

## Letter

## Efficient simplified orange and white phosphorescent organic light-emitting devices with reduced efficiency roll-off



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## ABSTRACT

Efficient orange phosphorescent organic light-emitting devices based on simplified structure with maximum efficiencies of 46.5 lm/W and 51.5 cd/A were reported. One device had extremely low efficiency roll-off with efficiencies of 50.6 cd/A, 45.0 cd/A and 39.2 cd/A at 1000 cd/m<sup>2</sup>, 5000 cd/m<sup>2</sup> and 10,000 cd/m<sup>2</sup> respectively. The reduced efficiency roll-off was attributed to more balanced carrier injection and broader recombination zone. The designed simplified white device showed much lower efficiency roll-off than the control one based on multiple emitting layers. The efficiency of simplified white device was 40.8 cd/A at 1000 cd/m<sup>2</sup> with Commission Internationale de l'Eclairage coordinates of (0.39, 0.46).

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## 1. Introduction

In the past two decades, performances of organic light-emitting devices (OLEDs) have been improved dramatically by developing new efficient materials and a variety of device architectures [1–5]. Phosphorescent OLEDs are more effective because phosphorescent materials can harvest both singlet and triplet excitons which leads to the potential to achieve 100% internal emission efficiency in principle [6–8]. Due to the strong exciton self-quenching in a neat film, the phosphorescent dyes are usually doped into appropriate host materials, that is to say, the emissive layer should adopt a host–guest doped system. In order to realize efficient exothermic energy transfer, the host material with wide band-gap should have higher triplet energy than phosphorescent dyes. Moreover, to suppress the undesired energy transfer from higher triplet excitons generated in emission layer to the lower nonradiative triplet state of hole/electron transporting layers (HTL/ETL), exciton blocking layer should be introduced between the emission layer and HTL/ETL.

In order to realize effective light emission, OLEDs with various configurations have been proposed, such as using a mixed host system [9] or multiple emitting layers [10], using vertically stacked multiple emissive units connected by charge generating layer

[11], using pin structures to obtain lower turn-on voltage [12], using the structures of double exciton blocking layer [13], stepwise multi-emissive layers [14] and balanced carrier injection [15] et al.

Though the above multiple structures could enhance efficiency or lifetime, such complicated architectures inevitably increase the manufacturing complexity cost and hamper their commercialization application in field of display or lighting. Therefore, it's necessary and important to simplify the OLEDs structure for the potential commercial use [16–20]. Recently, Wang et al. demonstrated trilayer green phosphorescent device with efficiency of 78.0 lm/W at 100 cd/m<sup>2</sup> [19]. Suh et al. reported efficient bi-layer green phosphorescent device with current efficiency of 33.6 cd/A by using 1,1-bis[(di-4-tolylami-no)phenyl]cyclohexane and 1,3,5-tri(p-pyrid-3-yl-phenyl) benzene as mixed host system [20]. But for phosphorescent devices with simplified structures, triplet–triplet annihilation and triplet–polaron quenching at high luminance are avoidless and much more severe than the devices with multiple structures [21,22], which leads to efficiency decreases rapidly and restrains phosphorescent OLEDs of their use in solid lighting application.

Here we reported efficient simplified orange OLEDs with reduced efficiency roll-off by introducing iridium (III) bis(4,6-difluorophenyl-pyridinato-N,C<sup>2'</sup>) (picolate) (Flrpic). The maximum efficiencies of 46.5 lm/W and 51.5 cd/A were achieved for the device with Flrpic doping concentration of 15%. The designed white device showed much lower efficiency roll-off with high efficiency of 40.8 cd/A at 1000 cd/m<sup>2</sup>.

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## 2. Experiment

Fig. 1 shows the device structure and the proposed energy diagram of the simplified OLEDs. 4,4',4''-tris(N-carbazolyl)-triphenylamine (TCTA) with good hole transporting characteristics (hole mobility  $\sim 10^{-4}$  cm<sup>2</sup>/Vs) was used as HTL. Moreover, TCTA was also used as the host for the phosphorescent dyes to further simplified the device structure. tris[3-(3-pyridyl)-mesityl]borane (3TPYMB) with electron mobility of  $10^{-5}$  cm<sup>2</sup>/Vs was used as ETL. iridium(III) bis(4-phenylthieno-[3,2-c]pyridinato-N,C<sup>2'</sup>) acetylacetonate (PO-01) doped into TCTA was used as orange emission layer. 100 nm Mg: Ag (10:1 by weight) was used as the cathode. All organic materials and the cathode were grown in succession by thermal evaporation without breaking vacuum ( $\sim 3 \times 10^{-4}$  Pa). The layer thickness and the deposition rate of materials were monitored in situ by an oscillating quartz thickness monitor. The electroluminescent (EL) spectra and Commission International de L'Eclairage (CIE) coordinates of the devices were measured by using a PR655 spectroscan spectrometer. The luminance–voltage and current–voltage characteristics were measured simultaneously with the measurement of the EL spectra by combining the spectrometer with a programmable Keithley 2400 source. All the devices were characterized without encapsulation and the measurements were carried out at room temperature under ambient conditions.

