Hybrid white organic light-emitting devices consisting of a non-doped thermally activated delayed fluorescent emitter and an ultrathin phosphorescent emitter

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ABSTRACT

Hybrid white organic light-emitting devices (OLEDs) are fabricated by employing non-doped emitting layers (EMLs), which are consisted of a blue thermally activated delayed fluorescent (TADF) emitter 9, 9-dimethyl-9, 10-dihydroacridine-diphenylsulfone (DMAC-DPS) and an ultrathin yellow iridium complex bis[2-(4-tertbutylphenyl)benzothiazolato-N,C^{2'}] iridium (acetylacetonate) [(tbt)₂Ir(acac)]. With thickness optimization of DMAC-DPS, a white OLED achieves maximum current efficiency, power efficiency, and external quantum efficiency of 34.9 cd/A, 29.2 lm/W, and 11.4 %, respectively, as well as warm white emission with relatively stable electroluminescence spectra. The results suggest that, bipolar charge carrier transport property and concentration independent property of DMAC-DPS, charge carrier trapping effect of the ultrathin (tbt)₂Ir(acac), and balanced self-emission process and energy transfer process between DMAC-DPS and (tbt)₂Ir(acac), contribute to high device performance.

Keywords: white organic light-emitting device, non-doped emitting layer, thermally activated delayed fluorescent, ultrathin phosphorescent emitter

1. Introduction

Organic light-emitting device (OLED) has aroused lots of interests due to its advantages of lightweight, high luminance, high efficiency, low driving voltage, unique flexibility, and so on. Among of which, white OLEDs (WOLEDs) have been considered as a potential alternative for applications in flat panel displays, solid-state lightings and backlights for liquid crystal display. In typically multilayer structured white OLEDs, the emitters are one of the most important components, including either red, green and blue emitters, or blue and yellow emitters, where the blue emitter is indispensable. The first generation fluorescent emitters and the second generation phosphorescent emitters have been extensively utilized[1,2]. As for the fluorescent ones, only 25% singlet (S₁) excitons can make contribution to light emission, restricting the device efficiency. In the case of phosphorescent emitters, both 25% singlet and

75% triplet (T_1) excitons can be efficiently used for radiative emission, so phosphors have been widely utilized. Moreover, the blue fluorescent emitters have relatively high stability but relatively low efficiency, while the well-known blue phosphorescent emitters suffer from short lifetime and low stability[3], although they present high efficiency. As well known, conventional organic emitters are subjected to aggregation-caused quenching[4], due to strong intermolecular interactions between the emitter molecules in solid state. To solve this problem, a host-guest doping system has been widely adopted[5,6], by distributing the emitters in one or more host matrixes. However, it is difficult to precisely control the doping ratios of the host and guest materials, and the repeatability and yield are restricted, leading to high production cost accordingly.

Up to the present, the third generation emitters, namely, the thermally activated delayed fluorescent (TADF) materials which were firstly introduced in OLEDs by Adachi group in 2009[7], have being received huge attentions[8-11]. The TADF emitters possess a small energy gap between S_1 and T_1 , resulting from the spatially separated the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) which are mainly located on the electron donor and acceptor moieties of the TADF molecules, respectively, and thus enhances reverse intersystem crossing (RISC) process from T_1 state to S_1 state[13], leading to delayed fluorescence. The additional singlet excitons production increases the internal quantum efficiency up to 100%, due to the complete utilization of both singlet and triplet excitons. Recently, a kind of high efficiency TADF emitters that are insensitive to the doping ratio have been reported[14-16], enabling high-efficiency OLEDs based on non-doped emitting layer (EML) structure. The non-doped EMLs provide an effective way to realize high device performance, and exhibit many merits of simple fabrication, high repeatability and low cost. Up to date, a few studies have been progressed on non-doped fluorescent or phosphorescent OLEDs, to develop high-efficiency monochromic or white OLEDs[5,17,18]. However, there is lack of work to fabricate hybrid white OLEDs by employing the second and third generation emitters with non-doped EML structure, which has great potential to realize high performance OLEDs, considering the advantages of the phosphorescent and TADF emitters.

Hence, in this work, we propose an easily realizable strategy to fabricate highly efficient white OLEDs. The hybrid white OLEDs based on non-doped EMLs have been designed, by employing a blue TADF emitter with concentration independent property and a yellow phosphorescent emitter with high efficiency feature, which has great potential to facilitate OLEDs' industry development.

