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Efficient management of excitons in red and white organic light-emitting diodes by employing blue thermally activated delayed fluorescent emitter based acridine/sulfone derivative as the host

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ABSTRACT

Highly efficient red and white organic light-emitting diodes (OLEDs) employing blue thermally activated delayed fluorescent (TADF) emitter of mSOAD based acridine/sulfone derivative as the host are realized by efficient management for singlet and triplet excitons. The red OLEDs reach a high maximum current efficiency, power efficiency and EQE of 32.7 cd A^{-1}, 31.0 lm W^{-1} and 20.3%, respectively. And the white OLEDs with different dopant concentrations exhibit ultra-high color stability of ± (0.008, 0.001), ± (0.000, 0.003) and ± (0.002, 0.005), respectively. Meantime, the high efficiencies are also achieved with maximum current efficiencies, power efficiencies and EQEs of 28.2–32.0 cd A^{-1}, 26.3–30.5 lm W^{-1} and 12.2–17.4%, respectively. The analysis of energy level and transient decay characteristics demonstrate the excellent device performances are mainly stemmed from the sufficient confinement for charges and excitons and efficient energy transfer from the TADF host to dopant.

1. Introduction

The thermally activated delayed fluorescent (TADF) emitter received more and more attention in recent years due to the huge advantages in organic light-emitting diodes (OLEDs) compared to traditional fluorescent and phosphorescent emitters. Traditional fluorescent emitters possess low cost and high stability, but the maximum internal quantum efficiency (IQE) of merely 25% prevents its further development [1,2]. Phosphorescent emitters could achieve 100% IQE [3–5], however, the high cost and resource scarcity caused by the existence of heavy metal (Ir or Pt) also request the exploitation of more efficient emitter. Thereby, pure organic small molecule materials based TADF emitter with almost 100% IQE become more and more important.

In generally, typical TADF emitters are combined with donor segment and acceptor segment. Furthermore, the corresponding highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy level are also decided by donor segment and acceptor segment, respectively. The space separate donor and acceptor segments also provide a small difference between singlet and triplet energy level split (ΔE_{ST}), which conducts an efficient reverse intersystem crossing (RISC) process of triplet excitons. So the 100% IQE could be realized in theory due to the RISC process, which is the intrinsic characteristic of TADF emitter. In fact, many high efficiency TADF emitters with 100% IQE based blue, green and red light-emitting had been reported since 2012, but almost all the OLEDs based TADF emitter had to adopt host-guest doped system [6–10]. Host-guest doped system increases the fabrication complexity and reduces the devices repeatability. Besides, similar to the phosphorescent emitters, highly efficient blue TADF emitters also face a tremendous challenge because of the broad band-gap and low RISC efficiency [11–13]. So the development of highly efficient and non-doped blue TADF emitters still have much work to do. On the other hand, the host application of TADF

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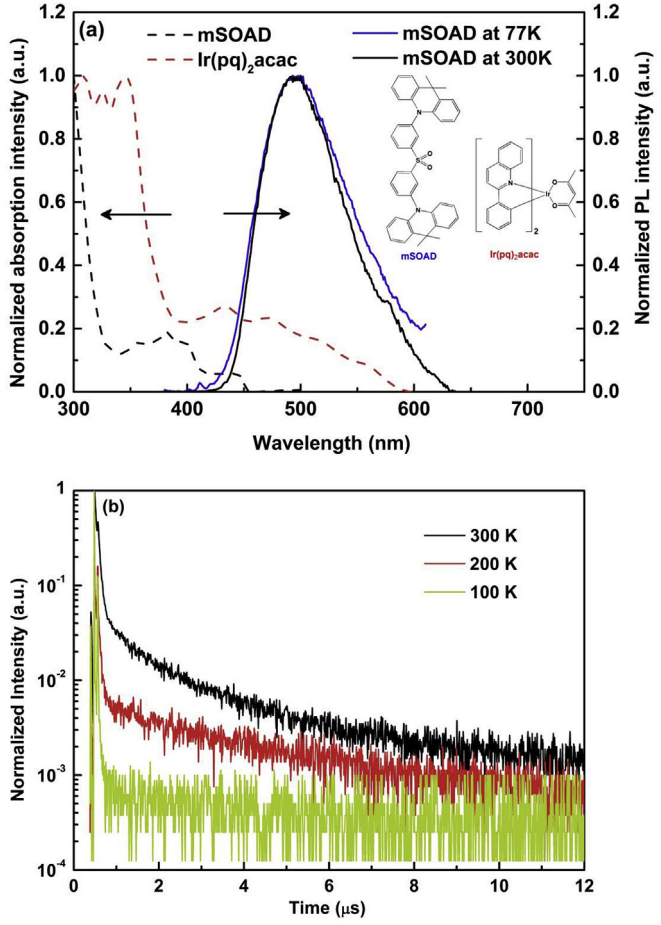

