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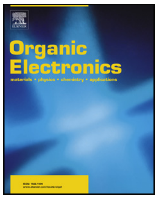

Structural design for highly efficient pure fluorescent warm WOLEDs by employing TADF molecule as blue emitter and exciplex donor

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ABSTRACT

In this work, highly efficient pure fluorescent white organic light-emitting diodes (WOLEDs) are realized through the structural design by employing thermally activated delayed fluorescence (TADF) molecule of mSOAD as blue emitter and exciplex donor, respectively. The combination of mSOAD: PO-T2T exhibits yellow-green exciplex emission with high EQE of 9.7%. Based on the application of this exciplex as the host, the high efficiency WOLEDs with current efficiency, power efficiency and external quantum efficiency (EQE) of 30.2 cd A^{-1}, 28.0 lm W^{-1} and 11.6%, respectively, are achieved finally. Meantime, stable warm white emission spectra with Commission Internationale de l'Eclairage (CIE) coordinates change as low as (0.030, 0.008) from 5 V to 8 V are also realized. The reasonable structural design and double functions application of TADF molecule are responsible for the high performances fluorescent WOLEDs.

1. Introduction

White organic light-emitting diodes (WOLEDs) have attracted much attention in solid-state lighting areas as the development and application of OLEDs in panel displays. In general, highly efficient WOLEDs are combined with hybrid fluorescent-phosphorescent emitters or full phosphorescent emitters [1–4]. However, although phosphorescent emitters could achieve high efficiency based the radiative transition of triplet excitons, but the heavy metal (Ir or Pt) in phosphorescent emitters also cause the problem of high cost and resource scarcity. While WOLEDs based traditional pure fluorescent emitters could only achieve maximum 25% internal quantum efficiency (IQE) due to the non-radiative transition of triplet excitons. Therefore, the exploitation to highly efficient pure fluorescent WOLEDs without phosphorescent emitters become very important.

In order to break the IQE limit of fluorescent emitters, the thermally activated delayed fluorescence (TADF) mechanism was proposed, and almost 100% IQE have been realized in monochrome fluorescent OLEDs by employing TADF emitters [5–7]. The key to TADF mechanism is the triplet excitons reverse intersystem crossing (RISC) process occurred from triplet excited state (T_1) to singlet excited state (S_1) to achieve the utilization of triplet excitons, then the followed singlet excitons radiative transition or energy transfer contribute to the improved photoluminescence (PL) or electroluminescence (EL) efficiency. TADF process could occur on one molecule of TADF emitter, but also could occur on exciplex. Exciplex is formed from the charge transfer between two molecules of donor and acceptor, and the OLEDs based exciplex emission could achieve high external quantum efficiency (EQE) of ~15% [8,9]. Although the high efficiency monochrome fluorescent OLEDs have been earned from the TADF emitters or exciplex emission by efficient triplet excitions RISC process, but the pure fluorescent WOLEDs that need precise structure design still face huge challenge even if there have some fluorescent WOLEDs were reported [10–12]. Full exciplexes with blue and orange emission were utilized to construct the fluorescent WOLEDs by tandem or spacers structure [13–15]. The TADF emitters with red, green and blue were also applied to fabricate the fluorescent WOLEDs by different device structures [16,17]. But the combination of TADF emitter and exciplex to achieve pure fluorescent WOLEDs is rare [18]. Therefore, we consider combining with the TADF emitter and exciplex to gain the highly efficient pure fluorescent WOLEDs through the reasonable device structure design.

In this manuscript, a new exciplex with yellow-green emission are obtained first by employing TADF emitter of bis(3-(9,9-dimethyl-9,10-dihydroacridine)phenyl)sulfone (mSOAD) as the donor, then followed a

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novel structure are designed for highly efficient pure fluorescent WOLEDs by utilizing mSOAD as the blue emitter and exciplex donor, respectively. The resulting pure fluorescent WOLEDs possess high current efficiency, power efficiency and EQE of 30.2 cd A^{-1}, 28.0 lm W^{-1} and 11.6%, respectively. Meantime, high spectral stability are also realized with the Commission Internationale de l'Eclairage (CIE) coordinates change as low as (0.030, 0.008) from 5 V to 8 V. Such a high performance pure fluorescence WOLEDs demonstrate the potential of novel strategy that double functions application of TADF molecule.

