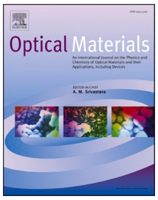
Optical Materials 110 (2020) 110510

Contents lists available at ScienceDirect

Optical Materials

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



Research Article

Triplet collection for highly efficient single-emitting-layer pure fluorescent WOLED based thermally activated delayed fluorescent host of acridine/sulfone derivative

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ARTICLE INFO

Keywords:
mSOAD
RISC
TADF host
Fluorescent OLED

ABSTRACT

Herein, highly efficient single-emitting-layer structure pure fluorescent white organic light-emitting diodes (WOLED) were designed by employing thermally activated delayed fluorescence emitter of mSOAD as host material to sensitize traditional fluorescent dopant of Rubrene and DCJTB, respectively. The WOLED based Rubrene dopant achieve a cool white emission with maximum current efficiency, power efficiency and external quantum efficiency of 33.0 cd/A, 34.5 l m/W and 12.2%, respectively. While a warm white emission with the efficiency of 20.9 cd/A, 20.1 lm/W and 9.0%, respectively, were realized in WOLED based DCJTB dopant. The study to energy level and photoluminescence transient decay lifetime demonstrate the sufficient confinement to charges and excitons, and efficient triplet collection by host triplet exciton reverse intersystem crossing and energy transfer to dopant are the reasons of high device efficiency and stable spectra.

1. Introduction

White organic light-emitting diodes (WOLED) received much more attention due to its huge potential in flexible and natural plane lighting source. In general, the emitters used in WOLED could be divided into traditional fluorescent, phosphorescent and thermally activated delayed fluorescence (TADF) materials. Traditional fluorescent materials possess low cost, high commercialization and stability, while low exciton utilization of only 25% singlet exciton restricts its further application [1,2]. Phosphorescent materials present high exciton utilization of 100% due to spin-orbit coupling of the existent of Ir/Pt heavy metal [3,4], however, high cost, low stability and unsustainable development compels researcher to exploit new emitter materials.

While high efficiency pure fluorescent materials conducted triplet exciton reverse intersystem crossing (RISC) characteristics were exploited successfully by Adachi group in 2012 [5–7], which was called TADF materials. Commonly, small singlet-triplet energy gap (ΔE_{ST}) is achieved in TADF materials through the design of spatially separated molecular donor and acceptor unit, which could promote RISC process of triplet exciton to singlet state energy level. After that, a series of highly efficient TADF materials and exciplex of red, green and blue were developed [8–15]. Besides as the emitter, the host application of TADF materials was also studied by doping phosphorescent and fluorescent emitter into TADF materials, and high efficiency dopant emission were realized through the energy transfer from TADF materials host to phosphorescent/fluorescent dopant.

Fukagawa et al. reported relatively low cost and long operational lifetime phosphorescent OLED by the utilization of TADF materials as the host materials, which employing only 1 wt% emitter concentration exhibits external quantum efficiency (EQE) of over 20% and a long operational lifetime of about 20 times than the reference device [16]. Zhao et al. achieved a high efficiency and low efficiency roll-off green

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https://doi.org/10.1016/j.optmat.2020.110510
Received 26 August 2020; Received in revised form 1 October 2020; Accepted 6 October 2020

OLED based fluorescent and phosphorescent dopant, respectively, by employing blue TADF material as the universal host [17]. Zhang et al. designed highly efficient hybrid WOLED with low roll-off and good color stability by adopting blue TADF material as host and orange phosphorescent material as dopant, while the phosphorescent material conducted high cost and unsustainable development as mentioned above [18]. Wu et al. demonstrated high-performance all fluorescent WOLEDs with EQE of 18.2% by strategic management of singlet and triplet excitons within an efficient emissive zone [19]; however, the design of three emitting layers (EMLs) also increased the device complexity. Therefore, although the monochrome OLED based TADF materials as host had exhibited some advantages, while the WOLED with TADF materials as host still need further studied, and high efficiency, low cost and simple structure WOLED need be exploited. We consider the combination of TADF materials as host and traditional fluorescent emitter as dopant to construct simple structure pure fluorescent WOLED is an efficient pathway in the future.

In this work, highly efficient single-emitting-layer structure pure fluorescent WOLED were realized successfully by employing blue TADF materials of bis(3-(9,9-dimethyl-9,10-dihydroacridine)phenyl)sulfone (mSOAD) as host and orange/red traditional fluorescent emitters of (5,6,11,12)-tetraphenyl-naphthacene (Rubrene)/4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) as dopant, respectively. The WOLED based Rubrene achieved the maximum EQE of 12.2% with cool white emission of 7747 K and (0.26, 0.43) at 6 V, while the warm white emission of 3624 K and (0.41, 0.41) at 6 V, maximum EQE of 9.0% were obtained in WOLED with DCJTB as dopant; meantime, high spectra stability was also achieved in the pure fluorescent WOLED. The high device efficiency and stable spectra could be attributed to efficiently host triplet exciton RISC and high energy transfer efficiency to dopant by Förster process.