Fig. 1. The optical characteristics of mSOAD and Ir $(pq)_2$acac film. (a) Absorption spectra of mSOAD and Ir $(pq)_2$acac and PL spectra of mSOAD at 300 K and 77 K with 10 m s delay. Inset is the molecule structure of mSOAD and Ir $(pq)_2$acac. (b) PL transient decay curves of mSOAD film under different temperatures.

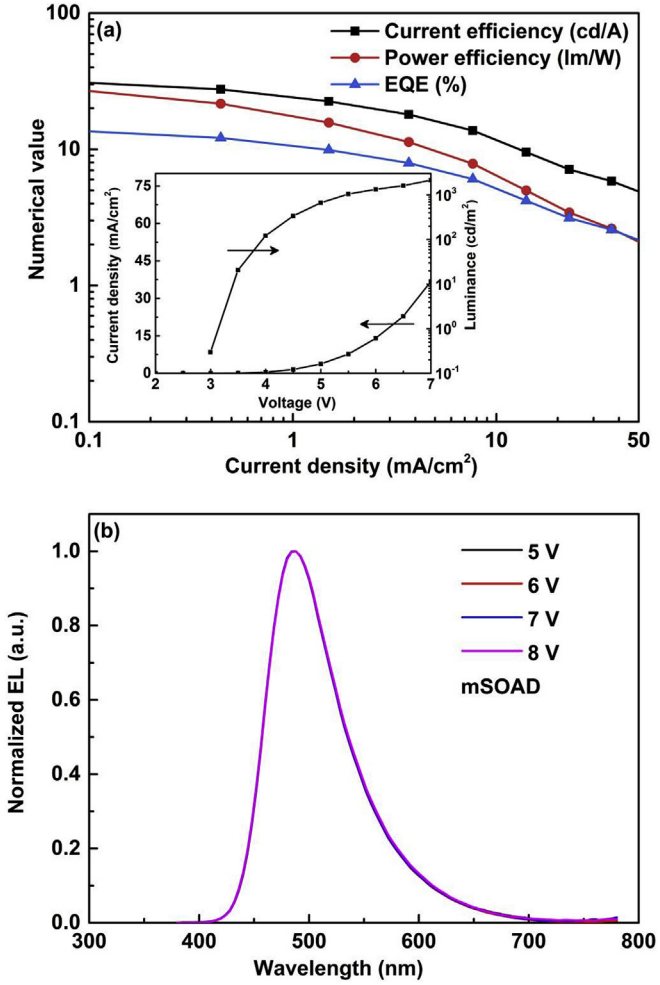

Fig. 2. The EL performances of blue OLEDs based mSOAD emitter. (a) Current efficiency, power efficiency and EQE-current density curves. Inset is the current density-voltage-luminance curves. (b) EL spectra from 5 V to 8 V. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

emitter is also under the research due to the excellent carriers transport bipolarity due to the existence of donor and acceptor segments. Duan et al. made many efforts to develop the TADF host and some outstanding results had been reported [14–17]. But utilizing TADF emitters as the host is still at the initial stage and many more deep research need to be investigated.

In this manuscript, the photoelectric properties of a newly synthesized blue TADF emitter of bis(3-(9,9-dimethyl-9,10-dihydroacridine) phenyl)sulfone (mSOAD) based acridine/sulfone derivative are studied. The simple non-doped blue OLEDs based mSOAD exhibit high external quantum efficiency (EQE) of 14%. More importantly, highly efficient red and white OLEDs with the blue TADF emitter as the host, phosphorescent emitter Iridium (III) bis(2-phenylquinoline) acetylacetonate (Ir $(pq)_2$acac) as the dopant, are also realized through the efficient management for excitons. The red OLEDs achieve the maximum current efficiency (CE), power efficiency (PE) and EQE of 32.7 cd A^{-1}, 31.0 lm W^{-1} and 20.3%, respectively. And all the white OLEDs with different dopant concentrations exhibit ultra-high color stability with the enhanced operation voltages, along with high maximum CEs, PEs and EQEs of 28.2–32.0 cd A^{-1}, 26.3–30.5 lm W^{-1} and 12.2–17.4%, respectively.