2. Experimental

All the devices were fabricated on clean ITO-coated glass substrates, which were pre-treated by oxygen plasma to increase the work function of anode. Then, the organic and metallic layers were subsequently coated via thermal deposition under a pressure of 3×10^{-4} Pa and 3×10^{-3} Pa, respectively. The film thickness and deposition rates were monitored in situ by oscillating quartz thickness monitors. Hybrid white OLEDs were fabricated with a structure of: ITO/TAPC (40 nm)/mCP (10 nm)/DMAC-DPS (X)/(tbt)₂Ir(acac) (0.5 nm)/Bphen (40 nm)/Mg: Ag (100 nm), wherein X was 1 nm, 4 nm, 7 nm, 10 nm, 13 nm, respectively. Among of which, 1, 1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) and N,N'-dicarbazolyl-3,

5-benzene (mCP) were used as hole transporting layer (HTL) and exciton blocking layer (EBL), respectively, while 4, 7-diphenyl-1, 10-phenanthroline (Bphen) was used as the electron transporting layer (ETL), and Mg: Ag alloy was the cathode. A TADF material with 9, 9-dimethyl-9, 10-dihydroacridine (DMAC) donor and diphenylsulfone (DPS) acceptor functioned as the blue emitter, while bis[2-(4-tertbutylphenyl)benzothiazolato-N,C^{2'}] iridium (acetylacetonate), denoted as (tbt)₂Ir(acac), acted as the yellow phosphorescent emitter. Device structure and energy levels of the materials are shown as Fig. 1.

Luminance-current density-voltage (L - J - V) characteristics were measured with a Keithley 4200 source, and electroluminescence (EL) spectra and Commission Internationale de l'Eclairage (CIE) coordinates were recorded with an OPT-2000 spectrometer. External quantum efficiency (η_{EQE}) was calculated with a computer program. All the devices without capsulation were tested in air at room temperature. Transient photoluminescence (PL) decay characteristics were recorded on a HORIBA Scientific Single Photon Counting Controller FluoroHub-B, the samples were excited at 370 nm using a NanoLED-370 as the excitation light source, and the emitted photons were detected by a TBX detector connected to a TBX-PS power supply.

3. Results and discussion

3.1. Electrical characteristics of the devices

Fig. 1 shows the L - J - V characteristics of the devices. With increasing the thickness of DMAC-DPS from 1 nm, 4 nm, 7 nm, 10 nm to 13 nm, it can be seen from the J - V curves that, the current density is gradually increased. This tendency is different from typical J - V curves of the OLEDs, where the current density usually descends as the total thickness of the organic layers increases, due to low conductivity of conventional organic semiconductor materials. Meanwhile, it is learned from the L - V curves, the devices show low turn-on voltage (voltage at a luminance of 1 cd/m²), which is declined from 3.1 V to 2.5 V as the thickness of DMAC-DPS increases, corresponding to the increasing tendency of the J - V curves. The L - V curves also illustrate that the highest luminance is achieved to be 16,430 cd/m² at 14 V, 18,210 cd/m² at 14 V, 12,420 cd/m² at 13.5 V, 12,960 cd/m² at 13.4 V and 15,940 cd/m² at 13.5 V for the 1-nm, 4-nm, 7-nm, 10-nm and 13-nm DMAC-DPS devices, respectively. Reasonable explanations for the current density and luminance behaviors of the devices will be explained as follows.

As learned from the energy levels in Fig. 1, DMAC-DPS has a HOMO energy level of 5.9 eV along with a LUMO energy level of 2.9 eV[19], which is the same as that of mCP and Bphen, respectively. These matched energy levels can greatly promote charge carrier transport into the EMLs, while the holes transport through the TAPC HTL prior to transport across the mCP EBL, and the electrons transport across the Bphen ETL. Moreover, DMAC-DPS has bipolar charge transfer property due to the electron donor and acceptor groups in its chemical structure, which facilitates both holes and electrons to transport and recombine in the EMLs. Although the thickness of (tbt)₂Ir(acac) EML is less than 1 nm, leading to an un-continuous film, it has been proved that thus an ultrathin layer has an important effect on charge carrier trapping[17], which can also be induced from its higher HOMO level as shown in Fig. 1. This kind of charge carrier trapping effect plays an obvious influence on charge carrier transport when the thickness of DMAC-DPS is small, such as 1

nm and 4 nm. When the thickness of DMAC-DPS is gradually increased, more holes and electrons can be effectively transported into the DMAC-DPS EML, while the carrier trapping effect of (tbt)₂Ir(acac) is relatively weakened. Therefore, the current density is enhanced with increasing the thickness of DMAC-DPS, and the driving voltage is reduced at the same luminance.