2. Experimental section

2.1. Materials preparation and device fabrication

Apart from the mSOAD synthesized in our laboratory, the other materials used in this manuscript were purchased from commercial sources and used without further purification. All the OLEDs were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω per square. 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 at a rate of 5.0 Å/s with a shadow mask, which defined the device area of $3 \times 3\,mm^2$.

2.2. Characterization

The 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. The 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 current density-voltage-luminance curve and spectra data. Besides, all the measurements were carried out at room temperature and under ambient conditions without any protective coatings.

3. Results and discussion

3.1. Yellow-green exciplex

The mSOAD is a highly efficient blue fluorescent emitter conducted TADF characteristics with emission peak around ~490 nm, which is designed and synthesized by our group. The relevant synthetic route, and other thermal, optical and EL characterization data had been described in other papers [19,20]. In the previous studies, the EQE of mSOAD could reach to 14% in the non-doped blue OLEDs with the device structure as follows: ITO/MoO_3 (3 nm)/mCP (25 nm)/mSOAD (20 nm)/DPEPO (5 nm)/TPBi (40 nm)/LiF (1 nm)/Al. Here we find mSOAD is not only a TADF emitter, but also could act as a donor molecule to form the exciplex. The molecules structure and PL spectra of mSOAD (donor), (1,3,5-triazine-2,4,6-triyl)tris(benzene-3,1-diyl))tris (diphenylphosphine oxide (PO-T2T) (acceptor) and mSOAD: PO-T2T mixed films are showed in Fig. 1a and b. As the Fig. 1b shows, the PL spectra of mSOAD: PO-T2T mixed film has an obvious red shift fixing emission peak position at 526 nm, which is different from the PL of donor mSOAD and acceptor PO-T2T. The appearance of the new emission peak with mixed film indicates the formation of exciplex between mSOAD and PO-T2T. The emission peak value energy of 2.6 eV is similar to the energy gap between the highest occupied molecular orbital (HOMO) of mSOAD (5.8 eV) and lowest unoccupied molecular orbital (LUMO) of PO-T2T (3.5 eV), which also demonstrates the formation of exciplex between mSOAD and PO-T2T. Besides, the typical two PL lifetimes of prompt and delayed under 526 nm with mSOAD: PO-T2T mixed film by the measurement of PL transient decay property present the TADF characteristic of the exciplex, which are showed in Fig. S1. We consider the TADF materials that could be acted as donor in exciplex system should conduct the suitable HOMO/LUMO energy level to form the exciplex with acceptor materials, high singlet and triplet excited state energy to confine exciplex excitons.

To study the EL performance of this novel exciplex, we fabricated the OLEDs defined as Device A with the structure as follows: ITO/MoO_3 (5 nm)/mCP (30 nm)/mSOAD: PO-T2T (1:1, 20 nm)/PO-T2T (60 nm)/LiF (1 nm)/Al and the EL results are showed in Fig. 1c and d. The low turn-on voltage of 2.3 V and high luminance of 25000 cd m^{-2} are realized with emission peak of 540 nm. The slight difference between the emission peak position of PL and EL may derive from the different formation process of exciplex under optical and electrical excitation [21–23]. Meanwhile the EL spectra of the exciplex exhibit a large coverage from 450 to 700 nm and the full width at half maximum (FWHM) is as high as ~100 nm, which result to a yellow-green emission with CIE coordinates of (0.36, 0.56). Furthermore, the maximum efficiencies could reach up to 31.4 cd A^{-1}, 36.6 lm W^{-1} and 9.7% for current efficiency, power efficiency and EQE, respectively. The wide spectral coverage and high efficiency indicate the WOLEDs could be constructed based the exciplex. So next we combine with the highly efficient exciplex of yellow-green emission and mSOAD of blue emission to design the pure fluorescent WOLEDs.