2. Experimental section

2.1. Device fabrication

All the OLEDs 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 (approximately 4×10^{-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 at a rate of 5.0 Å/s with a shadow mask, which defined the device area of 3×3 mm^2.

2.2. Characterization

The photoluminescence (PL) spectra were measured with FluoroMax-4 fluorescence spectrometer (HORIBA Jobin Yvon). The UV-Vis absorption spectrum was recorded with Hitachi U-3900 scanning spectrophotometer. 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. EQE was calculated from the current density-voltage-luminance curve and spectra data. All the organic materials were procured commercially without further

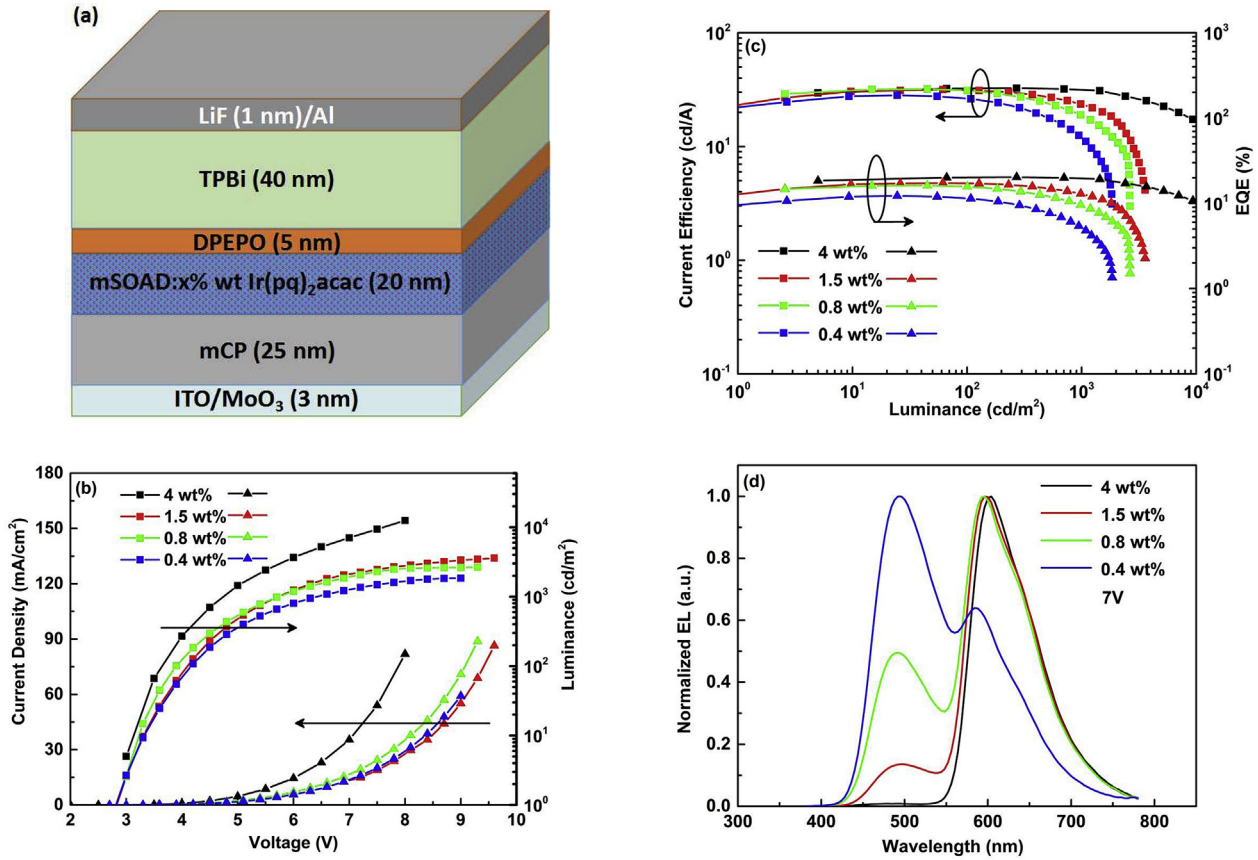

Fig. 3. The EL performances of red and white OLEDs with different dopant concentrations. (a) Device structure. (b) Current density-voltage-luminance curves. (c) Current efficiency-luminance-EQE curves. (d) EL spectra at 7 V with different concentrations. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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