2. Experimental section

All the WOLEDs were fabricated on Indium tin oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω/sq. The ITO substrates were cleaned first with acetone, deionized water, acetone sequential and then treated by ultraviolet-ozone for 15 min, after that the ITO substrates were loaded into high vacuum chamber ($\sim 4 \times 10^{-4}$ Pa) for subsequent deposition. The organic layers were deposited at a rate of 1.0 Å/s. The inorganic layers of MoO_3 and LiF at the deposition rate of 0.1 Å/s. Al cathode was deposited in the end with a shadow mask, which defined the device active area of 3×3 mm^2. The UV–Vis absorption spectrum was recorded with Hitachi U-3900 scanning spectrophotometer. The photoluminescence (PL) spectra were measured with 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. Electroluminescence (EL) spectra were measured through PR-655 spectra scan spectrometer with computer controlled. The current-voltage-luminance curves were measured with Keithley 2400 power supply combined with BM-7A luminance colorimeter. EQE was calculated from the EL spectra data and current density-voltage-luminance curve. All the organic materials were procured commercially without further purification except mSOAD, which was synthesized in our own laboratory. All the measurements were carried out at room temperature and under ambient conditions without any protective coatings.

3. Results and discussion

The single-emitting-layer structure WOLED is adopted complementary colors emission of blue and orange, which derived from host and dopant, respectively. The blue TADF host materials used in this work is mSOAD based acridine/sulfone derivative, which is synthesized in our laboratory. The detail of synthesis, optical physics, TADF characteristics and EL performance had been reported in other journals [20,21], so the host application of mSOAD is the focus here. The molecular design of donor and acceptor units in mSOAD shown in Fig. 1a indicates potential charge transport bipolarity and the hole-/electron-only device confirms the excellent bipolar transport ability of holes and electrons [22]. The conventional orange/red fluorescent emitters of Rubrene/DCJTB are selected as the dopant due to its high stability and low cost. Besides, the PL spectra of Rubrene and DCJTB shown in Fig. 1b present orange and red emission, which indicates that the white emission could be achieved by the simultaneous emission of blue host and orange/red dopant. Furthermore, the PL spectrum of mSOAD and absorption spectra of Rubrene/DCJTB are also exhibited in Fig. 1b to study the energy transfer process. A large overlap between host of mSOAD and dopants of Rubrene/DCJTB could be observed, which indicate highly efficient Förster energy transfer process could happen between singlet state energy level of mSOAD and Rubrene/DCJTB [23,24]. Therefore, traditional orange/red fluorescent emitters of Rubrene/DCJTB are adopted as dopant to construct white emission with the advantages of suitable spectra, high stability, low cost and high energy transfer efficiency from mSOAD host, which may break the 5% EQE upper limit of traditional fluorescent WOLED by the efficient collection and utilization of triplet excitons.

Based on the previous work, we first designed the WOLED structure based Rubrene dopant as follows: ITO/MoO_3 (3 nm)/mCP (25 nm)/mSOAD: x wt% Rubrene (20 nm)/DPEPO (5 nm)/TPBi (40 nm)/LiF (1 nm)/Al, where x = 0.4, 0.8 and 1.2, and the corresponding WOLED is defined as $W_{0.4}$, $W_{0.8}$ and $W_{1.2}$, respectively. The device structure is also showed in Fig. 2a, where *m*-bis(*N*-carba-zolyl)benzene (mCP) and 1,3,5-tris(*N*-phenyl-benzimidazol-2-yl)ben-zene (TPBi) is the hole and electron transport layer, respectively. mSOAD: x wt% Rubrene is the EML and bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO) is the exciton block layer to confine all the exciton in EML. The white emission would be constructed from blue host of mSOAD and orange dopant of Rubrene, while the emission intensity of blue and orange could be also adjusted by different doping concentrations. The WOLED spectra of $W_{0.4}$ to $W_{1.2}$ under various voltages are displayed in Fig. 2b–d. As the EL spectra show, the emission of host blue and dopant orange are all obtained from $W_{0.4}$ to $W_{1.2}$, and orange emission intensity enhanced with the increase of dopant concentration, which declare the efficient energy transfer from mSOAD to Rubrene. Besides, the little change of spectra under different voltages suggests relative stable white spectra were obtained. The WOLED of $W_{0.4}$ present a cool white emission with correlated color temperature (CCT) of 7747 K and Commission Internationale de l'Eclairage (CIE) coordinates of (0.26, 0.43) at 6 V, while the device of $W_{1.2}$ with high concentration exhibit a warm white emission with CCT of 3341 K and CIE coordinates of (0.45, 0.48) at 6 V. That means the white emission point could be modulated easily through the doping concentration optimization.