3.2. Pure fluorescence WOLEDs

The exciplex of mSOAD: PO-T2T exhibits very wide spectral coverage from 450 nm to 700 nm, so we consider the warm white emission could be earned by combining with the exciplex and red/blue emitter. Exciplex as the host to sensitize the dopant have been applied in OLEDs due to the advantages of charge barrier-free injection, wide recombination zone and efficient energy transfer [24–28]. So first we construct the WOLEDs by introducing conventional red fluorescent material of 4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) as the dopant to mix into the exciplex. The device structure designed as follows: ITO/MoO_3 (5 nm)/mCP (30 nm)/mSOAD: PO-T2T (1: 1): x wt% DCJTB (30 nm)/PO-T2T (60 nm)/LiF (1 nm)/Al. The x is 0.3, 0.8 and 1.3, which are denoted as Device B, C and D, respectively. As results, high efficiency and warm white emission OLEDs are obtained by controlling the concentration of DCJTB in exciplex host. The EL performances of the WOLEDs with different concentrations DCJTB are showed in Fig. 2.

The EL spectra of the WOLEDs with different concentrations DCJTB, showed in Fig. 2b, exhibit the emissions of exciplex host and DCJTB dopant. Meanwhile, the emission intensity of exciplex host decreases as the increase of DCJTB concentrations, which demonstrate the efficient energy transfer from the exciplex host to dopant. The CIE coordinates of Device B to D under 6 V are (0.369, 0.525), (0.444, 0.494) and (0.478, 0.482), present the warm white emission. Besides, the stable white emission EL spectra are also realized, that means the spectra show little change under the various voltages, which are displayed in Fig. S2. The slightly red shift of EL spectra with increased concentrations shown in Fig. 2b can be regarded as the strong intermolecular interaction between host and dopant. The current density-voltage-luminance curves and current efficiency-current density-EQE curves are showed in Fig. 2c and d. All the WOLEDs of Device B to D show low turn-on voltage of 2.7 V and high luminance of > 10000 cd m^{-2}, which could be attributed to the barrier-free charge injection/transport from electrode to emitting layer (EML) and efficient excitons recombination. And the high efficiencies are also obtained in the pure fluorescent WOLEDs. The current efficiencies, power efficiencies and EQEs of Device B to D are 28.5 cd A^{-1}/27.6 lm W^{-1}/9.4%, 26.8 cd A^{-1}/25.5 lm W^{-1}/9.5% and 21.4 cd A^{-1}/18.3 lm W^{-1}/8.4%, respectively. The high charges recombination efficiency, triplet excitons RISC efficiency from T_1 to S_1 energy level on exciplex host and high energy transfer efficiency from exciplex host to DCJTB dopant are the main reasons resulting in high