3.1. Non-doped blue OLEDs

The blue TADF emitter of mSOAD based acridine/sulfone derivative is synthesized in our laboratory, which is described briefly as follows: it was synthesized by the Buchwald-Hartwig amination reaction between 9,9-dimethyl-9,10-dihydroacridine and bis(3-iodophenyl)sulfone. The two 9,9-dimethyl-9,10-dihydroacridine segments were linked to the diphenylsulphone core via 3,3′-positions, presenting a highly twisted zig-zag configuration. The absorption and PL spectra of pure mSOAD film are showed in Fig. 1a and inset shows the molecular structure. Almost the same PL spectra in room temperature and 77 K with 10 m s delay demonstrate the ultra-small ΔE_{ST} between singlet and triplet excited state energy level, which could be estimated 0.02 eV (S_1 = 2.92 eV and T_1 = 2.90 eV) from the onset of the fluorescence and phosphorescence spectra [6,18]. The small ΔE_{ST} also indicates an efficient RISC of triplet excitons could be achieved in mSOAD emitter. The transient decay curves of mSOAD films under different temperatures are exhibited in Fig. 1b. The obvious double-exponential decay behavior and increased decay fluorescent lifetime as the enhancement of temperature confirm the TADF characteristics of mSOAD emitter [19,20]. At 300 K, the fitted value of prompt and delayed fluorescent lifetime are 51.8 ns and 2.11 μs, respectively. Therefore, the PL spectra and transient decay characteristics indicate mSOAD is a highly efficient TADF emitter. And the synthetic route and other thermal, optical and characterization data of mSOAD had been published in another paper [21].

Then we fabricate OLEDs based the blue emitter with simple non-doped structure as follows: ITO/MoO_3 (3 nm)/mCP (25 nm)/mSOAD (20 nm)/DPEPO (5 nm)/TPBi (40 nm)/LiF (1 nm)/Al. m-bis(N-carbazolyl)benzene (mCP) and 1,3,5-tris(N-phenyl-benzimidazol-2-yl)benzene (TPBi) act as hole and electron transport layer, respectively. Thin layer of bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO) is the exciton block layer (EBL) to confine mSOAD excitons in emitting layer (EML) due to the ultra-high triplet excitons energy of 2.98 eV [22–24]. The device performances of efficiency and EL spectra are showed in Fig. 2. The non-doped blue OLEDs achieve excellent device efficiencies with maximum CE, PE and EQE of 31.7 cd A^{-1}, 28.5 lm W^{-1} and 14%, respectively. It is almost one of the best blue TADF emitter with non-doped structure. Although many blue TADF materials had been reported, but most of them have to adopt the host-guest doped system. So high efficiency OLEDs based mSOAD with non-doped structure also promote the development of blue TADF materials. The sky blue emission is obtained and the same blue EL spectra under various voltages demonstrate the intrinsic light-emitting of mSOAD emitter without any noticeable contribution from charge transport layer. The high EL efficiency with non-doped structure confirms that mSOAD based acridine/sulfone derivative is an excellent blue TADF emitter.

3.2. Red and white OLEDs

Based on the excellent photoelectric performances of mSOAD emitter, we focus on the research to its host application in red and white OLEDs. Analogous to the non-doped blue OLEDs structure, the red and white OLEDs are performed as follows: ITO/MoO_3 (3 nm)/mCP

