



# Synthesis and characterization of highly efficient solution-processable orange Ir(III) complexes for phosphorescent OLED applications

Zhiming Zhong<sup>a,b</sup>, Xue Wang<sup>b,c</sup>, Yawei Ma<sup>b</sup>, Feng Peng<sup>a,b</sup>, Ting Guo<sup>b</sup>, Jia-Xing Jiang<sup>c</sup>,  
Lei Ying<sup>a,b,\*</sup>, Jian Wang<sup>b,\*\*</sup>, Junbiao Peng<sup>b</sup>, Yong Cao<sup>b</sup>

<sup>a</sup> South China Institute of Collaborative Innovation, Dongguan 523808, China

<sup>b</sup> Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, China

<sup>c</sup> Key Laboratory for Macromolecular Science of Shaanxi Province, School of Materials Science and Engineering, Shaanxi Normal University, Xi'an, Shaanxi 710062, PR China

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## ABSTRACT

Two efficient solution-processable orange Ir(III) complexes were developed based on the classical orange phosphorescent dye Ir(phq)<sub>3</sub>. The indenocarbazole and bipolar triazine-indenocarbazole moieties introduced into IrO1 and IrO2, respectively, afforded good solubility (> 10 mg mL<sup>-1</sup>) in chlorobenzene, making these emitters suitable for the fabrication of solution-processed devices. The obtained devices based on IrO1 with 4,4'-bis(carbazol-9-yl)biphenyl as the host material displayed a maximum luminance efficiency of 45.8 cd/A with CIE coordinates of (0.55, 0.45). The devices based on IrO2 exhibited a redshift of the spectrum relative to Ir(phq)<sub>3</sub> and displayed a maximum luminous efficiency of 27.0 cd/A with CIE coordinates of (0.59, 0.41). To the best of our knowledge, these are the highest performances reported to date for solution-processed Ir(phq)<sub>3</sub>-based light-emitting devices. These excellent performances demonstrate that IrO1 and IrO2 are promising candidates for high-performance orange phosphorescent organic light-emitting diodes.

## 1. Introduction

Organic light-emitting diodes (OLEDs) have become the most promising candidates for next-generation solid-state lighting sources [1–7]. The key to high-performance OLEDs is to recycle the nonradiative triplet excitons. Various advanced molecular systems have been developed to utilize both singlet and triplet excitons, such as fluorescence with triplet–triplet annihilation (TTA) [8–12], thermally activated delayed fluorescence (TADF) [13–16], phosphorescent emitters [17–19], and so forth [20,21]. Compared with the other approaches, applying phosphorescent emitters is the most straightforward and mature method [22,23]. Phosphorescent emitters have been widely applied in monochromatic (from near-infrared (NIR) to deep blue) and white OLEDs (WOLEDs) and can be used to easily achieve external quantum efficiencies (EQEs) exceeding 20% by vacuum evaporation processes [24]. Among these dyes, orange phosphorescent emitters are important building blocks for the fabrication of simplified dual-component WOLEDs with high luminance efficiencies and color rendering index (CRI) [25–28]. Compared with yellow emitters, as the complementary color of blue (especially the traditional sky-blue emitting dye, FIrpic), orange