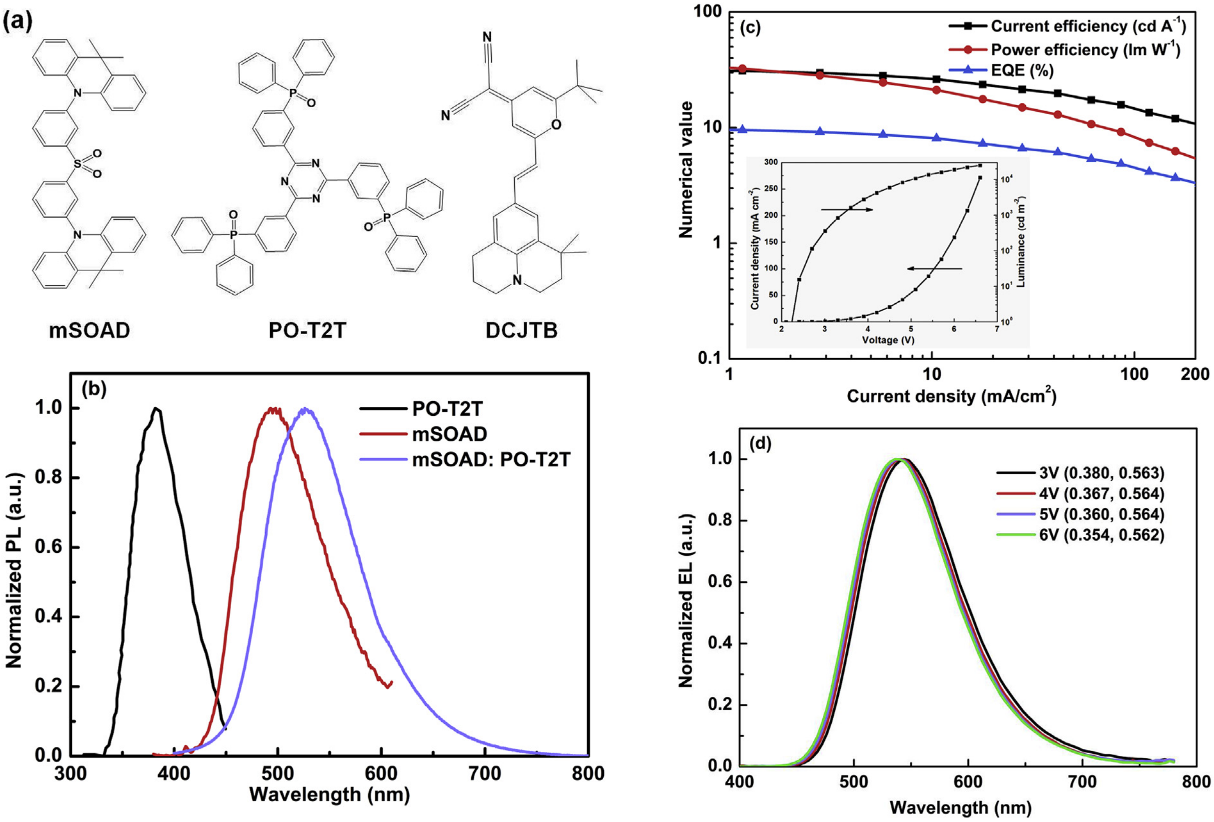

Fig. 1. (a) The molecules structure of donor mSOAD and acceptor PO-T2T. (b) The PL spectra of mSOAD, PO-T2T and mSOAD: PO-T2T mixed film. (c) The EL performances of Device A with current efficiency, power efficiency and EQE-current density curves. Inset is the current density-voltage-luminance curves. (d) EL spectra from 3 V to 6 V.