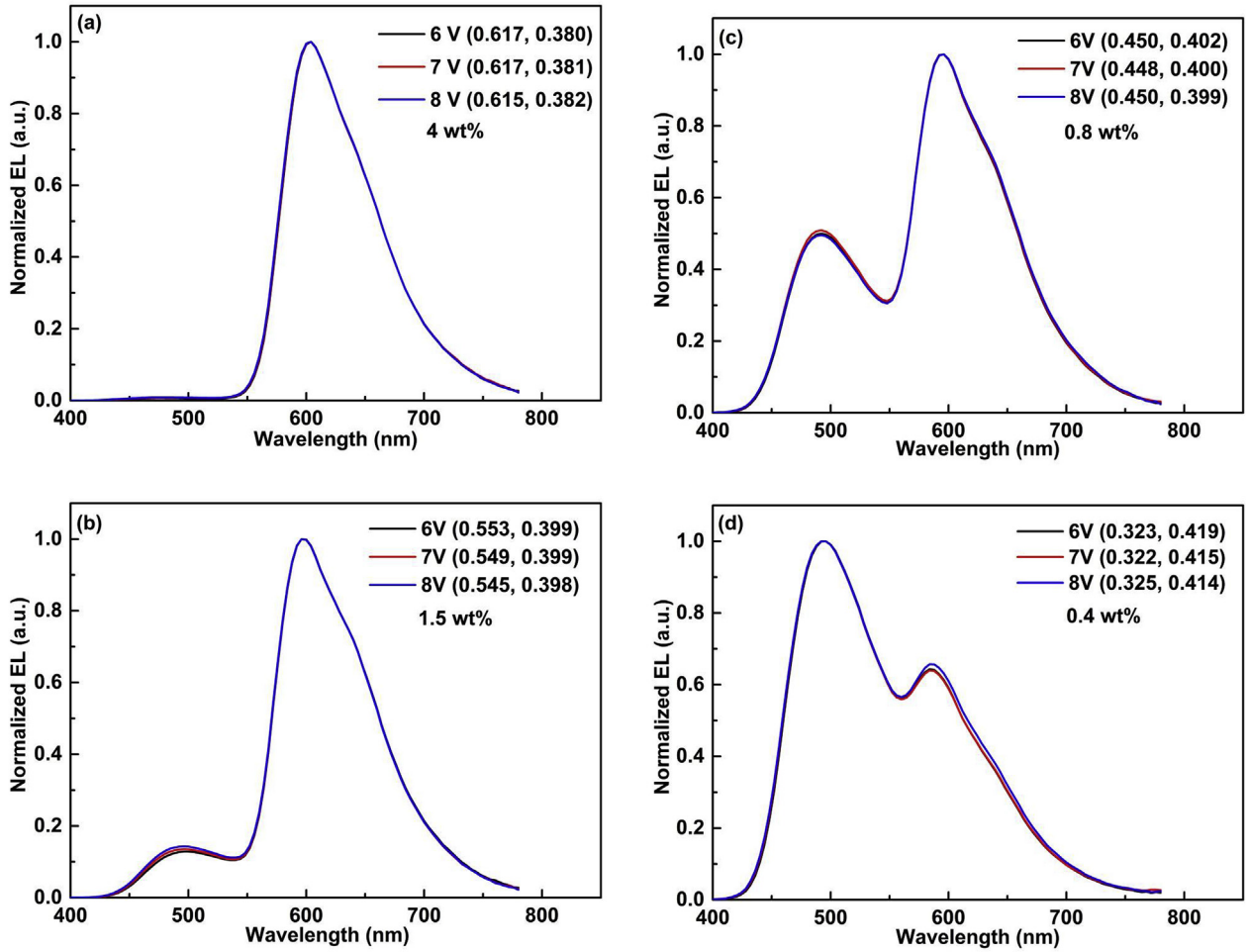

Fig. 4. The EL spectra of red and white OLEDs under different dopant concentrations from 6 V to 8 V. (a) 4 wt%. (b) 1.5 wt%. (c) 0.8 wt%. (d) 0.4 wt%. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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

	V_{on} [V]	$CE_{Max.}/PE_{Max.}/EQE_{Max.}$ [a] [cd A^{-1}/lmW^{-1}/%]	$CE_{1000}/PE_{1000}/EQE_{1000}$ [b] [cd A^{-1}/lmW^{-1}/%]	CIE coordinates [7 V]	ΔCIE coordinates
Non-doped blue	3.1	31.7/28.5/14.0	13.7/7.8/6.0	(0.181, 0.322)	–
4 wt%	2.9	32.6/31.0/20.3	17.3/7.3/10.8	(0.617, 0.381)	–
1.5 wt%	2.8	31.6/29.0/17.4	23.6/13.0/13.0	(0.549, 0.399)	(± 0.008, ± 0.001)
0.8 wt%	2.8	32.0/30.5/16.2	17.5/9.0/8.7	(0.448, 0.400)	(± 0.000, ± 0.003)
0.4 wt%	2.8	28.2/26.3/12.2	11.1/5.3/4.8	(0.0322, 0.415)	(± 0.002, ± 0.005)

[a] Maximum current efficiency (CE), power efficiency (PE) and EQE.
[b] CE, PE and EQE at 1000 cd/m^2.