The WOLED efficiency curves of current efficiency, power efficiency and EQE under different doping concentrations are showed in Fig. 3. All the three pure fluorescent WOLED achieved a low turn-on voltage of below 3 V, which indicates the efficient charges injection and recombination. And the maximum efficiency were obtained in $W_{0.4}$ with maximum current efficiency, power efficiency and EQE of 33.0 cd/A, 34.5 lm/W and 12.2%, respectively. While the efficiency in $W_{0.8}$ were 28.0 cd/A, 29.0 lm/W and 9.8%, a relative low efficiency but above 5% EQE were also realized in $W_{1.2}$ with maximum current efficiency, power efficiency and EQE of 15.5 cd/A, 15.8 lm/W and 5.2%, respectively. The high efficiency of pure fluorescent WOLED could be attributed to efficient charge injection/recombination, host triplet excitons efficient RISC process and high energy transfer efficiency from mSOAD to dopant [25–27]. The detailed discussion would be given below.

From the device energy level diagram shown in Fig. 3d, we can see that there almost have no injection barriers from transport layer to EML due to the deeper highest occupied molecular orbital (HOMO) energy level of mCP (6.1 eV) than mSOAD (5.8 eV) for holes and only 0.1 eV

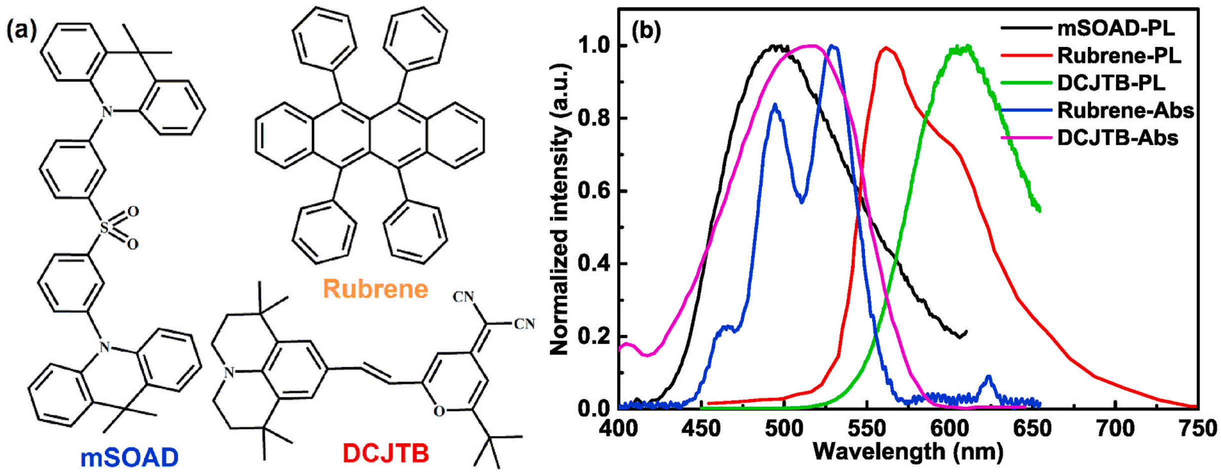

Fig. 1. The molecular structure, PL and absorption spectra. (a) Molecular structure of mSOAD, Rubrene and DCJTB. (b) PL spectra of mSOAD, Rubrene and DCJTB, absorption spectra of Rubrene and DCJTB.