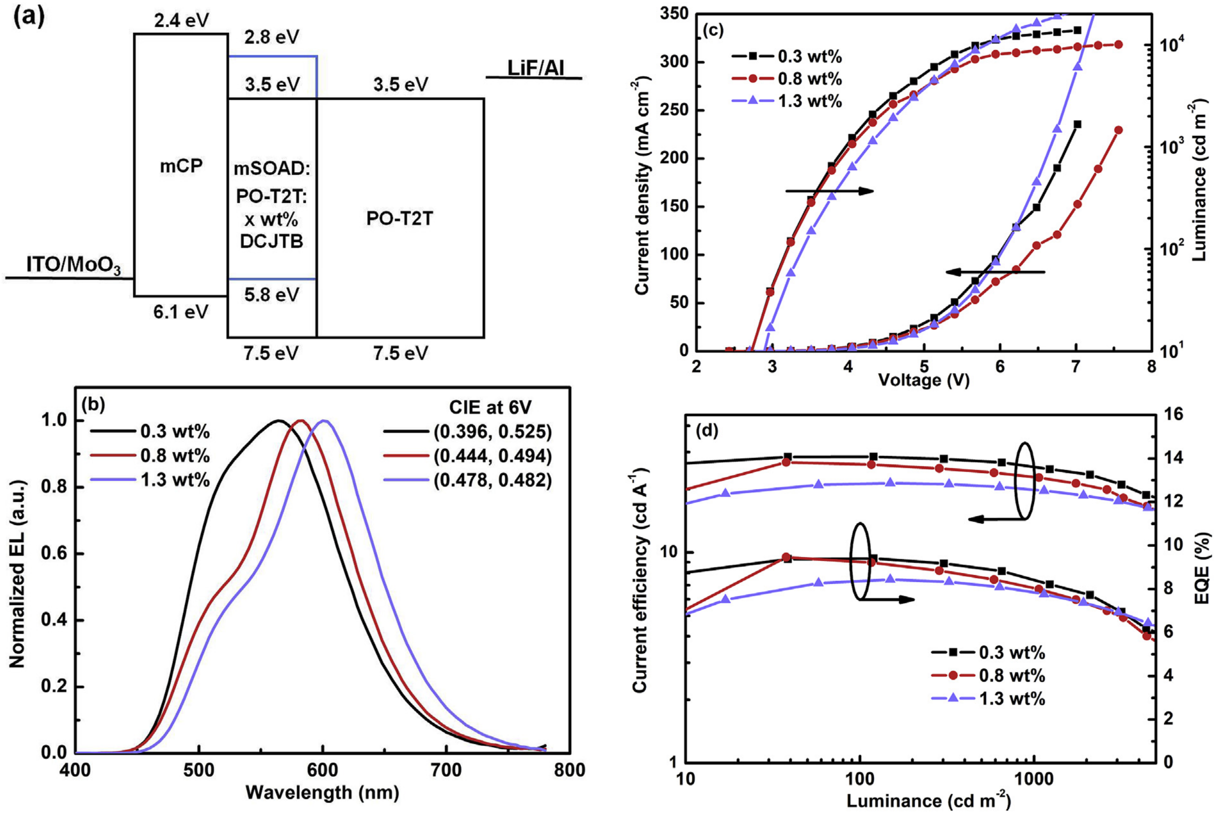

Fig. 2. The device structure and EL performances of Device B to D. (a) The device structure. (b) EL spectra at 6 V under different DCJTB concentrations. (c) Current density-voltage-luminance curves. (d) Current efficiency-luminance-EQE curves.