(25 nm)/mSOAD: x wt% Ir $(pq)_2$acac (20 nm)/DPEPO (5 nm)/TPBi (40 nm)/LiF (1 nm)/Al, where x = 4, 1.5, 0.8 and 0.4, respectively. Ir $(pq)_2$acac is the red phosphorescent dopant, so red or white light-emitting could be realized by controlling the dopant concentrations. The devices performances with different dopant concentrations are showed in Fig. 3. Under a high concentration of 4 wt%, the device shows pure red light-emitting EL spectra with only one emission peak fixed at around 604 nm. While the other three devices exhibit white light-emitting with two emission peaks, which come from mSOAD and Ir $(pq)_2$acac, respectively. Besides, the blue light-emitting intensity increases with the reduced concentrations of Ir $(pq)_2$acac, which suggests the efficient energy transfer from mSOAD to Ir $(pq)_2$acac occurs. A complete energy transfer under 4 wt% concentration results in the pure red light-emitting, while the incomplete energy transfer from mSOAD to Ir $(pq)_2$acac could realize the white light-emitting. The slight blue shift of EL spectra with reduced concentrations shown in Fig. 3d can be regard as the strong intermolecular interaction between host and dopant because of the change of concentrations. The large overlap between PL spectrum of mSOAD and absorption spectrum of Ir $(pq)_2$acac, showed in Fig. 1a, also indicates the high energy transfer efficiency from mSOAD to Ir $(pq)_2$acac.

The red and white OLEDs also demonstrate a low turn-on voltage below 3 V, which indicates the efficient charges injection and recombination in the devices. The red OLEDs with 4 wt% concentration earn a high maximum CE, PE and EQE of 32.7 cd A^{-1}, 31.0 lm W^{-1} and 20.3%, respectively. While the white OLEDs with 1.5 wt%, 0.8 wt% and 0.4 wt% concentrations achieve maximum CEs/PEs/EQEs of 31.6 cd A^{-1}/29.0 lm W^{-1}/17.4%, 32.0 cd A^{-1}/30.5 lm W^{-1}/16.2%, 28.2 cd A^{-1}/26.3 lm W^{-1}/12.2%, respectively. The high efficiency of red and white OLEDs could be attributed to efficient RISC process and high energy transfer efficiency from mSOAD to dopant, which would be discussed more below.

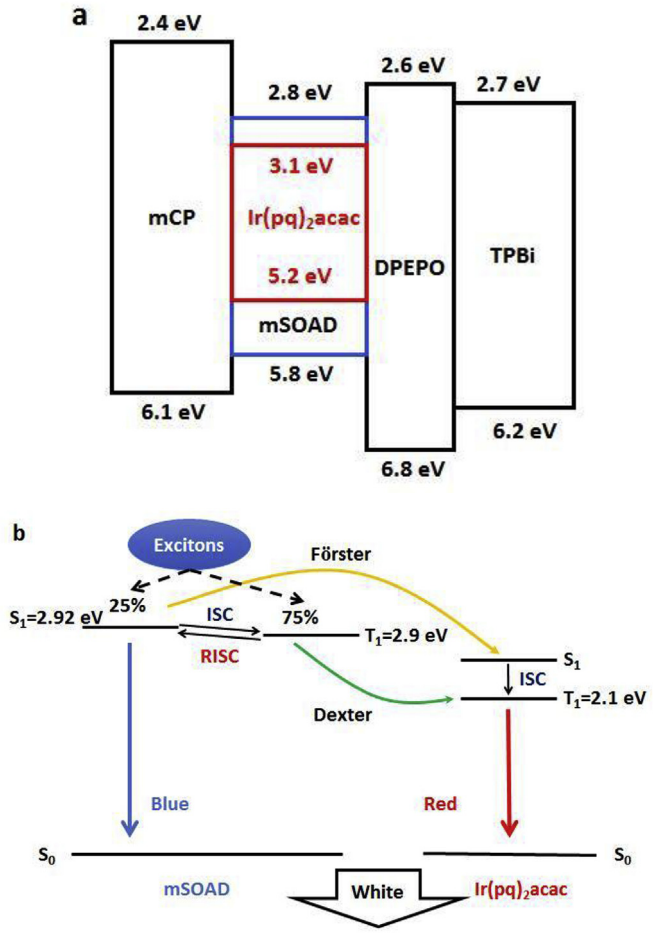

Fig. 5. The energy level and energy transfer schematic diagram. (a) Energy level diagram. (b) Energy transfer schematic diagram.