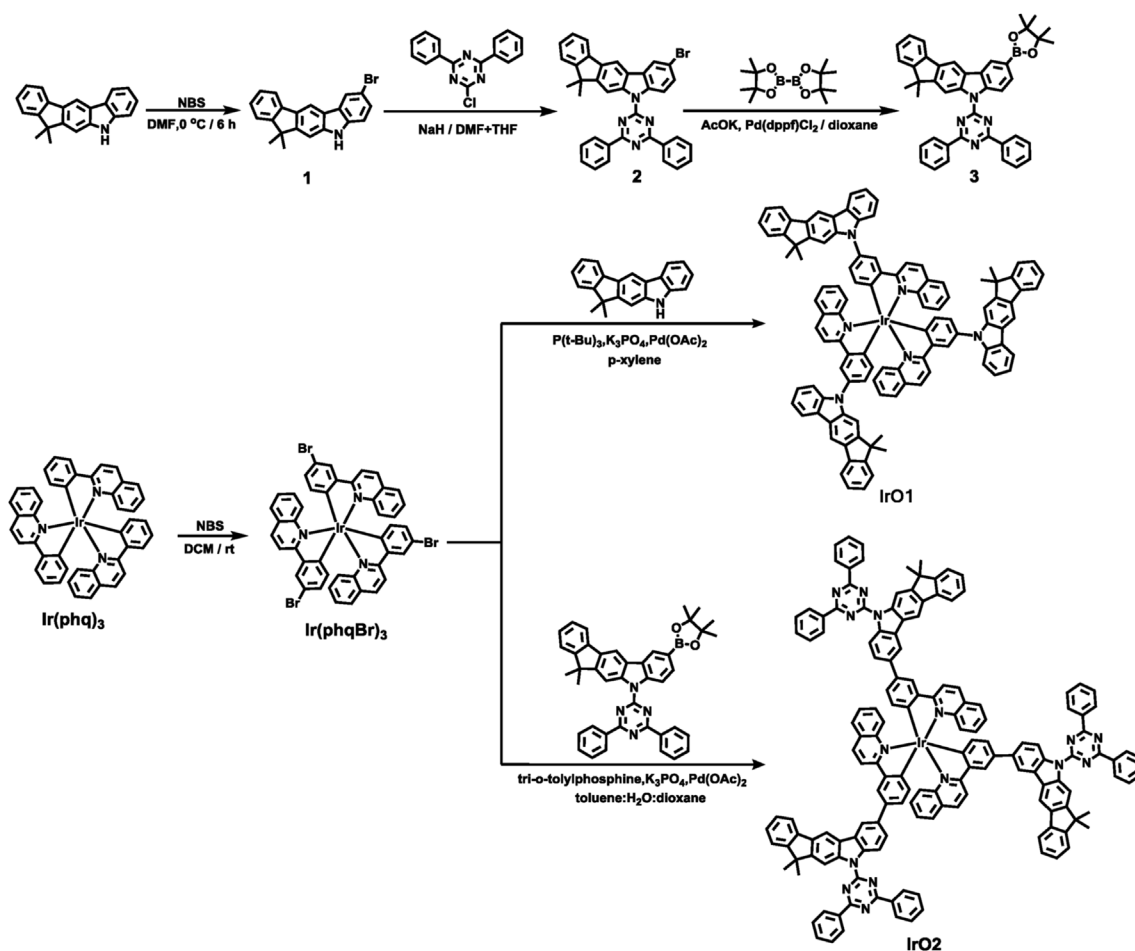
emitters possess an advantage in terms of CRI, because they exhibit a more intense long-wavelength emission and can thus perform better than yellow phosphorescent materials in WOLEDs [1,3,29]. Compared with red emitters, orange emitters generally display a greater efficiency. On the one hand, the narrower energy gaps of red dopants usually introduce severe charge-trapping effects in the OLED devices, leading to a higher working voltage and lower power efficiency [30,31]. On the other hand, the spectra of red emitters typically extend to the NIR region, resulting in wasted energy, especially in the case of donor–acceptor (D–A)-type red-light-emitting molecules with typically broad spectra [32].

Over recent years, many highly efficient orange OLEDs have been reported. However, most of these were evaporation processed under high vacuum rather than solution processed. To develop highly efficient orange OLEDs based on the low-cost solution-process technique, Wong et al. developed a set of Ir(III) complexes with 2-(9,9-diethylfluorene-2-yl)pyridine main ligand and various ancillary ligands, exhibiting a peak luminance efficiency of 62.1 cd/A with 1931 Commission Internationale de L'Eclairage (CIE) coordinates of (0.56, 0.43) when doped into the well designed exciplex-host-based device structure

\* Corresponding author. South China Institute of Collaborative Innovation, Dongguan 523808, China.