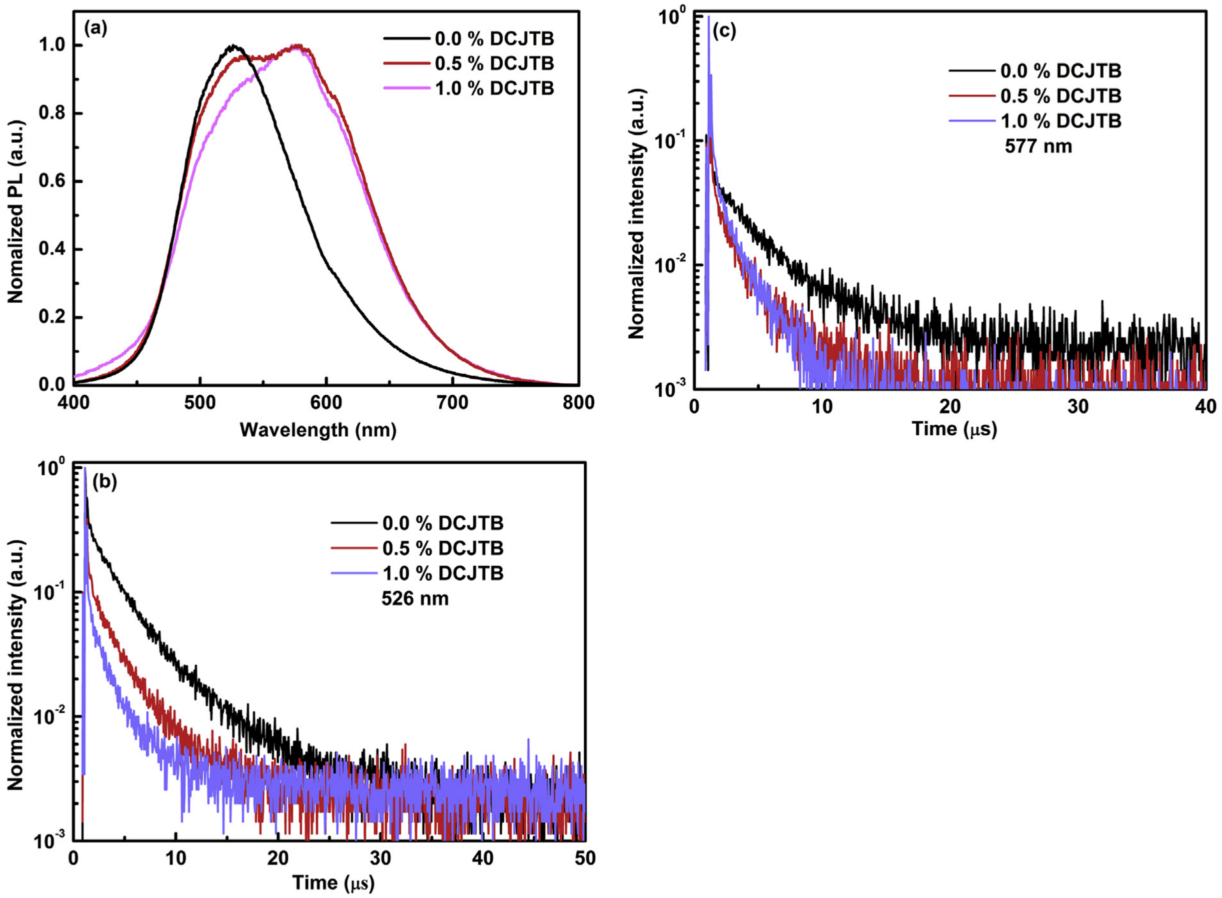

Fig. 3. The steady PL spectra and PL transient decay characteristics of different concentrations doping films of mSOAD: PO-T2T (1: 1): x wt% DCJTB. (a) Steady state PL spectra. (b) PL transient decay curves observed at 526 nm. (c) PL transient decay curves observed at 577 nm.

efficiency of the pure fluorescent WOLEDs. We also found the EQE of Device D (8.4%) is lower than Device B (9.4%) and Device C (9.5%), it should be caused by the different energy transfer paths in Device B to D. At a low concentration of DCJTB, the main energy transfer process from exciplex host to DCJTB is Förster energy transfer, while Dexter energy transfer process between exciplex and DCJTB would be enhanced under a high concentration, which would lead to the waste of triplet excitons due to the non-radiative transition from T_1 energy level of DCJTB. The steady PL spectra and PL transient decay characteristics of different concentrations doping films of mSOAD: x wt% DCJTB (x = 0, 0.5 and 1.0) are showed in Fig. 3, further demonstrate the energy transfer process from exciplex host to DCJTB dopant. The intensity of exciplex host emission decrease with increased dopant concentrations, which indicates the efficient energy transfer in the host-guest system [29]. Typical two lifetimes of prompt and delayed are observed under 526 nm with exciplex emission peak showed in Fig. 3b. Under optical excitation, only the singlet excitons could be produced on exciplex host and a dynamic cyclic process of ISC and RISC between singlet and triplet states could occur [30]. As the increase of doped concentrations, the energy transfer efficiency enhances, which leads to the reduction of prompt and delayed lifetime [31]. Similar to the 526 nm emission peak, the PL transient decay curves observed at 577 nm in Fig. 3c keep the same behavior, which demonstrates again the energy transfer mechanism in the doped films. The energy transfer efficiency increases with enhanced doped concentrations, which reduce the intersystem crossing (ISC) and RISC process and lower the transient lifetime in Fig. 3c.

The TADF molecule of mSOAD plays the role of exciplex donor in above section of WOLEDs structure. But we consider the excellent fluorescent emission characteristics of mSOAD could be applied to improve the device efficiency further. So we try to optimize the WOLEDs structure by employing TADF molecule of mSOAD as blue emitter and exciplex donor, respectively. To ensure the efficient emission of mSOAD, the spacer layer is utilized to separate the blue emitter and exciplex layer, and the WOLEDs designed are fabricated as follows: ITO/MoO_3 (5 nm)/mCP (30 nm)/mSOAD (20 nm)/spacer layer (x nm)/ mSOAD: PO-T2T (1: 1): 0.8 wt% DCJTB (30 nm)/PO-T2T (60 nm)/LiF (1 nm)/Al. The concentration of DCJTB is the same as Device C due to the high efficiency and suitable spectra. Three kinds of common materials of bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO), 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPb) and 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi) are selected as the spacer layers materials due to the suitable LUMO/HOMO energy level and high excitons energy. After optimizing the thickness of spacer layer, we obtain three best performance pure fluorescent WOLEDs for each material, defined as Device E, F and G, which corresponding to DPEPO (2 nm), TmPyPb (5 nm) and TPBi (5 nm), respectively. The relevant EL performances are showed in Fig. 4.

After adding the TADF molecule of mSOAD as blue emitter, all the WOLEDs with three different spacers achieve an improved EQE that exceeding 10%. The current efficiencies, power efficiencies and EQEs reach to 29.0 cd A^{-1}/25.7 lm W^{-1}/10.6%, 28.5 cd A^{-1}/26.5 lm W^{-1}/ 11.0% and 30.2 cd A^{-1}/28.0 lm W^{-1}/11.6% with DPEPO, TmPyPb and TPBi as the spacer layer, respectively. Meanwhile, a low turn-on voltage of < 3 V and high luminance of > 10000 cd m^{-2} are also realized, which are showed in Fig. S3. The enhancement of WOLEDs performances are attributed to the reasonable excitons distribution and efficient excitons restriction [32,33]. The WOLEDs of Device G with TPBi (5 nm) as spacer achieve the best EL performance with current efficiency, power efficiency and EQE of 30.2 cd A^{-1}, 28.0 lm W^{-1} and