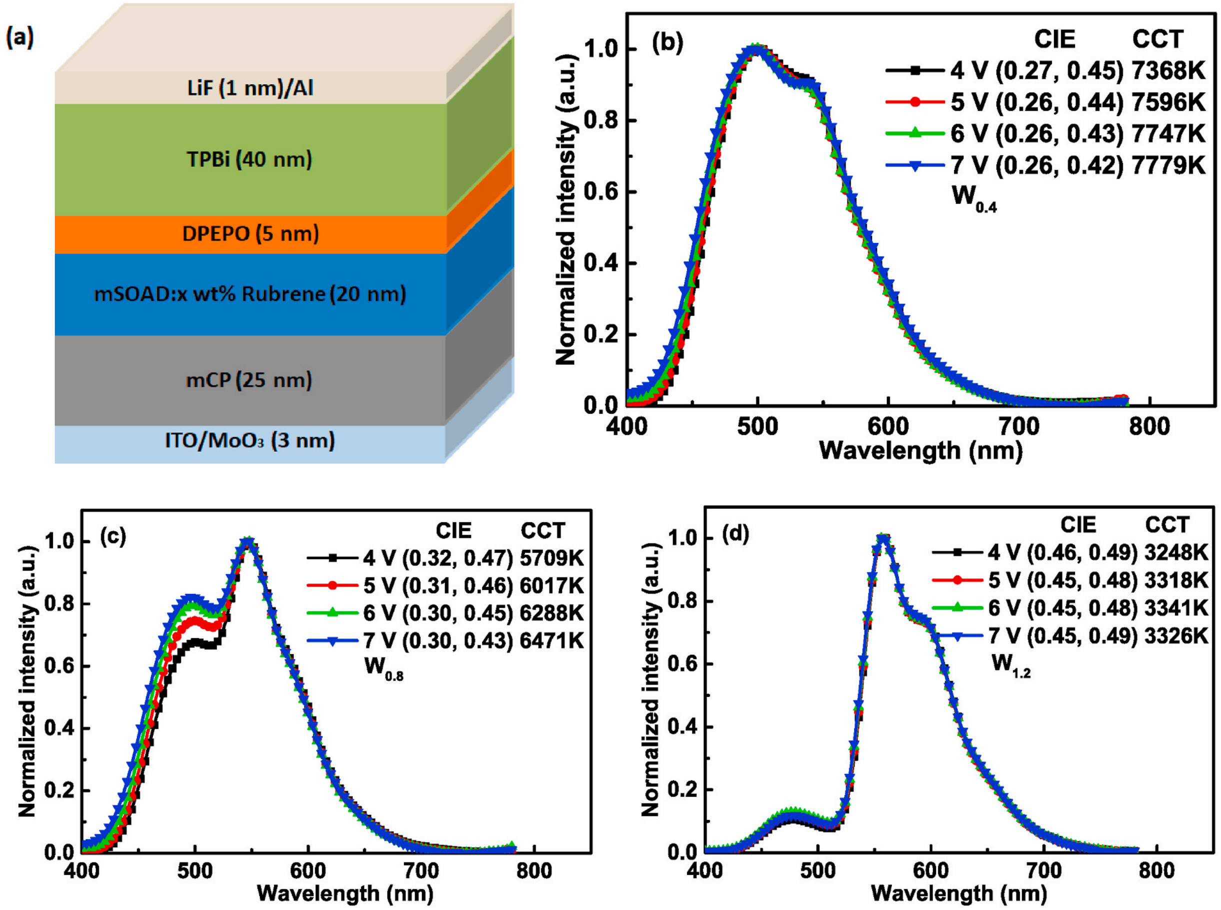

Fig. 2. The device structure and EL spectra of pure fluorescent WOLED based Rubrene dopant. (a) Device structure diagram. (b) EL spectra of $W_{0.4}$. (c) EL spectra of $W_{0.8}$. (d) EL spectra of $W_{1.2}$.

barrier in lowest unoccupied molecular orbital (LUMO) energy level from TPBi (2.7 eV) to DPEPO (2.6 eV) for electrons. And the low turn-on voltage of below 3 V is benefited from the barrier-free charge injection. Besides, the shallow LUMO energy level of mCP (2.4 eV) and deep HOMO energy level DPEPO (6.8 eV) could confine electrons and holes efficiently in mSOAD host, respectively, which also improve the charge recombination efficiency in EML. Except for charge injection, transport and recombination in OLED, the exciton behavior produced in EML could also influence the device performance seriously. Under electric excitation, 25% singlet excitons and 75% triplet excitons are formed on mSOAD host firstly. While the higher singlet and triplet excitons energy of mCP ($S_1 = 3.5$ eV, $T_1 = 3.0$ eV) and DPEPO ($S_1 = 3.94$ eV, $T_1 = 2.98$ eV) [28–30] than mSOAD ($S_1 = 2.92$, $T_1 = 2.9$ eV) could confine the excitons in EML efficiently and prevent the excitons migration to adjacent hole or electron transport layer. Therefore, the excitons in mSOAD host could be transferred to dopant efficiently and improve the device

