



Color stable and highly efficient hybrid white organic light-emitting devices using heavily doped thermally activated delayed fluorescence and ultrathin non-doped phosphorescence layers



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ABSTRACT

Blue/orange complementary fluorescence/phosphorescence hybrid white organic light-emitting devices with excellent color stability and high efficiency have been fabricated, which are based on an easily fabricated multiple emissive layer (EML) configuration with an ultrathin non-doped orange phosphorescence EML selectively inserted between heavily doped blue thermally activated delayed fluorescence (TADF) EMLs. Through systematic investigation and improvement on luminance-dependent color shift and efficiency deterioration, a slight Commission Internationale de l'Eclairage coordinates shift of (0.008, 0.003) at a practical luminance range from 1000 to 10000 cd/m², a maximum power efficiency of 45.8 lm/W, a maximum external quantum efficiency (EQE) of 15.7% and an EQE above 12% at 1000 cd/m² have been achieved. The heavily doped blue TADF emitters which act as the main charge transport channels and recombination sites in the host with high-lying lowest triplet excited state, take advantage of the bipolar transport ability to broaden the major charge recombination region, which alleviates triplet energy loss. The selectively inserted ultrathin non-doped orange EML makes its emission mechanism dominated by Förster energy transfer, which is effective to keep color stable under different drive voltages.

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1. Introduction

White organic light-emitting devices (WOLEDs) have been paid tremendous research attentions on their applications in full-color flat-panel displays and solid state lighting, due to their advantages such as light weight, high resolution, homogeneous large-area emission and potential application on flexible substrates [1,2]. In the past two decades, WOLEDs have been significantly developed by various approaches, including emitting material system innovation and device configuration optimization, etc [3,4]. Nowadays, three kinds of WOLEDs based on different emitting materials are reported, which are full-fluorescence WOLEDs, full-phosphorescence WOLEDs and fluorescence/phosphorescence hybrid WOLEDs. Since phosphors theoretically enable an internal quantum efficiency (IQE) of 100% by harvesting both singlet and triplet excitons [5,6]. The full-phosphorescence WOLEDs can obtain

efficiency fourfold higher than that of full-fluorescence ones [7,8]. However, the development of full-phosphorescence WOLEDs is limited by the lack of blue phosphors with both high efficiency and good stability, which results in the poor lifetime of WOLEDs [9,10]. Soon after, hybrid WOLEDs combining blue fluorophores with green and red phosphors have attracted considerable interest, which take advantage of the superior stability of fluorophores and high efficiency of phosphors simultaneously [11–13].

To achieve high-efficiency hybrid WOLEDs, triplet energy loss through the non-radiative triplet states of the blue fluorophores should be prevented. Although the employment of blue fluorophores with high-lying lowest triplet excited state (T1) facilitates the energy transfer from blue fluorophores to phosphors, resulting in an alleviated triplet energy loss [14,15], there are still some triplet excitons inevitably deactivated on blue fluorophores through non-radiative recombination during diffusion process, due to the short radius of Dexter energy transfer [16]. Recently, the blue thermally activated delayed fluorescence (TADF) emitters which were introduced by Adachi et al., are considered as promising alternatives to replace conventional blue fluorophores in hybrid

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WOLEDs [16–19], since blue TADF emitters with high T1 and high efficiency have been reported, due to their intrinsic small singlet-triplet splits and the potential of realizing unity IQE [20,21]. More important, the triplet energy loss on blue TADF emitters can be significantly decreased, since triplet excitons can convert to singlet ones through reverse inter-system crossing (RISC) process, and the latter can be used for blue fluorescence by radiative decay or transferred to phosphors via long-radius Förster energy transfer.

Besides, highly efficient and color stable hybrid WOLEDs are usually based on three typical device configurations, including multiple emissive layers (MEML), single emissive layer with multiple dopants (SEML-MD) and single emissive layer with single dopant (SEML-SD). However, the high-performance WOLEDs were obtained through complex device structures and/or precisely controlled fabrication process, which raise the fabrication cost and decrease reproducibility. For instance, to achieve high color stability, the MEML based WOLEDs need an additional charge or exciton-blocking interlayer [22,23], while the SEML-MD based WOLEDs require precisely controlled doping process for optimum doping concentration ratio between different dopants [19]. Furthermore, to alleviate the deteriorated effect of charge trapping on color stability and realize incomplete host-guest Förster energy transfer, the state-of-the-art blue/yellow or blue/orange complementary WOLEDs based on SEML-SD structure also need precise doping control to maintain an extremely low (≤ 0.5 wt%) doping concentration [12,24]. Hence, high-performance WOLEDs with simplified configuration and/or easy fabrication should be further developed.

Recently, a blue TADF emitter bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (DMAC-DPS) is highly attractive due to its high photoluminescence quantum yield (PLQY) of 0.88 in neat film and bipolar transport ability [20,25]. Since methyl groups on the DMAC units inhibit the intermolecular π - π stacking interactions between electron donors, the high PLQY of DMAC-DPS in solid state is attributed to the aggregation-alleviated ability, which enables the highly efficient OLEDs with easily fabricated heavily doped EML. Besides, DMAC-DPS exhibits a rather broad characteristic emission spectrum with a full width at half maximum about 80 nm, which is suitable as blue emission component in blue/orange complementary WOLEDs.