## 3. Results and discussions

Firstly, the orange devices with the structure of ITO/MoO<sub>3</sub> (2 nm)/TCTA:X% Flrpic:5% PO-01 (70 nm)/3TPYMB (30 nm)/Mg:Ag were fabricated. And the Flrpic doping concentration X was varied, being 0, 3, 5, 10, 15, 20 for devices A, B, C, D, E, F, respectively.

The current efficiency–luminance and power efficiency–voltage characteristics of the orange devices are shown in Fig. 2(a) and (b). In general, all the devices based on Flrpic co-doped with PO-01 into TCTA have higher efficiency than device A. Table 1 shows the performance characteristics of devices A–F. Device E with Flrpic doping concentration of 15% has the maximum power efficiency of 46.5 lm/W and maximum external quantum efficiency (EQE) of 15.6%. Note that the maximum power efficiency of device E is improved by more than 90% compared to device A, which is owing

to the lower turn-on voltage of device E (0.59 V lower than device A). The devices B–F with Flrpic co-doped show lower efficiency roll-off. For example, device E has efficiencies of 50.6 cd/A, 47.7 cd/A, 45.0 cd/A and 39.2 cd/A at 1000 cd/m<sup>2</sup>, 3000 cd/m<sup>2</sup>, 5000 cd/m<sup>2</sup> and 10,000 cd/m<sup>2</sup> respectively, corresponding to low efficiency roll-off of 1.7%, 7.4%, 12.6% and 23.4%. On the contrary, device A has much higher efficiency roll-off, such as 27.8% and 74.3% decreases in efficiency at 5000 cd/m<sup>2</sup> and 10,000 cd/m<sup>2</sup>, respectively.

The emission mechanism of the orange devices A–F is shown in Fig. 3. For device A without Flrpic doping, as can be seen from Fig. 1, the interfacial energy barriers at the interface of TCTA/3TPYMB for electrons injection are as large as 0.6 eV (PO-01) and 0.9 eV (TCTA), resulting in electrons accumulated at the interface and higher turn-on voltage [23]. We think direct excitons formation on PO-01 is negligible in device A due to the lack of electrons in emission layer. So most excitons are formed at the narrow zone near the interface and then are transferred to PO-01. According to the results of Erickson et al. [24], for device A with narrow recombination zone, the efficiency roll-off is dominated by triplet–triplet annihilation which was proportional to the square of triplet exciton density [21]. For device A, the triplet exciton density becomes higher with increasing luminance which leads to severe efficiency roll-off.

When Flrpic is co-doped with PO-01 into TCTA host for devices B–F, the electrons are first injected into Flrpic due to the much smaller offset of lowest unoccupied molecular orbital (LUMO) energy level (0.2 eV) between Flrpic and 3TPYMB, which makes excitons recombination zone expanding toward the anode side. Considering TCTA host with good hole-transporting property efficiently blocks electrons, the introduction of Flrpic into TCTA host will balance against the major carriers (the holes) in emission layer. Moreover, the main purpose of fixing Flrpic doping range in the entire TCTA film (70 nm) is to further decrease the hole current density due to the trapping effect of Flrpic which could be confirmed from the energy diagram in Fig. 1 [25]. Meanwhile, the trapped holes by Flrpic could also facilitate electron injection into emission layer [26,27]. So more balanced carrier injection into emission zone and broader recombination zone could be obtained, which contributes to high efficiency and low efficiency roll-off. Fig. 4(a) shows the current density–voltage characteristics of

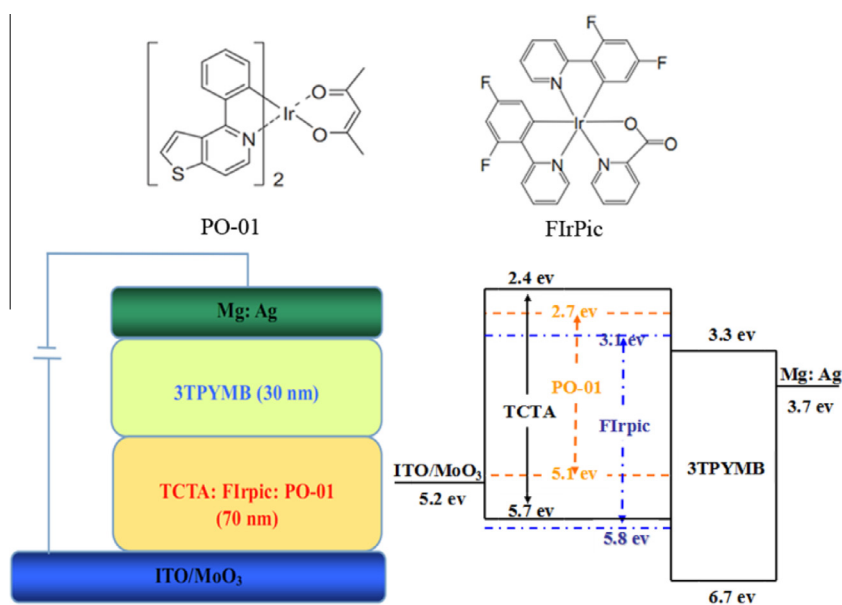


Fig. 1. The device structure and the proposed energy diagram of OLEDs.

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