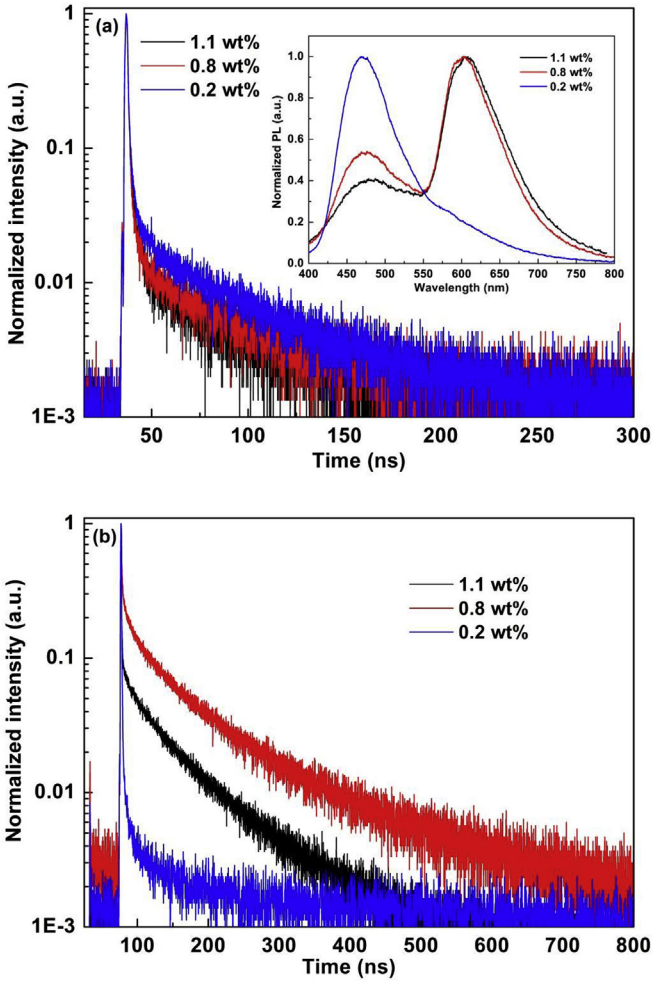

Fig. 6. The PL transient decay characteristics of different concentrations doped films of mSOAD: x wt% Ir (pq)2acac. (a) PL transient decay curves observed at 475 nm. Inset is the steady state PL spectra of the doped films. (b) PL transient decay curves observed at 604 nm.

The EL spectra of red and white OLEDs under different voltages are showed in Fig. 4. As mentioned above, the same EL spectra of red OLEDs with various voltages declare further the efficient and complete energy transfer from mSOAD to dopant. The mechanism that the relative intensity of dopant increases with the concentration from 0.4 wt % to 4 wt% could be explained as the mSOAD host transfer more excitons to dopant with the enhanced concentrations. But to our more surprising are the color stability of white OLEDs, all the white OLEDs show ultra-high color stability with the enhanced operation voltages. The Commission Internationale de l'Eclairage (CIE) coordinates changed ratios of the three white OLEDs from 6 V to 8 V are merely ± (0.008, 0.001), ± (0.000, 0.003) and ± (0.002, 0.005), respectively. We consider the ultra-high color stability could be attributed to the points showed below: The suitable HOMO and LUMO levels in the devices could facilitate charge injection and provide charge balance in EML. The high triplet energies of mCP and DPEPO could confine efficiently excitons in EML and further stabilize the recombination zone. The great overlap between the absorption spectrum of dopant and PL spectrum of host ensures the highly efficient energy transfer efficiency. A summary EL performance of all the OLEDs in this paper is showed in Table 1.