\*\* Corresponding author.

E-mail addresses: [msleiyang@scut.edu.cn](mailto:msleiyang@scut.edu.cn) (L. Ying), [jianwang@scut.edu.cn](mailto:jianwang@scut.edu.cn) (J. Wang).



Scheme 1. Synthesis of IrO1 and IrO2.

[33,34]. Liu et al. synthesized the bis[2-di(*p*-methoxyphenyl)-amino (9,9-diethylfluoren-2-yl)-5-(trifluoromethyl)pyridine][acetylacetonate] iridium(III) and also realized highly efficient solution-processed orange-red OLEDs with a maximum luminance efficiency of 26.3 cd/A and 1931 CIE coordinates of (0.60, 0.39) with the same device structure [35]. Nonetheless, the development of novel solution-processable orange phosphorescent emitters for highly efficient OLED devices remains a great challenge.

Herein, we report the fabrication of highly efficient solution-processable orange OLEDs using two newly synthesized Ir(pq)<sub>3</sub>-based emitters as phosphorescent dopants (Scheme 1). Ir(pq)<sub>3</sub> was selected as the core molecule owing to its short triplet-state lifetime, which was expected to contribute to a high phosphorescence yield and thus permit the realization of high-performance phosphorescent OLEDs (PhOLEDs). By introducing indenocarbazole (IrO1) or triazine-indenocarbazole moieties (IrO2) into the 3-position of the phenyl ring of the 2-phenylquinoline ligand, we expected to be able to tune the emission wavelength together with the solubility. The attachment of an inert *n*-hexyl group to the phenyl moiety of the 2-phenylquinoline ligand was previously reported to improve the film morphology and boost the performance from 2.7 cd/A to 17.0 cd/A after solution processing [36]. Solution-processed OLEDs based on the new iridium complex IrO1 exhibited bright yellow-orange emission with a maximum luminance efficiency of 45.8 cd/A and CIE coordinates of (0.55, 0.45). Furthermore, the attachment of the bipolar triazine-indenocarbazole moiety to the 2-phenylquinoline ligand led to a redshift of the spectrum of the resultant iridium complex IrO2 and a maximum luminance efficiency of 27.0 cd/A and CIE coordinates of (0.59, 0.41) for the OLEDs. These luminous efficiencies are the highest reported to date for solution-

processed Ir(pq)<sub>3</sub>-based OLEDs.

## 2. Results and discussion

### 2.1. Synthesis and characterization

The target emitters IrO1 and IrO2 were obtained by the synthetic routes outlined in Scheme 1. After bromination of the red phosphorescent Ir(pq)<sub>3</sub> core, the obtained Ir(pqBr)<sub>3</sub> was reacted with either 1,3-dihydro-3,3-dimethylindeno[2,1-*b*]carbazole through a C–N coupling reaction or 5-(4,6-diphenyl-1,3,5-triazin-2-yl)-7,7-dimethyl-2-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-5,7-dihydroindeno[2,1-*b*]carbazole (3) through a Suzuki coupling reaction to afford IrO1 and IrO2, respectively. The detailed synthetic procedures and characterization of the two emitters and their intermediates are provided in the Experimental section. As summarized in Table 1, IrO1 and IrO2 were found to exhibit decomposition temperatures (*T*<sub>d</sub>, defined as the temperature at which 5 wt% loss is reached) of 449 and 529 °C, respectively, as determined by thermogravimetric analysis. Neither of the emitters exhibited an obvious glass-transition temperature (*T*<sub>g</sub>) upon analysis by differential scanning calorimetry, suggesting that both of the compounds possess an amorphous morphology in the solid state.

### 2.2. Photophysical properties

Fig. 1a shows the absorption and photoluminescence (PL) spectra of IrO1 and IrO2 dissolved in *p*-xylene solution at room temperature. The absorption spectra can be divided into three parts. The strong absorption bands below 380 nm closely correspond to the spin-allowed ligand-

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