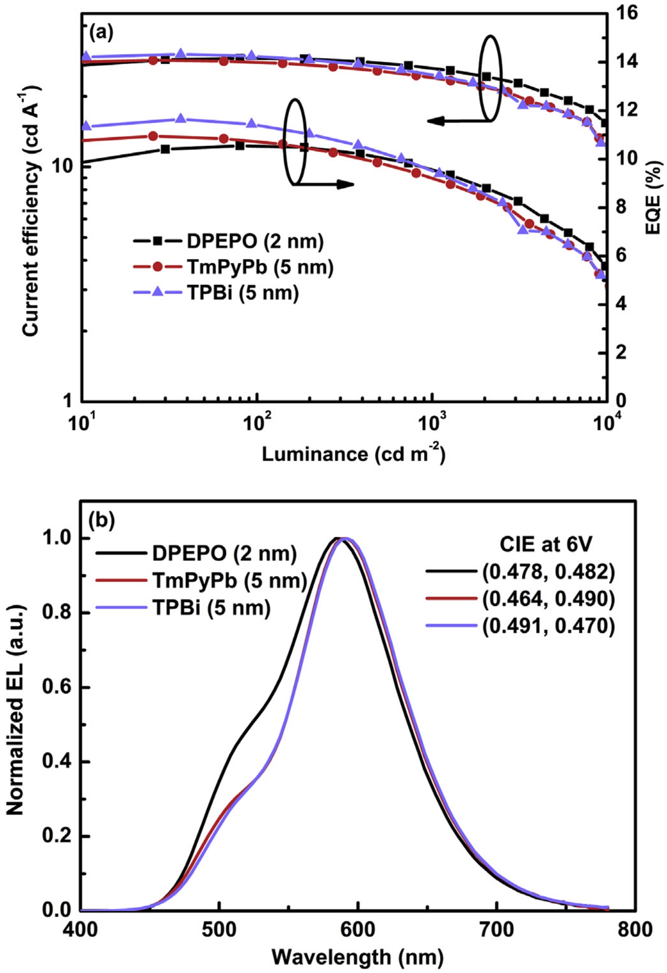

Fig. 4. The EL performances of Device E to G. (a) The current efficiency-luminance-EQE curves. (b) The EL spectra at 6 V.

11.6%, respectively. We consider the suitable energy level of HOMO/LUMO of TPBi is responsible for the high efficiency and the more discussions would be showed in detail below. The EL spectra of WOLEDs with DPEPO, TmPyPb and TPBi as the spacer shown in Fig. 4b exhibit warm white emission at 6 V with CIE coordinates of (0.478, 0.482), (0.464, 0.490) and (0.491, 0.470), respectively. And the EL spectra under various voltages are showed in Fig. S4, which present stable white emission spectra. Two emission peaks of ∼510 nm and ∼590 nm are showed in the EL spectra and construct the warm white emission. The emission of ∼590 nm is derived from the dopant of DCJTB. While we consider the emission peak of ∼510 nm is the overlap of blue emitter of mSOAD and yellow-green exciplex of mSOAD: PO-T2T due to the adjacent emission peak each other of mSOAD at 490 nm and exciplex at 540 nm.

In order to provide deep insight into the excellent device performances of high efficiency and stable white spectra, we depict the schematic diagram of energy transfer and light-emitting process in Fig. 5. Overall, the white emission could be divided into two sections, one is the blue emitter from the emission of pure TADF molecule of mSOAD, and the other is the simultaneous emission of exciplex host (mSOAD: PO-T2T) and dopant (DCJTB) due to the incomplete energy transfer. Under electric excitation, mSOAD layer emit blue emission and exciplex layer would transfer part of excitons to DCJTB by Förster energy transfer process for yellow-green and red emission. Herein, we focus on former emission and the application of spacer layer, while latter emission and the energy transfer process between exciplex host and dopant has been discussed above. The spacer layer could block holes and promote excitons formation in mSOAD emitting layer, then result in the blue emission. Besides, the more suitable HOMO energy level of TPBi (6.2 eV) than TmPyPb (6.7 eV) and DPEPO (6.8 eV) compared to mSOAD (5.8 eV) also ensure the efficient excitons recombination in latter mSOAD: PO-T2T: 0.8 wt% DCJTB emitting layer. The high pure fluorescent WOLEDs efficiency, one side deriving from the high singlet and triplet excitons energy of mCP, TPBi and PO-T2T, which could confine efficiently the excitons in emitting layer; on the other side coming from the efficient triplet excitons up-conversion of mSOAD molecule and mSOAD: PO-T2T exciplex host, which improve the triplet excitons utilization ratio. The EL performances of all the OLEDs in this paper are listed in Table 1.