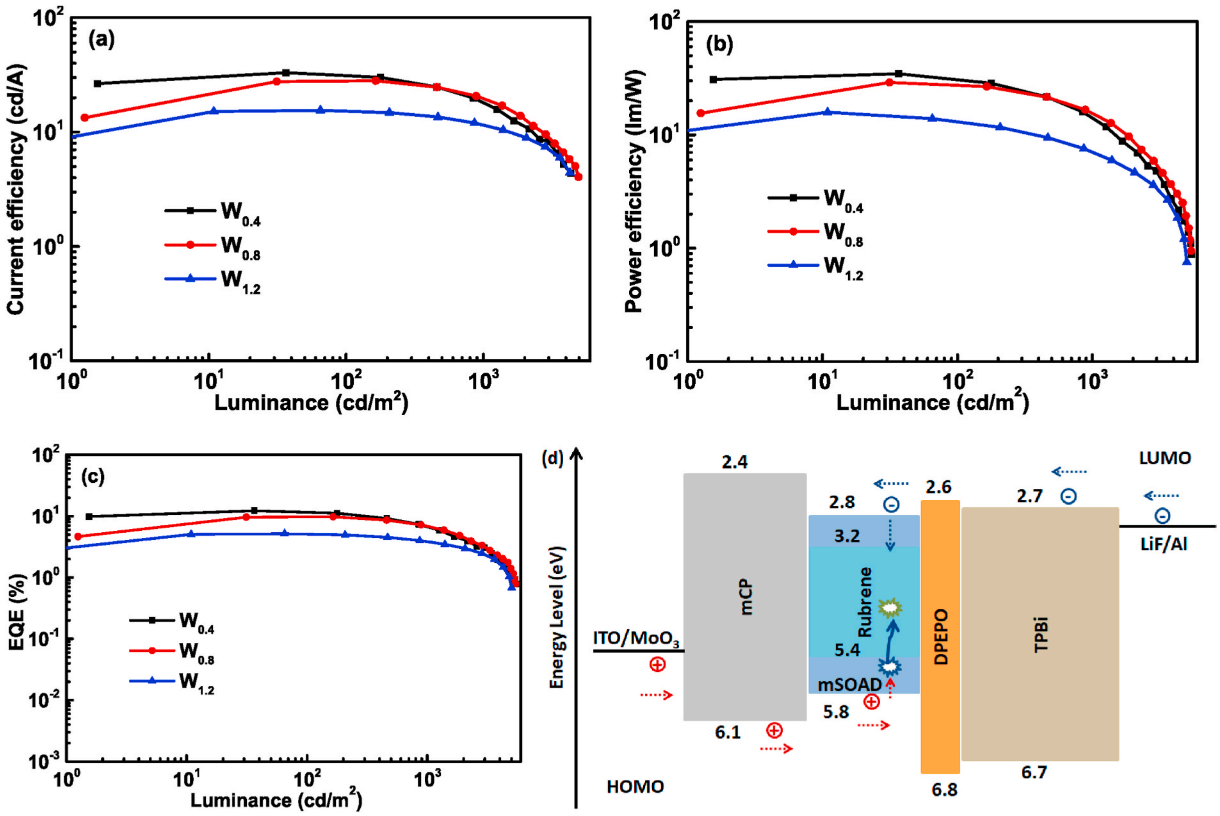

Fig. 3. The EL efficiency curves of pure fluorescent WOLED based Rubrene dopant and energy level diagram. (a) Current efficiency-Luminance curves. (b) Power efficiency-Luminance curves. (c) EQE-Luminance curves. (d) Device energy level diagram.

efficiency. The EL performances of all the WOLED in this paper are listed in Table 1.

In order to understand the energy transfer and luminance process in-depth, the steady PL spectra and transient PL decay behavior of mSOAD: x wt% Rubrene (x = 0.3, 0.5 and 0.8) films are studied in Fig. 4. As Fig. 4a shows, the emission intensity of Rubrene enhances as the concentration increases, which demonstrates again the energy transfer from mSOAD host to Rubrene dopant. Fig. 4b–c exhibits the transient PL decay curves with the monitor peaks of 485 nm and 545 nm, which is the emission peak of blue host of mSOAD and orange dopant of Rubrene, respectively. As expected, the TADF host emission of mSOAD exhibits prompt and delayed two fluorescent components. But to our surprised, the emission of Rubrene that doped into mSOAD exhibit similar behavior and also has two components of prompt and delayed fluorescence. We can also see that the lifetime decreases gradually as the increase of Rubrene concentration. Under optical excitation, the triplet excitons of mSOAD are produced by efficient intersystem crossing (ISC) process, and the delayed component of Rubrene is resulted from the energy transfer of up-converted host triplet excitons due to the small ΔE_{ST} of mSOAD. As the concentration increases, the ISC efficiency of mSOAD reduces and the RISC efficiency also reduces, which result in the delayed lifetime decreases. So under electric excitation, the 25% singlet excitons produced on mSOAD host would transfer to the singlet state of Rubrene by Förster process and the 75% triplet excitons would up-convert to singlet state through efficient RISC, then the new singlet excitons formed through up-conversion would also transfer to the singlet state of Rubrene. Therefore, high efficiency Rubrene emission could be achieved with efficient triplet excitons collection. While the Dexter energy transfer between host-guest triplet state energy level and direct charge trapping by Rubrene dopant could be suppressed due to the low doping concentration [16,18]. The energy transfer process schematic diagram discussed above is displayed in Fig. 4d. Hence, based on the discussion above, the consistent transient decay behavior of mSOAD host and Rubrene confirms the emission of Rubrene is derived from the Förster energy transfer of mSOAD host singlet excitons and up-converted triplet excitons.

