lm W^{-1} and 20.3% were obtained for the current efficiency (CE_{max}), power efficiency (PE_{max}), and EQE_{max}, respectively. It is worth noting that the value of EQE_{max} is obtained at the high luminance of 3347 cd m^{-2}, which is very

Table 1 Summary of the EL parameters of the OLEDs with different doping concentrations of PO-01

Doping concentration of PO-01	V_{on}[a] [V]	CE_{max}[b] [cd A^{-1}]	PE_{max}[c] [lm W^{-1}]	EQE_{max}[d] [%]	L_{max}[e] [cd m^{-2}]	V_{end}[f] [V]	CIE_{10000}[g]
3%	2.6	59.1	45.6	17.3	34559	6.8	(0.46, 0.52)
5%	2.6	62.1	48.7	18.2	41609	6.8	(0.47, 0.52)
8%	2.6	68.4	50.9	20.3	54013	6.8	(0.47, 0.52)
10%	2.6	51.9	45.3	15.6	54607	6.4	(0.48, 0.52)
12%	2.4	49.9	46.3	14.9	52607	6.4	(0.48, 0.52)

[a] V_{on}: Turn-on voltage at 1 cd m^{-2}. [b] CE_{max}: Maximum current efficiency. [c] PE_{max}: Maximum power efficiency. [d] EQE_{max}: Maximum EQE. [e] L_{max}: Maximum luminance. [f] V_{end}: Turn-off voltage. [g] CIE_{10000}: CIE coordinates at the luminance of 10,000 cd m^{-2}.

distinctive from other host materials.[25–27] Such excellent performance should be attributed to the high and balanced hole and electron mobilities of TPACFOXZ, which broadens the exciton recombination zone and suppresses the quenching effect of the triplet–triplet annihilation and triplet-polaron quenching. Meanwhile, all the devices show stable yellow emission with the CIE coordinates of (0.47–0.48, 0.52) at 10 000 cd m^{-2} (Fig. S2, ESI†).

As for the complementary-color WOLEDs, it requires the synergistic luminescence from TPACFOXZ and PO-01 for covering the whole visible region as much as possible. In order to develop highly efficient hybrid WOLEDs with a simple device architecture, on the basis of the yellow phosphorescent OLEDs, we firstly fabricated the single-EML WOLED (**W**) by realizing the incomplete energy transfer from TPACFOXZ to PO-01. The device configuration is ITO/PEDOT:PSS (40 nm)/TCTA (20 nm)/TPACFOXZ:PO-01 (1%, 10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm), where the doping concentration of PO-01 is as low as 1%. As shown in Fig. 3 and Fig. S3 and Table 2 (ESI†), the device **W** exhibits the maximum luminance, EQE_{max}, CE_{max} and PE_{max} of 17,662 cd m^{-2}, 12.1%, 39.5 cd A^{-1}, and 30.6 lm W^{-1}, respectively. More importantly, there is a negligible efficiency roll-off at the high luminance of 1500 cd m^{-2}. Meanwhile, the device **W** shows a desirable and tunable warm-white light. When the luminance increases from 1000 to 10 000 cd m^{-2}, the CIE coordinates, correlated color temperatures (CCTs), and CRIs shift from (0.43, 0.49), 3659 K, and 37 to (0.29, 0.33), 7508 K, and 63, respectively. According to the emission mechanism in Fig. 3a (inset), owing to the low doping concentration, holes and electrons are mainly recombined and formed the excitons on the TPACFOXZ molecules. Triplet excitons can be converted into singlet exitons *via* high-lying reverse intersystem crossing process, as well as T_1 excitons through the internal conversion process. The electro-generated and transformed singlet excitons can not only transfer their energy to PO-01 molecules for the yellow emission, but also decay to the ground state through emitting blue light. Due to the dense triplet energy levels of TPACFOXZ (Fig. S4, ESI†), the internal conversion process from high-lying triplet state to T_1 state is unavoidable. Thus, the lack of an efficient DET process from TPACFOXZ to PO-01 (caused by low doping concentration) may be the main reason for the waste of T_1 excitons and low EQE of device **W**.