4. Conclusion

In summary, the TADF molecule of mSOAD is employed to construct the pure fluorescent WOLEDs by acting as blue emitter and exciplex donor, respectively. Highly efficient fluorescent warm WOLEDs with reasonable structural design are realized with maximum current efficiency, power efficiency and EQE of 30.2 cd A^{-1}, 28 lm W^{-1} and 11.6%, respectively. Management to excitons distribution and efficient confinement to charges and excitons, and utilization to TADF molecule

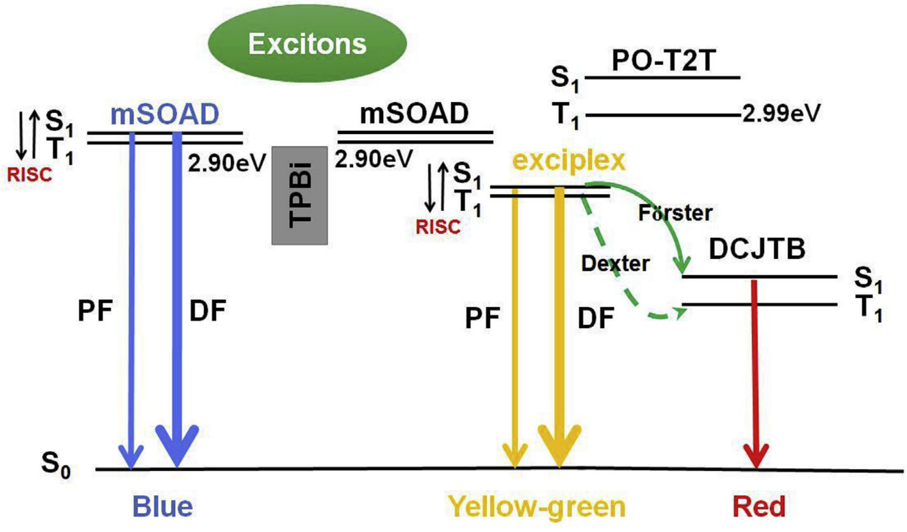

Fig. 5. The schematic diagram of energy transfer and light-emitting process.

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

Devices	V_{on}[a] [V]	$CE_{Max.}/PE_{Max.}/EQE_{Max.}$[b] [cd A^{-1}/lm W^{-1}/%]	$CE_{1000}/PE_{1000}/EQE_{1000}$[c] [cd A^{-1}/lm W^{-1}/%]	CIE at 6 V
A	2.4	31.4/39.0/9.7	28.6/25.1/9.0	(0.354, 0.562)
B	2.7	28.5/27.6/9.4	25.2/17.7/8.5	(0.396, 0.525)
C	2.7	26.8/25.5/9.5	22.7/18.5/8.0	(0.444, 0.494)
D	2.7	21.4/18.3/8.4	19.9/13.1/7.9	(0.478, 0.482)
E	2.7	29.0/25.7/10.6	25.9/16.1/9.5	(0.464, 0.490)
F	2.7	28.5/26.5/11.0	23.5/13.8/9.2	(0.491, 0.470)
G	2.7	30.2/28.0/11.6	24.5/15.2/9.6	(0.494, 0.470)

[a] Turn-on voltage.
[b] Maximum current efficiency (CE), power efficiency (PE) and EQE.
[c] CE, PE and EQE at 1000 cd m^{-2}.

are responsible to the high efficiency and stable white emission spectra. We believe the multi-function exploration to TADF molecules help to enlarge its application and contribute to the realization of high efficiency pure fluorescent WOLEDs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.orgel.2019.05.052.

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