Finally, we also conduct another traditional fluorescent emitter of DCJTB as dopant to construct the pure fluorescent WOLED by employing the same device structure mentioned above. So the EML based DCJTB device is mSOAD: x wt% DCJTB, where the x = 0.6 and 1.2, and the

Table 1
A summary EL performances of all the WOLED in this paper.

WOLED	Turn-on Voltage (V)	[a]CE$_{\mathrm{Max.n}}$ (cd/A)	[b]PE$_{\mathrm{Max.}}$ (lm/W)	[c]EQE$_{\mathrm{Max.}}$ (%)	CIE at 6 V	CCT at 6 V (K)
W$_{0.4}$	2.7	33.0	34.5	12.2	(0.26, 0.43)	7747
W$_{0.8}$	2.7	28.0	29.0	9.8	(0.30, 0.45)	6288
W$_{1.2}$	2.9	15.5	15.8	5.2	(0.45, 0.48)	3341
W1$_{0.6}$	2.9	20.9	20.1	9.0	(0.41, 0.41)	3624
W1$_{1.2}$	2.9	11.0	11.1	5.2	(0.52, 0.41)	2035

[a] Maximum current efficiency.
[b] Maximum power efficiency.
[c] Maximum EQE.

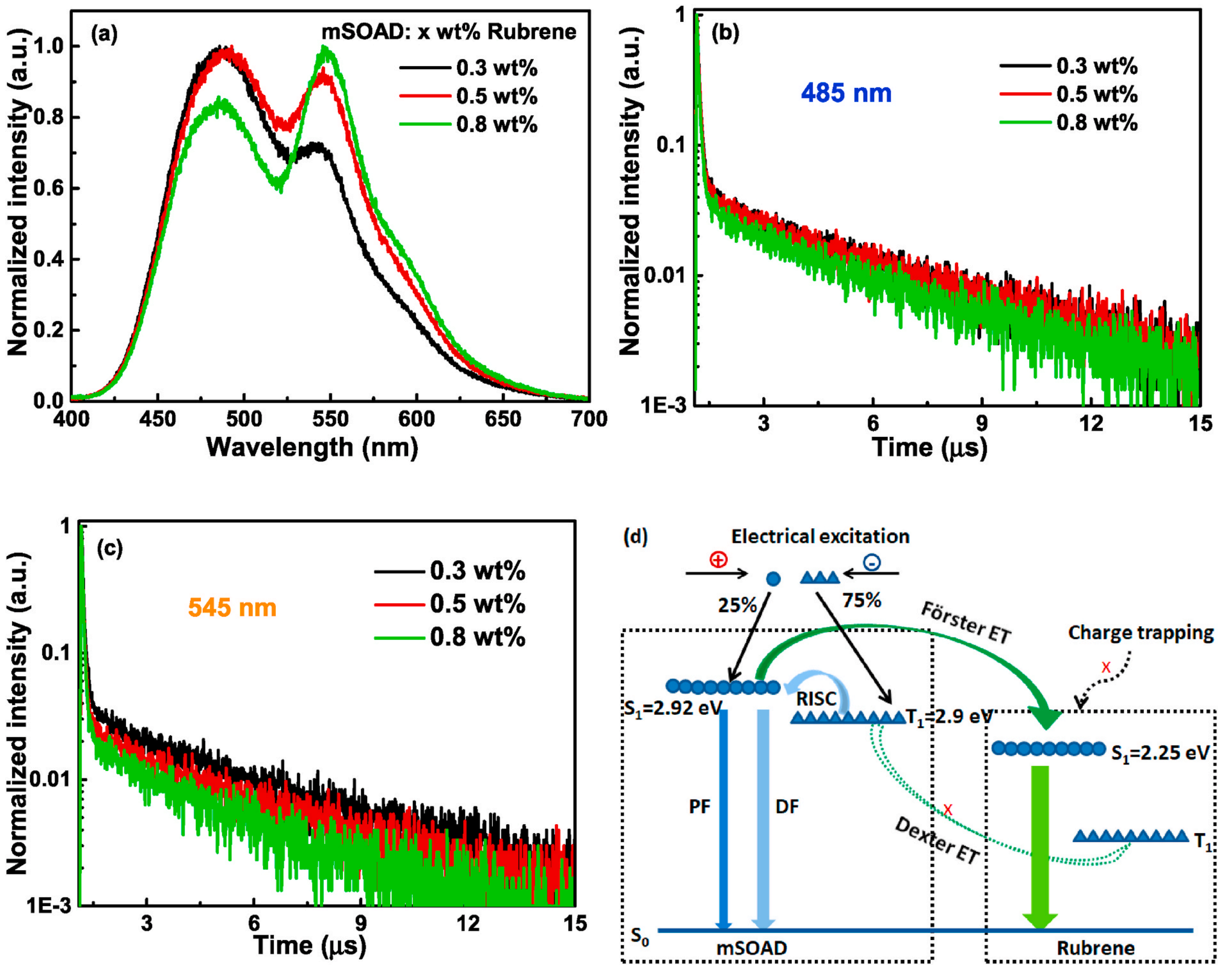

Fig. 4. The steady PL spectra and transient PL decay characteristics with different concentrations Rubrene in mSOAD films, and energy transfer schematic diagram. (a) Steady PL spectra. (b) PL transient decay behavior observed at 485 nm. (c) PL transient decay behavior observed at 545 nm. (d) Energy transfer process schematic diagram.

corresponding device is defined as $W1_{0.6}$ and $W1_{1.2}$, respectively. The device performances of efficiency and spectra are showed in Fig. 5. A high efficiencies are also achieved in $W1_{0.6}$ with maximum current efficiency, power efficiency and EQE of 20.9 cd/A, 20.1 l m/W and 9.0%, respectively. The EQE of 9.0% also exceed the fluorescent quantum upper limit of 5%. The transient PL decay characteristics of mSOAD: x wt % DCJTB film showed in Fig. 5c–d presents the similar behavior curves with mSOAD: x wt% Rubrene film, which demonstrates the same energy transfer and luminance process. We also consider the sufficient collection for triplet excitons through host triplet excitons RISC and efficient Förster energy transfer are responsible for the high device efficiency. While an above 5% EQE efficiency level was also obtained in $W1_{1.2}$, with maximum current efficiency, power efficiency and EQE of 11.0 cd/A, 11.1 l m/W and 