Therefore, in this work, we propose blue/orange complementary hybrid WOLEDs with an easily fabricated MEML structure consisting of heavily doped fluorescence EMLs using DMAC-DPS and an ultrathin non-doped phosphorescence EML employing an orange iridium complex bis(4-*tert*-butyl-2-phenylbenzothiazololato-*N,C*²)iridium(III) (acetylacetonate) [(tbt)₂Ir(acac)]. The MEML based WOLEDs without interlayer exhibit excellent color stability and high efficiency with a slight Commission Internationale de l'Éclairage (CIE) coordinates shift of (0.008, 0.003) from 1000 to 10000 cd/m² and a maximum external quantum efficiency (EQE_{max}) of 15.7%, respectively. Meanwhile, the charge transport process and emission mechanism have been systematically investigated by analyzing the electroluminescence (EL) spectra, characteristics and photoluminescence (PL) transient decay curves.

2. Experimental

Indium tin oxide (ITO) coated glass substrates with a sheet resistance of 15 Ω /sq which acted as anode, were pre-cleaned with detergent, acetone, deionized water and ethanol for 15 min at each ultrasonic step. Then oxygen plasma treatment was performed to further clean the ITO surface. Organic functional layers and metallic cathode were thermally evaporated in vacuum under a pressure of 3×10^{-4} Pa and 3×10^{-3} Pa, respectively. The device configurations in this research are listed in Table 1, and all devices have an active

Table 1
Device configurations of blue OLEDs and WOLEDs.

Device	MoO ₃	TAPC	mCP	mCP: DMAC-DPS				Bphen
B1				15 (10 wt%)				
B2	10	40	10	15 (30 wt%)				40
B3				15 (50 wt%)				
Device	MoO ₃	TAPC	mCP	mCP: B	Ora			Bphen
W1					1			
W2	10	40	10	15	0.3			40
W3					0.1			
W4					0.05			
Device	MoO ₃	TAPC	mCP	mCP: B	Ora	mCP: B	Bphen	
P1				10	0.1	5		
P2	10	40	10	5		10	40	
P3				0		15		
Device	MoO ₃	TAPC	mCP	DPEPO: B	Ora	DPEPO: B	Bphen	
W5				15		0		
W6	10	40	10	12	0.1	3	40	
W7				9		6		

The above are organic functional layers except ITO anode and 100 nm-thick Mg: Ag cathode, mCP: B = mCP: DMAC-DPS(50 wt%); Ora=(tbt)₂Ir(acac); DPEPO: B = DPEPO: DMAC-DPS(50 wt%). The unit for layer thickness is nm.

area of 0.25 cm². Therein, molybdenum trioxide (MoO₃) was used as hole injection layer, while 4,4'-cyclohexylidenebis[*N,N*-bis(4-methylphenyl)aniline] (TAPC) and 4,7-diphenyl-1,10-phenanthroline (Bphen) were employed as hole transport layer and electron transport layer, respectively. Besides, 3,5'-*N,N'*-dicarbazole-benzene (mCP) was inserted between TAPC and blue EML to inhibit exciplex formation at the interface of TAPC/DMAC-DPS. The blend films of DMAC-DPS fluorophore doped in mCP or bis[2-(diphenylphosphino)phenyl]ether oxide (DPEPO) host matrix acted as blue EMLs. The ultrathin non-doped film of (tbt)₂Ir(acac) phosphor was an orange EML. The energy level diagram of WOLEDs along with the chemical structures of the organic materials in EMLs are depicted in Fig. 1.

UV-vis absorption spectrum was characterized with a SHIMATZU UV-1700 spectrophotometer. PL spectra were recorded with a PerkinElmer LS55 spectrometer. EL spectra and CIE coordinates were measured with an OPT-2000 spectrometer. PL transient decay characteristics were recorded with a HORIBA Scientific Single Photon Counting Controller FluoroHub-B, in which the samples were excited at 370 nm using a NanoLED-370 excitation light source, and the emitted photons were detected by a TBX detector connected to a TBX-PS power supply. Current density-voltage-luminance (*J-V-L*) characteristics were tested with a Keithley 4200 source and a luminance meter. All the measurements were performed in air at room temperature without encapsulation except that the PL transient decay characteristics were recorded under nitrogen atmosphere.

3. Results and discussion

According to the UV-vis absorption spectrum of the (tbt)₂Ir(acac) neat film in Fig. 1(b), the absorption peaks from 300 nm to 350 nm, are mostly assigned to ligand-centered (LC) π - π^* transitions, while the absorption peaks located at 450 nm and 490 nm are originated from singlet and triplet metal to ligand charge transfer (¹MLCT and ³MLCT) transitions, respectively. The PL spectrum of the DMAC-DPS neat film which is excited at 370 nm using an excitation light source, shows a peak at 472 nm originated from singlet intramolecular charge transfer transition. There is a large spectral overlap between the PL spectrum of DMAC-DPS and the ¹MLCT absorption band of (tbt)₂Ir(acac). It indicates that efficient Förster

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