Figs. 3(a), (b) and (c) depict the current efficiency-luminance, power efficiency-luminance and external quantum efficiency-luminance characteristics of the devices, respectively. It is observed that, as the thickness of DMAC-DPS increases, the device efficiency is firstly enhanced and then decreased, and the 4-nm DMAC-DPS device performs better than the other devices. A maximum current efficiency and power efficiency of 27.9 cd/A and 19.8 lm/W, 42.4 cd/A and 33.2 lm/W, 34.9 cd/A and 29.2 lm/W, 25.8 cd/A and 21.2 lm/W, 20.1 cd/A and 13.5 lm/W, is obtained in the 1-nm, 4-nm, 7-nm, 10-nm, 13-nm DMAC-DPS devices, respectively. At the same time, Fig. 3(c) states that the highest η_{EQE} is achieved to be 8.75%, 15.45%, 11.4%, 10.2%, 8.1%, respectively. The high luminance and efficiency also certify the great importance of the ultrathin (tbt)₂Ir(acac), attributing to a small degree of molecular aggregation and concentration quenching.

DMAC-DPS exhibits nearly concentration independent property and high PL quantum efficiency in the neat film, due to its methyl groups that are favorable to restrict the interactions between the DMAC units[15]. Moreover, DMAC-DPS has bipolar charge transfer property, a broad electro-hole recombination zone is formed at both the blue and yellow EMLs rather than the narrow EMLs/ETL interface. Considering that DMAC-DPS has a T_1 of 2.9 eV, TAPC with a T_1 of 2.87 eV is adopted as the HTL and mCP with a T_1 of 2.9 eV[20] is used as the EBL. Meanwhile, Bphen ETL exhibits a higher T_1 of 2.59 eV[21] than that of (tbt)₂Ir(acac) (2.2 eV)[17]. Thereby, the formed excitons can be well confined in the DMAC-DPS and (tbt)₂Ir(acac) EMLs, which is favorable for performance improvement. It can also be seen from Fig. 3, the devices demonstrate low efficiency roll-off, and the device efficiencies maintain high values at a luminance such as 1,000 cd/m², which is good for applications in displays and room lightings. The high performance of these non-doped OLEDs suggests that the non-doped EML architecture allows the recombination zone to extend to the EMLs, thus the concentration quenching between the DMAC-DPS molecules plays little role on the device efficiency.

3.2. Emission mechanism and energy transfer

Prior to the EL spectra, the emission mechanisms of DMAC-DPS TADF emitter and (tbt)₂Ir(acac) phosphorescent emitter are explained as illustrated in Fig. 4. It is reported that the phosphorescence spectra of DMAC-DPS at 77 K shows no vibronic structure feature[22], indicating a charge-transfer (CT) character induced by intramolecular transitions[23], and the CT excitons include singlets ¹CT, S₁(¹CT), and triplets ³CT, T₁(³CT). After excitons are formed on DMAC-DPS molecules, firstly, prompt fluorescent is generated by deactivation of the directly formed ¹CT state to the ground state (GS). Then, the energy up-conversion transition, ³CT→¹CT, occurs through RISC because of a small ¹CT-³CT split, and results in delayed fluorescent by relaxing the up-converted ¹CT excitons to the ground state. The energy difference between E₀₋₀ (³CT) and E₀₋₀ (¹CT) determines the reverse energy transfer rate, where the zero-zero energy (E₀₋₀) can be estimated from the onset of the emission band [20].

In case of phosphorescent (tbt)₂Ir(acac), the metal d-orbital has an important effect on the electronic transitions, and it shows remarkable metal d contribution in the lowest lying transition like other metal complexes[24]. The excited (tbt)₂Ir(acac) has a CT nature which is called metal to ligand charge transfer (MLCT)[25], consisting of singlet MLCT, S₁(¹MLCT), and triplet MLCT, T₁(³MLCT). Meanwhile, it exhibits significant ³MLCT and triplet ligand-centered (³LC) admixture[26]. The energy transition happens rapidly from the ¹MLCT state to the ³MLCT/³LC mixed states through spin-orbit coupling induced by the heavy metal effect[27]. The phosphorescence is subsequently generated by relaxation the excited triplet states to the ground state via intersystem conversion (ISC).