Although the WOLED (**W**) with the single-EML structure can to a certain degree simplify technology and reduce cost, there are still major deficiencies in the performance. Compared to the single-EML WOLEDs, the multi-EML WOLEDs have the greater advantages of the flexible arrangement of each EML and precise regulation of the charge/exciton distribution, which provides a reliable alternative route to high efficiency and low efficiency roll-off hybrid WOLEDs with the full utilization of both singlet and triplet excitons.[23,28] According to the analysis of the HOMO/LUMO energy levels, the material PO-01 has a similar HOMO energy level with TPACFOXZ, while it shows a deeper LUMO energy level than TPACFOXZ, indicating that the electron-trapping effect of PO-01 may be possible in the EL process. To relieve this effect, a valid arrangement of EMLs with yellow EML + blue EML (YB-type) has been designed, where the yellow EML is located on the side of the hole-transporting layer (TCTA). As shown in Fig. S6 (ESI†), the T_1 of TPACFOXZ is 2.5 eV, while the T_1s of TCTA and TPBi are 2.72 and 2.6 eV, respectively. Therefore, the electro-generated triplet excitons can be well confined in the EML. Furthermore, compared to TPACFOXZ, the shallower LUMO energy level of TCTA and the deeper HOMO energy level of TPBi can confine the carriers in the EML. Thus, a series of double-EML white devices with the structure of ITO/PEDOT:PSS (40 nm)/TCTA (20 nm)/Yellow EML/Blue EML/TPBi (30 nm)/LiF (1 nm)/Al (100 nm) were designed and fabricated. Here, the thicknesses of blue EML (TPACFOXZ) are 5, 10, 15, and 20 nm for the devices **W1**, **W2**,

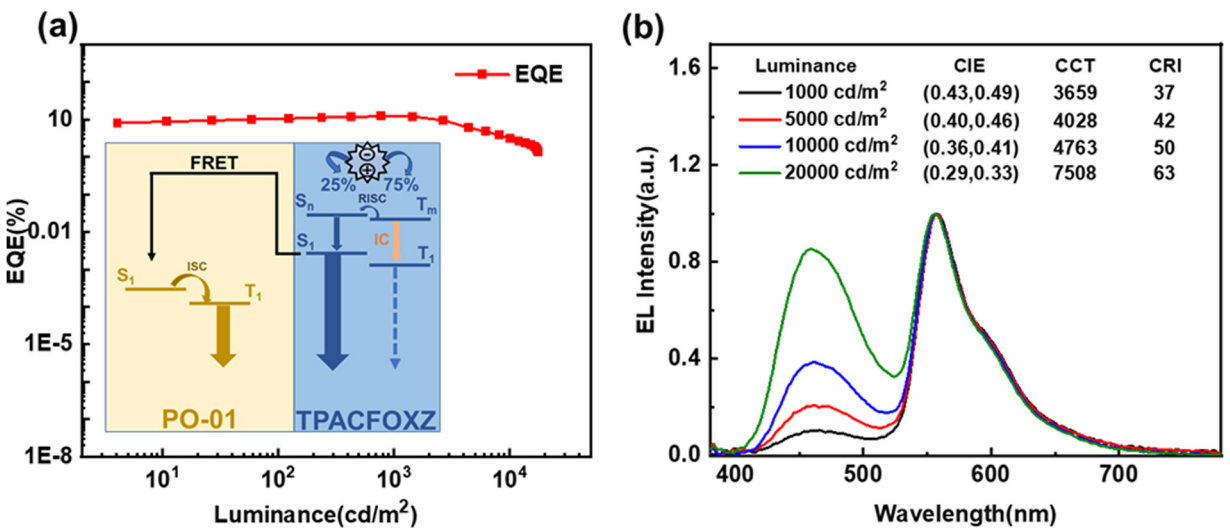

Fig. 3 (a) EQE of the device W and schematic diagram of the luminescence mechanism (inset), where FRET is the Förster energy transfer, IC is the internal conversion, ISC is the intersystem crossing, RISC is the reverse intersystem crossing, and T1 and S1 are the first triplet and singlet states, respectively. (b) EL spectra of device W at different luminance values.