5.2%, respectively. Besides the high device efficiency with optimized concentration of 0.6 wt%, excellent white EL spectra shown in Fig. 5e is also achieved in $W1_{0.6}$. From 5 V to 8 V, the spectra have little change with CIE coordinates of (0.41, 0.41) and CCT of ~3600 K, which is a warm white emission. However, the increased dopant concentration induces strong DCJTB emission due to more energy transfer from mSOAD to DCJTB, which results in weak host blue emission in $W1_{1.2}$. But although the same host material of mSOAD is adopted to fabricate WOLED, the maximum EQE of Rubrene device is 12.2%, which is higher than DCJTB device of 9.0%. We consider the different maximum EQE is derived from the different PL efficiency because of the same host material and similar overlap degree between host PL spectrum and guest absorption spectrum, which indicate the comparative energy transfer efficiency. So the PL efficiency becomes the main difference. Anyway, the TADF emitter of mSOAD could play the role of host material to sensitize traditional fluorescent dopant, which could break the 5% EQE upper limit through the harvest of triplet excitons by RISC process.

4. Conclusion

In summary, the highly efficient pure fluorescent WOLED with simple single-emitting-layer structure were realized by the application of blue TADF emitter of mSOAD as host material and traditional fluorescent emitter of Rubrene and DCJTB as dopant, respectively. The pure fluorescent WOLED based Rubrene dopant achieved the maximum EQE of 12.2% and cool white emission with CCT of 7747 K and CIE coordinates of (0.26, 0.43) at 6 V. While a warm white emission with CCT of 3624 K, CIE coordinates of (0.41, 0.41) and maximum EQE of 9.0% were obtained with DCJTB dopant. The PL transient decay measurement demonstrates the harvest to triplet excitons through TADF molecule RISC process and efficient Förster energy transfer play the main role to high device efficiency. The host role enlarge TADF molecule application and we also believe the TADF molecule could act more function in high efficiency pure fluorescent WOLED in the future.