Since DMAC-DPS possesses much higher singlet and triplet energies than that of (tbt)₂Ir(acac), Förster and Dexter energy transfer occurs from the TADF DMAC-DPS to the phosphorescent (tbt)₂Ir(acac), involving singlet and triplet excitons[28,29]. To confirm the presence of energy transfer, the PL transient decay characteristic of DMAC-DPS: 0.5wt% (tbt)₂Ir(acac) doped film is investigated. Fig. 5 shows the PL transient decay curves of the emission at 460 nm and 560 nm in the DMAC-DPS: (tbt)₂Ir(acac) blend film excited at a wavelength of 370 nm, corresponding to DMAC-DPS and (tbt)₂Ir(acac) emission, respectively. By using a third-order exponential decay fitting, the transient decay lifetime of the delayed fluorescence is estimated to be 1.1 μs for DMAC-DPS and 2.2 μs for (tbt)₂Ir(acac). With compared to the delayed lifetime of 3.0 μs for DMAC-DPS neat film[15], the decay lifetime of DMAC-DPS in the blend film is greatly shortened. This suggests that effective energy transfer is produced between DMAC-DPS and (tbt)₂Ir(acac), as the energy transfer process accelerates the RISC process of TADF materials[30], bringing to the shortened lifetime of the delayed fluorescence in DMAC-DPS. The energy transfer can be further evidenced by moderately overlapped PL spectrum of DMAC-DPS[15] with the absorption spectrum of (tbt)₂Ir(acac)[17], showing three peaks at 409 nm, 451 nm and 480 nm resulted from ¹MLCT, ³MLCT and ³LC[31], respectively, as presented in the inset of Fig. 5. Moreover, while the RISC process of the TADF material could be accelerated by virtue of the energy transfer from the TADF material to the phosphorescent material, the ISC process of the TADF material could be simultaneously retarded[32]. Therefore, the triplet density on TADF DMAC-DPS molecules will be decreased, and the triplets induced concentration can be greatly suppressed, contributing to alleviated efficiency roll-off as mentioned above.

3.3. EL spectral behaviors of the devices

Fig. 6 illustrates the EL spectra of the devices at a voltage of 5 V. All the spectra show the same (tbt)₂Ir(acac) emission with a peak at 556 nm and a shoulder at 598 nm. However, the blue emission intensity of DMAC-DPS centering at 487 nm is different from each other. As the thickness of DMAC-DPS increases, the blue intensity is obviously strengthened. On one hand, charge carriers are directly recombined on DMAC-DPS and (tbt)₂Ir(acac) molecules, and produces self-emission process of both emitters, leading to the blue and yellow emission parts of the spectra. On the other hand, the energy transfer process between the blue and yellow emitters plays an important role on the spectral behavior of the devices. It is known that Förster energy transfer (FRET) is a long-range process, of which an energy transfer radii (R_0) can reach up to 10 nm[33], while Dexter energy transfer is a short-range process with a R_0 at about 1 nm[34]. Thus, it is reasonably assumed that the effect of Förster

energy transfer on light emission is varied by changing the DMAC-DPS thickness from 1 nm to 13 nm, while the Dexter energy transfer, located at the blue-/yellow-EML interface, exists irrespectively of the DMAC-DPS thickness and shows similarly fair effect in all the devices. In the 1-nm and 4-nm DMAC-DPS devices, strong FRET occurs between DMAC-DPS and (tbt)₂Ir(acac), bringing to primary yellow emission. By keeping increase the thickness of DMAC-DPS to 7 nm, 10 nm and 13 nm, more excitons are formed on DMAC-DPS molecules, and the singlet excitons, generated either directly or through RISC, contribute to enhanced blue emission.