Table 2 Summary of the EL performance of WOLEDs

Devices	V_{on}[a] [V]	CE_{max}[b] [cd A^{-1}]	PE_{max}[c] [lm W^{-1}]	EQE_{max}[d] [%]	L_{max}[e] [cd m^{-2}]	CRI_{1000}[f]	CCT_{1000}[g] [K]	CIE_{1000}[h]
W	2.8	39.5	30.6	12.1	17662	37.2	3659	(0.43, 0.49)
W1	3.8	58.4	41.7	18.0	25386	38.2	3548	(0.44, 0.48)
W2	3.8	63.4	45.3	19.9	29768	41.6	3731	(0.42, 0.47)
W3	3.8	78.8	53.8	27.1	32453	48.7	4181	(0.39, 0.43)
W4	3.8	80.4	54.9	26.8	34753	47.3	3999	(0.39, 0.44)
WR	2.6	48.8	51.7	14.8	34684	39.5	3761	(0.42, 0.48)

[a] V_{on}: Turn-on voltage at 1 cd m^{-2}. [b] CE_{max}: Maximum current efficiency. [c] PE_{max}: Maximum power efficiency. [d] EQE_{max}: Maximum EQE. [e] L_{max}: Maximum luminance. [f] CRI_{1000}: Color rendering index at 1000 cd m^{-2}. [g] CCT_{1000}: Correlated color temperature at 1000 cd m^{-2}. [h] CIE_{1000}: CIE coordinates at 1000 cd m^{-2}.

W3, and **W4**, respectively. Meanwhile, to accelerate the DET process from TPACFOXZ to PO-01 and alleviate the concentration-quenching effect, TPACFOXZ:PO-01 (8%, 10 nm) is chosen as the yellow EML. The device structure, EL performance of the white devices are shown in Fig. 4, and summarized in Table 2. All the devices show the maximum luminance over 25 000 cd m^{-2}, which is higher than that of the single-EML device **W** (17 662 cd m^{-2}). When the thickness of TPACFOXZ is 5 nm, corresponding to the device **W1**, the maximum forward-viewing efficiencies of 58.4 cd A^{-1}, 41.7 lm W^{-1}, and 18.0% are achieved for CE, PE, and EQE, respectively. The EL performance is continually increased with increasing thickness of TPACFOXZ. The best forward-viewing CE, PE, and EQE of 78.8 cd A^{-1}, 53.8 lm W^{-1}, and 27.1% are obtained, respectively, in the device **W3** with the thickness of 15 nm for TPACFOXZ, which is one of the best performances for the HLCT-WOLEDs without using outcoupling enhancement. Assuming the light out-coupling efficiency is 20%–30%, the exciton utilization efficiency is nearly 100%. Moreover, the value of EQE is still as high as 23.8% at the luminance of 1000 cd m^{-2}, showing the low efficiency roll-off. Even when the thickness of TPACFOXZ increases to 20 nm, the EQE of device **W4** just slightly decreases to 26.8%. For comparison, the white device (**WR**) with the configuration of ITO/PEDOT:PSS (40 nm)/TCTA (20 nm)/TPACFOXZ (15 nm)/TPACFOXZ:PO-01 (8%, 10 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm) was fabricated where the yellow EML is located on the side of electron-transporting layer (TPBi). It can be seen from Fig. S5 (ESI†)