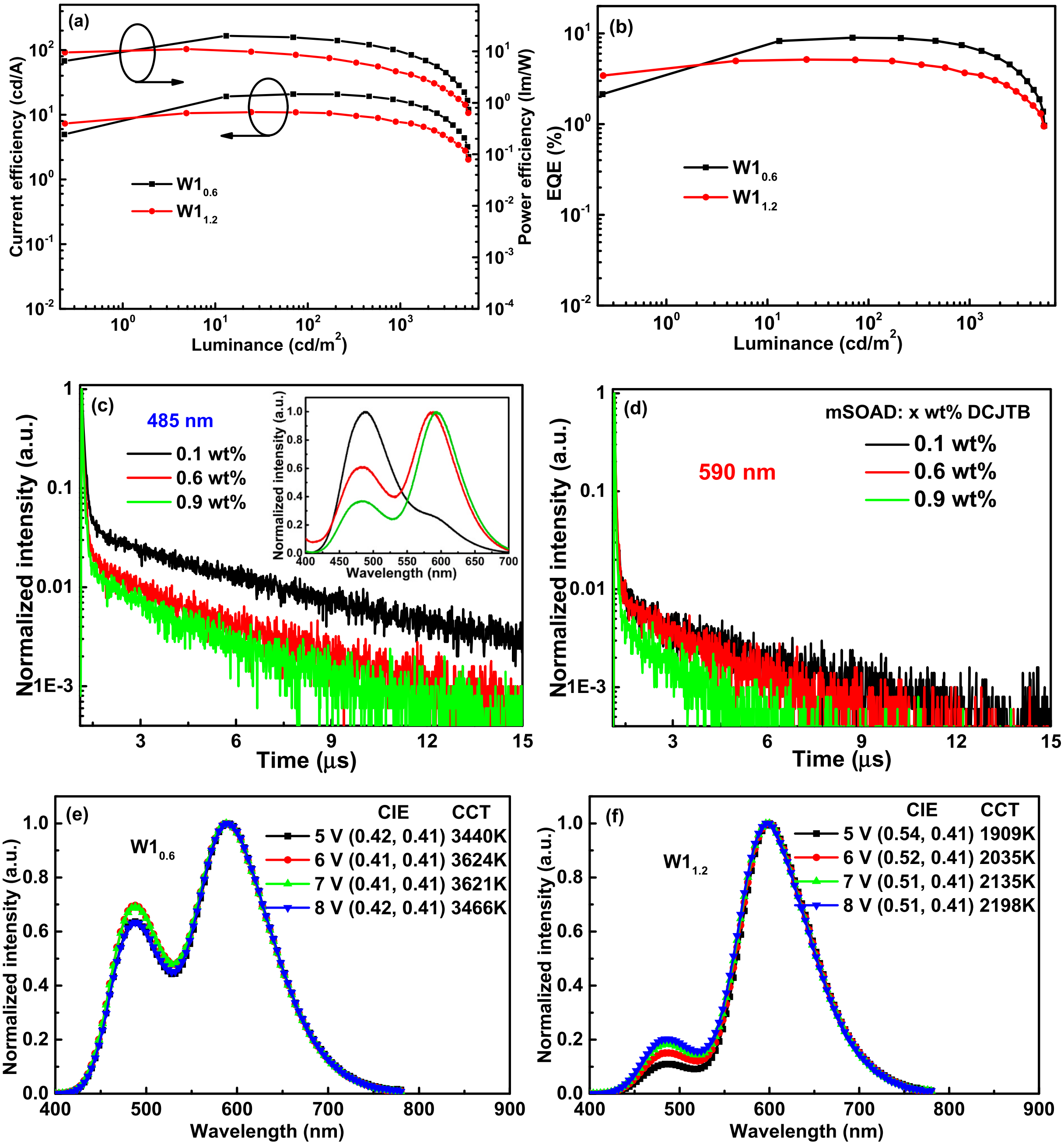

Fig. 5. The EL performances of pure fluorescent WOLED based DCJTB dopant and transient PL decay characteristics with different concentrations DCJTB in mSOAD films. (a) Current efficiency-Luminance-Power efficiency curves. (b) EQE-Luminance curves. (c) PL transient decay behavior observed at 485 nm. Inset is the steady PL spectra. (d) PL transient decay behavior observed at 590 nm. (e) EL spectra of W1$_{0.6}$. (f) EL spectra of W1$_{1.2}$.

Author contributions statement

L.Z.M. wrote the main manuscript and carried out most of the experiments and data analysis. D.B.Z. and X.Y.B. helped measure and collect the experimental data. Z.B. and L.J. guided the progress of experiments and manuscript writing. G.Y., C.H.F., W.H. and L.W.L. took part in the mechanism discussions. All authors reviewed the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was financially supported by National Natural Science Foundation of China (61605137 and 61775155), the publication fee by the Science & Technology Research Program of Chongqing University of Arts and Sciences (R2018SJG21 and R2018SDQ15); the Scientific and Technological Innovation Programs of Higher Education Institutions in Shanxi (STIP, 2020–104).

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