Figs. 7(a), (b) and (c) demonstrate the EL spectra of the 7-nm, 10-nm and 13-nm DMAC-DPS devices, respectively, as a function of applied voltages. In contrast, the 7-nm and 10-nm DMAC-DPS devices display relatively stable spectra variation, and the 13-nm DMAC-DPS device exhibits large spectra variation. By increasing the applied voltage from 4 V to 9 V, as learned from Figs. 7(a) and (b) the blue intensity is slightly decreased, and the CIE coordinates of the devices are ranged from (0.40, 0.46) to (0.40, 0.45) and (0.35, 0.43) to (0.37, 0.44), respectively, showing warm white emission. Compared to the 7-nm DMAC-DPS device, the 10-nm DMAC-DPS device shows higher blue intensity, due to more excited DMAC-DPS molecules generated in the thicker DMAC-DPS EML. As observed from Fig. 7(c), the device shows white emission arising from comparable blue and yellow emission intensity at low voltages, whereas the blue intensity is gradually reduced with increased voltages. That is to say, the yellow intensity is relatively enhanced, indicating that the recombination zone is shifted toward the yellow EML close to the cathode, since more charge carriers could effectively transfer across the DMAC-DPS EML under high electric fields, which is inferred from the *L-J-V* characteristics discussed above. As for the devices with 1-nm and 4-nm DMAC-DPS, they both show similarly strong yellow emission and weak blue emission, as well as stable EL spectra with varied voltages (not shown here). The reasons for the different spectral profiles of the devices will be given in the following.

By applying external voltages, holes and electrons in the non-doped OLEDs will be directly trapped by DMAC-DPS and (tbt)₂Ir(acac) molecules. Then, the recombination zone is extended to both blue and yellow EMLs, along with well confined excitons in the EMLs. When the thickness of DMAC-DPS is shorter (such as 1 nm and 4 nm) than the FRET R_0 , the energy transfer from DMAC-DPS to (tbt)₂Ir(acac) occurs efficiently; meanwhile, the number of excited DMAC-DPS molecules is low, and the self-emission from DMAC-DPS is inapparent, leading to dominated yellow emission. When the thickness of DMAC-DPS increases to 7 nm and 10 nm, which is comparable to the FRET R_0 , as displayed in Fig. 8(a), we reckon that the self-emission (process I) of DMAC-DPS and (tbt)₂Ir(acac), and the Förster energy transfer (process II) and Dexter energy transfer (process III) between DMAC-DPS and (tbt)₂Ir(acac) are significantly balanced, resulting in improved color stability. Furthermore, as the thickness of DMAC-DPS reaches 13 nm, which beyond the FRET R_0 , the low energy excited (tbt)₂Ir(acac) sites are saturated, either from direct charge trapping or energy transfer (processes II and III). Meanwhile, more excitons are formed on DMAC-DPS molecules, as depicted in Fig. 8(b), and the self-emission of DMAC-DPS (process I) is improved, contributing to the heightened blue intensity. Upon changing the voltage, the spectra variation can be explained by a field-dependent competition between direct charge trapping on the emitters and charge carrier transport across the EMLs. In the 13-nm DMAC-DPS device,

more charge carriers effectively transfer across the DMAC-DPS EML at high voltages, and the recombination zone shifts toward the yellow EML, which is not favorable for direct exciton formation on DMAC-DPS molecules, resulting in decreased blue emission intensity.

4. Conclusion

In this work, we have successfully constructed hybrid white OLEDs by employing a non-doped blue TADF emitter DMAC-DPS and an ultrathin yellow phosphorescent emitter (tbt)₂Ir(acac). The concentration independent property of DMAC-DPS enables it to be used as a neat emitting film, while its bipolar transport property allows a broad recombination zone to be located in the EMLs. Through thickness optimization of DMAC-DPS, balanced self-emission and energy transfer processes between the blue and yellow emitters is achieved, leading to improved color stability. The results indicate that the TADF DMAC-DPS could be effectively utilized to replace the conventional blue phosphorescent emitters which suffer from short lifetime and low stability. With our proposed strategy, it is feasible to simplify the fabrication process and reduce the production cost, and the realized high performance suggests the non-doped OLEDs have great potential to become the dominate type of OLEDs.

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Fig. 1 Device structure and energy levels of the materials

Fig. 2 Luminance-current density-voltage characteristics

Fig. 3(a) Current efficiency, (b) power efficiency, and (c) external quantum efficiency-luminance characteristics

Fig. 4 Emission mechanisms of DMAC-DPS and (tbt)₂Ir(acac)

Fig. 5 PL transient decay curves of the emission at 460 nm and 560 nm. Inset is the PL spectrum of DMAC-DPS and the absorption spectrum of (tbt)₂Ir(acac)

Fig. 6 EL spectra of the devices at a voltage of 5 V

Fig. 7 EL spectra of the device with (a) 7 nm, (b) 10 nm, and (c) 13 nm DMAC-DPS

Fig. 8 Emission processes in the devices with (a) 7 nm and 10 nm, and (b) 13 nm DMAC-DPS