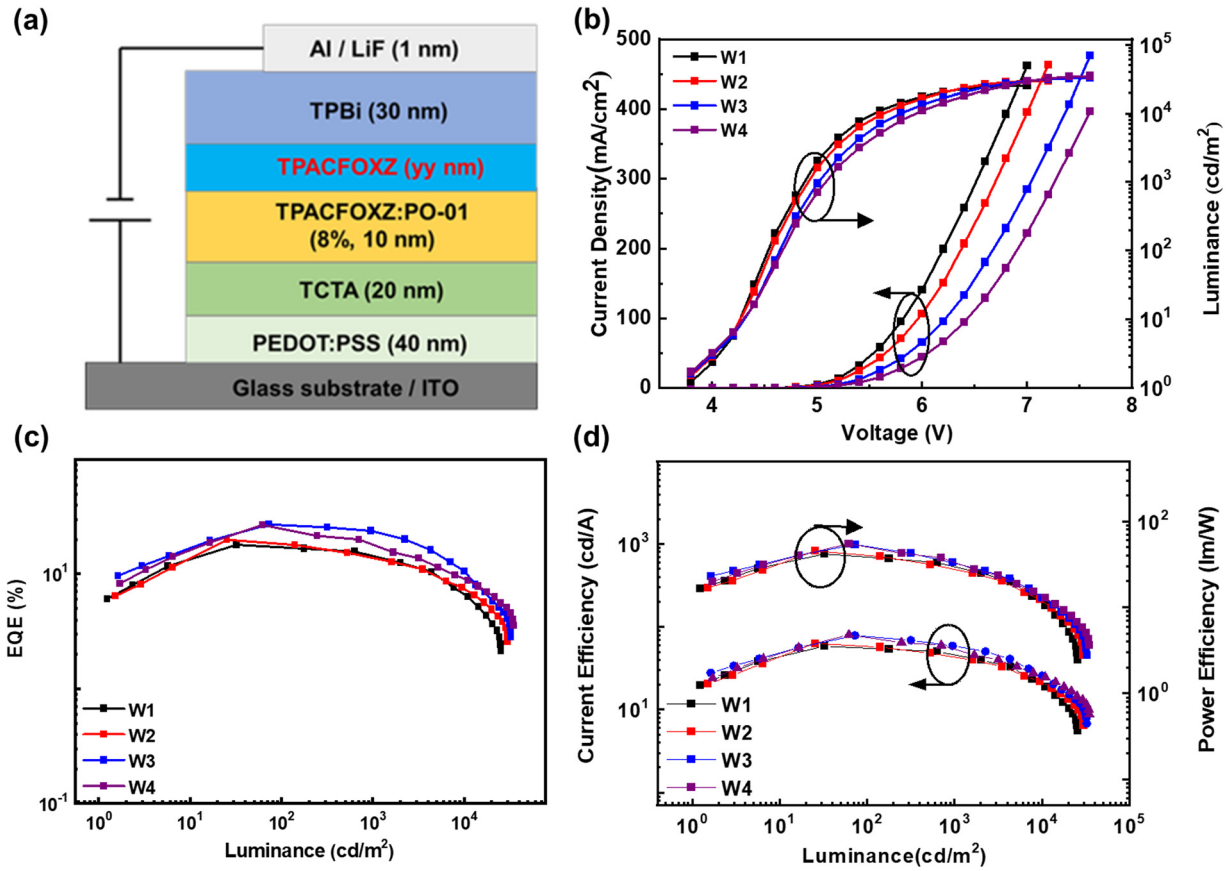

Fig. 4 (a) Device structure of YB-type WOLEDs (**W1**, **W2**, **W3**, and **W4**). (b) Current density-voltage-luminance curves. (c) EQE-luminance curves. (d) Current efficiency-luminance-power efficiency curves.