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 level and energy transfer process in Fig. 5. The HOMO energy level of mSOAD is estimated from the cyclic voltammetry and the LUMO energy level is estimated from HOMO and the optical bandgap (Eg), which is determined from the onset of the absorption band. One hand, the holes could be injected from anode into EML barrier-free because the HOMO energy levels of mCP (6.1 eV) is deeper than mSOAD (5.8 eV). The injected electrons also have a very small LUMO barrier of 0.1 eV between TPBi (2.7 eV) and DPEPO (2.6 eV). Almost non-barrier charges transport is one reason of low turn-on voltage (< 3 V). Besides, the shallow LUMO of mCP (2.4 eV) and deep HOMO of DPEPO (6.8 eV) could confine efficiently the injected electrons and holes to improve the charges recombination efficiency. On the other hand, the 25% singlet excitons and 75% triplet excitons could produce first on the mSOAD host under the electric excitation. The 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) are higher than mSOAD (S_1 = 2.92 eV, T_1 = 2.9 eV), so the excitons produced on mSOAD host could also be confined in EML and prevent the excitons migration to adjacent transport layer. Finally, the singlet and triplet excitons on mSOAD could be transferred to dopant by Förster and Dexter energy transfer process. The triplet excitons also could be changed into singlet excitons by efficient RISC process and then be transferred to dopant through Förster energy transfer. The red light-emitting could be obtained by the radiative transition of triplet excitons that transferred from Dexter process or the intersystem crossing (ISC) process. While the white light-emitting could be realized by the optimization of dopant concentrations to control the light-emitting intensity of host blue and guest red. The efficient management for singlet and triplet excitons achieves nearly complete excitons utilization, which is responsible for the high efficiency and the efficient confinement for charges and excitons stabilizes the recombination zone

[25–27], further contributes to the stable white EL spectra under different operation voltages. Furthermore, we consider the charge trapping don't exist in the white OLEDs. Although there have the large HOMO/HOMO (0.6 eV) and LUMO/LUMO (0.3 eV) difference between mSOAD and Ir $(pq)_2$acac, but the charges barrier-free injection into EML ensure the efficient recombination of excitons on mSOAD. Almost the consistent current density curves under different concentrations in Fig. 3b also confirm the light-emitting of red emitter is derived from the energy transfer rather than charges trapping [28,29].

Finally, the PL transient decay characteristics with different concentrations doped films of mSOAD: x wt% Ir $(pq)_2$acac are showed in Fig. 6. The steady PL spectra of the doped films are displayed in inset of Fig. 6a. The intensity of blue emitter increases with reduced dopant concentrations, which indicates the efficient energy transfer in the host-guest system. Typical two lifetimes of prompt and delayed are observed under 475 nm with mSOAD emission peak showed in Fig. 6a. Under optical excitation, only the singlet excitons could be produced on mSOAD host and a dynamic cyclic process of ISC and RISC between singlet and triplet states could occur. As the decrease of doped concentrations, the energy transfer efficiency reduces, which leads to the increase of prompt and delayed lifetime. Similar to the 475 nm emission peak, the PL transient decay curves observed at 604 nm in Fig. 6b keep the same behavior except for the curve of 0.2 wt% concentration, which demonstrates again the energy transfer mechanism in the doped films. The energy transfer efficiency decreases with reduced doped concentrations, which enhance the ISC and RISC process and increase the transient lifetime in Fig. 6b. The short lifetime of 0.2 wt% concentration in Fig. 6b may be also the mSOAD emission lifetime, because the ultra-low concentration results in very weak emission of Ir $(pq)_2$acac.

4. Conclusion

By employing a newly synthesized blue TADF emitter as the host, highly efficient red and white OLEDs are realized. The red OLEDs achieve a high maximum current efficiency, power efficiency and EQE of 32.7 cd A^{-1}, 31.0 lm W^{-1} and 20.3%, respectively. And the white OLEDs based different dopant concentrations exhibit ultra-high color stability, the CIE coordinates changed ratios are merely ± (0.008, 0.001), ± (0.000, 0.003) and ± (0.002, 0.005), respectively. The high efficiencies are also obtained simultaneously with maximum current efficiencies, power efficiencies and EQEs of 28.2–32.0 cd A^{-1}, 26.3–30.5 lm W^{-1} and 12.2–17.4%, respectively. The excellent device performances are mainly stemmed from the sufficient confinement for charges and excitons and efficient energy transfer. We believe the host application further enlarges the potential and promotes the development of TADF emitter.

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