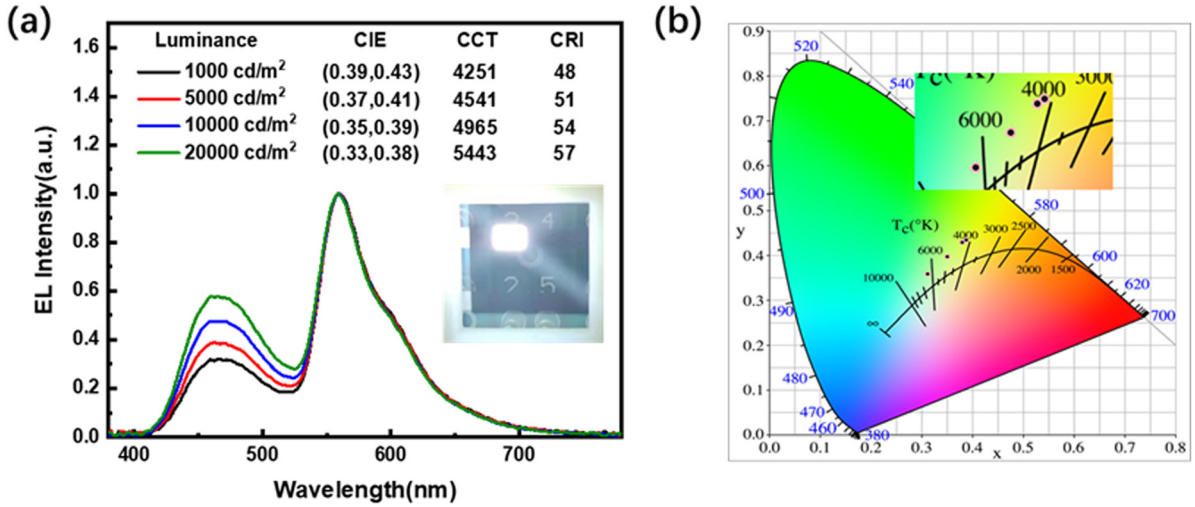

Fig. 5 (a) EL spectra and color coordinates (b) of device **W3** at different luminance values.

that the forward-viewing CE, PE, and EQE values were 48.8 cd A^{-1}, 51.7 lm W^{-1}, and 14.8%, respectively, which is well below that of **W3**. This further illustrates that the precise regulation of EMLs is essential for the development of high-performance HLCT-WOLEDs. Meanwhile, the single-carrier devices with and without doping PO-01 were fabricated based on the EML of device **W3** (Fig. S7, ESI†). Although the weaker carrier-trapping effects exist when doping the dopant PO-01, the hole and electron transporting keep relative balance, which is the key to obtain high performance for WOLEDs of this structure.

Normalized EL spectra of **W1-4** at the luminance from 1000 to 20 000 cd m^{-2} are shown in Fig. 5 and S8. All four devices (**W1–W4**) emit warm-white or white light with the spectra covering the region from 410 to 720 nm. At 1000 cd m^{-2}, the CIE coordinates are (0.44, 0.49), (0.42, 0.47), (0.38, 0.44) and (0.39, 0.44) for devices with the blue EML of 5, 10, 15 and 20 nm, respectively, and the CCTs ranges from 3455 to 4251 K, while the CRIs are as low as 37–48, due to the absence of longer wavelength emission. With increasing thickness of TPACFOXZ, the blue emission is almost increased gradually, indicating that more excitons are used by TPACFOXZ, which can be attributed to the broad exciton recombination zone and effective high-lying RISC process from triplet to singlet states. Moreover, the devices not only show the relatively stable emission spectra (the variation of CIE coordinates is (0.05, 0.05) for device **W3**) within the luminance ranging from 1000 to 20 000 cd m^{-2}, but also have the considerable bule emission component, which differs from previous high-performance two-color hybrid WOLEDs that are usually obtained when most excitons are transferred to high efficiency phosphors that lead to a weak blue emission.[10,29]

Conclusion

In summary, by combining the balanced-transporting blue HLCT material TPACFOXZ with the reasonable arrangement of the emissive layers, a high-performance F/P hybrid white device with higher maximum current efficiency (78.8 cd A^{-1}), maximum power efficiency (53.8 lm W^{-1}), maximum external quantum efficiency (27.1%), and maximum luminance (32453 cd m^{-2}) has been achieved. Compared with previous reports, our study shows that introducing blue HLCT materials can not only achieve reasonably balanced white light, but also realize full utilization of both singlet and triplet excitons at high current density. This further demonstrates the feasibility of WOLEDs with HLCT materials as both host and emitting materials, and provides a reliable path for the preparation of high-performance hybrid WOLEDs.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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